

FEB 17 2000
3

ENGINEERING DATA TRANSMITTAL

S

1. EDT

628251

2. To: (Receiving Organization) Distribution	3. From: (Originating Organization) Process Control	4. Related EDT No.: N/A
5. Proj./Prog./Dept./Div.: Criticality Safety Evaluation/Technical Operations/PC/Process Engineering	6. Design Authority/ Design Agent/Cog. Engr.: C. A. Rogers	7. Purchase Order No.: N/A
8. Originator Remarks: This document is being released into the supporting document system for retrievability purposes.	9. Equip./Component No.: N/A	
	10. System/Bldg./Facility: N/A	
11. Receiver Remarks: For release.	11A. Design Baseline Document? <input type="checkbox"/> Yes <input checked="" type="checkbox"/> No	
	12. Major Assm. Dwg. No.: N/A	
	13. Permit/Permit Application No.: N/A	
14. Required Response Date: 02/15/00		

15. DATA TRANSMITTED					(F)	(G)	(H)	(I)
(A) Item No.	(B) Document/Drawing No.	(C) Sheet No.	(D) Rev. No.	(E) Title or Description of Data Transmitted	Approval Desig- nator	Reason for Trans- mittal	Ori- gini- ator Dispo- sition	Receiv- er Dispo- sition
1	RPP-5296	N/A	0	Criticality Safety Evaluation of Hanford Site High-Level Waste Storage Tanks	S	2	1	1

16. KEY					
Approval Designator (F)		Reason for Transmittal (G)		Disposition (H) & (I)	
E, S, Q, D or N/A (see WHC-CM-3-5, Sec.12.7)		1. Approval	4. Review	1. Approved	
		2. Release	5. Post-Review	2. Approved w/comment	
		3. Information	6. Dist. (Receipt Acknow. Required)	3. Disapproved w/comment	
				4. Reviewed no/comment	
				5. Reviewed w/comment	
				6. Receipt acknowledged	

17. SIGNATURE/DISTRIBUTION (See Approval Designator for required signatures)											
(G) Rea- son	(H) Disp.	(J) Name	(K) Signature	(L) Date	(M) MSIN	(G) Rea- son	(H) Disp.	(J) Name	(K) Signature	(L) Date	(M) MSIN
		Design Authority									
		Design Agent									
2	1	Cog.Eng. C.A. Rogers	<i>C.A. Rogers</i>	2/15/00	R2-11	1	1	C. DeFigh-Price	<i>C. DeFigh-Price</i>	2-16-00	R2-12
2	1	Cog. Mgr. N.W. Kirch	<i>N.W. Kirch</i>	2/15/00	R2-11	1	1	C.S. Eberle	<i>C.S. Eberle</i>	2/15/00	8444
		QA				1	2	K.D. Fowler	<i>K.D. Fowler</i>	2/15/00	R2-11
2	1	Safety L.E. Thomas	<i>L.E. Thomas</i>	2/15/00	SP1						
		Env.									

18. A.E. Young <i>A.E. Young</i> Signature of EDT Originator	19. N/A Authorized Representative Date for Receiving Organization	20. N.W. Kirch <i>N.W. Kirch</i> Design Authority/ Cognizant Manager	21. DOE APPROVAL (if required) Ctrl. No. <input type="checkbox"/> Approved <input type="checkbox"/> Approved w/comments <input type="checkbox"/> Disapproved w/comments
--	---	--	---

Criticality Safety Evaluation of Hanford Site High-Level Waste Storage Tanks

C. A. Rogers

CH2M HILL Hanford Group, Inc., Richland, WA 99352
U.S. Department of Energy Contract DE-AC06-96RL13200

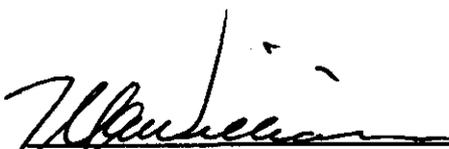
EDT/ECN: EDT-628251 UC: 2070
Org Code: 74B50 CACN/COA: 101965/BB30
B&R Code: EW 3120074 Total Pages: 224

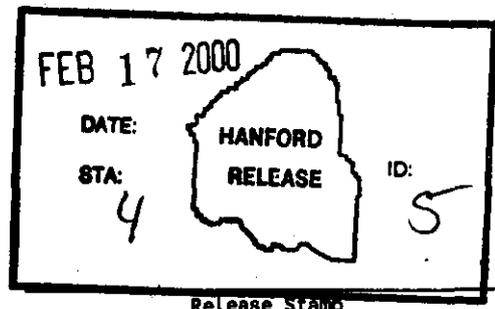
Key Words: Criticality Safety Evaluation, CSR, High-Level Waste, Storage Tanks, Tank Farms, Tank Waste, Plutonium, Double-Shell Tanks, DST, Single-Shell Tanks, SST, DCRT

Abstract: This criticality safety evaluation covers operations for waste in underground storage tanks at the high-level waste tank farms on the Hanford site. This evaluation provides the bases for criticality safety limits and controls to govern receipt, transfer, and long-term storage of tank waste. Justification is provided that a nuclear criticality accident cannot occur for tank farms operations, based on current fissile material and operating conditions.

TRADEMARK DISCLAIMER. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof or its contractors or subcontractors.

Printed in the United States of America. To obtain copies of this document, contact: Document Control Services, P.O. Box 950, Mailstop H6-08, Richland WA 99352, Phone (509) 372-2420; Fax (509) 376-4989.


Release Approval
Date: 2/17/00



Approved for Public Release

**CRITICALITY SAFETY EVALUATION OF HANFORD SITE
HIGH-LEVEL WASTE STORAGE TANKS**

C. A. Rogers
CH2M HILL Hanford Group, Inc.

Date Published
February 2000

Prepared for the U. S. Department of Energy
Office of River Protection

Approved for public release; distribution is unlimited

APPROVALS

Title: **Criticality Safety Evaluation of Hanford Site High-Level Waste Storage Tanks**

Prepared by: C. A. Rogers Feb. 15, 2000
C. A. Rogers
Criticality Safety Specialist
Process Engineering
Date

Reviewed by: C. S. Eberle Feb. 15, 2000
C. S. Eberle
Criticality Safety Specialist
Criticality and Shielding, FFS
Date

Approved by: John DeFigh-Price 2/16/00
John DeFigh-Price
Manager, Process Engineering
Date

Approved by: K. D. Fowler 2/15/00
K. D. Fowler
RPP Criticality Safety Representative
Process Engineering
Date

Approved by: L. E. Thomas 2/15/00
L. E. Thomas
Facility Safety Representative
Nuclear Regulatory Compliance Support
Date

This page intentionally left blank.

EXECUTIVE SUMMARY

A criticality safety evaluation is provided for waste stored in underground tanks at the high-level waste tank farms on the Hanford Site. This evaluation provides the bases for limits and controls to govern receipt, transfer, and storage that will ensure continued long-term criticality safety of tank waste. Justification is provided that a nuclear criticality cannot occur over the range of credible tank farm operations. New operations, such as waste retrieval and treatment, must be reviewed before initiation to determine if they are covered by this evaluation.

Criticality safety depends upon demonstrating inherent characteristics of waste that ensure subcriticality without intervention. The focus of the criticality safety program is to ensure that waste enters the tank farms with subcritical concentrations of fissile material and that tank farm activities do not adversely change the margin of subcriticality. An important characteristic that ensures subcriticality is a large mass ratio of neutron absorbing materials to plutonium. For each neutron absorber, there is a mass ratio with respect to plutonium that will ensure subcriticality for an unlimited volume of waste. For example, subcriticality is ensured when the iron/plutonium mass ratio exceeds 160, regardless of the degree of moderation or reflection, or when the overall solids/plutonium mass ratio exceeds 770 in homogeneous waste, regardless of the actual solids composition. The subcritical mass fraction, defined as the ratio of the actual absorber/plutonium mass ratio to the subcritical limit mass ratio, is used as a measure of subcriticality. Subcriticality is assured when the subcritical mass fraction exceeds unity.

The contents of both single-shell storage tanks (SSTs) and double-shell storage tanks (DSTs) have subcritical mass fractions that are at least an order of magnitude larger than required to ensure subcriticality. The tank-averaged solids/plutonium mass ratio for DST 241-SY-102 exceeds 10,000, and this mass ratio is the smallest for any storage tank.

The potential for increasing plutonium concentration through settling of suspended particulate, gravity segregation, chemical separation, selective precipitation, and evaporation of liquid was evaluated. Mechanisms capable of concentrating the plutonium are counteracted by the large solids/plutonium mass ratio and by the small particle size for waste solids.

Whyatt et al. (1996) concludes that under normal tank condition, plutonium forms tightly bound agglomerates with other waste solids, and these agglomerates prevent plutonium from increasing its concentration. Whyatt et al. (1996), Serne et al. (1996), and Bratzel et al. (1996) were not able to identify any mechanism in the tank waste environment capable of eroding the margin of subcriticality.

When the plutonium concentration is less than 2.6 g/L throughout the waste, subcriticality is assured, regardless of composition, moderation, or reflection. The median plutonium concentration for 200 analyzed samples of waste solids is about 0.01 g/L (0.04 g/gal), a value 260 times smaller than the minimum critical concentration (Braun et al. 1994). The plutonium saturation concentration in the supernatant liquid is less than this value. The highest reported concentration (this occurs in waste solids) is 0.89 g Pu/L, a value 2.9 times smaller than the minimum critical concentration (Lipke 1999).

A program of waste characterization and surveillance is conducted to increase knowledge of waste and to verify compliance to discharge criteria.

CONTENTS

EXECUTIVE SUMMARY	iii
1.0 INTRODUCTION.....	1-1
2.0 DESCRIPTION OF TANK FARM FACILITIES.....	2-1
2.1 ENTRY POINTS INTO TANK STORAGE SYSTEM.....	2-1
2.2 LONG-TERM STORAGE TANKS	2-2
2.2.1 Double-Shell Storage Tanks	2-3
2.2.2 Single-Shell Storage Tanks	2-3
2.2.3 Leakage Accumulation Locations.....	2-4
2.3 SHORT-TERM HOLDING TANKS.....	2-4
2.3.1 Double-Contained Receiver Tanks	2-4
2.3.2 Vault Tanks.....	2-5
2.3.3 Catch Tanks	2-6
2.4 242-A EVAPORATOR FACILITY	2-6
2.5 MISCELLANEOUS FACILITIES	2-7
2.5.1 204-AR Waste Unloading Facility.....	2-7
2.5.2 209-E Complex.....	2-8
2.5.3 Vertical Storage Units	2-9
2.5.4 213-W Dry Waste Compactor Facility	2-9
2.5.5 2727-WA Sodium Storage Facilities	2-9
2.5.6 Grout Treatment Facility	2-9
2.5.7 Cribs, Ponds, and Ditches	2-9
2.5.8 Ventilation Systems.....	2-10
2.6 WASTE TRANSFER SYSTEMS	2-10
2.6.1 Cross Site Transfer Lines	2-10
2.6.2 Interconnecting Lines	2-10
2.6.3 241-AX-152 Diverter Station.....	2-11
2.7 INACTIVE MISCELLANEOUS UNDERGROUND STORAGE TANKS	2-11
3.0 REQUIREMENTS DOCUMENTATION.....	3-1
3.1 REQUIREMENTS DOCUMENTS	3-1
3.2 TECHNICAL SAFETY REQUIREMENT	3-1
3.2.1 Applicability	3-3
3.3 DOUBLE CONTINGENCY PRINCIPLE	3-4
3.4 CRITICALITY SAFETY CRITERIA.....	3-4
3.4.1 Discharged Waste.....	3-4
3.4.2 Evaporators.....	3-5
3.4.3 Transfers Between Tanks	3-5
3.5 MARGIN OF SAFETY.....	3-5
3.5.1 Discussion.....	3-6
4.0 METHODOLOGY.....	4-1

5.0 DISCUSSION OF CONTINGENCIES	5-1
5.1 CONTROL FAILURES	5-3
5.1.1 Holdup Tanks	5-3
5.1.2 Breakdown of Controls	5-3
5.1.3 Special Case.....	5-4
5.1.4 Hypothetical Accident.....	5-4
5.1.5 Conclusion.....	5-5
5.2 TRANSFERS FROM PLUTONIUM FINISHING PLANT (PFP).....	5-5
5.2.1 Individual Control Failures	5-6
5.2.2 Control Failures Required For Criticality	5-8
5.3 PLUTONIUM DISTRIBUTION	5-9
5.3.1 Layers Having Different Compositions	5-10
5.3.2 Mixing.....	5-11
5.4 LEAKAGE OF SUPERNATE INTO GROUND.....	5-11
5.4.1 Comparison to Saltwell Pumping	5-11
5.4.2 Critical Plutonium Concentration in Hanford Soil	5-12
5.4.3 Conservative Model of Soil	5-12
5.4.4 Minimum Critical Plutonium Mass in Soil.....	5-13
5.4.5 Critical Slab Thickness.....	5-13
5.4.6 Conclusion.....	5-13
5.5 WASTE GENERATORS CRITICALITY SAFETY AUDIT PROGRAM.....	5-14
6.0 EVALUATION AND RESULTS	6-1
6.1 AVERAGE PLUTONIUM CONCENTRATION IN INCOMING WASTE	6-1
6.1.1 Basis for 0.04 g Pu/L Limit on Batch-Averaged Concentration	6-1
6.2 WASTE WITH LOW SOLIDS CONTENT.....	6-3
6.2.1 Tanks With Less Than 10 kg of Plutonium	6-3
6.2.2 Transfers With Plutonium Concentration Less Than 0.001 g/L	6-4
6.2.3 Batches With Less Than 15 g of Plutonium	6-4
6.2.4 Tanks With Waste Spreading Capability.....	6-4
6.3 LIMIT ON SOLIDS CONTENT	6-5
6.3.1 Plutonium Concentration in Settled Solids.....	6-5
6.3.2 Limit on Neutron Absorber/Plutonium Mass Ratio in Discharged Batch	6-5
6.3.3 Tank-Averaged Solids/Plutonium Mass Ratio	6-7
6.3.4 Saltcake.....	6-7
6.4 ASSURANCE OF ADEQUATE MIXING.....	6-8
6.5 INTERTANK TRANSFERS OF SUPERNATANT LIQUID	6-9
6.6 ACCOUNTING FOR URANIUM-235 ENRICHMENT	6-10
6.6.1 Uranium-235 and the Plutonium Inventory	6-10
6.6.2 Determining the Uranium/Plutonium Subcritical Fraction	6-10
6.7 LIMITS FOR DOUBLE-CONTAINED RECEIVER TANKS.....	6-10
6.7.1 Agitator Pump Operation	6-11
6.7.2 Neutron Monitors on Double-Contained Receiver Tank 244-TX	6-11
6.8 EVALUATION OF PLANNED OPERATIONS.....	6-11

7.0 DESIGN FEATURES AND ADMINISTRATIVELY CONTROLLED LIMITS AND REQUIREMENTS	7-1
7.1 FACILITY CLASSIFICATION	7-1
7.2 DEFINITIONS.....	7-2
7.3 ALLOWED OPERATIONS THAT CAUSE NEGLIGIBLE DISTURBANCE	7-3
7.3.1 Surveillance Activities	7-3
7.3.2 Addition of Liquids	7-3
7.3.3 Operations That Have Negligible Effect On Nuclear Reactivity	7-4
7.4 WASTE DISTURBING OPERATIONS.....	7-5
7.5 GENERAL SPECIFICATIONS	7-5
7.5.1 Requirements.....	7-5
7.5.2 Limits.....	7-5
7.6 WASTE TRANSFERS FROM OUTSIDE TANK FARMS	7-6
7.6.1 Requirements.....	7-6
7.6.2 Limits for Transfers From Outside Tank Farms.....	7-7
7.7 ADDITIONAL LIMITS FOR DOUBLE-CONTAINED RECEIVER TANKS.....	7-8
7.8 242-A EVAPORATOR.....	7-8
7.9 PROPOSED CHANGES TO OPERATIONS.....	7-9
7.10 SURVEILLANCE PROGRAM.....	7-9
7.10.1 Schedule of Surveillance.....	7-9
7.10.2 Data Obtained from Samples	7-10
7.10.3 Investigation Levels	7-10
7.11 RECOVERY PLAN FOR NONCONFORMANCE.....	7-11
7.11.1 Nonconformance to Investigation Level.....	7-12
7.11.2 Nonconformance to Criticality Prevention Specification.....	7-12
7.11.3 Potential Criticality or Criticality Accident	7-12
7.12 FIREFIGHTING.....	7-13
7.13 VERIFICATION OF CONFORMANCE.....	7-13
7.13.1 Criticality Data Sheet	7-13
7.13.2 Transfer Data Sheet	7-13
7.14 CRITICALITY ALARM COVERAGE	7-14
8.0 SUMMARY AND CONCLUSIONS.....	8-1
8.1 PHYSICAL AND CHEMICAL PROCESSES	8-1
8.2 NEUTRON ABSORBER/PLUTONIUM MASS RATIOS.....	8-2
8.3 PLUTONIUM CONCENTRATION	8-3
8.4 ENRICHED URANIUM.....	8-3
8.5 NEUTRON MONITORS	8-4
8.6 AUDITS AND APPRAISALS.....	8-4
8.7 SURVEILLANCE PROGRAM.....	8-4
8.8 CRITICALITY ALARM COVERAGE	8-4
9.0 REFERENCES.....	9-1
9.1 BIBLIOGRAPHY.....	9-4

APPENDICES

A.0	PAST LIMITS ON INCOMING WASTE	A-1
A1.0	TANK FARM CRITICALITY PREVENTION SPECIFICATION LIMITS	A-3
A1.1	Limits in CPS-T-149-00010, Rev. D-0	A-3
A1.2	Limit on Plutonium Mass	A-5
A1.3	Transfers Using Cadmium as a Control	A-6
A2.0	DOUBLE-CONTAINED RECEIVER TANK LIMITS	A-7
A2.1	Description of Double-Contained Receiver Tank	A-7
A2.2	Double-Contained Receiver Tank Limits.....	A-9
A2.3	Basis for Double-Contained Receiver Tank Limits	A-10
A2.4	Neutron Monitors Installed on DCRT 244-TX	A-13
A3.0	242-A EVAPORATOR FACILITY	A-15
A3.1	Description of Process	A-15
A3.2	Limits	A-16
A3.3	Basis for Limits	A-16
A4.0	FACILITY DISCHARGE LIMITS	A-17
A4.1	Limits at the Purex Plant.....	A-17
A4.2	Limits at the Plutonium Finishing Plant.....	A-18
A5.0	REFERENCES	A-20
B.0	PARAMETERS IMPORTANT TO CRITICALITY SAFETY	B-1
B1.0	THE CONSERVATIVE WASTE MODEL	B-3
B1.1	Basis for Conservative Model.....	B-3
B1.2	Meaning of Conservative	B-4
B1.3	Composition of Conservative Waste Model.....	B-4
B1.4	Optimal Moderation	B-5
B1.5	Density of Dry Solids.....	B-6
B2.0	PLUTONIUM CRITICAL PARAMETERS	B-7
B2.1	Conservative Assumptions.....	B-7
B2.2	Plutonium in Water	B-8
B2.3	Plutonium in Waste.....	B-8
B2.4	Parametric Studies.....	B-9
B3.0	ABSORBER-TO-PLUTONIUM RATIOS.....	B-13
B3.1	Uranium-To-Plutonium Mass Ratio	B-13
B3.2	Iron and Manganese-To-Plutonium Mass Ratios	B-14
B3.3	Hydrogen-To-Plutonium Atom Ratio.....	B-14
B3.4	Boron and Cadmium	B-14
B3.5	Additional Absorber-To-Plutonium Mass Ratios.....	B-15
B3.6	Materials with Low Neutron Absorption Cross Sections.....	B-15
B4.0	COMBINING THE EFFECT OF SEVERAL ABSORBERS	B-16
B4.1	Sum of Actual-To-Minimum Subcritical Mass Fractions.....	B-16
B4.2	Solids-To-Plutonium Mass Ratio	B-18
B5.0	URANIUM CRITICAL PARAMETERS.....	B-18
B6.0	ESTIMATED NEUTRON MULTIPLICATION CONSTANT	B-19
B7.0	REFERENCES.....	B-21

C.0	CHARACTERIZATION OF TANK CONTENTS	C-1
C1.0	SOURCES OF CHARACTERIZATION DATA	C-3
C1.1	Best-Basis Inventories of Chemicals and Radionuclides	C-3
C1.2	Plutonium Inventory Database	C-4
C1.3	Hanford Defined Waste Model	C-4
C1.4	Analytical Samples	C-5
C1.5	Other Sources of Information	C-6
C2.0	DESCRIPTION OF WASTE STREAMS	C-7
C2.1	History of 200 Area Tank Farms	C-7
C2.2	Properties of Waste Relevant to Criticality Hazard	C-10
C2.3	Basic Waste Types	C-10
C3.0	LIQUID CONTENT	C-12
C4.0	ESTIMATE OF TOTAL PLUTONIUM IN TANK FARMS	C-13
C5.0	ESTIMATE OF URANIUM INVENTORY	C-14
C5.1	Average Uranium-235 Enrichment	C-14
C5.2	Waste Streams with Enriched Uranium	C-15
C6.0	ESTIMATE OF URANIUM-233 INVENTORY	C-19
C7.0	TANK-AVERAGED ABSORBER/PLUTONIUM MASS RATIOS	C-19
C7.1	Mass Ratios for Selected Tanks	C-20
C8.0	UNIQUE ITEMS IN TANK STORAGE	C-21
C8.1	Diatomaceous Earth	C-21
C8.2	Portland Cement	C-23
C8.3	Unique Items Containing Fissile Material	C-23
C9.0	ENHANCED PLUTONIUM PRODUCTION AT SURFACE OF FUEL ELEMENTS	C-24
C10.0	REFERENCES	C-26
D.0	PHYSICAL AND CHEMICAL PROCESSES IN TANK WASTE	D-1
D1.0	SOURCES OF CHEMISTRY INFORMATION	D-3
D1.1	Plutonium Chemistry	D-3
D1.2	Uranium Chemistry	D-4
D2.0	ALKALINITY	D-4
D2.1	Limit on pH	D-5
D2.2	Expected pH	D-6
D3.0	SOLUBILITY OF PLUTONIUM IN WASTE	D-7
D3.1	Concentration of Dissolved Plutonium	D-7
D3.2	Effect of Sodium Hydroxide on Plutonium Solubility	D-8
D3.3	Effect of Carbonates on Plutonium Solubility	D-8
D3.4	Upper Limit on Solubility	D-9
D4.0	CHEMICAL SEPARATION AND CONCENTRATION OF COMPONENTS ...	D-9
D4.1	Conditions That Prevent Plutonium Accumulation	D-11
D4.2	Removal of Neutron Absorbers	D-11
D5.0	CHEMICAL SEPARATION OF PLUTONIUM FROM URANIUM	D-12
D6.0	EXCEEDING DISCHARGE LIMITS	D-13

D7.0	SETTLING OF SUSPENDED SOLIDS	D-14
D7.1	Gravity Segregation	D-15
D7.2	Formation of Layers as a Dispersal Mechanism	D-16
D7.3	Case Study of Stratification	D-17
D7.4	Flocculation	D-18
D8.0	EVAPORATION	D-19
D9.0	MIXING	D-19
D10.0	CONCLUSIONS	D-20
D11.0	REFERENCES	D-21
E.0	GENERATION OF FUTURE WASTE	E-1
E1.0	PUREX PLANT	E-4
E2.0	PLUTONIUM FINISHING PLANT	E-4
E3.0	B-PLANT/WASTE ENCAPSULATION AND STORAGE FACILITY	E-5
E4.0	S-PLANT	E-6
E5.0	T-PLANT	E-6
E6.0	242-A EVAPORATOR AND LIQUID EFFLUENT RETENTION FACILITY	E-6
E7.0	WASTE FROM SINGLE-SHELL TANKS	E-7
E7.1	Saltwell Liquid Pumping	E-7
E7.2	Single-Shell Tank Solids Retrieval	E-7
E8.0	DOUBLE-SHELL TANKS	E-8
E8.1	Aging Waste Double-Shell Tanks	E-8
E8.2	Non-Aging Waste Double-Shell Tanks	E-8
E9.0	WASTE SAMPLING AND CHARACTERIZATION FACILITY	E-8
E10.0	EFFLUENT TREATMENT FACILITY	E-9
E11.0	100 AREAS	E-9
E12.0	300 AREA	E-9
E13.0	400 AREA	E-10
E14.0	REFERENCES	E-10
F.0	"MOST REASONABLE" HYPOTHETICAL ACCIDENT	F-1
F1.0	REFERENCE	F-4
G.0	EVALUATION OF PLANNED OPERATIONS	G-1
G1.0	SIMPLIFIED APPROACH TO EVALUATIONS	G-3
G2.0	BASIS DOCUMENTS ISSUED FOR SPECIFIC OPERATIONS	G-4
G3.0	THERMAL EFFECTS	G-5
G4.0	SLUICING	G-5
G5.0	SALTWELL PUMPING	G-7
G6.0	MIXER PUMP OPERATION	G-8
G6.1	Mixer Pump in Double-Shell Tank SY-101	G-8
G6.2	Mixer Pump in Double-Shell Tank AZ-101	G-8
G6.3	Mixer Pump in Double-Shell Tank AN-107	G-8
G7.0	AIRLIFT CIRCULATOR	G-9
G8.0	SLURRY DISTRIBUTOR	G-9

G9.0	TRANSFER OF LIQUID WASTE.....	G-9
G9.1	Particulate Suspended in Pumped Liquid.....	G-10
G9.2	Receiver Tank Considerations.....	G-10
G10.0	TRANSFER OF WASTE SOLIDS	G-11
G10.1	Transfer of Solids from Single-Shell Tank C-106 to Double-Shell Tank AY-102.....	G-11
G10.2	Future Transfers of Solids.....	G-14
G11.0	REFERENCES	G-15

TABLES

Table 2-1. Facilities that Interface with Tank Farms	2-2
Table 2-2. Farms Where Double-Shell Tanks (DSTs) Are Located	2-3
Table 2-3. Farms Where Single-Shell Tanks (SSTs) Are Located	2-3
Table 2-4. Active Catch Tanks	2-6
Table 3-1. Minimum Neutron Absorber*/Plutonium Subcritical Mass Ratios	3-3
Table 5-1. Contingencies and Barriers.....	5-2
Table 7-1. Minimum Neutron Absorber/Plutonium Subcritical Mass Ratios	7-7
Table A-1. Particle Size Volume Distribution.....	A-14
Table B-1. Composition of Conservative Waste Model.....	B-5
Table B-2. Plutonium Subcritical Limits.....	B-9
Table B-3. Minimum Subcritical Absorber-to-Plutonium.....	B-15
Table B-4. Minimum Subcritical Absorber-to-Plutonium.....	B-16
Table C-1. Highest Plutonium Concentrations Determined From Sample Analysis of Each Waste Type.....	C-6
Table C-2. Global Radionuclide Inventory.....	C-13
Table C-3. Uranium Inventory.....	C-15
Table C-4. Acronyms for Waste Streams	C-16
Table C-5. Waste Streams With High Uranium Content or Enrichment.....	C-17
Table C-6. Waste Streams With Enriched Uranium	C-18
Table C-7. Total Quantity of Waste in All Tanks - July 1999.....	C-20
Table C-8. Unique Contents of Single-Shell Tanks.....	C-22
Table E-1. Average Monthly Waste Generation Rates.....	E-3
Table E-2. Plutonium Finishing Plant Waste Solids Content.....	E-5

FIGURES

Figure B-1 Sphere Minimum Critical Diameter	B-11
Figure B-2. Sphere Minimum Critical Mass.....	B-11
Figure B-3. Slab Minimum Critical Thickness.....	B-12
Figure B-4. Slab Minimum Critical Mass	B-12
Figure B-5. Maximum k_{∞} for Homogeneous Sludge as a Function of Iron Content.....	B-20

LIST OF TERMS

B&W	Babcock and Wilcox Hanford
Bq/L	Becquerel per liter
CAS	Criticality Alarm System
CDS	Criticality Data Sheet
CDS	Criticality Detection System
CHG	CH2M Hill Hanford Group, Inc.
Ci	Curie
Ci/g	Curies per gram
Ci/L	Curies per liter
cm	centimeter
cm ²	square centimeters
cm ⁻¹	per centimeter
cp	centipoise
CPS	Criticality Prevention Specification
CSER	Criticality Safety Evaluation Report
CSR	Criticality Safety Representative
CWM	Conservative Waste Model
DCRT	Double-Contained Receiver Tank
DOE	U.S. Department of Energy
DQO	Data Quality Objective
DST	Double-Shell Tank
EDTA	Ethylenediaminetetraacetic acid
FDH	Fluor Daniel Hanford, Inc.
FSAR	Final Safety Analysis Report
ft	feet
ft ²	square feet

ft/s	feet per second
g	grams
g Cd/g Pu	grams cadmium per gram plutonium
g/cm ³	grams per cubic centimeters
g/gal	grams per gallon
g/ft ²	grams per square feet
g/kg	grams per kilogram
g/L	grams per liter
g/m ²	grams per square meter
g Pu/gal	grams plutonium per gallon
g Pu/L	grams plutonium per liter
g Pu/ft ²	grams plutonium per square feet
g Pu/m ²	grams plutonium per square meter
gal	gallon(s)
gal/min	gallons per minute
gpm	gallons per minute
HDW	Hanford Defined Waste
HEDTA	Hydroxyethylethylenediaminetriacetic acid
HEPA	High Efficiency Particulate Air
hp	horsepower
IMUST	Inactive Miscellaneous Underground Storage Tank
in.	inches
kCi	kilocurie
kg	kilogram
kg/L	kilograms per liter
kg/m ²	kilogram per square meter
kg Pu/m ²	kilograms plutonium per square meter
kgal	kilogallon
kL	kiloliter
km	kilometers
L	liter
LANL	Los Alamos National Laboratory
LMHC	Lockheed Martin Hanford Corporation
L/min	liters per minutes
L/s	liters per second
lb.	pound
m	meters
m/s	meters per second
m ³	cubic meters
mg/gal	milligram per gallon
mg/L	milligram per liter
MISF	miscellaneous inactive storage facilities
MJ	Mega Joules
mL	milliliters
PFP	Plutonium Finishing Plant
psig	pounds per square inch (gauge)

PUREX	Plutonium-Uranium Extraction
rem	Roentgen-equivalent man
RHO	Rockwell Hanford Operations
RMC	Remote Mechanical "C"
REDOX	Reduction Oxidation Plant
RPP	River Protection Project
SAR	Safety Analysis Report
SST	Single-Shell Tank
TBP	tributyl phosphate
TDS	Transfer Data Sheet
TPA	Tri-Party Agreement
TSR	Technical Safety Requirement
TWRS	Tank Waste Remediation Systems
vol%	volume percent
W/m ³	Watta per cubic meter
WHC	Westinghouse Hanford Company
wt%	weight percent
X/Pu	Neutron absorber (X) to plutonium (Pu) mass ratio
%	percent
μCi/g	microcuries per gram
μm	micrometer or micron
°C	degrees Celsius
°F	degrees Fahrenheit

1.0 INTRODUCTION

High-level radioactive waste from nuclear fuels processing is stored in underground storage tanks in the high-level waste (HLW) tank farms located in the 200 East and 200 West Areas on the Hanford Site. The older single-shell tanks (SSTs) are inactive and no longer receive waste. The newer double-shell tanks (DSTs) are active and will continue to receive waste. In addition, there are five double-contained receiver tanks (DCRTs) and two double-contained vaults used to hold waste on a temporary basis.

An important safety concern is the prevention of a self-sustaining neutron chain reaction, also known as nuclear criticality, in the tanks. In practice, criticality is possible in the tank waste environment only with isotopes that fission with moderated neutrons (i.e., isotopes that are fissile). Only ^{233}U , ^{235}U , and ^{239}Pu are present in high enough quantity to be of concern, and, of these, ^{239}Pu has the smallest critical mass. As a conservative measure, all fissionable isotopes are treated as if they were ^{239}Pu . This evaluation uses the term "fissionable" to emphasize that all isotopes capable of fissioning are tracked and the term "fissile" to stress that moderation is required to credibly develop a potential for criticality.

This Criticality Safety Evaluation Report (CSER) provides justification that a nuclear criticality cannot occur over the range of credible tank farms operations. Limits and controls to govern the receipt, transfer, and storage of waste are spelled out, and the bases for continued long-term safety are provided.

In the past, when process facilities were operating at full capacity, sizeable volumes of waste were discharged on an almost daily basis. In the future, with most facilities shut down, the volume of new waste received, and the content of plutonium, will be greatly reduced. Nevertheless, transfers of waste between tanks will continue to be made.

Inside a storage tank, the composition and distribution of waste components become uncertain due to waste complexity, mixing of waste types, and the difficulty of characterization. Because of this uncertainty, criticality safety depends upon demonstrating inherent characteristics of waste that ensure subcriticality. Characteristics of primary importance to criticality safety are the large proportion of waste solids to fissile material and the relative uniform mixing of components.

Critical concentrations and dimensions used in the evaluation of tank waste are based upon the conservative waste model (CWM) defined by Rogers (1993a) to provide a hypothetical waste composition known to be conservative relative to actual waste. The CWM defines a waste composition that has a smaller absorption cross section than any actual waste, and critical parameters derived from the model may be applied to waste from any storage tank. More commonly neutron absorber/plutonium mass ratios are used as the basis for criticality safety, and these apply to all waste compositions.

In the past, waste-generating operations were usually governed by flowsheets that precisely specified every aspect of the process and defined the waste by-product. Flowsheet control ensured that the upper limit on fissile content in waste was known to a high degree of certainty. Most future waste will be generated during cleanup of older facilities, and these operations may prove more difficult to characterize. The fissile content generated in cleanup operations is expected to be low.

A program of waste characterization and surveillance increases our knowledge of tank waste and verifies compliance to discharge limits. Characterization data increases the assurance of the waste's high degree of subcriticality.

Several studies of the criticality safety of tank waste have been issued. *High-Level Waste Subcriticality Safety Assessment* (Braun et al. 1994) collects and analyzes available characterization data. *Tank Farm Nuclear Criticality Review* (Bratzel et al. 1996) justifies subcriticality of tank waste by knowledge of physical and chemical properties (i.e., by the phenomenological properties of the waste). These reports discuss the basis for criticality safety using process history, waste stream compositions, waste sample data, and waste chemistry. Serne et al. (1996) and Whyatt et al. (1996) provide detailed chemistry studies to support these evaluations.

This CSER provides the bases for criticality prevention specification (CPS) limits and controls used for the receipt of waste into tank farms, for its transfer between tank farm facilities, and for long-term storage.

In the past, operations with SSTs were covered by *CSER 94-001: Criticality Safety of Single Shell Waste Storage Tanks* (Rogers 1994a) and operations with DSTs were covered by *CSER 94-004: Criticality Safety of Double Shell Waste Storage Tanks* (Rogers 1994b). This CSER replaces both of those CSERs. In addition, the description is expanded to include all facilities under control of tank farms.

2.0 DESCRIPTION OF TANK FARM FACILITIES

For the purpose of criticality safety, tank farms in its entirety is classified as a Limited Control Facility. Alternately, each individual facility within tank farms that is permitted to contain fissionable material, such as an individual waste storage tank, may be viewed as an individual Limited Control Facility.

For the purposes of this report, individual tank farm facilities are categorized according to the following functions: (1) long-term storage tanks; (2) short-term (intermediate) holding tanks and vaults; (3) evaporator facilities; (4) inactive underground tanks; (5) miscellaneous facilities; and (6) waste transfer system.

A detailed description of tank farm facilities is provided in the River Protection Project (RPP) Final Safety Analysis Report (FSAR) (LMHC 1999a). A summary is included here to identify and describe locations where fissionable material might be found. Most locations described are places where radioactive waste is stored. However, locations are also described where fissionable material might be placed inadvertently or where waste might accumulate under abnormal conditions. This section is intended as a guide to understanding the overall tank farms criticality safety program.

Waste stored in tank farm facilities is a complex mixture of chemicals that contains low concentrations of fissile material. A description of tank waste is provided in Appendix C, and the chemical processes of importance to criticality safety are summarized in Appendix D.

Under foreseeable conditions, the form and distribution of waste components ensures subcriticality, even with unlimited quantity and unrestricted geometry. Since criticality is precluded under credible conditions, it is not important that the description of tank geometry or a facility configuration be precise. In addition, some information provided may not relate directly to criticality safety, but is presented to provide a better understanding of tank farms operations. For these reasons, this CSER need not be revised to incorporate a change to a facility configuration or to correct a discrepancy in description, unless the discrepancy might be capable of reducing the margin of subcriticality.

The following description emphasizes facilities where fissionable material might be present, either in the course of normal operations or as the consequence of an accident. More detailed descriptions can be found in the FSAR (LMHC 1999a) and in Stickney and Lipke (1998).

2.1 ENTRY POINTS INTO TANK STORAGE SYSTEM

Criticality safety of tank waste depends on the control exercised before the waste enters the tank storage system. This control must be exercised at the various processing facilities that discharge waste to the tank farms system. Table 2-1 provides a list of facilities that are at present sending or have in the past sent waste to tank farms (i.e., the waste generators). Facilities presently sending waste are identified as "active interfaces" and have an "active" status. Most major waste

generators have been shut down and now have an “inactive status.”

There are potential sources of waste not indicated in Table 2-1, such as waste from the cleanup of Plutonium-Uranium Extraction (PUREX) or from the 400 Area. This waste is expected to contain little or no fissionable material and would be sent to the 204-AR Unloading Facility by truck.

T-Plant has both active and inactive interfaces. Although T-Plant has been shut down, it occasionally still sends waste via tanker truck to the 204-AR Unloading Facility.

Table 2-1. Facilities that Interface with Tank Farms

Originating Facility	Comment
Active Interfaces	
204-AR Unloading	Railcar and tanker truck unloading point.
222-S/REDOX	Sends waste to 204-AR.
340 Building	Sends waste to 204-AR.
T-Plant	Sends waste via truck to 204-AR.
219-S	Goes to DCRT 244-S.
242-A Evaporator	Removes water. Waste from tank storage recycled.
PFP (Z-Plant)	Handles separated plutonium. Goes to DCRT 244-TX .
Inactive Interfaces	
B-Plant/WESF	
PUREX	Source of most waste in tank storage.
T-Plant	Transfer lines to tank farms are inactive.
U-Plant	
242-S Evaporator	
242-T Evaporator	
Unique	Unique material input through risers. (See Appendix C8.0)

Notes:

PFP = Plutonium Finishing Plant
 PUREX = Plutonium-Uranium Extraction
 REDOX = Reduction-Oxidation Plant
 WESF = Waste Encapsulation and Storage Facility

2.2 LONG-TERM STORAGE TANKS

Long-term storage tanks are classed as single-shell tanks (SSTs) or double-shell tanks (DSTs), according to their construction, but there is no real distinction between the waste in DSTs and SSTs tanks relative to criticality safety. The tanks are located under 1.8 to 2.4 m (6 to 8 ft) of soil to provide shielding from radioactivity.

2.2.1 Double-Shell Storage Tanks

All new waste is sent to DSTs, which have been in use since 1971. These tanks are 22.9 m (75 ft) in diameter with a nominal capacity of 3,800 kL (1,000 kgal). A DST has two steel containment shells with an annular air space between them to greatly reduce the possibility of leakage of liquid waste into the ground. The outer shell is surrounded by reinforced concrete. Table 2-2 shows the location of the 28 DSTs.

Table 2-2. Farms Where Double-Shell Tanks (DSTs) Are Located

200 East Area Farms		200 West Area Farms	
Farm	Number of DSTs	Farm	Number of DSTs
241-AN	7	241-SY	3
241-AP	8		
241-AW	6		
241-AY	2		
241-AZ	2		
Total	25	Total	3

2.2.2 Single-Shell Storage Tanks

Single-Shell Tanks (SSTs) are older than the DSTs and have only one steel containment shell. There are 133 SSTs that have a diameter of 22.9 m (75 ft) and nominal capacities of 2,000, 2,900, and 3,800 kL (530, 758, and 1,000 kgal). There are also 16 SSTs that have a diameter of 6.1 m (20 ft) and a nominal capacity of 208,000 L (55,000 gal). Table 2-3 shows the locations of the 149 SSTs.

Table 2-3. Farms Where Single-Shell Tanks (SSTs) Are Located

200 East Area Farms		200 West Area Farms	
Farm	Number of SSTs	Farm	Number of SSTs
241-A	6	241-S	12
241-AX	4	241-SX	15
241-B	16	241-T	16
241-BX	12	241-TX	18
241-BY	12	241-TY	6
241-C	16	241-U	16
Total	66	Total	83

2.2.3 Leakage Accumulation Locations

If a leak were to develop in a storage tank, waste might accumulate in process and service pits or in the annular volume surrounding a DST. Potential locations of accumulation are:

- **DST annular volume** - The secondary tank on a DST is 1.5 m (5 ft) larger in diameter than the primary tank, which forms a 0.76-m (2.5-ft) wide annular space.
- **Central pump pit** - This pit, which is typically 2.1 m (7 ft) square by 3.0 m (10 ft) deep, is approximately centered on a tank, and waste is normally pumped out of a tank through it.
- **DST annulus pump pits** - These pits located over annulus risers are typically 2.1 m (7 ft) square by 3.5 m (11.6 ft) deep with 0.3-m- (1-ft-) thick concrete walls.
- **Feed pump pits** - These typically measure about 2.1 m (7 ft) square by 2.51 m (8.25 ft) deep with 0.3-m- (1-ft-) thick reinforced concrete walls.
- **Leak detection pits** - These are installed in all DST farms and in the 241-AX Farm. These pits typically measure about 2.1 m (7 ft) square by 3.2 m (10.6 ft) deep with 0.3-m- (1-ft-) thick reinforced concrete walls.
- **Drain pits** - These are located on the following DSTs: AN-101, AP-103, AW-102, and SY-102. The drain pits are typically 2.4 by 2.1 m (8 by 7 ft) by 3.5 m (11.6 ft) deep.
- **Sluice pits (DSTs only)** - These are typically 4.3 m (14 ft) square by 3.84 m (12.6 ft) deep.
- **Flush pits and valve pits** - These are also located near SSTs.

2.3 SHORT-TERM HOLDING TANKS

Upon discharge from a processing facility, waste may be sent to a double-contained receiver tank (DCRT) or to a vault tank for temporary storage before being transferred to a DST.

2.3.1 Double-Contained Receiver Tanks

Waste leaving a processing facility is often routed through a DCRT for temporary storage before reaching a DST. Generally, a DCRT receives SST saltwell liquor and provides flexibility of waste transfer routing.

DCRT 244-A and DCRT 244-BX are located in the 200 East Area, and DCRTs 244-S, 244-TX, and 244-U are located in the 200 West Area. They are described as follows:

- **DCRT 244-A** - This DCRT is an approximately 4.6-m (15-ft) diameter by 5.5-m (18-ft) long, vertically oriented, cylinder with a capacity of 61,627 L (16,280 gal). The DCRT 244-A valve pit contains lines for the original cross-site and B-Plant transfer lines, and it also contains drains.
- **DCRT 244-BX** - This DCRT is similar in size and shape to DCRTs 244-S and 244-TX, but has a capacity of 117,350 L (31,000 gal). This tank provides short-term storage for jet pump solution from the B, BX, and BY Tank Farms. DCRT 244-BX has no sluicing nozzles or neutron monitors.
- **DCRT 244-S** - This DCRT is similar in configuration to the 244-A DCRT, but has a capacity of 76,768 L (20,280 gal). The 244-S DCRT provides short-term storage for solution from the S and SX Tank Farms and the 222-S Laboratory. Two 10.2-cm (4-in.) diameter dry wells are provided to permit neutron monitoring on special occasions.
- **DCRT 244-TX** - This DCRT is used for short-term storage of Plutonium Finishing Plant (PFP) waste and saltwell waste (jet-pump solution) from the T, TX, and TY Tank Farms. The DCRT 244-TX is 3.7-m (12-ft) diameter by 12.5-m (41-ft) long with a volume of 117,350 L (31,000 gal). This tank is oriented horizontally with a slight slant such that the inlet is at the high end and the outlet is at the low end.

DCRT 244-TX uses 11 sluicing jets to circulate and flush the contents to prevent solids buildup. In addition, three neutron monitors are located underneath the tank at regular spacing. These monitors were installed to provide capability to detect plutonium buildup should it occur. However, chemistry studies (Whyatt et al. 1996; Serne et al. 1996; Bratzel et al. 1996) provide a basis for discontinuing their use.

- **DCRT 244-U** - This DCRT is an 117,350-L (31,000-gal) tank used to receive saltwell jet-pumped liquor from the 241-U Tank Farm. This tank has the same design and operation as DCRT 244-BX. No sluicing nozzles or neutron monitors are provided.

2.3.2 Vault Tanks

Although the 244-AR and 244-CR vaults were designed to provide chemical treatment, they are no longer used for this purpose.

- **244-AR Vault Tank** - The 244-AR Vault consists of a concrete canyon facility above three belowgrade process cells. The vault was designed to remove, treat, and transfer PUREX sludge to B-Plant for fission product removal, provide interim storage for PUREX acid waste feed to B-Plant, and receive and distribute the neutralized high-level waste of B-Plant. The process cells contain a 163,333-L (43,148-gal) tank, a 164,431-L (43,438-gal) tank, and two 18,143-L (4,793-gal) tanks. The 244-AR Vault is currently inactive.

- **244-CR Vault Tank** - The 244-CR vault is a belowgrade, two-level, multicell, concrete structure. The process cells contain two 189,250-L (50,000-gal) tanks and two 56,775-L (15,000-gal) tanks. This vault is inactive except for TK-CR-003, a DCRT with an 189,300-L (50,000-gal) capacity available for C Farm saltwell transfers. In 1998 TK-CR-003 contained approximately 15,900 L (4,220 gal) of saltwell waste from SSTs C-102, C-107, and C-110, with an unknown amount of solids associated with it (Stickney and Lipke 1998).

2.3.3 Catch Tanks

Catch tanks are buried underground at locations where potential leaks are anticipated. These tanks are normally empty and are intended to function as collection points for liquid wastes that might drain from waste transfer systems and DST equipment. There are four basic catch tank designs: (1) direct buried, concrete; (2) direct buried, steel; (3) steel tanks contained in a vault or pit; and, (4) concrete vaults with steel liners. Inactive catch tanks are classed as inactive miscellaneous underground storage tanks (IMUSTs).

The FSAR (LMHC 1999a) lists 12 active catch tanks. These are shown in Table 2-4.

Table 2-4. Active Catch Tanks

Catch Tank	Volume	
	Liters	Gallons
200 East Area Tanks		
241-A-302A	31,940	8,438
241-A-350	2,940	776
241-A-417	167,315	44,200
241-AX-152	41,640	11,000
241-AZ-151	45,046	11,900
241-AZ-154	3,290	869
241-ER-311	66,934	17,682
200 West Tanks		
241-S-304	23,850	6,300
241-TX-302C	66,930	17,681
241-U-301B	133,530	35,275
241-UX-302A	66,934	17,682
241-EW-151	2,940	776

2.4 242-A EVAPORATOR FACILITY

At issuance of this CSER, the RPP does not have responsibility for the 242-A Evaporator. Nevertheless, no separate CSER has been written for it, and its description and evaluation are retained. Functionally, it is part of tank waste operations.

The 242-A Evaporator is used to reduce the volume of dilute waste solutions, thereby reducing the number of DSTs required for storage. The evaporator process employs a conventional forced circulation, vacuum evaporation system to concentrate waste solutions. The main process components of the evaporator-crystallizer system include the reboiler, vapor-liquid separator, recirculation pump and pipe loop, slurry product pump, primary condenser, jet vacuum system, condensate collection tank, and ion exchange system.

After a period of shutdown, the 242-A Evaporator was restarted on April 15, 1994, and remains in operation. The evaporator is not run continuously, but rather part time in a series of campaigns. During a typical month of operation the volume of liquid evaporated is about 1,900 kL (500 kgal).

A minimum of two dedicated DSTs is required for operation of the evaporator. Prior to operation, wastes are collected in dilute receiver tanks. Any solids in the initial waste will settle out. When these tanks are full, the supernate is transferred to the evaporator feed tank (DST AW-102). After DST AW-102 is full and Evaporator Data Quality Objective (DQO) requirements are met, supernate is transferred to the 242-A Evaporator for boil-down.

Heating feed slurry in the reboiler starts the evaporation process. The heated slurry is discharged from the reboiler to the vapor-liquid separator vessel, which is maintained at a reduced pressure. Water vapor is drawn off and taken into the primary condenser to return it to a liquid phase. The slurry becomes more concentrated and is pumped to the evaporator receiver tank (DST AW-106) and allowed to settle. Supernatant liquid is pumped back as feed to the evaporator, and the process to remove additional water is repeated. During each pass through the evaporator, the waste concentration increases. Upon reaching the desired concentration level, the waste is transferred to DST AW-106. This concentrated waste is referred to as double-shell slurry feed (DSSF).

The 13 million-gallon storage capacity of the Liquid Effluent Retention Facility (LERF) is used to store the evaporator process condensate until it can be treated.

Both the 242-S and the 242-T Evaporator Facilities are inactive. The purpose and operation of these facilities were the same as that of the 242-A Evaporator Facility.

2.5 MISCELLANEOUS FACILITIES

A listing is made of miscellaneous facilities of interest to criticality safety. Of these facilities, only the 204-AR Waste Unloading Facility could contain waste to be transferred to a SST or a DST. For some facilities the criticality safety interest is the confirmation that fissile contaminated material are not brought into them.

2.5.1 204-AR Waste Unloading Facility

The 204-AR Waste Unloading Facility, built in 1981, is a reinforced concrete structure where liquid waste may be received from rail tank cars and from tanker trucks. The fissionable content of this waste is low.

This facility has a chemical adjustment capability utilizing 4 chemical make-up tanks and a sluicing system to remove sludge from tank cars. The 204-AR-TK-1 catch tank, with a volume of 5,678 L (1,500 gal), is located beneath the floor to collect floor drainage. Liquids from the catch tank are periodically pumped to tank farms.

2.5.2 209-E Complex

The 209-E Complex is located north of the Plutonium-Uranium Extraction (PUREX) Plant in the 200 East Area. This complex consists of the 209-E Critical Mass Laboratory, the 90-Day Waste Storage Pad, the Intermediate Holding Area, and offices to hold administrative support personnel. Tiffany (1998) provides a detailed description of the 209-E Building.

2.5.2.1 209-E Critical Mass Laboratory

The 209-E Critical Mass Laboratory was built in 1960 to study critical configurations. In the late 1980s, the facility was shut down, and all fissile material was removed, except for residual material that could not be removed from the storage tanks and from surfaces. The Critical Assembly Room contains two critical assembly hoods (with contaminated equipment), a water tank, and 11 process tanks ranging from 38 L (10 gal) to 400 L (106 gal). In addition, the Mix Room contains contaminated equipment and 9 tanks ranging from 10 L (2.6 gal) to 320 L (85 gal). A drain tank (TK-111) is located underground adjacent to the south end of the facility. The Critical Assembly Room and the Mix Room are ventilated through high efficiency particulate air (HEPA) filters.

A non-destructive analysis (NDA) was performed in 1989 to quantify the fissile material remaining after the tanks were flushed. Stickney and Lipke (1998) list the remaining quantities of fissile materials. Custodianship of this fissile material is no longer the responsibility of the Criticality Safety Representative (CSR).

2.5.2.2 Less Than 90-Day Waste Storage Pad

The Less Than 90-Day Waste Storage Pad, which is 12 m wide by 18 m long (40 by 60 ft), is divided into a hazardous waste pad, a mixed-waste pad, and a low-level waste pad. Day-to-day maintenance and operations in the 200 East Area tank farms generate the waste stored on the pad. This waste includes batteries, transformers, solvents, oils, paints, and cleaning materials. Much of this waste comes from non-radiologically contaminated areas, but some comes from radiation zones. Contaminated equipment and materials that have come into contact with tank waste are stored here. This Complex is compliant to the *Resource Conservation and Recovery Act of 1976* (RCRA 1976). Waste may not be stored longer than 90 days.

Low-level radioactive wastes are collected in the Intermediate Holding Area, separated from the Less Than 90-Day Waste Storage Pad by a chain link fence, where they are characterized before being transported to a disposal site.

2.5.3 Vertical Storage Units

A vertical storage unit is a vertical pipe set up in a tank farm to store dry, radiologically contaminated equipment or soil. These units are constructed of 0.3 to 0.6-m (1 to 2-ft) diameter, vertical steel pipes of 1.8 to 2.1-m (6 to 7-ft) length. Most vertical storage units are located in 200 West Area SST farms, and all are now inactive. They do, however, contain radioactive waste material.

2.5.4 213-W Dry Waste Compactor Facility

The 213-W Dry Waste Compactor Facility is located south of the Central Waste Complex in the 200 West Area. Beginning in 1985 low activity solid wastes were compacted for disposal in the 200 West Area Low Level Waste (LLW) Burial Grounds, but compacting activity has been discontinued and will not be resumed. This building is now used for contaminated instrument repair. A catch tank located below ground was sampled and determined to be below the level of radiological concern.

2.5.5 2727-WA Sodium Storage Facilities

The 2727-WA Sodium Storage Facility is used to store miscellaneous equipment in addition to sodium. This facility does not contain any fissile materials. Criticality safety verifies that no fissionable contaminated waste is brought into this facility.

2.5.6 Grout Treatment Facility

The Grout Treatment Facility was designed to combine low-level radioactive waste material with grout. The facility was shut down in 1989 and placed on standby in 1994. One grout disposal vault contains low-level DST phosphate/sulfate grouted waste. All other structures, components, and vessels are empty of radioactive wastes and chemicals. There are no plans to restart the facility for its original purpose (Stickney and Lipke 1998).

2.5.7 Cribs, Ponds, and Ditches

A crib is a subsurface liquid distribution system used to dispose of liquid effluents so that low-level radionuclides (if any) are absorbed and held in the soil. Most cribs consist of gravel-filled trenches 3.0 to 4.6 m (10 to 15 ft) deep with a bottom about 3 m (10 ft) wide and with sides that slope outward at the top.

Wastewater from operating facilities in the 200 East and 200 West areas is discharged to B Pond. A contingency pond is located nearby. Gravel-lined ditches also receive wastewater discharges.

Effluents sent to cribs, ponds, and ditches do not contain significant concentrations of fissile materials, and these locations are not of criticality safety concern.

2.5.8 Ventilation Systems

Ventilation systems have been suggested as locations where fissionable material might accumulate. One potential mechanism for deposition of plutonium bearing particles in ventilation ducts or filters is aerosolization of dried waste. However, plutonium agglomerates with other solids, and the mass ratio of neutron absorbers to plutonium in deposited particulate should be very high, as occurs in typical tank waste. The diameter of a typical ventilation duct is 25.4 cm (10 in.). If a duct were totally blocked with solids, such that no airflow is possible, the concentration of plutonium would be far below 22 g/L, the minimum required to achieve criticality under the most idealized conditions in a duct of this diameter. There is no identified mechanism capable of achieving this high a plutonium concentration.

2.6 WASTE TRANSFER SYSTEMS

Waste transfers can be placed in seven categories: (1) to the 242-A Evaporator; (2) cross site transfer; (3) between process facilities and tank farms; (4) farm-to-farm; (5) within a farm; (6) catch tank to storage tank; and (7) condensate collection tank to storage tank. The FSAR (LMHC 1999a) provides a description of transfer lines and miscellaneous transfer equipment and structures associated with waste transfers. The transfer lines contain waste only when a transfer is in progress.

2.6.1 Cross Site Transfer Lines

Waste is transported between the 241-SY-A and 241-SY-B valve pits in the 200 West Area and the 244-A lift station in the 200 East Area through the cross-site transfer system. An older system was replaced by a new buried pipe-in-pipe system approximately 10.5 km (6.5 miles) long. The interior pipe is stainless steel with a 7.6-cm (3-in.) diameter, and the exterior pipe is galvanized carbon steel with a 15.2-cm (6-in.) diameter. Transfers of liquid waste in either one or two lines can be made in either direction using the older tank farm transfer pumps. A booster pump has been added to transfer waste of higher viscosity with up to 30% solids.

2.6.2 Interconnecting Lines

Waste transfers can be routed along dedicated or nondedicated interconnecting transfer lines. A dedicated route does not require valving or jumper changes, while a nondedicated route might require the repositioning of jumpers or realignment of valves. Most active transfer lines have a diameter less than 10.2 cm (4 in.), and no line is listed as over 25.4 cm (10 in.) in diameter (LMHC 1998b). From the viewpoint of criticality safety, these diameters are small.

Valve pits, diversion boxes, diverter stations, catch tanks, and DCRTs are used to control or direct waste transfers. These structures are potential collection points of waste under normal operating conditions and following a leakage of waste.

2.6.3 241-AX-152 Diverter Station

The 241-AX-152 Diverter Station is an underground reinforced concrete structure that is 13.4 m (44 ft) long, 3 m (10 ft) wide, and 7.6 m (25 ft) high. This station utilized a moveable spout to redirect the flow of waste to any one of several exit pipes. Below the exit pipes is a 41,600-L (11,000-gal) catch tank.

2.7 INACTIVE MISCELLANEOUS UNDERGROUND STORAGE TANKS

Inactive miscellaneous underground storage tanks (IMUSTs) include a variety of radioactive underground tanks located throughout the 200 East and 200 West Areas. Tanks classified as IMUSTs include catch tanks, treatment tanks in vaults, neutralization tanks, decontamination drain tanks, and settling tanks. The IMUST classification is identified simply to track miscellaneous tanks that are not located within a major building or facility.

There are 70 IMUSTs in the 200 Areas, and RPP is responsible for the majority of these tanks. IMUST capacities range from 189 L (50 gal) to 189,000 L (50,000 gal). Most IMUSTs are isolated and sealed. The contents of most IMUSTs are alkaline and generally typical of SSTs. A few contain decontamination solutions, and a few may be acidic. All IMUSTs are posted and barricaded. In a recent survey (Stickney and Lipke 1998), there was no indication of any new or unique hazard associated with these tanks.

Chiao (1996) provides a preliminary criticality safety review of 62 IMUSTs. The conclusion is that only non-intrusive operations are to be permitted. Bratzel et al. (1996) and the more recent evaluation of process histories for IMUSTs leads to the conclusion that the contents of those IMUSTs which are alkaline contain wastes generally typical of SSTs. These IMUSTs are bounded by this CSER. All other IMUSTs require an evaluation to determine if proposed intrusive work is bounded or whether a new CSER is required.

This page intentionally left blank.

3.0 REQUIREMENTS DOCUMENTATION

The limited ability to control or monitor waste after it has entered tank storage makes it necessary to exercise criticality safety control over the waste before it is received. Limits and controls are applied before discharge to ensure waste has inherent characteristics that guarantee subcriticality into the indefinite future without supervision.

3.1 REQUIREMENTS DOCUMENTS

DOE Order 5480.24, *Nuclear Criticality Safety* (DOE 1992), mandates that a program be established ensuring that criticality safety is comprehensively addressed and receives an objective review, with all identifiable risks reduced to acceptably low levels and management authorization of the operation is documented.”

- Section 7c requires that the “nuclear criticality safety program be fully documented.”
- Section 7b (3) and (4) provide the requirements for operation without a Criticality Alarm System (CAS) or a Criticality Detection System.

In the future, DOE Order 420.1, *Facility Safety* (DOE 1995) will replace DOE Order 5480.24 as the requirements document for RPP. This change will have no impact on this CSER.

Administrative controls for RPP nuclear criticality safety are found in Chapter 5.7 of *Tank Waste Remediation System Technical Safety Requirements* (LMHC 1996). Implementation of the technical safety requirement that a criticality safety program be maintained is provided by Chapter 5.7 of HNF-IP-1266, *Tank Farms Operation Administrative Controls* (CHG 2000a). Classification as a Limited Control Facility requires a documented criticality safety evaluation that demonstrates “that a criticality is prevented by the form or distribution of fissionable material, after allowing for credible accidents.”

This CSER has been formatted according to the format recommended by DOE-STD-3007, *Guidelines for Preparing Criticality Safety Evaluations at Department of Energy Non-Reactor Nuclear Facilities* (DOE 1998).

3.2 TECHNICAL SAFETY REQUIREMENT

Administrative controls for nuclear criticality safety are found in Chapter 5.7 of *Tank Waste Remediation System Technical Safety Requirements* (LMHC 1996). The Technical Safety Requirement (TSR) for criticality safety and the key elements against which the program is evaluated are:

Requirement for Nuclear Criticality Safety

A program shall be maintained for ensuring waste remains subcritical.

Program Key Elements

- a. Criticality limits and controls shall be documented in Criticality Safety Evaluation Reports (CSERs) and implemented in Criticality Prevention Specifications (CPSs) and procedures.
- b. Procedures shall be established for recovery from a nonconformance.
- c. Criticality safety training shall be provided for operations and technical personnel.
- d. For transfers into the tank farms from non-tank farm facilities (e. g., Plutonium Finishing Plant, B-Plant), the following pretransfer conditions shall be met:
 - The pH shall be ≥ 8 .
 - The plutonium concentration shall be < 0.04 g/L averaged over the entire volume of the transferred waste.
 - The neutron absorber (X) to plutonium (Pu) mass ratio (X/Pu) shall be greater than at least one of the minimum subcritical ratios shown in Table 3-1. This condition is not required if the plutonium concentration is ≤ 0.001 g/L.
 - Compliance with the above conditions may be demonstrated by sample analysis or calculation of final conditions based on initial sample analysis, but requires INDEPENDENT VERIFICATION.
- e. Activities in Miscellaneous Inactive Storage Facilities (MISFs) (except for sampling/monitoring) are prohibited without characterization of the waste and, if required, preparation of a CSER and CPS.
- f. Transfers via the Replacement Cross Site Transfer System, which will intentionally transfer high solids contents, will require characterization of the waste, preparation and approval of a CSER and, if required, a CPS prior to initiating the transfer.

Table 3-1. Minimum Neutron Absorber*/Plutonium Subcritical Mass Ratios

Neutron Absorber (X)	Minimum Neutron Absorber/Plutonium Subcritical Mass Ratio (X/Pu)
Chromium (Cr)	135
Iron (Fe)	160
Manganese (Mn)	32
Nickel (Ni)	105
Total Uranium (U)	770

*Insoluble neutron absorbers.

3.2.1 Applicability

The criticality safety program applies to DSTs, Aging Waste Facility (AWF) tanks, DCRTs, catch tanks, and 244-AR Vault that would receive transfers from a non-tank farm facility (i.e., transfers from outside the tank farms into the tank farms). This includes all waste transfers made through direct transfer piping systems, or received by bulk via the 204-AR Waste Unloading Facility. In addition, the criticality safety program applies to MISFs for which no new waste will be received. The IMUSTs are included in the MISFs.

The applicability statement for the TSR focuses on transfers from outside tank farms and does not mention SSTs. In fact, the requirement to maintain a criticality safety program applies to all operations in tank farms, including those associated with SSTs.

The limiting conditions for transfers provided in key element "d" do not apply to waste transfers to or from the 242-A Evaporator facility during an evaporation campaign, or to any transfers made between tank farm facilities. These conditions also do not apply to caustic or water additions to tank farm facilities. The pH condition, as stated in key element "d," does apply to chemical shipments received from non-tank farm facilities or bulk shipment.

Safety is based upon ensuring that waste is sent to tank storage with a large margin of subcriticality. Inside a storage tank, subcriticality is maintained by characteristics inherent to the waste, and these characteristics are not altered when waste is transferred between storage tanks. Characteristics of tank waste that ensure continued subcriticality include: (1) a small particle size that inhibits gravity segregation; (2) a tendency to agglomerate; (3) a tendency for plutonium to sorb onto solids; (4) a large mass ratio of solids to plutonium; and (5) a low plutonium solubility.

3.3 DOUBLE CONTINGENCY PRINCIPLE

The basic criterion for criticality safety is the double contingency principle, stated as follows:

At least two unlikely, independent, and concurrent changes (contingencies) in processing and/or operating conditions must happen before criticality becomes possible.

A contingency is an unlikely action, event, or occurrence that causes a change in operating conditions such that a prescribed safe operating condition or limit is exceeded.

Usually, the double contingency principle requires two independent parameters be controlled to ensure subcriticality. However, at times subcriticality depends on control of only a single parameter. In that situation, two independent administrative controls are required to ensure that the single parameter limit is not violated. For example, when plutonium concentration is very low, criticality is not possible regardless of the waste composition, the degree of moderation, or the degree of reflection. In this case, only plutonium concentration need be limited, but at least two controls must be exercised to prevent violation of the concentration limit. In practice, plutonium concentration is controlled by requiring a minimum quantity of neutron absorbing solids and by requiring analytical verification of their presence. A discussion of contingencies is provided in Section 5.0.

Limits and controls are spelled out in a CPS used by personnel responsible for the discharge of waste. The bases for those limits and controls are provided in this CSER.

3.4 CRITICALITY SAFETY CRITERIA

Criteria used to ensure criticality safety at tank farms fall into three categories: (1) criteria for discharge of waste into tank farms; (2) criteria for sending waste through an evaporator; and (3) criteria for transferring waste between tanks.

3.4.1 Discharged Waste

The following criteria are used to develop specific limits and controls for waste discharged from generating facilities.

1. Discharged waste shall conform to one of the following limits.
 - a) The plutonium concentration shall be less than the minimum critical concentration for an infinite system times a safety factor.

or

- b) The mass ratio of neutron absorbers to plutonium shall be greater than the minimum mass ratio that can be made critical in an infinite system times a safety factor.
2. A safety factor shall be used to ensure that a critical parameter will not be exceeded. This safety factor will be large enough to compensate for uncertainties associated with a parameter and for the likelihood of exceeding the limit value.
3. Discharged waste shall be sufficiently well mixed to preclude localized violations of limits.
4. The pH of discharged waste will be controlled to ensure alkalinity.
5. Waste is maintained alkaline in tank storage.

3.4.2 Evaporators

Waste sent through an evaporator shall remain subcritical over the entire range of possible water content, including completely dry.

3.4.3 Transfers Between Tanks

Transfers between tanks shall meet the following criteria.

1. Transfer of supernatant liquid may be made between storage tanks.
2. Transfers of solids may be made between waste storage tanks, if covered by this CSER or a CSER specific to that operation.

3.5 MARGIN OF SAFETY

The operating limit for a parameter is derived from the subcritical limit on that parameter and a safety factor. The subcritical limit is the largest value that can be assured of being subcritical. The safety factor is a multiplier used to ensure an adequate margin of safety. The operating limit is obtained by multiplying the subcritical limit by the safety factor.

For limits that are maximum values, the safety factor is less than unity, while for limits which are minimum values, the safety factor is greater than unity. The safety factor shall take into account:

1. The range of possible changes in composition and distribution within the tank storage environment, including variations caused by transferring waste between tanks and mixing.
2. The possibility that drainable liquids, along with dissolved components, might be removed from the waste.

3. The range of possible violations of controls on discharged waste.

The safety factor applied to a subcritical concentration or subcritical mass limit will not be larger than 0.5, while the safety factor applied to a subcritical component/plutonium mass ratio limit will not be less than 2.0. In practice, this safety margin is usually increased by 10% to ensure that doubling a limit quantity cannot result in criticality.

3.5.1 Discussion

Before criticality is possible, one of two scenarios is required. The first scenario is a single violation that exceeds the operating limit by a large factor (i.e., much greater than 2). The RPP criticality safety program and also the criticality safety programs of waste generators provide assurance that discharge of a single batch capable of criticality is essentially impossible. The second scenario is for a limit to be exceeded repeatedly by a relatively small factor over many waste discharges.

The margin of safety for tank waste is provided by a large mass ratio of neutron absorbing material to plutonium. Normally, a single batch of waste contains only a small quantity of plutonium, and its effect on the margin of subcriticality is small. A low neutron absorber content in one discharge will not significantly affect criticality safety, and the effect of this lack of neutron absorbers will be offset by a high content of neutron absorbers in another batch. For this reason, the absorber/plutonium mass ratio for an individual discharge need not be as large as that required for the entire contents of a tank. In the course of many discharges, individual batches will mix together and mitigate the impact of occasional batches with a low absorber content.

Waste is discharged with a large enough margin of safety to ensure subcriticality over the range of possible variations in composition. The complexity of tank waste makes it difficult to precisely assess the margin of subcriticality. Waste transfers and mixing change the distribution of components. When several different waste streams are sent to a tank, layers of distinctly different composition may form. Waste composition may vary both vertically and horizontally.

Waste characterization studies are used to verify waste composition. However, the volume of waste analyzed in characterization samples represents only an extremely small fraction of the total volume of waste. The uncertainty associated with such a small fraction of analyzed waste makes it necessary to use conservative assumptions when modeling waste for criticality safety evaluation.

When a discharge of waste is made to tank farms, the criticality safety program of the facility sending the waste and the tank farms criticality safety program work together to provide high assurance that no CPS limit is exceeded. In the unlikely event that a limit is exceeded, the margin created by the safety factor provides assurance that the subcritical limit will not be exceeded. In reality, the true margin of subcriticality is considerably greater than that provided by the safety factor. The safety factor is intended to prevent violation of the subcritical limit, but the subcritical limit itself has an additional safety margin derived from conservative assumptions made in its derivation.

If newly discharged waste is not chemically compatible, a chemical explosion might occur. Although it is important that an explosion be prevented, an explosion would not produce a waste configuration capable of criticality. No criterion for chemical compatibility or for flammable gas control is required for criticality safety.

This page intentionally left blank.

4.0 METHODOLOGY

This CSER provides descriptive information drawn from published studies of tank waste. Parameters are identified that are to be controlled to ensure that the waste remains subcritical into the indefinite future under all credible operating and accident conditions. A discussion is provided of barriers that maintain the basic safety parameters within acceptable limits.

Section 2.0 provides a description of tank farm facilities. This is intended to identify and describe locations where fissionable material may be found.

Specific limits are provided for the control of waste coming into the tank storage system. An evaluation of these limits is made to demonstrate that a high degree of subcriticality will be maintained after this waste is received. A surveillance program is prescribed to monitor the storage tanks to verify that the waste is well subcritical.

Since the present condition of tank waste is determined by past history, a review of criticality safety limits and controls that were used in CPSs and in facility operating procedures in the past is provided in Appendix A.

No calculations were performed in the process of writing this report. For this reason, there are no descriptions of criticality codes or their validation. However, many parameters have been developed in other documents that are important to this evaluation. Appendix B, "Parameters Important to Criticality Safety," contains a compilation of these parameters and provides information on their basis.

Sources of characterization data are described, and a summary of waste streams is provided, in Appendix C. Emphasis is given to information and parameters most relevant to criticality safety.

Within the tank waste environment physical and chemical processes capable of changing uranium and/or plutonium concentrations are of concern. Appendix D summarizes published studies of physical and chemical processes of importance to criticality safety. No new chemistry information is developed in this evaluation report.

A survey of anticipated waste sources is provided in Appendix E, based on estimates made in 1998. The information provided is descriptive and intended to help understand the need of the tank farms criticality safety program.

Appendix G describes the process of evaluating new operations and outlines a simplified approach to evaluating activities that may disturb the settled waste solids.

This page intentionally left blank.

5.0 DISCUSSION OF CONTINGENCIES

A contingency is a possible, but unlikely, change in a condition or control that would, if it occurred, reduce the number of barriers that prevent a nuclear criticality accident. This section discusses contingencies required for criticality safety in tank farms and the barriers that prevent them.

Criticality in a waste storage tank would be the culmination of a series of implausible events, including repeated process failures and violation of controls and procedures. Table 5-1 provides a list of contingencies and the associated barriers. Each contingency has several barriers associated with it.

The key elements of the criticality safety program are the barriers that prevent a contingency. A major problem with establishing clearly defined barriers is the high level of complexity surrounding the creation and storage of tank waste. In the past, "control failures," rather than contingencies, were used in the evaluation of criticality safety. A control can be precisely defined, and its failure can be identified. Due to the processes by which tank waste is generated, no credible single failure of a control is a contingency. In fact, a contingency requires multiple control failures. For example, criticality might occur if a storage tank were filled with dry waste containing several grams of plutonium per liter. However, to achieve this configuration would require dozens, and perhaps hundreds, of repeated violations of one or more controls.

A survey of Hanford Site waste generators is provided in the FSAR (LMHC 1999a). Only PFP was determined to have a sufficient quantity of plutonium to cause concern that a criticality might result at tank farms. T-Plant possesses plutonium, but the quantity and concentration are insufficient to be of concern. Other Hanford Site processing plants (B-Plant, PUREX, Reduction Oxidation Plant (REDOX), bismuth-phosphate, and uranium recovery) are shut down.

Chemistry is important to criticality safety. Chemical processes are a determining factor in the ability of waste components to be rearranged, removed, or concentrated. These processes are capable of altering the probability of continued subcriticality. Knowledge of the chemical processes in tank waste increases the confidence that neutron absorbing waste components will not separate from the plutonium. A detailed discussion of tank waste chemistry is provided in Appendix D.

Table 5-1. Contingencies and Barriers

Contingency		Barriers
No.	Description	
1	Absorber/plutonium mass ratio	<p>Process controls at generator.</p> <p>CPS limits and controls at generator.</p> <p>Transfer data sheets filled out and verified.</p> <p>Limited availability of separated plutonium.</p> <p>Requirement that solids/plutonium exceed 5,000 when plutonium inventory exceeds 10 kg.</p>
2	Distribution of components	<p>Particle size distribution.</p> <p>Agglomeration of solids.</p> <p>Waste alkaline (pH > 8).</p> <p>Mixing before discharge.</p> <p>Formation of layers (fluidity).</p> <p>Average plutonium concentration in incoming batch.</p> <p>Maximum plutonium in incoming batch.</p>
3	²³⁵ U Enrichment	<p>Little uranium available with enrichment > 1.25 wt%</p> <p>Most uranium irradiated (reduced ²³⁵U content).</p>
4	Plutonium in soil or in DST annulus	<p>Containment shell.</p> <p>Low solubility of plutonium in supernatant liquid.</p> <p>Low solids content in leaking liquid.</p> <p>Filtering of particulate by soil near source of leak.</p>
5	Deliberate discharge of excessive quantity of plutonium	<p>Difficulty of entry into tank.</p> <p>Special Nuclear Materials (SNM) Accountability.</p>

5.1 CONTROL FAILURES

The composition of tank waste is primarily the product of discharges made when processing plants were in full operation. The quantity of plutonium now in tank storage is far greater than the quantity expected to arrive in the future. Although many processing facilities are now shut down, the PFP is still in operation. This section is written in the present tense, but it also applies to operations during the period in which most waste was received.

Several independent events of low probability are required to create a critical configuration.

5.1.1 Holdup Tanks

Waste generated at a processing facility is accumulated in a "holdup tank" until discharged. Typically, a holdup tank is cylindrical with a volume of about 18,900 L (5,000 gal). Although small when compared to a DST or a SST, the diameter of a holdup tank is large from the perspective of criticality safety. For this reason, holdup tanks, and the processes producing waste, are carefully monitored to ensure compliance to CPS limits. Although a low plutonium concentration and an adequate quantity of solids are produced under normal operating conditions, the operating personnel are always on guard against off-normal (upset) conditions that might send larger quantities of plutonium to the waste stream. Criticality safety at tank farms is related to how well waste holdup tanks are controlled. Controls are placed on holdup tanks to ensure that waste discharged to tank farms is inherently safe.

The following discussion looks at hypothetical worst case discharges that might have been made at anytime since 1945. No criticality has ever occurred within a holdup tank. If criticality had occurred, the waste would not have been discharged. This fact places an upper limit envelope around plutonium quantities and concentrations in discharges to tank farms.

5.1.2 Breakdown of Controls

Most waste at tank farms originated from processes governed by flowsheets for which every stage was carefully monitored and controlled. An abnormal condition of the waste would be reflected in an abnormal product. A drop in the quantity or purity of the product, plutonium would have been recognized, and an investigation would have been undertaken. Multiple violations of process controls must occur before discharge of a nearly critical batch of waste is possible.

No records of past abnormal discharges have been found. It is extremely unlikely that a highly abnormal condition could pass undetected. It is difficult to explain how a large quantity of plutonium, an extremely valuable material and the object of the entire production process, could have been improperly routed to waste without detection.

The possibility is considered that separated plutonium might be accidentally routed to the holdup tank. Hypothetically, one might assume that an 18,900-L (5,000-gal) holdup tank is filled with waste containing plutonium at a concentration of 4 g/L. This concentration is hundreds of times greater than expected. The total quantity of plutonium would then be 75 kg, the quantity normally found in several million liters of waste.

When discharged into the 22.9-m (75-ft) diameter storage tank, this waste forms a layer less than 5 cm (2 in.) thick. The areal density would be only 0.18 kg/m² (17 g/ft²), as compared to the minimum critical areal density of 2.6 kg/m² (240 g/ft²) for plutonium in water. At least 14 additional identical discharges are required to reach the minimum critical areal density, and neutron absorbers would still ensure subcriticality.

5.1.3 Special Case

One special case, however, needs closer examination. When incoming waste falls into deep supernatant liquid, it is possible for it to spread into a quasi-spherical or cylindrical volume. Criticality becomes possible when the plutonium concentration exceeds 7 g/L (26.5 g/gal). The potential for criticality exists while the discharged plutonium is suspended in the supernatant liquid. When the settling solids reach the surface of the sludge, they will spread out into a thin layer on top of the already settled solids.

Because incoming waste is diluted with supernatant, the plutonium concentration upon entering the storage tank must exceed 7 g/L, if criticality is to be possible. Since pumping over a long distance tends to disperse the plutonium, the concentration before discharge would have to be even greater, probably twice the minimum critical concentration. In addition, the quantity of plutonium must be large enough to permit criticality. The holdup tank is stirred prior to discharge, and criticality would be expected to occur in that tank. For this discharge to be made, at least three independent CPS violations would be required: exceeding the batch limit; failure to provide neutron absorbing solids; and failure to stir.

The CPS limit of 0.04 g Pu/L for waste received at tank farms would have to be exceeded by a factor of more than 200 for criticality to be possible. At the same time, the quantity of plutonium at this extremely elevated concentration would have to exceed 520 g. Normally, the quantity of plutonium in a holdup tank is less than 50 g. Multiple undetected errors would be required to exceed a critical quantity.

Waste is discharged with an adequate quantity of solids to ensure subcriticality. Usually the quantity of solids far exceeds that required to ensure subcriticality. Even a low quantity of neutron absorbing solids, however, would increase both the critical plutonium concentration and critical mass.

5.1.4 Hypothetical Accident

To assess the consequences of a hypothetical criticality accident it was necessary to construct a scenario that might lead to criticality. A scenario identified as the "most reasonable" configuration that might result in criticality is described in Appendix F. The initiating waste transfer was chosen to be 500 L (132 gal) of a 10 g Pu/L solution pumped at a rate of 4.7 L/s.

Even though this was determined as a “most reasonable” scenario, it was also considered to be “beyond extremely unlikely.” Multiple running failures would have to occur without being identified and without interruption by other operations.

5.1.5 Conclusion

If waste is subcritical in a holdup tank, the probability that its discharge could achieve criticality in a storage tank is extremely low. Regardless of the initial plutonium concentration, discharged waste would spread out in the storage tank, and the resulting plutonium areal density would be small. If criticality does not occur immediately upon entering tank storage, the probability that it would occur later is very small. Even the discharge of multiple, highly abnormal batches of waste would be unlikely to create a configuration approaching criticality in a storage tank. In conclusion, creation of a region near critical in a waste storage tank would require repeated failures of process controls, and the probability that such a region exists is very small.

5.2 TRANSFERS FROM PLUTONIUM FINISHING PLANT (PFP)

Plutonium Finishing Plant (PFP) is the only fuel processing plant still sending waste to tank farms, and a detailed discussion of the limits and controls used at PFP is now provided. It is recognized that projects at PFP are changing, and operations are being shut down. The following description is not expected to remain current, and some described operations are at present no longer in use. However, knowledge of past operation is important to understanding the present state of tank waste, and this section is written as much to understand the past as to anticipate the future.

The present state of tank waste is the product of waste transfers made over many years. Although theoretically possible, it is highly unlikely that a control failure when PFP was in full production might have resulted in an “off-normal” discharge of waste to tank farms. For a potential for a future criticality to exist, the deviation from normal of a past discharge (or series of discharges) had to be large. No record of any such failure exists. For such large control failures to go undetected is extremely unlikely, if not impossible. Reviews of the present state of stored waste and the history of waste discharges have been published, and these studies concluded that tank waste is well subcritical (Braun et al. 1994; Bratzel et al. 1996). In recent years, PFP has made only infrequent transfers of waste to tank farms, and transfers in the future will continue to be infrequent. The possible impacts of control failures in the future are now examined.

Plutonium Finishing Plant documents use the terms, receiver tank, sample tank, and holdup tank interchangeably. For this discussion, “holdup tank” is used throughout.

The FSAR (LMHC 1999a) identifies four waste streams as having some probability of operation in the future:

- **Analytical laboratory waste** -- Analytical laboratory waste is generated in batches typically 30 to 100 mL in volume, and these are collected into 4-L (1-gal) batches. In a transparent 4-L bottle, the liquid color is a sensitive visual indicator of plutonium concentration. A high plutonium concentration is seen as green, and this color darkens with higher plutonium content. Wastes containing a high concentration of plutonium are stored and then returned to the originating process. Waste containing only low concentrations of plutonium is taken to a drain leading to holdup tank TK-D8. Prior to disposal, waste is sampled and analyzed, and the results are reviewed and approved by Process Engineering (i.e., the 241-Z Cognizant Engineer), the Environmental Compliance Officer, and the Operations Manager. Upon final approval, the waste is transferred to holdup tank TK-D8.
- **Vertical calciner waste** -- Vertical calciner waste comes from the offgas system and contains negligible plutonium, even after a process upset. This waste is gathered into batches whose volume is between 15 and 30 L.
- **Remote Mechanical "C" (RMC) ion exchange filtrate** -- The filtrate from the ion exchange process contains the wash solution from the RMC line. Plutonium is eluted (removed using a solvent) from the column before washing, and the wash solution contains minimal plutonium. Only an undetected process upset during elution could result in plutonium remaining in the column and being removed in the wash solution to become part of the waste. The waste is sent to a separate holding tank.

Operations and the nuclear materials control group would identify a loss of plutonium in the washing process rather than in the elution process. Remote Mechanical "C" line waste is sampled, and the analysis results are reviewed and approved by the shift manager before transfer to holdup tank TK-D8.

- **Persulfate process waste** -- The feed to the persulfate process is ash containing low concentrations of plutonium. Waste comes from the caustic wash step. The wash solution is filtered to remove any ash carried over during the wash. The waste is stored in a waste holdup tank.

5.2.1 Individual Control Failures

Only one or two discrete transfers of waste to tank farms per year are likely in the foreseeable future, and these will pass through holdup tank TK-D5 in the 241-Z Building. Procedures and evaluation reports for this facility relevant to criticality safety are:

- ZO-101-010, *Transfer TK-D5 to Tank Farms* (B&W 1999b)
- ZO-101-019, *Transfer TK-D7 Solutions to TK-D5* (B&W 1996)
- ZO-101-020, *Transfer TK-D8 Contents to TK-D5* (B&W 1997a)
- ZO-101-023, *Sample Tank D-4, D-5, D-7, or D-8* (B&W 1997b)

- CPS-Z-165-80741, *241-Z Solution Disposal Facility* (B&W 1999a)
- OSD-Z-184-00010, *Operating Specifications for 241-Z Waste Facility* (B&W 1997c)
- HNF-SD-CP-OCD-036, *OSD-Z-184-00010 Supporting Document* (Funston and Meng 1997)
- WHC-SD-SQA-CSA-20213, *CSAR 79-044: 241-Z Solution Disposal Facility* (Chiao 1980)

CPS-Z-165-80741 (B&W 1999a) limits the total plutonium inventory in any 241-Z tank to a maximum of 400 g. The plutonium concentration in solutions transferred to tank farms has been limited in the past to 0.033 g/L (0.125 g/gal). This CSER provides the basis for increasing this limit to 0.04 g/L for future waste batches sent to tank farms. A running inventory is maintained for holdup tank TK-D5 using the Transfer Data Sheet (TDS) which records the specific gravity, volume, and plutonium concentration for each addition. This information is used to calculate the final plutonium concentration for transfers of waste to DCRT 244-TX.

Procedure OSD-Z-184-00010 (B&W 1997c) provides operating specifications for the 241-Z Building, including criticality safety limits required by CPS-Z-165-80741 (B&W 1999a) for PFP and CPS-T-149-00010 (LMHC 1998c) for tank farms. This procedure requires that:

- Plutonium concentration is 0.033 g/L (maximum). This limit ensures a total plutonium mass less than 400 g in holdup tank TK-D5.
- Solids content is between 1 to 3 vol% as determined by centrifuge analysis.
- Nitrate, nitrite, and hydroxide concentrations ensure conformance to pH limit.
- Tank agitator is run for at least 10 minutes before sampling.
- There is no separable organic phase in liquid transferred.
- Sample line is recirculated for at least 20 minutes before a sample is drawn.

The maximum plutonium concentration permitted in waste received at tank farms is 0.04 g/L, and the minimum solids content permitted in transfer to DCRT 244-TX is 0.7 vol% solids. The probability of violating tank farms limits on maximum plutonium concentration, minimum solids content, or particle size is now discussed.

5.2.1.1 Exceeding Criticality Prevention Specification (CPS) Concentration Limit

Plutonium concentration is determined by dividing the mass of plutonium in a batch by the total volume of solution. The holdup tank TK-D5 inventory is found by summing the plutonium contributions from individual Transfer Data Sheets (TDS). The volume of solution is determined from weight factor instrumentation and an average specific gravity. The average specific gravity is calculated from specific gravity and volume entries on the individual TDSs. A transfer in

which the plutonium concentration actually exceeds the limit would require one of the following errors: (1) a recorded plutonium inventory smaller than actual; (2) a calculated volume larger than actual; or (3) a math error in dividing mass by volume. The probability of one, or more, of these errors occurring and violating the concentration limit is small. A conservative estimate is that such an error might occur once in 10 to 20 transfers.

5.2.1.2 Sending Insufficient Solids

A smaller than actual solids volume would require one of the following errors: (1) failure to agitate sufficiently (bad sample); (2) failure to recirculate sample line (bad sample); (3) analytical error in determining solids content; or (4) recording error. Again, a conservative estimate is that such an error might occur once in 10 to 20 transfers.

5.2.1.3 Particle Size Larger than Expected

Waste is received into the 241-Z Building in solution, and the particle size for this waste is very small. Plutonium entering the waste stream passes through filters that provide added assurance of a small particle size. Waste placed in DSTs is expected to have a large weight fraction of primary particles less than 10 μm in diameter, and this criterion is met for 241-Z waste under normal operating conditions. Larger particles would indicate failure of a filter or an upset in operating conditions. Either condition is unlikely, and the upset condition would have to persist for a long time before a significant quantity of plutonium would accumulate in holdup tank TK-D5.

5.2.2 Control Failures Required For Criticality

All areas within PFP must comply with strict material balance controls administered by the nuclear material control group. Waste streams are sampled prior to approval for transfer to holdup tank TK-D8.

The waste stream from the analytical laboratories has the highest probability of accumulating enough plutonium to be of concern. Nevertheless, an accumulation of several hundred liters of solution with several kilograms of plutonium is required for criticality, and this represents the combined plutonium inventory of hundreds of 4-L waste containers. The following repeated failures would be required:

- Failure by the Nuclear Materials Control group to identify the loss of inventory in material balance reports.
- Failure to recognize that the quantity of plutonium was close to or over the Operational Safety Requirements (OSRs) for inventory control.
- Failure of laboratory personnel to notice the green to dark color of the solution (waste containers are transparent, and typical waste is colorless).

- The Process Engineer, the Operations Manager, and the Environmental Control Officer must fail to notice the high plutonium quantity entered on 4-L laboratory waste containers. Operators must enter an incorrect low concentration, rather than the correct high concentration, on numerous containers.
- Operators must incorrectly record the daily running laboratory waste inventory totals for many 4-L bottles released to the waste receiver tank.
- Failure on many occasions to send high plutonium samples back for reprocessing. In addition, failure of the process group to notice that samples had not been returned as anticipated.

Remote Mechanical "C" (RMC) ion exchange filtrate has a lower probability than analytical laboratory waste of sending enough plutonium to holdup tank TK-D8 to be of criticality safety concern. Repeated process upsets would be required where plutonium is not eluted from the column prior to wash. The following repeated failures would be required for this accumulation to occur:

- Operators would not notice the process upsets.
- Nuclear materials control would not notice the loss of material.
- Equipment failures, operator errors, or management errors would have to occur on each sample of RMC waste from the process upset, such that discharge of a large quantity of plutonium was not recognized.
- No significant dilution in holdup tank TK-D8 from other streams can occur during the time of multiple process upsets on the RMC line.

A transfer of waste to tank farms requires completion of a waste manifest to satisfy the State of Washington Department of Ecology and other regulators. This includes a record of the transfer, including completion of a TDS. Included in the record are the water content, concentration of hydroxide, nitrite, nitrate, percent solids, and the iron/plutonium mass ratio.

For a criticality to occur in a DST from a PFP transfer, there must be a failure to sample and record the transfer, and the mass and concentration of the plutonium must be far greater than normal. At the same time, the waste must have maintained a subcritical configuration while in holdup tank TK-D8, while in DCRT TX-244, and through the process of being transferred to tank farms.

5.3 PLUTONIUM DISTRIBUTION

The effect of plutonium distribution and variations in waste composition on critical parameters is now examined. Minimum critical parameters derived from the CWM are shown to remain conservative, after waste streams have been mixed.

The minimum critical plutonium concentration is 2.6 g/L, and the minimum critical areal density is 2,582 g/m² (240 g/ft²) (Rogers et al. 1996). When plutonium is assumed uniformly distributed, the minimum plutonium mass required for criticality in a 22.9-m (75-ft) diameter tank is 1,060 kg. Roetman et al. (1994) determined the total quantity of plutonium in tank farms to be less than 981 kg. If the entire tank farms plutonium inventory were to be uniformly distributed in a single tank, the tank would remain subcritical.

5.3.1 Layers Having Different Compositions

Waste in a storage tank may be derived from several waste streams, each forming a distinct layer.

When plutonium is homogeneously distributed over a large volume, the minimum critical concentration is small, and the minimum critical mass is large. When the distribution of waste components is not uniform, the conditions under which criticality is possible become more complex. A configuration optimized to produce the smallest possible critical mass will have a plutonium concentration several times larger than the minimum critical concentration for a homogeneous system. For example, the minimum critical plutonium concentration in a large, homogenized volume is about 7.2 g/L, and the critical mass at this concentration is infinite. However, when the plutonium concentration is 30 g/L, the minimum critical plutonium mass drops to 520 g, and the critical volume becomes only 17 L. This is the smallest possible critical mass, but the plutonium concentration is 4 times greater.

The minimum critical plutonium concentration of 2.6 g/L occurs when the configuration is homogeneous. No matter the degree of heterogeneity (non-uniformity), criticality is not possible unless the plutonium concentration exceeds this value over a sizeable volume.

Waste was sent to tank storage in a series of waste streams, each comprised of a large number of batches. Under idealized conditions, the solids in a waste stream form a single layer of more or less uniform composition and thickness on top of the existing sludge. In the absence of waste disturbing activities, the sludge is composed of a series of relatively uniform layers.

The macroscopic (total) neutron absorption cross section for a kilogram of conservative waste model (CWM) solids is less than that for a kilogram of actual waste taken from any waste stream. If CWM solids replace each layer, then each layer becomes more conservative. In this way, CWM solids can replace all sludge in a tank, and the resultant configuration will be more conservative than the original combination of waste types. It is conservative to replace the entire contents of a storage tank by the CWM.

Criticality is possible in the CWM only by increasing the plutonium concentration above 2.6 g/L. This minimum critical plutonium concentration is conservative when applied to actual sludge, regardless of the number of layers of varying composition there are within the sludge layer.

5.3.2 Mixing

Although each waste stream is different, each can be replaced by the CWM, and the resulting waste is conservative when compared to the original. Every small volume within the sludge contains waste with a macroscopic neutron cross section larger than the CWM. When different waste types are mixed, regions of higher and lower macroscopic neutron absorption cross sections blend to produce a more uniform composition. The macroscopic neutron absorption cross section for the mixture will be the weighted-average of the original sludge types. This weighted-average will be larger than the smallest of the original values, and that value was larger than that of the CWM.

In conclusion, the macroscopic neutron absorption cross section of settled solids taken from any location in the sludge will exceed that of the CWM, regardless of the degree of mixing. After mixing, the minimum critical plutonium concentration will not be less than that of the CWM, or 2.6 g/L.

5.4 LEAKAGE OF SUPERNATE INTO GROUND

The possibility of criticality following a leak of liquid waste into the ground is examined. Leakage from a storage tank is compared to leakage into a saltwell, and a discussion is provided of the quantity and distribution of plutonium required to create a critical configuration in the soil.

The second containment shell of a DST is designed to ensure that leakage from the inner wall will be contained within the outer wall. No liquid can leak from a DST into the soil.

5.4.1 Comparison to Saltwell Pumping

A saltwell is a perforated steel pipe inserted vertically to the bottom of the sludge to allow liquid to drain from the sludge. Collected liquid is then pumped out of the tank. The dynamics of liquid waste draining through a crack at the bottom of a storage tank is the same as liquid draining into a saltwell.

When liquid drains from sludge into a saltwell, there is initially some solids contained in the liquid, but the amount quickly decreases (see Appendix G5.0 for more details). Bratzel et al. (1996) states that "the bulk solids surrounding the saltwell pipe begin to act as a filter media to prevent additional finely divided solids from migrating into the well." Bratzel et al. (1996) concludes that "saltwell pumping is a localized and benign activity that does not cause significant separation and/or concentration of solid materials."

Serne et al. (1996) studied the effects of saltwell pumping on plutonium distribution. Waste was assumed to have a 30% porosity, a 12 cm^2 permeability, a 1.4 g/cm^3 density, and a 12.5 cp viscosity. A pump rate of 7.6 L/min (2 gal/min) was assumed. The maximum flow velocity was found to be 0.002 m/s at the edge of the saltwell, and the velocity dropped off rapidly with distance from the pump. This is much less than the velocity required to suspend plutonium, and redistribution is of little concern. In conclusion, the quantity of solids removed through saltwell pumping is too small to be of concern.

Only a small quantity of solids would be transported from the tank, and these solids would be inhibited from further transport by filtering by the soil itself. The quantity of plutonium reaching the soil would be small, and this plutonium would penetrate only a short distance into the soil. The quantity of plutonium that penetrates more than 30 cm into the soil would be negligible.

5.4.2 Critical Plutonium Concentration in Hanford Soil

In the unlikely event that liquid waste drains into soil, criticality would be highly unlikely. The quantity of plutonium required to achieve criticality would far exceed the upper limit of the quantity likely to be transported into the soil. An assessment is now made of the critical parameters for plutonium in soil.

Ridgway and Carter (1972) calculated critical parameters for plutonium mixed with soils with two different void fractions: 30 and 40 vol%. They assumed a dry Hanford soil with a density of 2.43 g/cm³. The primary constituents of this soil were SiO₂ (81.0 wt%), Al₂O₃ (6.0 wt%), CaO (4.0 wt%), Fe₂O₃ (2.0 wt%), MgO (2.0 wt%), K₂O (1.0 wt%), and Na₂O (1.0 wt%). The plutonium was assumed to contain 3 wt% ²⁴⁰Pu. Calculations were made for fully saturated soil for both void fractions. The minimum critical concentration is reported as 2 g Pu/L when the water content is only a small volume percent.

The Ridgway and Carter (1972) critical parameters were determined for a specific soil composition not proven to be conservative for other soil types likely to be encountered. For this reason, new parameters were determined using a soil model that is conservative in comparison to any soil type.

5.4.3 Conservative Model of Soil

Silicon dioxide is a major component of sandy soil and one of the least neutron- absorbing materials occurring in nature. The macroscopic neutron absorption cross section for silicon dioxide is less than that of actual soil composition. The minimum concentration of plutonium that can be made critical mixed with silicon dioxide is smaller than the minimum that can be made critical in soil of any composition. A model that assumes soil is composed entirely of silicon dioxide is conservative in comparison to any Hanford soil types and is more conservative than the CWM.

Sanchez et al. (1996) provides a detailed study of critical parameters for plutonium-silicon dioxide-water systems. The minimum critical plutonium concentration in silicon dioxide is 1 g/L and occurs when completely dry. Altschuler and Doto (1991) provide a more conservative minimum critical plutonium concentration of 0.8 g/L. When an Upper Safety Limit (USL) on k_{∞} of 0.95 at a 95% confidence level is used, the corresponding USL on plutonium concentration becomes 0.7 g Pu/L, based on Altschuler and Doto (1991).

The minimum critical plutonium concentration in silicon dioxide sand occurs when the sand contains no water. The neutron absorption in water exceeds that for silicon dioxide, and addition of any water, or any other naturally occurring soil component, increases neutron absorption. The silicon dioxide soil model is conservative when compared to any real soil.

5.4.4 Minimum Critical Plutonium Mass in Soil

Plutonium concentration alone does not determine criticality. The quantity of plutonium must also be large enough. In systems of plutonium, water, and silicon dioxide, the minimum critical plutonium mass is achieved when the water content is high and the silicon dioxide content is low. Addition of silicon dioxide dilutes the water and reduces the degree of moderation. This increases the mass of plutonium, and also the volume, required for criticality.

In dry silicon dioxide, the plutonium minimum critical mass is about 36.5 kg. This requires a plutonium concentration of about 3.1 g/L in a volume of 11,700 L (Sanchez et al. 1996). The corresponding Si/²³⁹Pu atom ratio is about 2,812. When the Si/²³⁹Pu atom ratio is held constant at 2,812 and the water content is increased to 4.5 wt%, the minimum plutonium critical mass drops to 11.0 kg. Criticality then will occur at a concentration of 2.8 g Pu/L in a volume of 3,910 L.

If the Si/²³⁹Pu atom ratio is reduced to 752, the dry plutonium critical mass increases to 47.2 kg, and this occurs at 11.6 g Pu/L in a volume of 4,060 L. When the Si/²³⁹Pu atom ratio is maintained at 752, the addition of water will lower the plutonium critical mass until it reaches a minimum of 2.0 kg at 7.5 g Pu/L in a volume of 273 L (Sanchez et al. 1996).

If tank waste is replaced with silicon dioxide, the minimum critical concentration will not fall below the CWM minimum of 2.6 g/L until the Si/²³⁹Pu atom ratio is about 2,812. The critical mass will then be at least 11.0 kg of plutonium and will require a volume of 3,910 L. To achieve criticality at a lower plutonium concentration, the mass of plutonium and the volume required both become larger. For criticality to occur when the plutonium concentration is 1.3 g/L requires a sphere whose volume is 162,000 L (i.e., very large). This sphere would be composed entirely of silicon dioxide, and the mass of plutonium would be greater than 209 kg. This is more than twice the largest plutonium inventory in any storage tank.

5.4.5 Critical Slab Thickness

The critical thickness of a slab of plutonium is smallest when the water content is high. The minimum critical areal density of 2.6 kg Pu/m² (240 g Pu/ft²) for plutonium in pure water is conservative when applied to configurations of plutonium in soil.

At the subcritical limit on plutonium concentration in silicon dioxide of 0.7 g/L, the critical slab thickness is infinite. For criticality to occur at the minimum critical areal density of 2.6 kg Pu/m² that applies for the CWM, the slab thickness in silicon dioxide would have to exceed 1 m (a conservative estimate). For any credible soil composition the critical slab thickness would exceed 2 m, when the plutonium concentration is less than 2.6 g/L.

5.4.6 Conclusion

A leak would carry only a small quantity of solids out of a tank, and these solids would contain only a small proportion of plutonium. Some additional plutonium would be dissolved at a low concentration in the supernatant liquid. Regardless of the actual volume of leakage, however, the total quantity of plutonium reaching the soil would be small, and the quantity that would penetrate more than 30 cm would be much smaller yet (see Sections 5.4.1 and Appendix G5.0).

It would be extremely unlikely for the plutonium concentration outside of the tank to reach 1 g/L at any location. For criticality to occur, the plutonium concentration would have to exceed 1 g/L over a large volume. It is not possible for a leak to result in criticality in the soil, assuming conditions that now exist in tank storage.

5.5 WASTE GENERATORS CRITICALITY SAFETY AUDIT PROGRAM

From the beginning of nuclear fuel production in the 1940's, all nuclear facilities have had a criticality safety program. An important part of a facility's criticality safety program has been surveillance by operators and periodic audits by the operating group and by an independent safety organization.

Removal of plutonium from nuclear fuel was a complex process requiring precision. The majority of waste in the tank farms was generated at the Plutonium-Uranium Extraction (PUREX) Plant. Criticality safety was maintained through adherence to a detailed description of the processes. Every step of the process was thoroughly understood, carefully planned, and monitored to ensure conformance to the process flowsheet. A careful overview was maintained to ensure a pure product and to prevent accidents. Precise control of the processes not only produced high quality reactor fuel, but also resulted in a well defined waste product having a high neutron absorber content in relationship to the quantity of plutonium.

In 1961 Tomlinson (1961) reviewed the safety of manufacturing and chemical processing facilities at Hanford. His report demonstrates that an active criticality safety program was conducted during the period when waste was actively sent to tank farms. Tomlinson (1961) described the formal safety audits and reviews as follows:

Engineers involved with process technology, but not having line responsibilities for plant operation, have assigned responsibilities to perform routine audits of the plant process operations, thereby insuring operation within the limits defined by the process specifications. In addition, nuclear safety practices are formally reviewed once per year by a Senior Engineer specializing in nuclear safety and twice a year by a qualified engineer within the Technology Operation.

Plutonium discharged to waste storage tanks has always been combined with a large proportion of neutron absorbing solids. Failure to meet the required limits would have required a breakdown in procedural control. The level of control provided by the criticality safety program, using both administrative procedures and engineered controls, makes it highly unlikely that an off-normal condition occurred in past operations without detection. If a violation were detected, the procedures required that operations be stopped until the condition was corrected. No waste would have been discharged until after all conditions were brought into conformance with CPS limits. It is unlikely that even a single batch of waste with an excessive quantity of plutonium was discharged without detection, and it is extremely unlikely that multiple discharges of off-normal batches were ever made.

An effort was made to locate records of violations of criticality safety specifications applying to discharge of waste from processing plants, but none were found. Although this does not prove that no noncompliant discharges were ever made, all of the people interviewed agreed that the number of such discharges must have been either very low or nonexistent. Throughout the examination of characterization records, the studies of chemistry, and the review of discharge records, the possibility was considered that noncompliant discharges might have occurred in the past. No evidence was found for any. It is worth noting, nevertheless, that the process of transferring a batch of waste to tank farms has a very strong tendency to mitigate any hypothetical criticality safety irregularity by diluting the nonconforming batch with a large volume of other waste.

Tank farms has a program of periodic criticality safety audits of facilities. However, since tank waste is not directly observable, these audits are not able to directly determine compliance to in-tank criticality safety limits. These audits are primarily concerned with programmatic issues. Assessment of the margin of subcriticality of the stored waste is accomplished through a program of surveillance.

This page intentionally left blank.

6.0 EVALUATION AND RESULTS

The bases are provided for limits and controls to be used in the Criticality Prevention Specification (CPS). Limits used in the past are discussed in Appendix A. Parameters important to criticality safety are discussed in Appendix B.

The "plutonium concentration," unless otherwise qualified, refers to the highest plutonium concentration that exists in any small subvolume (i.e., less than 1 L) of the waste. The "average plutonium concentration" refers to the total quantity of plutonium divided by the total volume of waste under consideration.

6.1 AVERAGE PLUTONIUM CONCENTRATION IN INCOMING WASTE

A limit is placed on the average plutonium concentration for a batch of incoming waste. The batch-averaged plutonium concentration is the total plutonium in a batch divided by the volume of the batch. This is a secondary limit that is set much lower than the subcritical limit concentration. Its purpose is to lower the probability that the maximum concentration in settled solids will ever exceed the "safe" concentration limit. When plutonium enters the tank at a low average concentration, it enters along with a large volume of highly fluid liquid that spreads out over the entire surface of the existing waste in the tank. Since the tank cross sectional area is large, this spreading of the waste is a dispersal mechanism that ensures a low areal density of plutonium. The primary limit to ensure subcriticality is the limit on the neutron absorber/plutonium mass ratio (see Section 6.3).

Before 1994, the limit on average plutonium concentration in incoming waste was 0.013 g/L (0.050 g/gal) for all tanks, except for two tanks using a discharge limit of 0.033 g/L (0.125 g/gal). In 1994 the CPS was changed to extend the larger limit of 0.033 g/L to all DSTs. This CSER provides justification to increase the limit on the batch-averaged plutonium concentration to 0.04 g/L (0.15 g/gal) for all tanks.

6.1.1 Basis for 0.04 g Pu/L Limit on Batch-Averaged Concentration

The following limit applies to a batch of waste entering tank storage from outside of tank farms:

The total quantity of plutonium in a batch of waste during discharge into a storage tank divided by the batch volume (i.e., the batch-averaged plutonium concentration) shall not exceed 0.04 g/L (0.15 g/gal).

This limit does not apply inside of a storage tank. Inside a storage tank, waste solids will settle into a thin layer on top of the existing sludge, and the plutonium concentration within the settled solids will be higher than the batch-averaged value. In fact, it might be an order of magnitude greater, or more, than the batch-averaged limit of 0.04 g/L.

The rationale for picking a limit of 0.04 g/L on the average plutonium concentration in an incoming batch of waste is:

- (1) The primary limit for criticality safety is the limit on the minimum solids-to-plutonium mass ratio. The limit on average plutonium concentration is a secondary limit whose violation cannot by itself lead to criticality. This limit is intended to mitigate a violation of the limit on the minimum solids-to-plutonium mass ratio, which is the primary protection against criticality. A low average plutonium concentration ensures that when the incoming plutonium will be spread more widely over the area of the storage tank. For each batch of waste, this ensures a plutonium areal density much smaller than the minimum critical areal density, regardless of the solids-to-plutonium mass ratio.
- (2) The actual limit on average plutonium concentration is arbitrary, so long as it is substantially lower than the minimum critical concentration. Historically, a limit of 0.013 g/L was used, but this value is sometimes difficult to ensure unless water is added for dilution. A higher limit of 0.04 g/L reduces the possibility of noncompliance and is, therefore, desirable.
- (3) For criticality to occur the plutonium concentration must increase from 0.04 g/L to more than 2.6 g/L. This is a factor of at least 65 before criticality is possible. Usually, the average plutonium concentration in incoming waste is much smaller than 0.04 g/L, and the factor increase would have to be much greater than 65. This provides a high assurance that incoming waste will spread into a layer of low plutonium areal density.
- (4) A high fluidity is required for incoming tank waste to permit it to be pumped through 3-in. diameter pipes for a long distance to reach tank farms. This level of fluidity is independent of the average plutonium concentration and will not be reduced when the limit is 0.04 g/L, and there should be no reduction in the tendency to spread into a thin layer.

If the tank inventory is assumed to be 215 kg of plutonium, a quantity greater than considered possible, the plutonium concentration would have to exceed 4 g/L (15.1 g/gal) before criticality is possible. This is 100 times greater than the limit on incoming waste. Even at this high concentration, the probability of criticality would remain very low because: (1) a large mass of plutonium would be required; and (2) the region of high plutonium concentration would have to be large. For instance, when the plutonium concentration is 4.0 g/L, criticality requires a spherical volume of 53,700 L (14,200 gal). A cube of this volume is 3.8 m (12.3 ft) on a side. The probability that 215 kg of plutonium would ever be in a storage tank is extremely low, and the probability that it would be confined to a volume this small is much lower yet.

The primary control for criticality safety is the ratio of solids to plutonium. This is discussed in Section 6.3. Sufficient solids are required to ensure subcriticality in the settled solids. Over the period when most of the waste was received at tank farms, an upper limit was placed on the plutonium concentration in settled solids of 1 g/L. This limit has been replaced by a limit on the neutron absorber/plutonium mass ratio. The presence of solids inhibits the plutonium from

concentrating and also provides neutron absorption. When agglomeration of the particles into large tightly bound masses is taken into account, subcriticality is guaranteed to a very high degree of certainty.

6.2 WASTE WITH LOW SOLIDS CONTENT

In an addendum to CSAR 79-007 (Carter 1979), Carter (1983) addressed the discharge of waste containing little or no solids. The primary concern with low solids content is the reduced ability to inhibit plutonium from concentrating when waste dries out, forms layers, or is filtered in solids. Carter (1983) addresses this concern, as follows:

In the new waste tanks, mechanisms exist (airlift circulators and slurry distributors) which distribute the plutonium over a large area (and over a large volume in the case of DST 102-AW). Accumulations over a large area (slab) require a minimum critical areal density of at least 240 g/ft² for low-density wastes (Carter et al. 1969, p. III.A.8.100-3). If the slab is limited to only a 10-ft radius (unlikely in the 75-ft diameter tank), then 75 kg would be the minimum amount of plutonium required for critical and this is at a concentration of 15 g/L. If a slab is limited to 3.64-ft radius, the minimum critical mass is approximately 10 kg, but also at a minimum critical concentration of about 15 g/L. The critical areal density increases rapidly as the concentration approaches 7 g/L; at 10 g/L it is 290 g/ft². Limitation of the plutonium to a small area is not considered credible in these tanks because of the very liquid wastes plus the capability to spread the wastes. Layers of different concentrations will tend to increase the critical areal density. With the spreading mechanisms, localized accumulations are no longer considered a potential problem.

When waste contains little or no solids and also little or no plutonium, it is difficult to determine the ratio of solids to plutonium. An example of such a waste type is wash water. In practice, the uncertainty in the solids/plutonium mass ratio will be large. For this reason, special consideration has been given to waste with low solids and low plutonium content.

6.2.1 Tanks With Less Than 10 kg of Plutonium

When the content of solids and of plutonium in liquid waste is low, it is very difficult to accurately determine the ratio of solids to plutonium. After examining transfers with low solids content, Carter (1983) concluded that "restrictions on in-tank plutonium concentrations are unnecessary below 10 kg of tank inventory." Carter (1983) concluded that 10 kg is too small a quantity of plutonium to ever achieve criticality when spread over an area the size of a storage tank. The areal density of 10 kg of plutonium spread uniformly over the area of a 22.9-m (75-ft) diameter tank is less than 1% of the minimum critical areal density.

If the tank plutonium inventory is less than 10 kg, transfers can be made into that tank without regard to the solids/plutonium mass ratio in the transfer. When the total quantity of plutonium in a tank reaches 10 kg, no further waste may be added until it is assured that the solids/plutonium mass ratio is at least 5,000 for waste in that tank.

6.2.2 Transfers With Plutonium Concentration Less Than 0.001 g/L

When the plutonium concentration is less than 0.001 g/L, waste may be transferred without regard to absorber content, either in the transferred waste or in the receiver tank waste. This plutonium concentration is deemed sufficiently small as to have a negligible impact on reactivity. This limit is a key element of the Technical Safety Requirement (TSR) (LMHC 1996).

In the past, waste with low solids content could be sent to tank storage if the plutonium concentration was less than 0.013 g/L (0.050 g/gal). This liquid waste would spread over a wide area and would not increase the plutonium areal density significantly. If an entire SST or DST is assumed filled with waste having a plutonium concentration of 0.013 g/L, the plutonium areal density would only be about 118 g/m² (11 g/ft²). This is less than 5% of the minimum critical areal density. When the plutonium concentration is 0.001 g/L, the areal density when the tank is completely filled is less than 10 g/m² (1 g/ft²). This is less than 0.5% of the minimum critical value.

When the plutonium concentration is less than 0.001 g/L, the actual solids/plutonium mass ratio is likely to, nevertheless, be large, and the margin of safety is greater than described in the previous paragraph.

6.2.3 Batches With Less Than 15 g of Plutonium

If the total quantity of plutonium in a batch is 15 g, or less, the plutonium is less than 3% of the minimum that can be made critical, and that batch will not influence the margin of safety, regardless of solids present. These transfers are permitted without additional restrictions. These small quantities are, nevertheless, added to the plutonium inventory for the tank.

When the total plutonium in a tank reaches 10 kg, an assessment of the solids volume is to be made to provide assurance that the tank-averaged solids/plutonium mass ratio is at least 5,000.

6.2.4 Tanks With Waste Spreading Capability

Airlift circulators and slurry distributors are sometimes used to spread incoming waste over a wide area of the tank. When these devices are used, discharge of larger batches of low solids content waste is permitted. Carter (1983) specifies the following limits for waste with low solids content when an airlift circulator or a slurry distributor is used:

1. The airlift circulator or the slurry distributors shall be operating to spread out the plutonium-bearing waste.
2. The maximum amount of plutonium for a single transfer is 200 g (all wastes).
3. Such transfers may continue until a total of 10 kg is in the tank.

4. Upon reaching 10 kg, the plutonium concentration in the tank(s) shall be determined by dividing the total tank plutonium inventory by the total solids volume in the tank or by some other appropriate method. This concentration shall be determined after each transfer.

The quantity of plutonium discharged at any single location using a slurry distributor is limited to 200 g. After the distributor has been moved to a new location, another transfer can be made. The fluidity of waste assures that it spreads out into a thin layer, and the plutonium areal density will increase only slightly. The plutonium concentration in all discharged waste is considerably less than the minimum critical concentration.

The total quantity of plutonium in a batch is less than 40% of the minimum critical mass for a plutonium-water system. For the mass of plutonium to exceed the minimum critical mass, the distribution mechanism would have to fail. Before criticality becomes possible, the plutonium would at the same time have to be placed in a compact, localized configuration. Such a configuration is not possible until the depth of waste reaches several feet. In addition, there would have to be a failure to mix the waste uniformly before discharge. These factors combine to make criticality extremely unlikely, if not impossible.

6.3 LIMIT ON SOLIDS CONTENT

The content of neutron absorbing solids is of primary importance to criticality safety. For criticality to be possible, the solids/plutonium mass ratio must be less than the minimum subcritical limit over a sizeable volume of contiguous waste. The volume required to achieve a critical configuration depends on how far the mass ratio of neutron absorbing solids falls below the subcritical limit.

Confidence that the neutron absorber/plutonium mass ratio will remain above the limit value following changes to the waste configuration (i.e., after mixing, combining waste types, transfer, etc.) is provided by knowledge of waste chemistry. A chemical property that greatly enhances confidence in the long-term stability is the tendency of the waste components to form tightly bound agglomerates. This property, and other important chemical properties, that contribute to long-term stability and continued subcriticality are discussed in Appendix D.

6.3.1 Plutonium Concentration in Settled Solids

Most plutonium in tank farms arrived under a plutonium limit of 1 g/L in settled solids. All 216 solids samples reported in WHC-SD-WM-ER-400 (WHC 1995) are less than 10% of the minimum critical plutonium concentration of 2.6 g/L. In the future no limit will be placed on plutonium concentration in settled solids, but rather a lower limit will be placed on the solids/plutonium mass ratio.

6.3.2 Limit on Neutron Absorber/Plutonium Mass Ratio in Discharged Batch

A key element of the TSR (LMHC 1996) requires that the mass ratio for at least one of five specified neutron absorbers in relationship to plutonium be greater than the minimum subcritical

ratio (see Section 3.2). The five neutron absorbers identified in the TSR are chromium, iron, manganese, nickel, or natural (or depleted) uranium. The bases for these minimum subcritical mass ratios are provided in Appendix B.

The TSR (LMHC 1996) requirement may be incorporated into the CPS using a limit, such as the following:

For a batch of waste received into tank farms, the component/plutonium subcritical mass ratio for at least one component shown in Table 4-1, as determined by analysis or predictive calculation, shall not be less than the corresponding subcritical mass ratio.

In order to protect the TSR (LMHC 1996) key element, the CPS should also include the following limit:

If the plutonium equivalent content in a batch of waste received into tank farms exceeds 50 g, the sum of the component subcritical mass fractions, as determined by analysis or predictive calculation, shall be at least 2.

In practice, the overall solids/plutonium mass ratio for the contents of a waste tank is expected to be much higher than the minimum ratio required by the above limits. The CPS limit permits the solids content in a batch of waste to be less than the expected content to allow for an occasional batch with a low solids content.

The use of a relatively low neutron absorber mass ratio limit in the CPS is justified by the following considerations:

1. Waste typically has a solids/plutonium mass ratio between 10,000 and 50,000. It is unlikely that the mass ratio in a batch will even approach a value as low as 1,000. The high average value over many discharges will more than compensate for the few batches with a low solids content.
2. Normally, the plutonium content in a batch is less than 50 g. When the plutonium exceeds 50 g, the CPS increases the required subcritical fraction for a batch to 2. Under the most idealized conditions, the minimum plutonium required for criticality is 520 g. Even if no neutron absorbing solids were present, more than ten 50-g batches would be required before criticality is possible.
3. The average plutonium concentration in a batch is limited to no more than 0.04 g/L, a value more than 65 times smaller than the minimum for which criticality is possible.
4. Violation of the mass ratio limit for several batches in succession would not create a potential for criticality because of the low average plutonium concentration and of the tendency for waste to spread into a thin layer.

5. When the storage tank plutonium content exceeds 10 kg, the solids/plutonium mass ratio for the tank as a whole is required to exceed 5,000. This value is less than half of the solids/plutonium mass ratio for waste in any of the 177 storage tanks in tank farms and is low enough to ensure easy compliance (see Section 6.3.3). At the same time it is more than 10 times the solids/plutonium mass ratio that ensures subcriticality and is large enough to ensure a large margin of subcriticality.

6.3.3 Tank-Averaged Solids/Plutonium Mass Ratio

For the tank farms as a whole, the average value of the solids/plutonium mass ratio has been conservatively estimated at 73,600. DST SY-102, which contains PFP waste, has an estimated solids/plutonium mass ratio of 12,555. This is the smallest solids content for any tank, and it exceeds 20 times the subcritical limit for homogeneous waste. The solids content in future waste is not expected to be less than in past waste.

Below a plutonium inventory of 10 kg, no limit is placed on the mass ratio due to the small areal density. Above a plutonium inventory of 10 kg, a minimum limit of 5,000 is placed on the tank-averaged solids/plutonium mass ratio. This mass ratio exceeds the minimum subcritical limit of homogeneous CWM solids by a factor of 10 and is large enough to ensure subcriticality for nonuniform distributions of waste components (within the range of credible variations).

The *Plutonium Inventory Database* (LMHC 1998a) keeps track of the plutonium inventory, total solids volume, and solids/plutonium mass ratio for DSTs. The total solids mass is obtained by multiplying the measured depth of solids by the tank area and the estimated density. No transfers of waste are made to SSTs, and no comparable plutonium inventory database is available for them.

The tank-averaged mass ratio is recalculated after each transfer from outside tank farms by adding the incoming plutonium to the inventory. However, the estimated solids content remains unchanged until a new measurement of the depth of solids is made. In this way, the estimated tank-averaged solids/plutonium mass ratio will decrease with each transfer until the solids content is remeasured. If the tank-averaged solids/plutonium mass ratio is found to approach the investigation level of 5,000, a detailed evaluation will be undertaken to characterize the waste.

6.3.4 Saltcake

Saltcake is soluble, and it generally forms the top layer when other solids types are present (i.e., above sludge). Saltcake sometimes forms a crust above the liquid layer. When water is added to saltcake, it dissolves. This allows it to be easily removed from other solids. See Appendix C2.3.3 for more information on saltcake.

As saltcake crystallizes, dissolved plutonium remains in solution and is not found within the saltcake. Saltcake may contain a small fraction of trapped sludge, either internal or sorbed on the surface, but the contribution of plutonium from this sludge is too small to be significant. The highest plutonium concentration associated with saltcake is found in the interstitial liquor. Bratzel et al. (1996) state that "analyses of the interstitial liquor show that plutonium

lower than the maximum solubility limit of 0.2 g Pu/L.” Braun et al. (1994, p.14) draws the conclusion that “saltcake waste is highly subcritical because of the very small quantity of plutonium and the very large amount of salt-type neutron absorbers.”

6.4 ASSURANCE OF ADEQUATE MIXING

A facility sending waste to tank farms must ensure that the limit on which criticality safety is based (i.e., either plutonium concentration or mass ratio) is satisfied throughout the waste. This requires that the incoming waste be “adequately” mixed.

To be “adequately” mixed for criticality safety does not require that waste be uniform throughout the settled sludge. Uniform mixing of individual batches with each other is difficult to achieve and difficult to verify. Criticality safety does not require a criterion this strict for mixing. For criticality safety, all that is required is that the neutron absorber/plutonium mass ratio exceed its corresponding CPS limit throughout the waste. This permits strata with greatly different compositions, so long as the neutron absorber/plutonium mass ratio for each is greater than the limit value.

Verification of mixing is best accomplished through analyses of waste samples. The waste is adequately mixed for the waste sampled if the neutron absorber/plutonium mass ratio exceeds the CPS limit. However, the volume of a sample is very small in relationship to the total volume of waste in a storage tank, and analysis of a limited number of waste samples does not provide definitive verification of mixing. Enhanced assurance of mixing is provided by knowledge of the processes of waste generation. Two characteristics of waste of particular importance to guaranteeing that waste remains mixed are small particle size and the tendency of waste particles to agglomerate. The fuel processing plants generated waste with a very small average particle size. Once these small particles are well mixed separation by mechanical means is almost impossible. In addition, the chemistry in the waste storage environment causes neutron absorbers to agglomerate with plutonium. The bonding between particles is strong and cannot be easily broken. Once tank waste has been mixed, it will remain mixed under any conditions that are permitted in the tank storage environment.

There are specific circumstances that are known to provide a state of adequate mixing. Waste is adequately mixed when:

- a. Waste components are in solution. Dissolved components diffuse to form a uniform concentration.
- b. Waste whose particles are less than 50 μm in size and which was well mixed before discharge. Mixing can be accomplished by operation of a mixer device (pump, rotary blade, etc.) prior to discharge for sufficient time to ensure mixing.
- c. The waste originates in a waste storage tank. Solids were sent to tank storage well mixed, and the alkaline storage environment keeps the waste components agglomerated.

6.5 INTERTANK TRANSFERS OF SUPERNATANT LIQUID

Supernatant liquid waste may be pumped from one tank to another. The upper bound on plutonium solubility in alkaline salt solutions (i.e., at saturation) is 0.0017 g/L (see Appendix D), and this establishes an upper limit on plutonium concentration in the liquid. However, a plutonium concentration above the saturation boundary is possible due to the presence of flocculent agglomerates with some adsorbed plutonium. The volume fraction occupied by suspended solids in liquid pumped from one tank to another is small. Therefore, the plutonium content in supernatant liquid is also small.

When the saltwell pumping method is used (applies only to SSTs), the liquid is passed through a screen to remove large particulate. Nevertheless, even after solids have been allowed to settle for a long time before being screened, the liquid will still contain a small content of suspended solids. The composition of suspended solids should closely resemble that of the waste solids in general. Serne et al. (1996) conclude that the quantity of solids removed during saltwell pumping is too small to be of concern. It would be highly unlikely for the plutonium concentration in the pumped liquid to exceed 0.01 g/L. Further discussion on saltwell pumping is provided in Appendix G5.0.

Double-shell tank AN-107 provides an instructive example of the plutonium concentration in flocculent material transferred in supernate. This tank has never received waste directly from a plutonium separation plant. All of its waste originated from transfers of supernatant liquid from other tanks. The settled solids, therefore, originated from flocculent material carried along in the liquid. The volume of settled solids in DST AN-107 is 852,000 L (225,000 gal), and the plutonium concentration is reported as 0.002 g/L.

A larger proportion of solids might be transferred if the waste were stirred prior to transfer and not enough time was allowed for solids to settle out. In this case, the content of suspended plutonium might be increased. However, sludge is composed largely of agglomerates in which the plutonium is tightly bound with a large proportion of neutron absorbing solids. Even with considerable stirring and agitation of the waste liquid before and during transfer, the plutonium in these agglomerates would not be separated from other solids. The proportion of neutron absorbing solids in relationship to the plutonium will remain high in the transferred waste.

Each proposed waste transfer operation must be reviewed to determine if mobilized solids are contained in the liquid. Transfer of supernatant and saltwell liquid is acceptable when the solids content is consistent with liquids that have been left undisturbed for a long period of time. The pH of liquid in the source tank and in the receiver tank must remain greater than 8. If mobilized (suspended) solids are present in the transferred waste or if the waste in the receiver tank will be significantly disturbed by these operations, a review and/or evaluation are to be completed according to requirements in Appendix G10.0 for transfer of waste solids.

6.6 ACCOUNTING FOR URANIUM-235 ENRICHMENT

In a homogenized uranium-water system, criticality is not possible until the ^{235}U enrichment exceeds 1.03 wt%. HNF-IP-1266, *Tank Farms Operation Administrative Controls* (CHG 2000), states that “for homogeneous uranium-water mixtures (and uranium compounds), systems enriched up to 1.00 wt% ^{235}U are not subject to a mass limit.”

6.6.1 Uranium-235 and the Plutonium Inventory

Most uranium in tank storage was received under the following limit from CPS-T-149-0010, Rev. D-0 (WHC 1988):

One gram of ^{233}U or ^{235}U in excess of 1 wt% enrichment shall be assumed equivalent to one gram of Pu, unless otherwise restricted by a specification. The amount of ^{235}U less than or equal to the equivalent of 1 wt% enrichment may be excluded in concentration and total mass calculations.

There are no chemical or physical means by which the ^{235}U can be separated from the ^{238}U in tank waste. Therefore, no increase in the ^{235}U enrichment is possible, except when combined with other uranium of higher enrichment. For these reasons, uranium enriched between 0.72 wt% and 1.0 wt% may be sent to tank storage without accounting for the ^{235}U content on the plutonium inventory.

6.6.2 Determining the Uranium/Plutonium Subcritical Fraction

The uranium/plutonium mass fraction may be used as a measure of subcriticality. This fraction is found by dividing the uranium/plutonium mass ratio by the uranium/plutonium subcritical limit mass ratio. However, the uranium/plutonium subcritical limit mass ratio of 770 assumes the uranium contains 0.72 wt% ^{235}U (i.e., it is natural uranium). When the subcritical mass fraction is determined, it is, therefore, necessary to treat the ^{235}U content in excess of 0.72 wt% as if it were plutonium. If the uranium is known with certainty to be depleted, the subcritical mass fraction can be directly obtained. If the possibility exists that the ^{235}U content might exceed 0.72 wt%, an isotopic analysis is required to determine the ^{235}U enrichment and the plutonium content before a subcritical mass fraction can be determined.

6.7 LIMITS FOR DOUBLE-CONTAINED RECEIVER TANKS

Additional limits are specified specifically for DCRTs 244-TX, 244-S, 244-U, and 244-TX. A limit of 2,000 g plutonium is used to ensure a low average plutonium concentration. A detailed discussion of the basis for limits placed on DCRTs is provided in Appendix A2.2.

6.7.1 Agitator Pump Operation

To prevent the buildup of plutonium, an agitator pump and sluicing nozzles are operated prior to a transfer from DCRT 244-TX into DST SY-102. Operation of the agitator pump is performed according to Tank Farm Operating Procedure TO-430-480, Section 5.2.19 (LMHC 1999c). This procedure requires that the agitator pump be operated for a minimum of 1/2 hour before initiating waste flow to DST SY-102.

6.7.2 Neutron Monitors on Double-Contained Receiver Tank 244-TX

Three neutron monitors are installed on DCRT 244-TX to monitor the radiation level. At the time the monitors were installed, there was no definitive information showing that an accumulation of plutonium could not occur. Even though the available information indicated that an accumulation would be unlikely, monitors were installed as a precautionary measure. An evaluation of the need for these neutron monitors is provided in Appendix A2.4.

To prevent buildup of plutonium, an agitator pump and sluicing nozzles are available. Prior to a transfer from DCRT 244-TX into DST 102-SY, the agitator pump and sluicing nozzles are operated. During a transfer in the past, at least 2 neutron monitors were used to record the radiation levels. Monitor readings after transfer were compared to readings prior to receipt to ascertain whether the radiation level had increased. An increase in background radiation would have indicated an accumulation of plutonium.

A computer simulation by Whyatt et al. (1996) and chemistry study by Bratzel et al. (1996) support the conclusion that an accumulation of plutonium in DCRT 244-TX cannot occur. Even without considering chemical bonding, the particle size distribution ensures that significant accumulation of plutonium is highly unlikely. When the tendency to agglomerate is taken into account, the ability for plutonium to settle out of the waste solids is reduced even more. Formation of a plutonium rich layer on the bottom and its subsequent failure to be flushed out when waste is transferred from DCRT 244-TX is not credible.

Neutron monitors on DCRT 244-TX do not contribute to an enhanced level of criticality safety, and their use may be discontinued.

6.8 EVALUATION OF PLANNED OPERATIONS

When a new operation is planned, a review will be made to determine if a criticality safety evaluation is needed. If the proposed operations fall within the envelope of existing evaluations, then no additional evaluation is required. Justification that the operation needs no further evaluation is to be documented in writing. When a criticality safety evaluation is performed for an operation previously evaluated in a different tank, information in this CSER may be relevant and can be referenced instead of copied into the new evaluation. Appendix G describes the process of evaluating new operations and outlines a simplified approach to evaluating activities that may disturb the settled waste solids.

This page intentionally left blank.

7.0 DESIGN FEATURES AND ADMINISTRATIVELY CONTROLLED LIMITS AND REQUIREMENTS

Limits and controls provided in this section are intended to be used in Criticality Prevention Specifications (CPS) to govern receipt of waste into tank farms from waste generating facilities and its transfer between storage tanks. These limits and controls meet criteria for criticality safety (see Section 3.4) and comply with the key elements of the Technical Safety Requirements (see Section 3.2).

It is acceptable to change the descriptive material and the format used in the CPS without revision to this CSER. New activities may be added to the list of specific activities, and obsolete activities may be removed, provided they fall within the description of activities provided.

A revision to this CSER is required when the scope of operations is expanded outside limits provided in this section. For instance, a new CSER (or revision) would be required if the Limited Control Status of RPP were to be challenged. A revision of this CSER is required before a limit may be increased above a maximum specified value or below a minimum specified value.

The surveillance program monitors the state of stored waste. When a parameter is outside the boundary defined by an investigation level, further investigation is undertaken to establish the quality of the analytical measurements or predictive computations used to identify the suspected infraction. Then, if an investigation level is exceeded, recovery actions are initiated.

Before each transfer, chemical compatibility between incoming waste and waste already stored is verified by tank farms engineering.

7.1 FACILITY CLASSIFICATION

According to HNF-IP-1266, *Tank Farms Operation Administrative Controls*, (CHG 2000a), classification as a Limited Control Facility requires a documented criticality safety evaluation that demonstrates “that a criticality is prevented by the form or distribution of fissionable material, after allowing for credible accidents.” This CSER provides the basis for classifying the entire tank farms as a Limited Control Facility. Alternately, each facility at tank farms may be thought of as an individual Limited Control Facility. Conditions that justify this classification must be maintained.

7.2 DEFINITIONS

The following definitions are for terms used in this CSER and which might be used in the CPS.

Plutonium Equivalence

One gram of ^{235}U in excess of 1.0 wt% enrichment or one gram of ^{233}U shall be assumed equivalent to one gram of ^{239}Pu , unless otherwise restricted by a specification. This fissile uranium is to be added to the plutonium inventory and included in the plutonium concentration. Natural and depleted uranium are excluded from inventory control.

Plutonium Equivalent Inventory

The plutonium equivalent inventory is the total quantity of plutonium plus the plutonium equivalence of the ^{235}U and ^{233}U .

Saltcake

Saltcake is the waste material not evaporated in an evaporator (called evaporator bottoms) that crystallizes after being pumped back to a storage tank and allowed to cool. Saltcake is soluble, and it generally forms the top layer when other solids types are present (i.e., above sludge). Saltcake sometimes forms a crust above the liquid layer. The plutonium concentration in saltcake is very low.

Sludge

Sludge is a mixture of metal precipitates (with interstitial liquid) which results from the neutralization and settling of chemical separation wastes. Sludge generally consists of metal oxides, hydroxides, carbonates, silicates, phosphates, sulfates, and other insoluble species. Some of these compounds are fairly soluble, but many are insoluble.

Slurry

“Double-Shell Slurry” refers to a mixture of inorganic salt crystals in a high salt mother liquor, with a small amount of metal hydroxide solids.

Solids

“Solids” refers to the non-liquid component of waste that settles when left undisturbed and which can be separated from the liquid using a centrifuge or by draining the waste through a fine mesh. Saltcake, sludge, and slurry are all forms of solids. For criticality safety, a distinction is made between “soluble” and “insoluble” solids.

Supernate

Supernate is the liquid portion of waste above the settled sludge. The terms “supernate” and “liquid” are usually used interchangeably. Supernatant liquid may contain dissolved saltcake.

Subcritical Mass Fraction

The component subcritical mass fraction is found by dividing the actual absorber-to-plutonium mass ratio by the corresponding minimum subcritical mass ratio. The total subcritical mass fraction is obtained by summing the component subcritical mass fractions.

7.3 ALLOWED OPERATIONS THAT CAUSE NEGLIGIBLE DISTURBANCE

The only control required for the operations that cause negligible disturbance in any waste tank is an assurance that only the specified operations are conducted. Inclusion or removal in the CPS of specific operations within the following categories may be made without revisions to this CSER.

7.3.1 Surveillance Activities

Surveillance activities in the dome space do not change the quantity, composition, or distribution of waste materials.

The following specific activities that do not affect nuclear reactivity are permitted:

- a. Liquid and solids level monitoring,
- b. Liquid Observation Well (LOW) scans,
- c. Temperature readings,
- d. Dome surveys,
- e. Dry well and lateral scans,
- f. Tank vapor space monitoring,
- g. Installation and repair of monitoring equipment,
- h. Operation and testing of active and passive ventilation systems, and
- i. Photography.

7.3.2 Addition of Liquids

Regardless of the pH of waste in a tank, water and/or chemicals may be added, provided they have a pH of 7.0, or greater, and do not contain fissionable material. Each time the total volume of additions increases by 500 gal or when the pH in liquid to be added is less than 7.0, verification shall be made that the pH for waste in the tank will not fall below 8.

Operations that require addition of water and/or chemicals include:

- a. Installation of monitoring equipment (such as thermocouple trees) using the water lance method,

- b. Instrument flushes,
- c. Temperature control,
- d. Contamination control (at the entrance and inside pits, risers, vents, etc.),
- e. Process condensate from tanks and active ventilation,
- f. Neutron poison additions (no restrictions on volume or quantity),
- g. Operating dip tubes,
- h. Priming and flushing pumps and transfer lines,
- i. Flushing deentrainers,
- j. Pressure checking transfer lines,
- k. Transfer of non-fissile aqueous solutions into waste tanks, and
- l. Routine maintenance.

No control is required on pH (alkalinity) in the following 242-A Evaporator processes, regardless of the volume of liquid:

- a. Diverted process condensate and steam condensate,
- b. Process pumps PB-1 and PB-2 seal pot drains, and
- c. Floor drains, including decontamination operations.

7.3.3 Operations That Have Negligible Effect On Nuclear Reactivity

Operations that come into direct contact with waste, or have the potential to come into direct contact, are permitted when it can be easily shown that there is no significant potential to reduce the margin of safety.

Specific permitted operations that have a negligible impact on criticality safety are:

- 1. Minor disturbances to the waste, similar to the following examples:
 - a. Scraping the surface of the waste
 - b. Drilling holes up to 15.2 cm (6 in.) in diameter
 - c. Inserting rods into the waste
 - d. Stirring supernate
 - e. Using cleaning solvents
- 2. Tank sampling activities.
 - a. Core sampling (push mode or rotary mode)
 - b. Auger sampling

- c. Supernatant sampling (including bottle-on-a-string)
 - d. Tank Vapor Space sampling
- 3. Neutron poison addition
 - 4. Neutron activation studies
 - 5. Operation and maintenance of airlift circulators (ALCs)

7.4 WASTE DISTURBING OPERATIONS

Operations that disturb settled solids must be reviewed to determine if a new criticality safety evaluation is required. The requirements for this review and the process used to complete an evaluation, if required, are described in Appendix G, "Evaluation of Planned Operations," of this CSER. Before a transfer of waste is approved, assurance is required that conditions in the receiver tank will remain within "existing waste conditions." This requires that waste in the sending and the receiving tanks are chemically similar and compatible.

Although not anticipated, a waste disturbing operation would be permitted in a SST, provided the requirements that would apply to the same operation in a DST are met. This includes maintaining an auditable continuous inventory of plutonium and an assessment of the plutonium concentration.

Only operations that cause negligible disturbance (see Section 7.3) are permitted in an IMUST. An operation that will disturb the waste requires that a CSER be prepared to cover it.

7.5 GENERAL SPECIFICATIONS

7.5.1 Requirements

- 1. An auditable continuous inventory of plutonium and an assessment of the plutonium concentration both in the solids and supernate shall be maintained for each DST and DCRT.

Unless otherwise stated, the plutonium inventory of a tank refers to the plutonium equivalent inventory. The plutonium equivalence for a batch is determined before discharge into a storage tank.

7.5.2 Limits

- 1. When a batch contains enriched uranium, the ^{235}U in excess of 1.00 wt% of the uranium shall be added to the plutonium inventory.
- 2. When the plutonium concentration is less than 0.001 g/L, waste may be transferred without regard to absorber content, either in the transferred waste or in the receiver tank waste.

3. For transfers of supernatant and interstitial liquids between tanks within tank farms, no requirements are placed upon pH, the mass ratio of neutron absorbers to plutonium, or the plutonium concentration. No limit is placed on caustic or water addition.
4. When the plutonium inventory in a tank reaches 10 kg, the tank-averaged solids/plutonium mass ratio for settled solids shall exceed 5000 before additional plutonium may be added.

7.6 WASTE TRANSFERS FROM OUTSIDE TANK FARMS

7.6.1 Requirements

1. Before each transfer (receipt) of waste from outside tank farms, demonstration of compliance shall be documented, and an independent verification shall be made.
2. A facility sending waste to tank farms will provide reasonable assurance that the limit on which criticality safety is based (i.e., either plutonium concentration or mass ratio) is satisfied throughout the waste. This requirement is satisfied for:
 - a. Waste in solution.
 - b. A batch containing less than 1 vol% solids.
 - c. A batch with its entire contents of solids in suspension at the time of discharge. Suspension of solids can be accomplished by operation of a mixer device (pump, rotary blade, etc.) prior to discharge for sufficient time.
 - d. Solids transferred between storage tanks. Solids in tank storage are considered mixed.
3. One of the following methods may be used to determine the plutonium concentration and/or the solids/plutonium mass ratio:
 - a. A computation based upon the plutonium mass, the solids volume, and the density of settled solids. Compensation shall be made for uncertainties to ensure that the calculated concentration or mass ratio is conservative.

The average plutonium concentration in precipitated (settled) solids can be found by dividing the total plutonium mass by the total solids volume.

The solids/plutonium mass ratio is found by dividing the density of settled solids (not including contained water) by the plutonium concentration.

$$\text{Solids/plutonium mass ratio} = \frac{\text{Density of Settled Solids (g/L)}}{\text{Plutonium Concentration (g/L)}}$$

If the measured density of settled solids is not available, assume a solids density of 1,200 g/L.

- b. A laboratory analysis of tank samples. Waste characterization data that provides component concentrations may be used.
- c. The minimum solids fraction and/or the maximum plutonium concentration possible as determined from a flowsheet governing the generation of the waste may be used to determine compliance.

[Note: There are few remaining processes, if any, for which this method is used.]

7.6.2 Limits for Transfers From Outside Tank Farms

1. Discharged waste shall be alkaline (basic) with a pH of at least 8.0, unless a criticality safety evaluation shows that alkalinity is not required.
2. For a batch of waste received from outside tank farm facilities, the plutonium concentration averaged over the total volume (both solids and liquid) of the batch shall not exceed 0.04 g/L (0.15 g/gal).
3. For a batch of waste received into tank farms, the neutron absorber/plutonium mass ratio for at least one component shown in Table 7-1 shall not be less than the corresponding subcritical mass ratio. This is not required when the plutonium concentration is less than 0.001 g/L (0.004 g/gal).

Table 7-1. Minimum Neutron Absorber/Plutonium Subcritical Mass Ratios

Neutron Absorber (X)	Minimum Neutron Absorber/Plutonium Subcritical Mass Ratio (X/Pu)
Chromium (Cr)	135
Iron (Fe)	160
Manganese (Mn)	32
Nickel (Ni)	105
Total Uranium (U)	770

4. If the plutonium equivalent content in a batch of waste received into tank farms exceeds 50 g, the sum of the component subcritical mass fractions, as determined by analysis or predictive calculation, shall be at least 2.

7.7 ADDITIONAL LIMITS FOR DOUBLE-CONTAINED RECEIVER TANKS

1. For DCRTs 244-A, 244-BX, 244-S, 244-TX, and 244-U, the maximum plutonium in each tank shall not exceed 2,000 g.
2. The solids content in a waste batch transferred from Z-Plant to DCRT 244-TX shall comprise at least 0.7% by volume.
3. After each transfer of Z-Plant solutions from DCRT 244-TX to DST SY-102, the transfer line shall be flushed.
4. For transfer of solution from DCRT 244-TX into DCRT 244-S:
 - a. The solution in DCRT 244-TX shall be allowed to settle for at least 24 hours prior to transfer.
 - b. The sluicing system in DCRT 244-TX shall not be operated prior to or during the transfer.
 - c. Prior to transfer, the Criticality Safety Representative (CSR) for tank farms shall be notified.

[Note: The sluicing system is not operated to permit solids to settle so as not to be transferred with the liquid to DCRT 244-S.]

5. For at least 25 minutes prior to transfer from DCRT 244-TX to DST SY-102, the agitator pump and sluicing nozzles must be operated, and they must continue to operate during the transfer.

[Note: It is intended that all solids be transferred. Sluicing ensures that solids are more easily pumped out.]

7.8 242-A EVAPORATOR

Requirements on pH, plutonium concentration, and neutron absorber/plutonium mass ratio applied to discharges from a waste generating facility do not apply to transfers to or from the 242-A Evaporator facility during an evaporation campaign. The 242-A Evaporator operations shall conform to the following:

1. Plutonium concentration
in feed solution: < 0.005 g/L (0.019 g/gal)
2. Before an evaporator campaign begins, a determination shall be made and documented of the expected quantity and concentration of plutonium that will be in the feed and slurry tanks at completion of the run.

3. After an evaporator campaign, a determination shall be made of the actual quantity and concentration of plutonium in the feed and slurry tanks, either by analysis or calculation.
4. No limit is placed on caustic or water addition.

7.9 PROPOSED CHANGES TO OPERATIONS

A proposed change to facility-operating procedures or to equipment that involve fissionable material shall conform to the following:

- The change shall be reviewed for compliance to applicable CPS limits and controls and approved by the independent criticality safety organization and the tank farm Criticality Safety Representative (CSR).
- If the proposed new operation is not within the existing criticality safety basis, a CSER will be completed and a CPS will be issued (if required) to cover the operation before the operation is initiated.

7.10 SURVEILLANCE PROGRAM

The goal of the criticality safety program is to establish limits and controls that ensure that waste will remain subcritical into the indefinite future. This goal requires an assurance that waste has a large margin of safety when received into the tank farms. Nevertheless, because of the high degree of uncertainty in waste composition and distribution, stored waste needs to be monitored to verify the margin of subcriticality.

A surveillance program shall be in place to monitor waste and to compare characterization data to investigation levels. The goal of a criticality safety surveillance program is to provide adequate information to verify that the waste is well subcritical. This section describes the elements of the surveillance program.

7.10.1 Schedule of Surveillance

The goal of surveillance is to provide reassurance of safety. This reassurance is important because of the great complexity and the difficulty of direct observation of tank waste. The surveillance program establishes a schedule by which to monitor safety:

1. The Criticality Safety Representative conducts two criticality safety audits of tank farms per year.
2. The U.S. Department of Energy conducts periodic assessments.

3. A criticality data sheet (CDS) is completed for each batch of waste received into tank farms and for each transfer within tank farms. This data confirms that CPS limits are met.
4. Each transfer of waste is entered into the Plutonium Inventory Database (LMHC 1998a), which keeps a current record of the plutonium inventory and total tank solids content.
5. An active characterization program obtains and analyzes waste samples for their chemical compositions. Analytical data provides reassurance that the margin of safety is large.

After waste has been stabilized, the need for surveillance is greatly diminished. The distribution of fissile material in dry waste will remain unchanged in the future.

7.10.2 Data Obtained from Samples

Waste samples are chemically analyzed to verify an adequate margin of safety. Parameters of interest to criticality safety include:

- **pH** - Alkalinity provides assurance that the plutonium is insoluble.
- **Specific Gravity** - The specific gravity is needed to convert from the units used in laboratory analysis to criticality safety limits.
- **Density of Solids** - The density of the solids is used to determine the mass ratio of solids to plutonium.
- **Plutonium Concentration** - The plutonium concentration is compared to the minimum critical concentration to provide a measure of subcriticality. In addition, it is used to determine component/plutonium mass ratios.
- **Neutron Absorber Concentrations** - The concentrations of components, such as uranium, iron, and manganese, provide data to calculate mass ratios. From these, the fraction of the subcritical limit mass ratio can be obtained.
- **^{235}U and/or ^{233}U Enrichment** - When uranium with ^{235}U enrichment greater than 0.72 wt% might be present or when ^{233}U might be present, their concentrations should be obtained.

7.10.3 Investigation Levels

An investigation level is a criterion of acceptability defined as the point at which concern arises that the margin of subcriticality may be eroding. Verification of the margin of subcriticality is accomplished by comparing selected measured parameters to investigation levels. An investigation level is set low to permit detection and correction of unexpected conditions before violation of a key element of the criticality safety program.

When one of the following Investigation Levels is found to be exceeded, an investigation shall be undertaken to verify the validity of the measurements and to ascertain the true condition of the waste:

- The plutonium concentration in settled (precipitated) solids exceeds 1.0 g/L (3.8 g/gal).
- When the tank plutonium inventory is less than 10 kg or the depth of supernatant liquid is less than 30 cm (12 in.), there is no restriction on pH.

When the tank plutonium inventory exceeds 10 kg and the depth of supernatant liquid exceeds 30 cm (12 in.), the investigation level for pH is 8.0, or less.

[Note: For criticality safety, pH is a secondary control. A low pH cannot by itself cause criticality.]

- When the plutonium inventory for a storage tank exceeds 10 kg, the tank-averaged solids/plutonium mass ratio is less than 5,000.

A tank-averaged solids/plutonium mass ratio is determined by dividing the best estimate of the total solids mass by the best estimate of the tank plutonium inventory.

[Note: This limit is satisfied if the volume of settled solids exceeds 42,000 L (11,000 gal) for each 10 kg of plutonium.]

Under certain conditions, when there is a high degree of certainty that criticality is not possible, surveillance requirements for a tank may be relaxed or waived. No surveillance is required when:

- A tank contains little, or no, liquid waste and whose contents have been stabilized.
- The plutonium inventory for a 22.9-m (75-ft) diameter tank is verified to be less than 10 kg.
- No operations are being conducted in the tank.

7.11 RECOVERY PLAN FOR NONCONFORMANCE

Suspected investigation level violations and CPS infractions are classed as nonconformances. When a nonconformance is uncovered, action is taken to bring the waste into conformance to (agreement with) criticality safety requirements.

7.11.1 Nonconformance to Investigation Level

If sample analysis shows an investigation level to be crossed, the Criticality Safety Representative (CSR) shall investigate the finding to determine if the apparent reduction in safety margin is real. If it is determined that a CPS limit has been violated or that the margin of subcriticality has been reduced to a level of concern, a recovery plan shall be prepared according to the requirements for a nonconformance to a CPS.

7.11.2 Nonconformance to Criticality Prevention Specification

If a suspected nonconformance is detected to the range of applicability or to a limit or control in the CPS, then an investigation will be conducted according to HNF-IP-0842, *RPP Administration* (CHG 2000b), Vol. IV, Section 6.3, *Criticality Safety Nonconformance Response*. A suspected nonconformance shall be immediately brought to the attention of the immediate manager who, in turn notifies the CSR and obtains other technical assistance as required to determine if a nonconformance has occurred. At this time an immediate review is made to determine if a limit [or an investigation level] has been violated. If violation of a limit or an investigation level cannot be ruled out, the immediate manager makes notifications as specified in the procedure.

An evaluation shall be made "to determine the appropriate method of recovery." The recovery plan shall be agreed upon and signed by the manager of the independent safety review organization (or delegate), the manager of the operating organization and, as appropriate, the engineering manager (or delegate). When recovery actions begin, the written plan shall be followed.

The CSR shall document the technical basis for the evaluation of the nonconformance. Documentation shall include an evaluation of the data used to reach a conclusion about the extent of the nonconformance.

7.11.3 Potential Criticality or Criticality Accident

Tank farms does not have a criticality alarm system (CAS) because the frequency of a criticality accident has been judged to be less than 10^{-6} /year. Also, waste tanks lie under 2.4 m (8 ft), or more, of soil, making it unlikely that any personnel would receive a 25-rem dose from a hypothetical criticality.

Although a criticality at tank farms is not considered credible, HNF-IP-1266, Section 5.7, "Nuclear Criticality Safety" (CHG 2000a), describes recovery actions that would be undertaken whenever conditions arise that have characteristics of a possible criticality accident. Occasional events have occurred, such as local high temperature readings, release of gases, or radiological releases, that exhibit some characteristics of a criticality, even though the causes are quite different.

The requirements for the RPP Emergency Preparedness Program are addressed by Administrative Control (AC) 5.14 in the TSR (LMHC 1996). Implementation of the Emergency Preparedness Program is addresses in Chapter 5.14 of HNF-IP-1266 (CHG 2000a).

Procedures for responding to an emergency are provided in *Tank Farms Building Emergency Plan*, WHC-IP-0263-TF (LMHC 1999b). Section 5.5, "Criticality," of LMHC (1999b) states that tank farms is classed as a limited control facility and a criticality is not credible due to the form and distribution of the fissionable material.

A hypothetical accident would be detectable only as a result of contamination spread, and it would be responded to according to the procedures for that type of accident. Upon notification of an accident, the tank farms Shift Manager (analogous to the Building Emergency Director) would form a task team of the appropriate personnel to immediately put the emergency plan into action. At this time there would be no way to know if a criticality accident had occurred, and no immediate way to find out. The response to a chemical or heat induced explosion would be the same as for criticality. An investigation into the source of the contamination would include an assessment as to whether it could have been caused by criticality.

7.12 FIREFIGHTING

No restrictions are placed on firefighting.

7.13 VERIFICATION OF CONFORMANCE

Before a transfer of waste is made, a Criticality Data Sheet (CDS) is filled out to verify conformance to CPS limits. A Transfer Data Sheet (TDS) is used to maintain a record of the transfer.

7.13.1 Criticality Data Sheet

Before a transfer of waste is made, a Criticality Data Sheet (CDS) must be filled out. The CDS provides a checklist of CPS requirements and requires that values be specified for parameters limited by the CPS. A CDS is intended to ensure conformance with the CPS.

There are three versions of the CDS, one used by the generator and two used by RPP engineering. The generator verification version is filled out by the organization that generates the waste and requires the signatures of a verifier and an independent verifier. One RPP engineering version is for transfer into any DCRT and the other version is for transfer into any DST. These CDSs are signed by the cognizant engineer.

7.13.2 Transfer Data Sheet

The Transfer Data Sheet (TDS) is used to keep track of waste transfers and to ensure compliance to rules and regulations. The TDS is designed to provide a description of the waste in a transfer and to provide a record so that waste can be tracked. A TDS is completed before waste is

transferred into a holdup tank in a processing facility and before each transfer to tank farms. In addition, a TDS is completed before each transfer between tanks at tank farms. Information from the Criticality Data Sheet (CDS) is generally used to complete the TDS. Data from the TDS are entered into the Plutonium Inventory Database (LMHC 1998a) to update the inventory after the transfer.

The organization making the transfer is responsible for determining values for all parameters required by the CPS. Waste samples are analyzed to determine values of parameters controlled by the CPS. For transfers between tanks, the composition is established using earlier sample analyses or from predictive computations. Calculations must be independently verified, and approval by the CSR must be obtained before the transfer is made.

At the waste generating facility, an inventory of fissile content is maintained for each holdup tank. When a transfer is made from the holdup tank, an estimate of the fissile content in the transfer is shown on the TDS, and this information is used to update the inventory for the storage tank. An estimate of plutonium inventory is derived from the measured plutonium concentration in settled solids and the total volume of solids. For conservatism, the maximum measured plutonium concentration is used to determine the inventory. In effect, the inventory is the upper limit estimation based on available plutonium concentration measurements.

7.14 CRITICALITY ALARM COVERAGE

DOE Order 5480.24 (DOE 1992), *Nuclear Criticality Safety*, Sections 7b (3) and (4), provide the following requirements for a Criticality Alarm System (CAS) or a Criticality Detection System. This requirement is also contained in DOE Order 420.1 (DOE 1995), *Facility Safety*, Section 4.3.3 e(3) with slightly different wording.

- (3) In those cases where the mass of fissionable material exceeds the limits established in paragraph 4.2.1 of ANSI/ANS-8.3, but a criticality accident is determined to be impossible due to the physical form of the fissionable material, or the probability of occurrence is determined to be less than 10^{-6} per year (as documented in a DOE approved SAR), neither a CAS nor a criticality detection system is required. In addition, neither a CAS nor a criticality detection system is required to be installed underwater when fissionable material is handled or stored beneath water shielding that is adequate to protect personnel; however, a means to detect fission gases or other volatile fission products should be provided in occupied areas immediately adjacent to such underwater storage areas except for fuel systems where no fission products are likely to be released. Also, neither a CAS nor a criticality detection system are required for fissionable material during shipment of fissionable material packaged in approved shipping containers, or fissionable material in approved shipping containers awaiting transport provided no other operation involving fissionable material not so packaged is permitted on the dock or in the shipment area.
- (4) The decision to install a criticality detection system rather than a CAS, and the decision that neither a CAS nor a criticality detection system is necessary, must be

justified based upon a documented DOE-approved safety analysis.

High-level waste tanks are categorized as limited control facilities, as defined in HNF-IP-1266, *Tank Farms Operation Administrative Controls*, (CHG 2000a). Controlling the form and distribution of fissionable material ensures criticality safety. In practice, the low concentration of plutonium and the high proportion of neutron-absorbing solids ensures a form that is highly subcritical regardless of the distribution of the waste, the degree of neutron moderation, and the presence of reflectors.

An assessment of the radiological doses and the toxicological exposure which might result from a postulated criticality assuming no controls is provided in *Tank Waste Remediation System Final Safety Analysis Report (FSAR)* (LMHC 1999a). The frequency of a criticality accident is considered to be less than 10^{-6} /year, or, in other words, beyond "extremely unlikely." Both the onsite and offsite radiological doses are found to be acceptable for an extremely unlikely accident.

This CSER provides documentation that the criteria permitting operations at tank farms without coverage of a criticality alarm system are met.

This page intentionally left blank

8.0 SUMMARY AND CONCLUSIONS

This CSER provides the technical bases for criticality safety limits and controls that govern receipt, transfer, and long-term storage of tank waste. Limits and controls provided in Section 7.0 may be used directly, or in a modified form, in the CPS for tank farms. Tank farms in their entirety are classified as a Limited Control Facility, and conditions that justify this classification must be maintained.

The safety philosophy is to require a margin of safety for new waste sufficient to maintain subcriticality for an unlimited period of time in the tank storage environment. The goal is to ensure subcriticality without placing unnecessary restrictions on transfers within tank farms. The technical bases for achieving this goal are provided.

Criteria are provided against which to evaluate tank waste. The basic safety criterion is the Double Contingency Principle that requires that at least two unlikely, concurrent, and independent contingencies occur before criticality is possible. In fact, multiple discharges of waste well outside CPS limits for both the generator facility and for tank farms would be required to create a condition for which criticality would be credible.

Most plutonium at tank farms was received while the processing plants were in operation. Criticality safety is more dependent upon the past history of waste discharges than it will be on future discharges. Roetman et al. (1994) provides an upper limit on the total plutonium inventory in all Hanford Site waste storage tanks to be 981 kg, based upon conservative assumptions of waste generation. Waste received in the future is not expected to increase the total plutonium inventory by much.

When new sources of waste are proposed, they must be reviewed. If they do not fall within the scope of this evaluation, a criticality safety evaluation must be made before receipt at tank farms.

An unlimited volume of tank waste will remain subcritical, and dimensions of tanks, piping, and equipment do not govern criticality safety. This CSER need not be revised when descriptive details change, unless the change is important to criticality safety. Most tank and equipment dimensions provided are expected to remain unchanged.

8.1 PHYSICAL AND CHEMICAL PROCESSES

Criticality safety of tank waste is dependent upon chemistry. Bratzel et al. (1996) concludes that "no physical or chemical phenomena or mechanism has been identified that could concentrate fissile material at sufficient quantities or concentrations to result in an accidental nuclear criticality." However, this conclusion depends on maintaining "existing waste tank conditions."

Maximum plutonium concentration is an important criticality safety parameter controlled by chemistry. For an increase in plutonium concentration to occur, it is first necessary to transport plutonium from a solid phase to an aqueous liquid phase, then to transport it back into a solid phase, and finally to deposit it into a compact geometry. Dissolution would require addition of large volumes of organic or inorganic plutonium complexants, oxidants, reductants, or acid. Acidification would cause vigorous reactions with nitrite and carbonate in the waste and also with the carbon steel in the tank walls and interconnecting pipes. These chemical reactions would generate a large volume of gas, leading over time to failure of the carbon steel pipelines and steel liner.

“Gravity segregation” is the process by which waste solids form into stratified layers according to particle size after settling through a layer of liquid. When particle size exceeds 100 μm , stratification is possible, but as particles become smaller, stratification becomes increasingly unlikely (Serne et al. 1996). When particle size is less than 10 μm , the ability to separate components into layers is small. Based on a large mass-fraction of tank waste having a particle diameter less than 10 μm , Serne et al. (1996) concluded that the plutonium concentration will not increase by a factor greater than 2.5 during settling.

Upon entering a large storage tank, waste spreads out to form a layer on top of the existing waste layer. At the same time, the process of mixing disperses plutonium and combines it with neutron absorbing solids. The tendency to spread and to mix prevents buildup of waste within a restricted volume.

Under normal tank conditions, the plutonium and other solids form tightly bound agglomerates that greatly inhibit the ability to increase the concentration of plutonium (Whyatt et al. 1996). Iron is a good neutron absorber present in most waste in a large mass ratio with respect to plutonium, and it has an affinity to agglomerate with plutonium. The formation of agglomerates is a powerful mechanism ensuring that the plutonium will remain combined with neutron absorbers. Studies by Whyatt et al. (1996), Serne et al. (1996), and Bratzel et al. (1996) did not identify any mechanism capable of eroding the margin of subcriticality.

Tank waste is maintained alkaline (i.e., has a high pH) to prevent dissolution and to ensure that plutonium remains combined with settled solids. A low pH would create a complex chemical environment difficult to evaluate.

8.2 NEUTRON ABSORBER/PLUTONIUM MASS RATIOS

Waste is received well mixed, and additional mixing may occur within the storage tank. A waste layer may be composed of several waste streams mixed together.

For each component of waste, there is a mass ratio relative to the plutonium content that ensures subcriticality. The subcritical mass fraction is defined as the component/plutonium mass ratio divided by the subcritical limit mass ratio. The contributions of various waste components may be combined by adding their subcritical mass fractions. Subcriticality is assured when the sum of fractions exceeds 1.0.

Hanlon (1999) gives the total volume of slurries and sludge in tank farms as 49,600 kL (13,100 kgal). If all tank farm plutonium is assumed uniformly distributed in slurries and sludge, a conservative estimate of the average plutonium concentration at tank farms is 0.020 g/L (0.076 g/gal). The solids/plutonium mass ratio for all waste in the tank farms is estimated to be at least 60,600. DST SY-102 has a tank-averaged solids/plutonium mass ratio of 10,000, the smallest for any storage tank. Nevertheless, the mass ratio for DST SY-102 is at least an order of magnitude larger than required to ensure subcriticality.

8.3 PLUTONIUM CONCENTRATION

The “plutonium concentration,” unless otherwise qualified, refers to the highest plutonium concentration that exists in any small subvolume (i.e., less than 1 L) of the waste. The “average plutonium concentration” refers to the total quantity of plutonium divided by the total volume of waste under consideration.

Rogers (1993a) defines a conservative waste model (CWM) with reduced neutron absorption for which critical parameters are conservative relative to actual waste. The complexity of an unknown heterogeneous distribution of waste types can be overcome by replacing the entire contents of a tank by the CWM.

Based upon the CWM, the subcritical limit on plutonium concentration is 2.6 g/L. For plutonium concentrations below 2.6 g/L, the greatest reactivity occurs when the waste is completely dry. The median plutonium concentration for 200 analyzed samples of waste solids from many different DSTs and SSTs was found to be 0.01 g/L (0.04 g/gal) (Braun et al. 1994). The highest reported plutonium concentration is 0.89 g/L for a sample from SST TX-118 (Lipke 1999).

For a batch of waste received from outside tank farm facilities, the plutonium concentration averaged over the total volume (both solids and liquid) shall not exceed 0.04 g/L (0.15 g/gal). After discharge into a storage tank, the plutonium concentration can increase as a result of solids settling into a layer. The plutonium concentration after settling is limited by the ratio of solids to plutonium.

Saltcake may contain a small fraction of trapped sludge, either internal or sorbed on the surface, but the contribution of plutonium from this sludge is too small to be significant. The highest plutonium concentration associated with saltcake is found in the interstitial liquor, and this is far below the solubility limit of 0.2 g Pu/L (Bratzel et al. 1996). Braun et al. (1994) concludes “saltcake waste is highly subcritical because of the very small quantity of plutonium and the very large amount of salt-type neutron absorbers.”

8.4 ENRICHED URANIUM

Kupfer et al. (1999) gives the best-basis estimate of the total inventory of uranium at tank farms to be 965,000 (see Appendix C5.0). Of this, 6,730 kg is estimated to be ²³⁵U, and the average ²³⁵U enrichment is 0.697 wt%. According to Agnew (1997), more than 90% is in SSTs.

According to the Hanford Defined Waste (HDW) Model, Revision 4 (Agnew 1997), about 4% of the total uranium in tank farms, is slightly enriched with a ^{235}U content between 0.72 and 0.86 wt%. The largest inventory of enriched uranium is 16,200 kg of 0.85-wt%-enriched uranium in SST S-107. Criticality is not possible until the enrichment exceeds 1.03 wt% ^{235}U (see Appendix B5.0).

When uranium is enriched to less than 1.0 wt%, the contained ^{235}U need not be included in the "plutonium-equivalent" inventory. Since virtually all uranium in tank waste has enrichment less than 1.0 wt%, only a small fraction of the ^{235}U has been included in the fissile inventory.

8.5 NEUTRON MONITORS

Three neutron monitors installed on DCRT 244-TX are used to compare radiation levels before and after waste transfers. An increase in background radiation would indicate that plutonium is accumulating. Whyatt et al. (1996) and Bratzel et al. (1996) support the conclusion that an accumulation of plutonium in DCRT 244-TX cannot occur. This conclusion is based on the small particle size and the tendency for waste components to agglomerate. The use of neutron monitors on DCRT 244-TX may be discontinued.

8.6 AUDITS AND APPRAISALS

Since the beginning of fuel handling operations on the Hanford Site, periodic audits and appraisals have been conducted to ensure that all operations, including the production and disposal of radioactive wastes, are in compliance to limits and controls applied to them.

8.7 SURVEILLANCE PROGRAM

A surveillance program is required to monitor the stored waste to ensure compliance with discharge criteria and to verify the accuracy of assumptions used in this evaluation. This program establishes specific criteria for determining which analytes are to be measured and when further investigation is required.

8.8 CRITICALITY ALARM COVERAGE

This CSER provides documentation that the criteria permitting operations at tank farms without coverage of a criticality alarm system are met.

9.0 REFERENCES

- Agnew, S. F., 1997, *Hanford Tank Chemical and Radionuclide Inventories: HDW Model Rev. 4*, LA-UR-96-3860, Los Alamos National Laboratory, Los Alamos, New Mexico.
- Altschuler, S. J., and P. C. Doto, 1991, *Minimum Critical Safety Limits For Nuclear Waste*, Transactions of the American Nuclear Society, Vol. 64, pp. 339-340.
- B&W, 1996, *Transfer TK-D7 to Solution to TK-D5*, PFP Procedure ZO-101-019, Rev/Mod C-0, Babcock & Wilcox Hanford, Richland, Washington.
- B&W, 1997a, *Transfer TK-D8 Contents to TK-D5*, PFP Procedure ZO-101-020, Rev/Mod C-1, Babcock & Wilcox Hanford, Richland, Washington.
- B&W, 1997b, *Sample TK-D4, TK-D5, TK-D7, or TK-D8*, PFP Procedure ZO-101-023, Rev/Mod D-1, Babcock & Wilcox Hanford, Richland, Washington.
- B&W, 1997c, *Operating Specifications for Plutonium Finishing Plant, 241-Z Waste Facility*, OSD-Z-184-00010, Rev/Mod G-0, Babcock & Wilcox Hanford, Richland, Washington.
- B&W, 1999a, *241-Z Solution Disposal Facility*, CPS-Z-165-80741, Revision. B-2, Babcock & Wilcox Hanford, Richland, Washington.
- B&W, 1999b, *Transfer TK-D5 to Tank Farms*, PFP Procedure ZO-101-010, Revision D, Change 1, Babcock & Wilcox Hanford, Richland, Washington.
- Bratzel, D. R., W. W. Schulz, R. Vornehm, and A. E. Waltar, 1996, *Tank Farm Nuclear Criticality Review*, WHC-SD-WM-TI-725, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- Braun, D. J., L. D. Muhlestein, T. B. Powers, and M. D. Zentner, 1994, *High-Level Waste Tank Subcriticality Safety Assessment*, WHC-SD-WM-SARR-003, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- Carter, R. D., G.R. Kiel, and K. R. Ridgway, 1969, *Criticality Handbook*, ARH-600, Vol. II, p. III.A.8.100-3, Atlantic Richfield Hanford Company, Richland, Washington.
- Carter, R. D., 1979, *Underground Waste Storage Tanks and Associated Equipment*, CSAR 79-007, SD-SQA-CSA-20108, Rockwell Hanford Operations, Richland, Washington.

- Carter, R. D., 1983, *Underground Waste Storage Tanks and Associated Equipment*, Addendum 1, CSAR 79-007, SD-SQA-CSA-20109, Rockwell Hanford Operations, Richland, Washington.
- CHG, 2000a, *Tank Farms Operation Administrative Controls*, HNF-IP-1266, CH2MHill Hanford Group, Inc., Richland, Washington.
- CHG, 2000b, *RPP Administration*, HNF-IP-0842, CH2MHill Hanford Group, Inc., Richland, Washington.
- Chiao, T., 1980, *CSAR 79-044: 241-Z Solution Disposal Facility*, WHC-SD-SQA-CSA-20213, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- Chiao, T., 1996, *Criticality Safety Review for Inactive Miscellaneous Underground Storage Tanks*, WHC-SD-SQA-CSA-30004, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- DOE, 1992, *Nuclear Criticality Safety*, DOE Order 5480.24, U. S. Department of Energy, Washington, D. C., Reviewed October 13, 1997.
- DOE, 1995, *Facility Safety*, DOE Order 420.1, U. S. Department of Energy, Washington, D. C., Reviewed October 13, 1997.
- DOE, 1998, *Guidelines for Preparing Criticality Safety Evaluations at Department of Energy Non-Reactor Nuclear Facilities*, DOE-STD-3007, Change Notice No. 1, U.S. Department of Energy, Washington, D. C.
- Funston, G. A., and C. D. Meng, 1997, *OSD-Z-184-00010 Supporting Document*, HNF-SD-CP-OCD-036, Rev. 1, B & W Hanford and Global Technologies, Inc., Richland, Washington.
- Hanlon, B. M., 1999, *Waste Tank Summary Report for Month Ending July 31, 1999*, HNF-EP-0182-136, Fluor Daniel Hanford, Inc., Richland, Washington.
- Kupfer, M. J., A. L. Boldt, K. M. Hodgson, L. W. Shelton, B. C. Simpson, R. A. Watrous; M. D. LeClair; G. L. Borsheim; R. T. Winward; B. A. Higley, R. M. Orme; N. G. Colton, S. L. Lambert, D. E. Place; and W. W. Schultz, 1999, *Standard Inventories of Chemicals and Radionuclides in Hanford Site Tank Wastes*, Lockheed Martin Hanford Corporation, Richland, Washington.
- Lipke, E. J., 1999, *Above Average Sample in 241-TX-118*, (Letter 74F00-EJL-99028 to N. L. Hulse, July 22), Lockheed Martin Hanford Corporation, Richland, Washington.
- LMHC, 1996, *Tank Waste Remediation System Technical Safety Requirements*, WHC-SD-WM-TSR-006, Rev. E, Lockheed Martin Hanford Corporation, Richland, Washington.

- LMHC, 1998a, *Plutonium Inventory Database*, HISI 11350, Lockheed Martin Hanford Company, Richland, Washington.
- LMHC, 1998b, *Tank Waste Remediation System Basis for Interim Operation*, HNF-SD-WM-BIO-001, Rev. 1, Lockheed Martin Hanford Corporation, Richland, Washington.
- LMHC, 1998c, *Waste Storage Tanks and Associated Equipment*, CPS-T-149-00010, Rev. I-0, Lockheed Martin Hanford Corporation, Richland, Washington.
- LMHC, 1999a, *Tank Waste Remediation System Final Safety Analysis Report*, HNF-SD-WM-SAR-067, Rev. 0, Lockheed Martin Hanford Corporation, Richland, Washington.
- LMHC, 1999b, *Tank Farms Building Emergency Plan*, HNF-IP-0263-TF, Rev. 4, Lockheed Martin Hanford Corporation, Richland, Washington.
- LMHC, 1999c, *244-TX DCRT to 241-SY-102 Slipstream Transfer via 244-S DCRT*, TO-430-480, Lockheed Martin Hanford Corporation, Richland, Washington.
- RCRA, 1976, *Resource Conservation and Recovery Act of 1976*, as amended, 42 USC 6901 et seq.
- Ridgway, K. R., and R. D. Carter, 1972, *Criticality Prevention Parameters of Plutonium in Soils*, ARH-2622, Atlantic Richfield Hanford Company, Richland, Washington.
- Roetman, V. E., S. P. Roblyer, and H. Toffer, 1994, *Estimation of Plutonium in Hanford Site Waste Tanks Based on Historical Records*, WHC-EP-0793, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- Rogers, C. A., 1993a, *CSER 92-009: An Analytical Model For Evaluating Subcritical Limits For Waste in Hanford Site Storage Tanks*, WHC-SD-SQA-CSA-20356, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- Rogers, C. A., 1994a, *CSER 94-001: Criticality Safety of Single Shell Waste Storage Tanks*, WHC-SD-SQA-CSA-20363, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- Rogers, C. A., 1994b, *CSER 94-004: Criticality Safety of Double Shell Waste Storage Tanks*, WHC-SD-SQA-CSA-20368, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- Rogers, C. A., K. N. Schwinkendorf, and H. Harris, 1996, *Criticality Parameters for Tank Waste Evaluation*, WHC-SD-SQA-CSA-507, Westinghouse Hanford Company, Richland, Washington.

- Sanchez, R., W. Myers, D. Hayes, R. Kimpland, P. Jaegers, R. Paternoster, S. Rojas, R. Anderson, and W. Stratton, 1996, *Criticality Characteristics of Mixtures of Plutonium, Silicon Dioxide, Nevada Tuff, and Water*, LA-UR-96-3738, Los Alamos National Laboratory, Los Alamos, New Mexico.
- Serne, R. J., G. A. Whyatt, S. V. Mattigod, Y. Onishi, P. M. Doctor, B. N. Bjornstad, M. R. Powell, L. M. Liljegren, J. H. Westsik, Jr., N. J. Aimo, K. P. Recknagle, G. R. Golcar, T. B. Miley, G. R. Holdren, D. W. Jeppson, R. K. Biyani, and G. S. Barney, 1996, *Fluid Dynamics, Particulate Segregation, Chemical Processes, Natural Ore Analog and Tank Inventory Discussions that Relate to the Potential for Criticality in Hanford Tanks*, WHC-SD-WM-TI-757, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- Stickney, R. G., and E. J. Lipke, 1998, *Authorization Basis Status Report (Miscellaneous TWRS Facilities, Tanks and Components)*, HNF-2503, Rev. 0, Duke Engineering and Services Hanford, Inc., Richland, Washington.
- Tiffany, M. S., 1998, *Authorization Basis for the 209-E Building*, Fluor Daniel Hanford Inc., Richland, Washington.
- Tomlinson, R. E., 1961, *Safety Review of Manufacturing Facilities - Chemical Processing Department*, HW-69586, General Electric Company, Richland, Washington.
- WHC, 1988, *Waste Storage Tanks and Associated Equipment*, CPS-T-149-00010, Rev. D-0, Westinghouse Hanford Company, Richland, Washington.
- WHC, 1995, *Tank Waste Source Term Inventory Validation*, WHC-SD-WM-ER-400, Westinghouse Hanford Company, Richland, Washington.
- Whyatt, G. A., R. J. Serne, S. V. Mattigod, Y. Onishi, M. R. Powell, J. H. Westsik, Jr., L. M. Liljegren, G. R. Golcar, K. P. Recknagle, P. M. Doctor, V. G. Zhirnov, J. Dixon, D. W. Jeppson, and G. S. Barney, 1996, *The Potential for Criticality in Hanford Tanks Resulting from Retrieval of Tank Waste*, PNNL-11304, Pacific Northwest National Laboratory, Richland, Washington.

9.1 BIBLIOGRAPHY

The following references contain information relevant to the criticality safety of tank waste, but are not referenced in the main body of this report.

- Altschuler, S. J., 1980, *CSAR 80-015: 244-TX and 244-S Salt Well Receiver Tanks*, SD-SQA-CSA-20110, Rev. 0, Rockwell Hanford Operations, Richland, Washington.

- Altschuler, S. J., 1991, *CSER 91-011: Minimum Critical Safety Limits for Nuclear Waste*, WHC-SD-SQA-CSA-20339, Westinghouse Hanford Company, Richland, Washington.
- Anderson, J. D., 1990, *A History of the 200 Area Tank Farms*, WHC-MR-0132, Westinghouse Hanford Company, Richland, Washington.
- ANS 1983, *Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors*, American National Standard ANSI/ANS-8.1-1983, American Nuclear Society, La Grange Park, Illinois.
- B&W, 1997d, *241-Z Solution Disposal Facility*, CPS-Z-165-80741, Rev. B-1, Babcock & Wilcox Hanford, Richland, Washington.
- Bailey, J. W., 1993, *Functional Design Criteria - Tank 241-C-106 Waste Retrieval Project W-320*, WHC-SD-W320-FDC-001, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- Blyckert, W. A., 1981, *Waste Evaporators 242-A, 242-S*, CSAR 81-022, SD-SQA-CSA-20112, Rev. 0, Rockwell Hanford Operations, Richland, Washington.
- Blyckert, W. A., 1983, *Operation of Tank 244-BX*, Addendum 1 to CSAR 81-015, SD-SQA-CSA-20111, Rev. 0, Rockwell Hanford Operations, Richland, Washington.
- Brady Raap, M. C., and T. S. Vail, 1999, *TWRS Nuclear Criticality Inspection and Assessment Plan*, HNF-3323, Rev. 0, EDT 619460, Duke Engineering Services Hanford, Richland, Washington.
- Brevick, C. H., L. A. Gaddis, and E. D. Johnson, 1995, *Tank Waste Source Term Inventory Validation*, 3 Volumes, WHC-SD-WM-ER-400, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- Carter, R. D., 1987, *Operation of Tank 244-U*, Addendum 2, CSAR 80-015, SD-SQA-CSA-063, Rev. 0, Rockwell Hanford Operations, Richland, Washington.
- Carter, R. D., 1994, *Addendum 2A, CSAR 79-007: Underground Waste Storage Tanks and Associated Equipment*, WHC-SD-SQA-CSA-20365, Rev. 0, Westinghouse Hanford Company, Richland, Washington. Written in 1984, but transmitted as EDT 157914 in 1994.
- Clemmons, J. S., and A. Q. Goslen, 1992a, *Minimum Safe Ratios of Fe and Mn to U-235 in an Infinite System (U)*, Nuclear Criticality Safety Analysis, WM-92-3, WER-MER-921143, Westinghouse Savannah River Company, Aiken, South Carolina.

- Clemmons, J. S., and A. Q. Goslen, 1992b, *Minimum Safe Ratios of Fe and Mn to Pu-239 in an Infinite System (U)*, Nuclear Criticality Safety Analysis, WM-92-5, WER-MER-921353, Westinghouse Savannah River Company, Aiken, South Carolina.
- Daling, P. M., T. S. Vail, G. A. Whyatt, D. I. Kaplan, K. M. Krupka, S. V. Mattigod, J. R. Serne, C. A. Rogers, D. G. Erickson, K. N. Schwinkendorf, and E. J. Lipke, 1997, *Feasibility Report of Criticality Issues Associated with Storage of K Basin Sludge in Tank Farms*, HNF-SD-WM-ES-409, Rev. 0, Duke Engineering & Services, Richland, Washington.
- Delegard, C. H., and S. A. Gallagher, 1983, *Effects of Hanford High-Level Waste Components on the Solubility of Cobalt, Strontium, Neptunium, Plutonium, and Americium*, RHO-RE-ST-3 P, Rockwell International, Richland, Washington.
- Delegard, C. H., 1985, *Solubility of $PuO_2 \cdot xH_2O$ in Alkaline Hanford High-Level Waste Solution*, RHO-RE-SA-75 P, Rockwell International, Richland, Washington.
- DiCenso, A. T., L. C. Amato, W. I. Winters, 1995, *Tank Characterization Report for Double-Shell Tank 241-SY-102*, WHC-SD-WM-ER-366, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- DOE, 1988, *Site Characterization Plan, Reference Repository Location, Hanford Site, Washington*, DOE/RW-0164, Vol. 3, U. S. Department of Energy, Washington, DC, pp. 4.1-106.
- FDH, 1999, *Tank Waste Remediation System Basis for Interim Operation*, HNF-SD-WM-BIO-001, Rev. 1A, Fluor Daniel Hanford Company, Richland, Washington.
- Hess, A. L., 1991, *Review of Records for Source of ^{233}U in Tank 104-C* (Letter 29210-91-030 to S. D. Godfrey, August 19), Westinghouse Hanford Company, Richland, Washington. This is Appendix B in Rogers (1994b).
- Hey, B. E., 1993, *Dose Consequence for Postulated 350 MJ Hanford Site Underground Radioactive Liquid Waste Tank Criticality*, WHC-SD-WM-TI-527, Westinghouse Hanford Company, Richland, Washington.
- Hobbs, D. T., T. B. Edwards, and S. D. Fleischman, 1993, *Solubility of Plutonium and Uranium in Alkaline Salt Solutions (U)*, WSRC-TR-93-056, Savannah River Technology Center, Westinghouse Savannah River Company, Aiken, South Carolina.
- LMHC, 1998, *Operating Specifications For the 241-AN, AP, AW, AY, AZ & SY Tank Farms*, OSD-T-151-00007, Rev. H-21, Lockheed Martin Hanford Corporation, Richland, Washington, March 6, 1999.

- Paxton, H. C., and N. L. Pruvost, 1987, *Critical Dimensions of Systems Containing ²³⁵U, ²³⁹Pu, and ²³³U*, LA-10860-MS, Los Alamos National Laboratory, Los Alamos, New Mexico.
- Reardon, W. A., 1987, *Operation of Tank 244-U, CPS-T-149-00061*, Addendum 2 to CSAR 80-015, Rockwell Hanford Operations, Richland, Washington.
- Reynolds, D. A. and D. L. Herting, 1984, *Solubilities of Sodium Nitrate, Sodium Nitrite, and Sodium Aluminate in Simulated Nuclear Waste*, RHO-RE-ST-SWD-024, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- Schwinkendorf, K. N., 1994a, *Criticality Analysis of Homogeneous Pu-Water-Waste Systems*. This is Appendix A.6.0 of Braun et al. (1994), *High Level Waste Tank Subcriticality Safety Assessment*.
- Schwinkendorf, K. N., 1994b, *Criticality Analysis of Various Idealized Waste Geometries*. This is Appendix A.7.0 of Braun et al. (1994), *High Level Waste Tank Subcriticality Safety Assessment*.
- Schwinkendorf, K. N., 1994c, *Analysis of Radial Isotopic Profile for Hanford Site Aluminum-Clad Fuel Elements*. This is Appendix A.8.0 of Braun et al. (1994), *High Level Waste Tank Subcriticality Safety Assessment*.
- Schwinkendorf, K. N., 1995, *Criticality Safety Evaluation for 300 Area N Reactor Fuel Fabrication and Storage Facility*, WHC-SD-NR-CSER-010, Rev. 1, Westinghouse Hanford Company, Richland, Washington.
- Schwinkendorf, K. N., 1997, *Criticality Safety Evaluation for Spent Nuclear Fuel Processing and Storage Facilities*, HNF-SD-SNF-CSER-005, Rev. 3, Fluor Daniel Northwest, Richland, Washington.
- Sederberg, J. P., 1994, *Chemical Compatibility of Tank Wastes in Tanks 241-C-106, 241-AY-101, and 241-AY-102*, WHC-SD-WM-ES-290, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- Stratton, W. R., 1983, *The Myth of Nuclear Explosions at Waste Disposal Sites*, LA-9360, UC-70, Los Alamos National Laboratory, Los Alamos, New Mexico, October 1983.
- Toran, L. E., C. M. Hopper, M. T. Nancy, C. V. Parks, J. F. McCarthy, B. L. Broadhead, and V. A. Colten-Bradley, 1997, *The Potential for Criticality Following Disposal of Uranium at Low-Level Waste Facilities*, NUREG/CR-6505, Vol. 1, ORNL/TM-13323/V1, "Uranium Blended With Soil," Oak Ridge National Laboratory, Oak Ridge, Tennessee.
- Tusler, L. A., 1995, *Double-Shell Tanks Pu Inventory Assessment*, WHC-SD-WM-TI-640, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- Vail, T. S., 1993, *Desk Instructions for Maintaining Pu Tracking System for Double Shell Tanks*, WHC-SD-WM-TI-555, Rev. 0, Westinghouse Hanford Company, Richland, Washington.

This page intentionally left blank.

APPENDIX A

PAST LIMITS ON INCOMING WASTE

This page intentionally left blank.

A.0 PAST LIMITS ON INCOMING WASTE

Nearly all plutonium expected to ever be sent to tank farms has been received. Although significant volumes of waste are expected to be discharged in the future, the total plutonium content is expected to be much less than in the past. This appendix reviews criticality safety limits and controls that were used in Criticality Prevention Specifications (CPSs) and in facility operating procedures. Procedures used in the past are responsible to a greater degree for the present state of tank waste than procedures now in use.

A1.0 TANK FARM CRITICALITY PREVENTION SPECIFICATION LIMITS

Past discharges to tank farms were governed by limits and controls specified in CPS-T-149-00010, *Waste Storage Tanks and Associated Equipment* (WHC 1988). This CPS applied to the waste evaporators, to double-shell tanks (DSTs), to the vaults (244-A, 244-AR, and 244-CR), and to the Rail Car Unloading Facility 204-AR. The bases for the original version are found in CSAR 79-007 (Carter 1979, Carter 1983). CSAR 94-004 (Rogers 1994) replaced CSAR 79-007 as the basis document in 1994. At that time extensive revisions were made to CPS-T-149-00010. This appendix describes revision D-0 in effect until 1994 with limits evaluated by Carter in 1979.

CPS-T-149-00060 (WHC 1980) and CPS-T-149-00061 (WHC 1987) provided limits for double-contained receiver tanks (DCRTs) 244-TX, 244-S, 244-U, and 244-BX. These CPSs are discussed in Appendix A2.2.

Waste was required to be mixed before discharge to provide assurance that localized plutonium concentrations would not form. Discharged waste was maintained alkaline to prevent plutonium from dissolving and to keep the plutonium mixed with solids. Before 1994 there was no limit in the CPS on pH because alkalinity was guaranteed by corrosion control, and a separate limit for criticality safety was not considered necessary.

A1.1 LIMITS IN CPS-T-149-00010, REV. D-0

The following limits are taken from CPS-T-149-00010, Rev. D-0 (WHC 1988):

²³⁵U Enrichment in Uranium

One gram of ²³³U or ²³⁵U in excess of 1 wt% enrichment is considered equivalent to one gram of plutonium, unless otherwise restricted by a specification. The amount of ²³⁵U less than or equal to the equivalent of 1wt% enrichment may be excluded in concentrations and total mass calculations.

Concentration Limits

Average concentration in feed solution to
241-A and 242-S Evaporators: <0.01 g/gal

Maximum plutonium concentration in mixtures routed to:

Waste Storage Tanks (except DST SY-102): 0.05 g/gal

244-AR and 244-CR vault tanks: 0.05 g/gal

DST SY-102: 0.125 g/gal

No transfers may be made into tanks which contain 10,000 or more grams plutonium or vaults unless the maximum plutonium density possible after in-tank concentration is shown to be as follows (except as permitted with cadmium addition):

DST SY-102 ≤ 2 g/L plutonium in solids

All Other Waste Tanks ≤ 1 g/L plutonium in solids

Total Plutonium Mass Limits

Total plutonium in any 75-ft diameter storage tank:

All tanks except SST C-104, SST C-106,
and DST SY-102 < 50 kg

SST C-104, SST C-106, and DST SY-102 < 125 kg

Cadmium Addition Limits

Cadmium addition applies only to transfers into DST SY-102.

<u>Plutonium concentration in Solids, g/L</u>	<u>Minimum required cadmium concentration (as precipitated solids) g Cd/g Pu</u>
0 < conc. \leq 2	0.00
2 < conc. \leq 30	0.33

Tanks Containing Less Than 10,000 g of Plutonium

Transfers into tanks that contain less than 10,000 g of plutonium are required to meet one of the following conditions:

1. The transfer contains not more than 15 g of plutonium.
2. An airlift circulator is operating in the tanks, and the transfer contains not more than 200 g of plutonium.
3. The transfer is made through a slurry distributor, and the total plutonium added to the waste at any single position does not exceed 200 g.
4. The maximum plutonium density is controlled in accordance with the requirements for tanks containing greater than 10,000 g of plutonium.

Tanks Containing More Than 10,000 g of Plutonium

To demonstrate compliance with in-tank plutonium concentration limits, the plutonium concentration in the tank or vault solids shall be determined by one of the following methods:

1. Dividing the total tank or vault plutonium inventory by the total solids volume in the tank, provided that the plutonium content can be considered distributed.
2. A predictive computation.
3. A laboratory analysis of tank samples.
4. The use of the minimum solids fraction possible as determined from the flowsheet.

This concentration shall be determined after each transfer. The concentration shall be shown on an auditable continuous inventory maintained for each tank.

A1.2 LIMIT ON PLUTONIUM MASS

Prior to 1994, CPS-T-149-00010, Revision D-0, specified a total plutonium mass limit of 125 kg for SSTs C-104 and C-106 and for DST SY-102. A plutonium concentration limit of 2 g/L was used in conjunction with the 125-kg inventory limit (Carter 1979). Justification for this combination of limits was based on three facts. First, when the plutonium concentration is 4 g Pu/L, the sphere critical mass for the CWM is 215-kg plutonium. A sphere containing 125 kg of plutonium will remain well subcritical at twice the limit plutonium concentration of 2 g/L. Second, it would be extremely unlikely for a large fraction of the plutonium inventory to

be confined to a small spherical volume. Third, the bonding of plutonium to solids (i.e., agglomeration and adsorption) makes it very difficult, if not impossible, to concentrate the plutonium (Bratzel et al. 1996). See Appendix D4.0 for a discussion of mechanisms of concentration.

A mass limit of 50 kg plutonium was used for all other storage tanks because they had a low inventory, and there was no need for a higher plutonium limit.

Chemistry studies have been completed that provide strong justification for the conclusion that the plutonium cannot be separated from the neutron absorbing solids to the extent that a criticality accident would be possible (see Appendix D). For this reason, the CPS limits on plutonium inventory (mass) and concentration have been replaced by a single limit on the neutron absorber/plutonium mass ratio. This limit assures subcriticality for an unlimited volume of waste, regardless of the plutonium concentration.

A1.3 TRANSFERS USING CADMIUM AS A CONTROL

During final cleanout of tanks between January and April 1995, PUREX transferred 18 batches of waste to DST AW-105 using limits requiring the addition of cadmium. These 18 transfers, which form the top layers of the settled solids in DST AW-105, contain the highest plutonium concentration of any of the waste in this tank. These were the only transfers ever made to tank farms in which cadmium (or any other neutron poison) was added as a criticality safety control.

As a result of these transfers, the plutonium inventory increased from 17,230 g to 22,843 g. A large part of this 5,613-g increase was actually ^{235}U added to the plutonium inventory as "plutonium-equivalent" material. The average quantity of plutonium in a batch was 311 g, and the average plutonium concentration was 0.018 g/L. The largest quantity of plutonium in any of these batches was 446 g, and the highest plutonium concentration in a single batch was 0.031 g/L.

Carter (1994) provides justification for a plutonium concentration as high as 30 g/L in precipitated solids provided that at least 1 g of cadmium is present for every 3 g of plutonium. For the CPS, a limit was placed on plutonium concentration of 6 g/L in the precipitated (settled) solids, and a limit was placed on the plutonium mass in a single batch of 500 g.

A cadmium/plutonium mass ratio greater than 0.13 ensures subcriticality. In these discharges, the cadmium/fissile atom ratios were between 2.6 and 11, far more than required to ensure subcriticality. An added safety margin was provided by the content of ^{238}U , which was sufficiently high to ensure subcriticality in the absence of the cadmium. This meant that subcriticality was ensured, even if the cadmium did not remain with the fissile material after discharge. Since cadmium is soluble, it was necessary to provide assurance of subcriticality, even if the cadmium is assumed to separate from the waste batch inside of the tank.

The $^{235}\text{U}/^{238}\text{U}$ and plutonium/ ^{238}U mass ratios are expected to remain unchanged. If it is assumed that fissile material does separate from other solids, the total quantity of plutonium in a batch is not enough to cause criticality. The solids in each batch would spread out to form a thin layer with enhanced fissile concentration, but with an areal density far less than the minimum required for criticality. The entire 5,613 g for all 18 batches provides an average areal density of only 13.6 g/m^2 (1.3 g/ft^2), a value only 0.5% of the minimum critical value. Waste already in DST AW-105 contains far more solids than required to ensure subcriticality, and neutronic interaction is negligible.

A2.0 DOUBLE-CONTAINED RECEIVER TANK LIMITS

Waste leaving processing facilities was first routed to a double-contained receiver tank (DCRT) before being sent to a single-shell tank (SST) or to a double-shell tank (DST). An off-normal discharge of plutonium from the Plutonium Finishing Plant (PFP) would first be routed into a DCRT where an accumulation of plutonium would most likely occur.

A2.1 DESCRIPTION OF DOUBLE-CONTAINED RECEIVER TANK

A DCRT has a volume less than 150,000 L (40,000 gal). Criticality safety for a DCRT is largely dependent on whether it receives waste from PFP, since PFP is the only plant capable of discharging separated plutonium. PFP waste has been sent to DCRTs 244-S and 244-TX. No waste from PFP has ever been sent to DCRT 244-U or 244-BX.

Operations in DCRTs include short-term storage of Z-Plant and jet pump solution from the 241-T, 241-TX, 241-TY, 241-S and 241-SX tank farms.

CPS T-6 (RHO 1980) was issued to cover operations of DCRTs. The limits in CPS T-6 were later transferred to CPS-T-149-00060 (WHC 1980) and CPS-T-149-00061 (WHC 1987).

A2.1.1 DCRT 244-S

Altschuler (1980) evaluates the criticality safety of DCRTs 244-TX and 244-S. For DCRT 244-S, he attaches an internal letter which provides a basis for not requiring neutron monitors. Controls exist which prevent the discharge of batches larger than the limit (i.e., an overbatch) from entering from PFP holdup tank D-5. Long-term buildup was not considered to be credible for DCRT 244-S, since plutonium-bearing solutions would only be sent to it infrequently. In addition, neutron monitors on the D-5 holdup tank and on DCRT 244-TX would detect an overbatch before it could be sent to DCRT 244-S. Nevertheless, as a precaution, two 4-in. diameter dry wells were provided to permit neutron monitoring on special occasions.

A2.1.2 DCRT 244-TX

Waste from PFP is routed directly to DCRT 244-TX. Since DCRT 244-TX will be routinely flushed, it is unlikely that sludge containing significant quantities of plutonium would remain after transfer. DCRT 244-TX has 11 sluicing nozzles and an agitator pump to prevent plutonium solids buildup. Nevertheless, three neutron monitors were installed to detect any accumulation should it occur.

Neutron flux readings for DCRT 244-TX are recorded on the Transfer Data Sheet (TDS). The monitor readings after waste has been transferred out are compared to the value before the waste was received. In this way it can be determined if the neutron background is increasing. An increase in the neutron background would indicate that an increase in the plutonium inventory has occurred in the tank. The actual mass of plutonium in the tank, however, cannot be determined.

A2.1.3 DCRT 244-BX

Blyckert (1983) addressed the operation of DCRT 244-BX, which was used to "receive saltwell jet-pumped liquor from the 241-B, 241-BX, and 241-BY tank farms." DCRT 244-BX has no sluicing nozzles or neutron monitors. The expected plutonium concentration in the solution pumped through DCRT 244-BX is 3.5×10^{-4} g/L, based upon analytical results from operation of this tank. This system was judge acceptable based on the low plutonium concentration.

A2.1.4 DCRT 244-U

Reardon (1987) and Carter (1987) address the operation of DCRT 244-U, used to receive saltwell jet-pumped liquor from the 241-U tank farm. This tank is of the same design as DCRT 244-BX, and operations associated with it are the same. No sluicing nozzles or neutron monitors are provided. The expected plutonium concentration is the same as for DCRT 244-BX, or 3.5×10^{-4} g/L. This system was judged acceptable to criticality safety.

A2.1.5 DCRT 244-A

DCRT 244-A operates in a pass-through mode and is not used to store waste. Waste passes through DCRT 244-A and does not stop, the expected fissile inventory at any one time is less than 15 g. Because of this low fissile inventory, it is concluded that DCRT 244-A does not need to be included in the CPS covering operation of DCRTs.

In the unlikely event that the plutonium inventories were to exceed 15 g, the facility would still comply with the designation of Limited Control Facility that applies to tank farm facilities. In addition, the material that passes through this facility must comply with limits in the CPS that cover the originating tank and the destination tank. Nevertheless, to avoid misunderstanding, DCRT 244-A may be added to the list of DCRTs in the CPS.

A2.2 DOUBLE-CONTAINED RECEIVER TANK LIMITS

Altschuler (1980) provides limits for CPS T-6 (RHO 1980) to cover transfer of saltwell liquid into the saltwell receiver vessel DST SY-102. This liquid was first transferred to DCRT 244-TX. DCRT 244-S was used as a catch tank to receive excess liquid should DCRT 244-TX not have sufficient volume. These limits were later transferred to CPS-T-149-00060 (WHC 1980) and CPS-T-149-00061 (WHC 1987). The limits are as follows:

Maximum plutonium concentration in solution
routed to the tanks: ≤ 0.05 g/gal

Maximum plutonium in each tank: 2000 g

A transfer line water flush shall be made after each DCRT 244-TX to DST SY-102 transfer of Z-Plant solutions.

Should it be necessary to transfer solution from DCRT 244-TX into DCRT 244-S, the DCRT 244-TX solution shall be allowed to settle and the sluicing system shall not be operated during the transfer. The Manager, Criticality and Radiological Analyses shall be notified of the plan to move the transfer prior to being made (WHC 1980; WHC 1987).

Limits and controls for DCRTs were transferred to CPS-T-149-00010 (LMHC1998b) covering operations in DSTs. The limits and controls on waste received from outside tank farms also apply to DCRTs. In addition, the following limits apply specifically to DCRTs:

1. For DCRTs 244-TX, 244-S, 244-U, and 244-BX, the maximum plutonium in each tank shall not exceed 2,000 g.
2. The solids content in a waste batch transferred from Z-Plant to DCRT 244-TX shall comprise at least 0.6% by volume.
3. After each transfer of Z-Plant solutions from DCRT 244-TX to DST SY-102, the transfer line shall be flushed with water.
4. For transfer of solution from DCRT 244-TX into DCRT 244-S:
 - a. The solution in DCRT 244-TX shall be allowed to settle.
 - b. The sluicing system in DCRT 244-TX shall not be operating during the transfer.
 - c. Prior to transfer, the Criticality Safety Representative for tank farms shall be notified.

5. Neutron monitors installed for DCRT 244-TX are capable of indicating plutonium accumulation. To prevent the buildup of plutonium, an agitator pump and sluicing nozzles are available.
 - a. Prior to a transfer from DCRT 244-TX into DST SY-102 the agitator pump and sluicing nozzles must be operated.
 - b. During the transfer from DCRT 244-TX into DST SY-102, at least 2 neutron monitors must be functioning.

It is intended that liquids transferred from DCRT 244-TX to DCRT 244-S be free of solids. For this reason, the solids are allowed to settle and sluicing jets are not used prior to a transfer to DCRT 244-S.

A2.3 BASIS FOR DOUBLE-CONTAINED RECEIVER TANK LIMITS

Altschuler (1980) describes the operation of DCRT 244-TX and DCRT 244-S as saltwell receiver vessels:

Both vessels will be used to collect the solutions from saltwells in the process of 'drying up' the underground waste storage tanks containing saltcake and sludges. In addition to receiving this type of solution, Tank 244-TX will also receive neutralized Z-Plant waste solutions from 241-Z Tank D-5. The criticality concern in the 244-TX tank arises from the receipt of this latter solution.

Contingencies in the 244-TX tank arise from exceeding the concentration or amount of fissile material charged from the D-5 tank at Z-Plant or continual accumulation of fissile material because of settling or precipitation. Analysis using well-referenced data (mainly ARH-600) (Carter et al. 1969) shows that the triple contingency criteria of RHO-MA-136 are met if a large deposit in one spot can be prevented from accumulating. The possibility of criticality arises not from the accumulation itself but by a sudden ineffective dispersal of the solids by a recirculation jet which enlarges and increases the moderation of the accumulation before sufficient dispersal has taken place in the 244-TX tank. The presence of the recirculation sluice jets will ensure removal of the solids when the solution is transferred out of the tank and a more uniform accumulation on the bottom than if the solids are left in the tank. Thus it is shown that the value of k-effective plus two standard deviations is less than 0.95 for all credible conditions.

Routine use of the 244-S tank does not involve the transfer of 244-TX solutions (containing Z-Plant plutonium wastes) into the tank, although the solution does go through a valve pit on top of the tank in making transfers to Tank 102-SY. The amount of plutonium transferred into the tank from the saltwells will be extremely small; thus, little or no buildup of plutonium is expected in the tank. If under special circumstances the 244-TX solution is to be transferred into or through Tank 244-S, special information, such as evaluating the amount of plutonium in the solutions being transferred and

determining the amount of plutonium hold up in the 244-S tank, will be requested by Criticality Engineering and Analysis (CE&A) prior to the transfer in order to evaluate the criticality potential of Tank 244-S. Thus, the criticality safety controls for the 244-S tank are judged to meet the contingency requirement of RHO-MA-136.

Altschuler (1980) concludes that a criticality safety evaluation should be made if solution is to be transferred from DCRT 244-TX into DCRT 244-S.

Altschuler (1980) investigated five contingencies. The following information is taken directly from his analysis.

Contingency 1: Exceeding the amount of fissile material allowed in DCRT 244-TX .

The D-5 Tank, which batch feeds the 244-TX tank, has two dry wells for neutron monitors and a limit of 400 g Pu. Therefore, exceeding 400 g is a violation of at least one contingency. Even were the 400 g optimally moderated (in ~13 L of water), optimally shaped (as a sphere), and totally reflected, it would still be critically safe ($k_{\text{eff}} < 0.95$).

There should be a minimum batch volume discharged in the 244-TX tank that will ensure a fissile concentration well below the minimum required for criticality, 7.0 g/L. Failure to transfer at least the minimum batch volume would be a second contingency so that the two contingency criteria are met.

Contingency 2: Exceeding the safe slab areal density or cylinder mass limit.

- (a) As a slab - The minimum critical areal density is 240 g/ft². Forty-five percent of this value, 108 g/ft² is considered safe. Assuming that the material settles evenly on the bottom of the 244-TX Tank, which has a projected area of 420 ft², the allowable inventory is 45.4 kg ²³⁹Pu. The 244-S tank bottom has an area of 176 ft² and the safe slab is 19.0 kg.
- (b) As a cylinder - If, instead of depositing evenly on the entire bottom surface of the tank, the material was deposited in a cylindrical segment at the bottom directly beneath the axis, the critical volume occupied by the fissile material at the minimum critical areal density could be as much as 2025 L. The allowable amount of plutonium in this cylinder is estimated to be 13.7 kg.

Contingency 3: Precipitation or settling in one spot.

Should the fissile material precipitate or settle out locally in one spot, it is expected to do so as a sludge of no more than 1.2 g/cm³ which is hemispherical in shape. (A hemisphere gives a more conservative result than would a sphere.)

A conservative critical volume and mass for a hemisphere is estimated to be 6.27 L and 7.5 kg, respectively. Forty-five percent of these values, 2.8 L and 3.4 kg, is considered safe.

Contingency 4: Resuspension of a uniform precipitate.

Unless the method of resuspension first assembles the allowable amount of fissile material for a uniform precipitate into a more reactive geometry, the system is safe since the conditions of the areal density are unchanged.

Contingency 5: Resuspension of a precipitate that has been accumulated in one spot.

This is the most limiting of the accidents considered. While 1.2 g/cm^3 is a rather wet sludge, it is quite far from optimum moderation (30 g Pu/L). Resuspension in such a manner by a jet of water that puffs up the accumulation, spreading it and increasing its moderation without immediately dispersing it, is potentially unsafe. Less than 20 L of such an optimally moderated mixture is critical and the presence of 3.8 kg allows considerable potential for criticality even if all of it is not optimally moderated and shaped. The 244-TX tank has 11 sluicing jets, three at each end and five along the center axis of the tank, just above the bottom. The use of these jets will minimize the possibility of any long-term accumulations and will keep the precipitate uniformly distributed. In addition, neutron monitors are in place, which permit detection of long term buildup.

The 244-S tank has no sluicing jets nor a permanent neutron monitor. Two drywells are provided so that portable neutron monitoring equipment may be used periodically or upon request. Operation of this tank is restricted to solutions that are low in precipitates and plutonium concentration. The contents of TK-244-TX will not normally be transferred into TK-244-S although the TK-244-TX to TK-102-SY transfer will drain into the tank. After each transfer, the line will be flushed to reduce the amount of Pu solids collected in TK-244-S. Under certain conditions, it may be necessary to transfer TK-244-TX solution into the 244-S tank. In this case the tank solution shall be allowed to settle and sluicing jets in TK-244-TX shall not be operated in order to reduce the amount of precipitate that will be transferred into TK-244-S.

Altschuler (1980) did not take into account neutron-absorbing characteristics of solids within the waste. The assumption of no neutron absorption by solids is very conservative.

Altschuler (1980) assumed the plutonium concentration in incoming waste is limited to 0.013 g/L (0.05 g/gal), the limit used for waste discharged from processing plants. If the 136,000 L (36,000 gal) DCRT 244-TX were filled at this incoming concentration, the plutonium inventory would be 1,800 g. The limit of 2,000 g was placed on the plutonium content to allow for a full tank at maximum concentration plus a small accumulation. However, in practice there is no way to determine the total inventory of the tanks with any certainty. The true basis for criticality safety is the assurance that waste discharged from the processing plants is in a form that cannot be made critical.

A2.4 NEUTRON MONITORS INSTALLED ON DCRT 244-TX

Neutron monitors were installed on DCRT 244-TX to provide the ability to detect plutonium buildup. Under normal operating conditions the potential for plutonium accumulation is extremely small, and accumulation may not actually be possible. Since waste in DCRT 244-TX comes from PFP where separated plutonium is handled, an off-normal condition leading to an unexpected discharge of plutonium was not ruled out when the evaluation was done in 1980 (Altschuler 1980). If particles can settle to the bottom and remain behind after waste is pumped out, the possibility must be considered that the same process might be repeated. Over time this might lead to an accumulation of plutonium in the tank. The inability to rule out plutonium accumulation or the possibility of a significant off-normal discharge of plutonium prompted installation of neutron monitors to indicate any increase in neutron background.

The neutron flux generated by plutonium would vary over a wide range depending on the distribution of plutonium, waste solids, and structural components. Because the distributions of plutonium and waste components are unknown, calibration of the neutron monitors to indicate the quantity of plutonium is not possible. For this reason, the goal of the monitor is to determine whether the background neutron flux has increased relative to earlier readings. An increase in neutron flux over time would indicate an increase in plutonium.

To prevent the buildup of plutonium, an agitator pump and sluicing nozzles are operated prior to a transfer from DCRT 244-TX into DST SY-102. Operation of the agitator pump is performed according to Tank Farm Operating Procedure TO-430-480, Section 5.2.19 (LMHC 1999). This procedure requires that the agitator pump be operated for a minimum of 1/2 hour before initiating waste flow to DST SY-102.

A2.4.1 Potential For Plutonium Segregation

Whyatt et al. (1996) examined the potential for segregation (separation) of plutonium from other waste solids in DST SY-102. This study is directly applicable to DCRT 244-TX. The degree to which the plutonium will segregate upon settling depends upon the sludge particle size distribution, the plutonium particle size distribution, and the plutonium particle density. The distribution of particle sizes in samples of DST SY-102 sludge is shown in Table A-1, based upon DiCenso et al. (1995). The distribution of plutonium particle sizes should be much the same as the distribution of sludge particles as a whole.

**Table A-1. Particle Size Volume Distribution
for Double-Shell Tank SY-102 Sludge¹**

Size, μm	Volume %
100 - 175	3
60 - 100	25
50 - 60	30
40 - 50	19
30 - 40	8
20 - 30	8
10 - 20	7
0 - 10	0.5

Note:

¹Whyatt et al. (1996)

If a high proportion of plutonium particles were to fall in a narrow range of sizes, a plutonium rich layer might form. Assuming a sludge particle size distribution provided by DiCenso et al. (1995), Whyatt et al. (1996) concluded that the maximum increase in plutonium concentration occurs when the PuO_2 particle size is 15 μm . A concentration factor, defined as the plutonium concentration after settling divided by the average plutonium concentration upon entering the tank, is used to describe the increase in plutonium. For PuO_2 particles with densities of 11.4 g/cm^3 and 6.0 g/cm^3 , the maximum concentration factors are given as 25.7 and 15.2, respectively. The initial plutonium concentration was assumed to be 0.154 g/L . If a uniform distribution of plutonium particle sizes is assumed over the range from 0 - 137 μm , the maximum increase in plutonium concentration at any depth in the settled sludge is 3. Whyatt et al. (1996) concluded that the actual concentration factor should not exceed this value.

Two conclusions can be drawn. First, for the expected sludge and plutonium particle sizes, the settled layers with the highest plutonium concentration should be close to the top. Second, the plutonium concentration factor in any layer should not exceed 3. These two conclusions indicate that it would be unlikely for plutonium to remain in DCRT 244-TX after the waste is pumped out.

A2.4.2 Reevaluation of Need for Neutron Monitors

To form a plutonium rich layer on the bottom, the average size of plutonium particles would have to exceed 140 μm . Such a layer might remain behind after the waste is pumped out. However, the actual plutonium particle sizes are considerably smaller than 140 μm , and only a small fraction of the plutonium would be expected to lie near the bottom of the sludge.

In computer simulations, Whyatt et al. (1996) models particles as tiny spheres without any association with each other. This does not provide a realistic assessment of the behavior of particles because it ignores the chemical form of the plutonium precipitate. The model used by Whyatt et al. (1996) provides a conservative envelope over the range of possible concentration factors. Bratzel et al. (1996) concluded that waste particles form into tightly bound agglomerates that prevent any significant segregation of plutonium. This indicates that the actual plutonium concentration factor is actually much smaller than the value of 3 reported by Whyatt et al. (1996). The chemistry of tank waste is discussed in Appendix D.

In conclusion, the computer simulation by Whyatt et al. (1996) and the chemistry study by Bratzel et al. (1996) clearly show that an accumulation of plutonium in the bottom of DCRT 244-TX cannot occur. Even without considering chemical bonding of particles, the particle size distribution ensures that only insignificant accumulation of plutonium is likely to occur near the bottom of the sludge. When the tendency to form agglomerates is taken into account, the ability to form plutonium rich layers is greatly diminished below the already small value guaranteed by the small particle sizes. The potential for plutonium to remain behind is very small. Based upon the extensive chemistry information now available, there is no longer a need to provide neutron monitors on DCRT 244-TX.

A3.0 242-A EVAPORATOR FACILITY

A3.1 DESCRIPTION OF PROCESS

Blyckert (1981) wrote the criticality safety evaluation for the 242-A and 242-S Evaporator facilities. He describes the evaporation process used to remove water from the waste.

The 242-A and 242-S evaporator-crystallizers utilize the same evaporation process. A conventional forced circulation vacuum-evaporation system is used to concentrate radioactive waste solutions. Feed to the evaporator is pumped to the reboiler where it is heated. The heated slurry is discharged from the reboiler to the vapor-liquid separator vessel, which is maintained at a reduced pressure. Under this process condition, a fraction of the water in the salt slurry concentrate flashes to vapor and is drawn into the primary condenser. As evaporation takes place, the concentration of salt increases in the slurry. The slurry is then pumped to underground storage tanks, allowed to settle, and the supernatant liquid is pumped back as feed to the evaporator.

Feed solutions to the evaporators are sampled and analyzed to assure that the fissile material concentration does not exceed 0.01 grams per gallon. Continuous operation of the evaporator-crystallizers concentrate the solution by a factor of two. (A batch would be required to concentrate the solution to twenty percent of its original volume, but this is difficult to accomplish.) Allowable content of fissile material for feed to storage tanks is 0.05 g/gal.

A3.2 LIMITS

Only the 242-A Evaporator remains in operation. The following limits for the 242-A Evaporator facility are included in CPS-T-149-00010 for the tank farms. CSAR 81-022 (Blyckert 1981) provides the basis for these limits.

1. Plutonium concentration
in feed solution: < 0.005 g/L (0.019 g/gal)
2. Before an evaporator campaign begins, a determination shall be made and documented of the expected quantity and concentration of plutonium which will be in the feed and slurry tanks at completion of the run.
3. After an evaporator campaign, a determination shall be made of the actual quantity and concentration of plutonium in the feed and slurry tanks, either by analysis or calculation.

A3.3 BASIS FOR LIMITS

Blyckert (1981) evaluated evaporator operation and concluded that there is no potential for criticality.

Fissile material content in the feed is not more than 0.01 g/gal. If the concentration of fissile material in the feed to the evaporator were double that of the allowable concentration, normal operation would not result in evaporator bottoms in excess of 0.05 g/gal, while if the system were operated in a batch mode the fissile material content of that limited volume could reach 0.1 g/gal. While this fissile material content would be double the allowable concentration for feed to storage tanks, it is less than three percent of that required to be reported and less than two-thirds of one percent of fissile material concentration in waste which might become critical, 4.3 g/L (Carter 1979). There is no known way for feed available to the evaporators to be handled to cause the system to become critical. Consequently, there are no other criticality safety restrictions for the evaporator operations. It should be noted that because of the high pH (>8) of the solutions, the plutonium content of the waste being concentrated is low. It should also be noted that criticality safety with 4.3 g/L of plutonium would require over 200 kg of plutonium.

With the maximum concentration of fissile material in the feed solution limited to 0.01 g/gal, we conclude there is no criticality potential and the evaporators meet the safety requirements of RHO-MA-136 (RHO 1981)

Blyckert (1981) examined the question whether plutonium might plate out in the evaporator and concluded that the quantity of plutonium that might collect would be too small to create a potential for criticality.

Plating or scaling of waste on the surface of the evaporator does not occur except at high specific gravity solution (>1.67) where 80 percent of the solids are sodium nitrate; the remainder is composed mainly of carbonates and phosphate. The plutonium concentration would not be increased greater than an order of magnitude. From an experimental run, 3,000 pounds of scale were formed which would give 0.6 g of plutonium based on the feed concentration of 0.0001 g of plutonium per liter of slurry. If the feed contained the maximum concentration permitted (0.01 g Pu/gal), 16 g of plutonium would be expected to be incorporated in the scale. This amount of plutonium is of no criticality safety concern, nor is double the amount. The scale easily dissolves in water. Plating out of scaling is therefore not a criticality safety problem.

When averaged over all waste in tank farms, the mass ratio of solids to plutonium is estimated to be 60,600 (see Appendix C7.0). The smallest solids/plutonium mass ratio is found in waste from PFP, but even this waste has a mass ratio greater than 10,000. If all liquid were to evaporate from the supernate, the suspended and dissolved portion of the waste would be deposited as solids. It is expected that the mass ratio of solids to plutonium in this deposit would be similar to that for the sludge in the tank from which the supernate is taken. The solids/plutonium mass ratio for any supernate is expected to be at least 10,000. This is 20 times larger than required to ensure subcriticality in homogeneous waste.

In practice, the solids/plutonium mass ratio in supernate may not be well known. It is this uncertainty that lead to the use of a limit of 0.005 g/L on plutonium concentration in the evaporator feed solution. This limit is small enough to ensure subcriticality when there is little neutron absorbing solids and after most liquid has been removed.

A4.0 FACILITY DISCHARGE LIMITS

The Plutonium-Uranium Extraction (PUREX) Plant and the PFP were the two primary sources of waste now in tank storage. This section discusses waste discharge limits used by these facilities.

A4.1 LIMITS AT THE PUREX PLANT

The PUREX Plant is shut down and no longer sends waste to tank farms. The primary control used at PUREX to ensure criticality safety was limitation of plutonium concentration to a maximum of 0.013 g/L (0.05 g/gal). This low concentration provided a margin of safety to allow for an increase in concentration caused by settling in the storage tanks. If the plutonium inventory in the receiver tank is assumed to be 200 kg (a conservative assumption), the minimum critical concentration (see Figure B-2) is about 3.7 g Pu/L, assuming CWM solids. A concentration of 0.013 g/L is 280 times smaller than the minimum concentration that can be made critical. The solids portion of waste provides assurance that settling and liquid removal cannot cause the plutonium to concentrate to unsafe levels. The volume of the solids after all liquid has been removed establishes a lower limit on the volume in which the plutonium can be concentrated.

The PUREX Plant is the source of the majority of waste in tank storage. Processes used at the PUREX Plant guarantee that waste will have a large proportion of solids. These processes have always been governed by flowsheets that are well regulated and ensure predictability. The proportion of solids in this waste is far greater than necessary to ensure subcriticality. This remained true even following upset conditions. Since the required quantity of solids was guaranteed by the process to always be present, there was no need to specify a limit in the CPS for the solids content. Plutonium at the PUREX Plant has always been associated with a large proportion of ^{238}U .

The large margin of subcriticality in PUREX Plant waste has been verified by chemical analyses of tank waste samples. The highest plutonium concentration yet reported for waste originating at PUREX, based upon chemical analysis, is around 0.3 g/L, a value one-ninth of the minimum critical concentration. When taken as a whole, the solids/plutonium mass ratio for the tank farms is at least 60,600 (see Appendix C7.0), far more than required to assure subcriticality. This large mass ratio provides a high level of assurance that PUREX Plant waste in tank storage will remain well below subcritical criteria.

A4.2 LIMITS AT THE PLUTONIUM FINISHING PLANT

The Plutonium Finishing Plant (PFP) handles plutonium in a purified (separated) form. The possibility that plutonium might be discharged with a low proportion of solids is a consideration. For this reason, PFP discharge limits include a limit on the minimum solids content.

A4.2.1 CPS Limits

Operations in the 241-Z Solution Disposal Facility are governed by the following limits provided in CPS-Z-165-80741, Rev. B-2 (B&W 1999a):

Total plutonium for any 241-Z tank (maximum):	400 g
Plutonium concentration of solutions transferred to Tank Farms (maximum):	0.0330 g/L 0.125 g/gal

Iron hydroxide neutron absorber shall be greater than 0.7% by volume (centrifuged OR calculated) in solutions transferred to Tank Farms.

The Plutonium Finishing Plant procedure ZO-101-010, *Transfer TK-D5 to Tank Farms* (B&W 1999b), ensures compliance to the CPS.

Prior to 1999, the plutonium concentration limit for solution transferred to tank farms was 0.05 g/gal (maximum), and the solids content was required to exceed 0.6 vol%. At that time, the iron concentration was required to exceed 35 g/L of solids.

A4.2.2 Iron/Plutonium Mass Ratio

NaOH, Fe(NO₃)₃, and NaNO₂ are added to TK-D5, and these cause the precipitation of iron hydroxide. Iron hydroxide has a specific gravity of 3.4 and a molecular weight of 89.86. When iron hydroxide occupies 0.7 vol%, its concentration is 23.8 g/L, and the iron concentration is 14.8 g/L. The minimum allowable iron/plutonium mass ratio is found by dividing the minimum iron concentration by the maximum plutonium concentration. In other words, when 14.8 g iron/L is divided by 0.125 g Pu/L, the iron/plutonium mass ratio is found to be 118, and this is the smallest value possible without violating limits in the PFP CPS.

When the plutonium concentration is less than 0.001 g/L, the CPS for tank farms allows transfers without any consideration of solids content. When the plutonium concentration exceeds 0.001 g/L, the CPS requires an iron/plutonium mass ratio greater than 160. This is greater than the minimum permitted by the PFP CPS. In practice, however, the plutonium concentration will usually be much less than 0.125 g/L, and the iron/plutonium mass ratio will be larger than 160.

To ensure compliance to the tank farms CPS, PFP fills out a Criticality Data Sheet (CDS) before discharge to tank farms. The iron/plutonium mass ratio is calculated and recorded. If the ratio is less than 160, tank farms will not receive the waste. When the plutonium mass exceeds 50 g, the tank farms CPS also requires the sum of mass ratios for waste components to exceed 2.

PFP waste is sent to DST SY-102. For this tank the solids content has been reported to be 9.8 vol%, and the solids/plutonium mass ratio is reported to be 12,555 (LMHC 1998a), a value 21 times larger than the subcritical limit (see Appendix C7.1.1). Therefore, the average solids content in batches of PFP waste is considerably greater than the minimum permitted under the CPS limits.

A4.2.3 Mixing

PFP procedure ZO-101-010 (B&W 1999b) provides warning statements concerning mixing before discharge, as follows:

TK-D5 agitator shall be operating and has been operating for at least 10 minutes prior to start of transfer.

TK-D5 agitator shall be operating during transfer to Tank Farms when WF is 10 divisions or higher.

The Weight Factor (WF) is a measurement of solution depth using a calibrated sight glass. These warning statements require the agitator be started more than 10 minutes before beginning the transfer and to remain operating until after the WF falls below the 10th division on the sight glass. At this time the agitator is turned off. Operation of the agitator keeps the solids suspended and mixed during the transfer. This ensures a relatively uniform plutonium concentration throughout the transfer and prevents localized volumes of high concentration.

A5.0 REFERENCES

- Altschuler, S. J., 1980, *CSAR 80-015: 244-TX and 244-S Salt Well Receiver Tanks*, SD-SQA-CSA-20110, Rockwell Hanford Operations, Richland, Washington.
- B&W, 1999a, *241-Z Solution Disposal Facility*, CPS-Z-165-80741, Rev. B-2, Babcock & Wilcox Hanford, Richland, Washington, June 9, 1999.
- B&W, 1999b, *Transfer TK-D5 to Tank Farms*, PFP Procedure ZO-101-010, Rev. D, Change 1, Babcock & Wilcox Hanford, Richland, Washington, October 4, 1999.
- Blyckert, W. A., 1981, *Waste Evaporators 242-A, 242-S*, CSAR 81-022, SD-SQA-CSA-20112, Rockwell Hanford Operations, Richland, Washington.
- Blyckert, W. A., 1983, *Operation of Tank 244-BX*, Addendum 1 to CSAR 81-015, SD-QA-SA-0111, Rockwell Hanford Operations, Richland, Washington.
- Bratzel, D. R., W. W. Schulz, R. Vornehm, and A. E. Waltar, 1996, *Tank Farm Nuclear Criticality Review*, WHC-SD-WM-TI-725, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- Carter, R. D., G.R. Kiel, and K. R. Ridgway, 1969, *Criticality Handbook*, ARH-600, Vol. I, p. III.A.8.100-3, Atlantic Richfield Hanford Company, Richland, Washington.
- Carter, R. D., 1979, *Underground Waste Storage Tanks and Associated Equipment*, CSAR 9-007, SD-SQA-CSA-20108, Rockwell Hanford Operations, Richland, Washington.
- Carter, R. D., 1983, *Underground Waste Storage Tanks and Associated Equipment*, Addendum 1, CSAR 79-007, SD-SQA-CSA-20109, Rockwell Hanford Operations, Richland, Washington.
- Carter, R. D., 1987, *Operation of Tank 244-U*, Addendum 2, CSAR 80-015, SD-QA-SA-63, Rockwell Hanford Operations, Richland, Washington.
- Carter, R. D., 1994, *Addendum 2A, CSAR 79-007: Underground Waste Storage Tanks and Associated Equipment*, WHC-SD-SQA-CSA-20365, Westinghouse Hanford Company, Richland, Washington. Written in 1984, but transmitted as EDT 157914 in 1994.

- DiCenso, A. T., L. C. Amato, and W. I. Winters, 1995, *Tank Characterization Report for Double-Shell Tank 241-SY-102*, WHC-SD-WM-ER-366, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- LMHC, 1998a, *Plutonium Inventory Database*, HISI 11350, Lockheed Martin Hanford Corporation, Richland, Washington.
- LMHC, 1998b, *Waste Storage Tanks and Associated Equipment*, CPS-T-149-00010, Rev. I-0, Lockheed Martin Hanford Corporation, Richland, Washington.
- LMHC, 1999, *244-TX DCRT to 241-SY-102 Slipstream Transfer Via 244-S DCRT*, TO-430-480, Lockheed Martin Hanford Corporation, Richland, Washington.
- Reardon, W. A., 1987, *Operation of Tank 244-U*, CPS-T-149-00061, Addendum 2 to CSAR 80-015, Rockwell Hanford Operations, Richland, Washington.
- RHO, 1980, *Criticality Prevention Specification T-6: Operation of Tanks 244-TX and 244-S*, Rev. 0, Rockwell Hanford Operations, Richland, Washington.
- RHO, 1981, *Nuclear Criticality Safety Standards*, Rockwell Hanford Operations, Richland, Washington.
- Rogers, C. A., 1994, *CSER 94-004: Criticality Safety of Double-Shell Waste Storage Tanks*, WHC-SD-SQA-CSA-20368, Westinghouse Hanford Company, Richland, Washington.
- WHC, 1980, *Operations of Tanks 244-TX and 244-S*, CPS-T-149-00060, Rev. A-0, Westinghouse Hanford Company, Richland, Washington.
- WHC, 1987, *Operation of Tanks 244-U and 244-BX*, CPS-T-149-00061, Rev. B-0, Westinghouse Hanford Company, Richland, Washington.
- WHC, 1988, *Waste Storage Tanks and Associated Equipment*, CPS-T-149-00010, Rev. D-0, Westinghouse Hanford Company, Richland, Washington.
- Whyatt, G. A., R. J. Serne, S. V. Mattigod, Y. Onishi, M. R. Powell, J. H. Westsik, Jr., L. M. Liljegren, G. R. Golcar, K. P. Recknagle, P. M. Doctor, V. G. Zhirnov, J. Dixon, D. W. Jeppson, and G. S. Barney, 1996, *The Potential for Criticality in Hanford Tanks Resulting from Retrieval of Tank Waste*, PNNL-11304, Pacific Northwest National Laboratory, Richland, Washington.

This page intentionally left blank.

APPENDIX B

PARAMETERS IMPORTANT TO CRITICALITY SAFETY

This page intentionally left blank.

B.0 PARAMETERS IMPORTANT TO CRITICALITY SAFETY

When a parameter is controlled for criticality safety, the range over which that parameter can be varied while maintaining the system subcritical must be known. The value at which the system is just critical is referred to as its critical value.

The subcritical limit is the largest (or sometimes the smallest) value of a parameter that assures subcriticality, after taking into account uncertainties in the method used to determine the value of the parameter. In this evaluation a "critical" parameter generally is taken to mean the subcritical limit and, therefore, describes a system that is actually subcritical. For this evaluation, the neutron multiplication constant (either k_{eff} or k_{∞}) for a subcritical limit will not exceed 0.95 with a 95 percent confidence level.

When a critical parameter is reported in an authoritative document, such as an American Nuclear Society Standard or the *Nuclear Safety Guide* (Thomas 1978), that parameter may be used as a subcritical limit, even though the corresponding neutron multiplication constant might exceed 0.95. Two important examples are the experimentally determined plutonium minimum critical concentration of 7.2 g/L in water (Paxton and Pruvost 1987) and the minimum critical plutonium areal density provided by Carter et al. (1969) as 240 g Pu/ft² (2,582 g/m²).

B1.0 THE CONSERVATIVE WASTE MODEL

Tank waste is a complex mixture of components, and our knowledge of the distribution of components is incomplete. The high degree of uncertainty in composition introduces considerable complexity into the determination of critical parameters. For this reason, a waste model, known as the conservative waste model (CWM), has been developed (Rogers 1993). This model is used as the basis for calculating conservative subcritical limits. Rogers (1993) provides subcritical limits used as the basis for limits and controls in this CSER.

B1.1 BASIS FOR CONSERVATIVE MODEL

The conservative waste model composition was developed by tabulating available waste analyses and selecting the highest concentration for components that are good neutron scatterers and poor neutron absorbers, and the lowest concentration of components that are good neutron absorbers. The conservative waste model is based upon 28 waste compositions analyzed for 16 SSTs. The primary goal of this waste model is to provide a waste composition known to have a smaller macroscopic absorption cross section than that of any actual waste. Critical parameters were calculated using optimal water moderation. In this way, subcritical limits calculated using the conservative waste model are conservative relative to the actual critical values for any actual waste.

B1.2 MEANING OF CONSERVATIVE

As used in this report, conservative values, estimates, or assumptions are ones that produce "worse" (more limiting or severe in some way) results than the actual case. Conservative estimates can be used to set limits or to simplify analyses of criticality safety. Such limits and analyses are used to make sure that, in spite of uncertainties, the real case is always as safe or safer than the model. Thus, if the model can be shown to be safe, the safety of the actual situation has been shown.

The CWM is intended to

- Simplify the evaluation of tank waste.
- Address features of the waste that are significant with respect to criticality.
- Always produce conservative analysis results (that is, operating limits derived from the model are as safe or safer than the limits would be if derived from the actual waste).

B1.3 COMPOSITION OF CONSERVATIVE WASTE MODEL

Table B-1 shows the composition of the CWM (Rogers 1993). Only the waste solids are included (with a dry density of 1,200 g/L). The concentrations of plutonium and water are independent variables not part of the waste model.

For waste to be bounded by the CWM envelope, the neutron absorption in one liter after all water has been removed must exceed the neutron absorption in 1,200 g of conservative model waste solids. The macroscopic absorption cross section for the CWM solids is 0.01096 cm^{-1} . Actual waste is expected to have greater absorption. Although theoretical models of waste having less absorption than this are possible, it is expected that all actual tank waste will satisfy this criterion.

Although only samples from SSTs were used to derive the CWM, critical parameters derived from the model may be applied to all tank waste. This is because DST waste is similar to that in SSTs and there is considerable conservatism included in the waste model. The high proportion of oxygen and the absence of hydrogen in the CWM also provide additional conservatism for all waste types likely to be present in the tanks, even those having a considerably different makeup from the analyzed samples used to develop the model.

Table B-1. Composition of Conservative Waste Model¹

Component	wt%
Oxygen (O)	40.7
Phosphorus (P)	6.9
Silicon (Si)	3.8
Sodium (Na)	21.5
Aluminum (Al)	7.2
Nitrogen (N)	0.0
Iron (Fe)	19.9

Note:

¹Rogers (1993)

B1.4 OPTIMAL MODERATION

Although not actually water, the liquid portion of waste generally contains a large water fraction. From a criticality safety perspective, it is conservative to treat this liquid as pure water. This is true because water is a better neutron moderator and a poorer neutron absorber than the other liquid components. In this discussion it will be called water.

At low plutonium concentrations characteristic of tank waste, the water content that results in a minimum critical plutonium concentration is very low. In fact, according to Rogers et al. (1996) at the subcritical limit concentration of 2.6 g Pu/L with CWM solids, optimal moderation means the solids are completely dry. In other words, at the lowest possible critical plutonium concentration, waste contains no water at all. The large fraction of solids provides moderation, while hydrogen in the water increases neutron absorption. With addition of water, the fraction of neutrons absorbed in elements other than plutonium is increased, and this loss of neutrons lowers the neutron multiplication constant.

At 2.6 g Pu/L, criticality is possible only with CWM solids that are completely dry. When the plutonium concentration is 3 g/L, the water content must be less than 7 wt% for criticality to be possible. As the plutonium content increases, so does the maximum water content at which an infinite volume of waste can go critical. At 4 g Pu/L, criticality is not possible when the water content exceeds 25 wt%. At plutonium concentrations above 6.5 g/L, much higher than found in tank waste, optimal water occurs when the waste is fully water-saturated.

B1.5 DENSITY OF DRY SOLIDS

For CWM solids, a bulk dry solids density of 1,200 g/L is assumed after all water (liquid) has been removed. This density was selected because it is a lower density than occurs for most real waste solids. However, the possibility is considered that the bulk density (i.e., particle density times packing fraction) of settled, loosely-packed, dense particles might under some circumstances be less than 1,200 g/L.

When suspended in water, the density of solids is reduced. However, solids cannot be suspended unless the void space is filled with water. The presence of the water ensures a higher plutonium critical concentration. When solids are suspended in water, criticality would only be possible for a plutonium concentration significantly greater than 2.6 g/L. In pure water the subcritical limit plutonium concentration, based on a k_{∞} of 0.935, is 6.5 g/L.

In deriving the plutonium minimum critical concentration of 2.6 g/L, several conservative assumptions were made. First, a conservative upper safety limit (USL) of 0.935 on k_{∞} was used when determining the subcritical limit on plutonium concentration. In other words, a calculated k_{∞} of 0.935 was assumed to be critical, thereby resulting in a calculated minimum critical concentration smaller than the actual value. Second, the total macroscopic neutron absorption cross section of 0.01096 cm^{-1} for CWM solids is less than that of an equal mass of actual waste. Third, actual waste is expected to have a dry solids density greater than 1,200 g/L. Fourth, optimal moderation for low plutonium concentrations occurs when the waste is extremely dry. In practice, a water content less than 10 wt% is extremely unlikely, whereas a 10 wt% water content would be sufficient, if added to CWM solids, to increase the plutonium minimum critical concentration to more than 3 g/L.

Per unit mass, neutron absorption in water is more than twice that of CWM solids. Because the plutonium minimum critical concentration of 2.6 g/L occurs for completely dry solids, any water in real waste can actually be thought of as being equivalent to an increase in the dry solids density. In fact, 100 g of water increases the minimum critical concentration as much as 200 g of dry solids.

These conservative assumptions ensure that the dry solids density would have to fall well below 1,200 g/L, and the water content would have to be extremely low before the minimum critical plutonium concentration would actually fall below 2.6 g/L. The probability of any actual waste achieving a composition capable of lowering the plutonium minimum critical concentration below this value is extremely small.

B2.0 PLUTONIUM CRITICAL PARAMETERS

Criticality Parameters for Tank Waste Evaluation (Rogers et al. 1996) is a source of information for evaluating tank waste.

B2.1 CONSERVATIVE ASSUMPTIONS

Plutonium in tank waste is distributed non-uniformly, and there is considerable uncertainty as to the distribution of waste components. Conservative assumptions are used to compensate for the lack of information.

The fissionable isotopes potentially present in waste are ^{233}U , ^{235}U , ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu , and ^{242}Pu . The plutonium isotopes ^{238}Pu , ^{240}Pu , and ^{242}Pu are fissionable, but not fissile. The presence of even a small quantity of moderating material increases the critical mass of these isotopes rapidly until criticality is not possible with any of them alone. In the tank waste environment, ^{238}Pu , ^{240}Pu , and ^{242}Pu act as mild poisons, and their presence increases the quantity of ^{239}Pu required to achieve criticality. Even though ^{241}Pu is more reactive than ^{239}Pu , it comprises only a couple percent of the plutonium, and its importance to criticality is more than offset by the presence of the non-fissile isotopes. It is conservative to assume that all plutonium isotopes are lumped together and replaced by ^{239}Pu . The term "plutonium," unless specifically stated otherwise, refers to all plutonium isotopes replaced by an equal quantity of ^{239}Pu .

Because ^{235}U is almost always combined with a ratio of ^{238}U sufficient to ensure that the uranium is subcritical, the ^{235}U can often be ignored (see Section B5.0 for more details). However, the ^{235}U content in excess of 1.00 wt% is added to the plutonium inventory, unless a specific analysis provides a basis for doing otherwise. In addition, when the uranium/plutonium subcritical mass ratio is used to demonstrate subcriticality, the ^{235}U in excess of 0.72 wt% must be added to the plutonium quantity.

^{233}U appears only in a few tanks, and when present it can be treated as if it were ^{239}Pu , and also added to the quantity of plutonium. That is to say, the ^{233}U and ^{235}U may be considered to be equivalent to ^{239}Pu . For these reasons the nuclear criticality safety of tank waste is usually discussed solely in terms of plutonium, and the plutonium content is understood to mean the "plutonium equivalent" content.

At fissile concentrations above 50 g/L, ^{233}U can have a smaller critical mass than ^{239}Pu . However, in a large volume the minimum critical concentration for ^{233}U is larger than for ^{239}Pu . Over the range of fissile concentrations found in waste, the replacement of ^{233}U with ^{239}Pu is conservative.

B2.2 PLUTONIUM IN WATER

Criticality is not possible for plutonium in a plutonium-water mixture unless the plutonium concentration exceeds 7.2 g/L. Supernate is composed largely of water, has a density that always exceeds that of water, and contains effective neutron absorbers. The plutonium critical concentration in supernate should never be less than that for pure water.

Rogers (1993) concludes that the minimum critical mass of plutonium in waste is 520 g when the density of waste solids is less than 1,200 g/L, and at least 750 g when the density of waste solids is more than 1,200 g/L. Although the minimum critical mass is often an important parameter in assessing criticality safety, it is of limited use when evaluating tank waste because the quantity of plutonium in a tank usually exceeds it.

To achieve criticality with a mass of plutonium equal to the minimum possible value of 520 g would require that the plutonium concentration be 30 g/L (113 g/gal). There are no known mechanisms capable of concentrating plutonium in waste tanks to such a high value.

B2.3 PLUTONIUM IN WASTE

A summary of critical parameters for plutonium in CWM and in water solution is shown in Table B-2.

For criticality to be possible in tank waste, it is necessary for the plutonium concentration to be at least 2.6 g/L (9.84 g/gal) over a large volume. This value is based upon the CWM and occurs in waste solids that are completely dry. For criticality to occur in a small volume, the plutonium concentration must be much greater than this value.

The minimum subcritical solids/plutonium mass ratio for the CWM is 476. Section B4.2 provides a discussion of this subcritical limit and recommends that the more conservative subcritical limit mass ratio of 770 be used as a generalized limit for solids.

To achieve criticality with waste having a plutonium concentration of 4.0 g/L requires a 4.7-m (15.4-ft) diameter, 53,750-L (14,200-gal) sphere containing 215 kg of plutonium.

For a slab of waste to go critical with 4 g Pu/L, it would have to be at least 2.1 m (7.0 ft) thick. This is a conservative value obtained by applying the formula for buckling conversion to the critical sphere.

At 10 g Pu/L, the critical mass in tank waste is approximately 3.0 kg of plutonium in a volume of 300 L (79.3 gal).

Table B-2. Plutonium Subcritical Limits

Parameter	Subcritical Limit
Conservative Waste Model	
Infinite critical concentration	2.6 g Pu/L (max.)
Solids/Plutonium mass ratio	770 (min.) ^a
Plutonium/Solids mass ratio	2.1 g/kg (max.)
Critical mass at 4.0 g Pu/L	215. kg Pu (max.)
Critical mass at 10.0 g Pu/L	3.0 kg Pu (max.)
Sphere diameter at 4.0 g Pu/L	4.7 m (15.4 ft) (max.)
Slab thickness at 4.0 g Pu/L	2.1 m (7.0 ft) (max.)
Critical areal density	240 g/ft ² (max.)
Plutonium-Water Solution	
Infinite critical concentration	7.2 g Pu/L (max.)
Critical areal density	240 g/ft ² (max.)

Note:

^aThis is the subcritical limit for the natural uranium/plutonium mass ratio and is more conservative than the CWM solids/plutonium subcritical limit mass ratio of 476.

max. = maximum

min. = minimum

The areal density includes all plutonium above a unit area of floor, including plutonium located all the way to the top surface. The minimum critical areal density of plutonium in water is 2,582 g Pu/m² (240 g Pu/ft²) (Carter et al. 1969). This is the smallest areal density that can be made critical.

B2.4 PARAMETRIC STUDIES**B2.4.1 k_{∞} for Plutonium in CWM Solids**

Schwinkendorf (1994a, 1994b) calculates k_{∞} for plutonium in CWM solids for plutonium concentrations over the range of 1 to 8 g/L. In addition, he calculates critical dimensions and masses for spheres, hemispheres, and cylinders. Solids content is varied between 0 and 75% of the total waste density with the remaining volume filled with water. k_{∞} is found to be larger when the solids content is larger. Above 7 g Pu/L, criticality is possible over the entire range of solids content, including pure water without any solids. When the solids are very dry, k_{∞} can exceed 1.2 at 7 g Pu/L.

B2.4.2 Sphere Minimum Critical Parameters

Figures B-1 through B-4, taken from Rogers et al. (1996), show minimum critical parameters as a function of plutonium concentration for three postulated waste compositions:

(1) CWM solids with water optimized; (2) pure water; and (3) a solution of sodium compounds intended to simulate "actual" waste as a saturated supernatant liquid, based on a composition described by Reynolds and Herting (1984). A 25-cm-thick close-fitting water reflector is assumed. Water content is varied over the entire range between 0 and 100 wt%.

Figure B-1 shows the sphere minimum critical diameter as a function of plutonium concentration. The asymptote for the plutonium concentration in water is shown to be 6.5 g/L, based on a subcritical limit on k_{∞} of 0.95. This is more conservative than the published subcritical limit of 7.2 g/L based on experiments.

Figure B-2 shows the sphere minimum critical mass as a function of plutonium concentration. The plutonium subcritical limit concentration in dry CWM solids is 2.6 g/L. When water is added, the plutonium minimum critical concentration increases. When the plutonium concentration exceeds 8 g/L, critical parameters for the CWM become the same as an optimal water system.

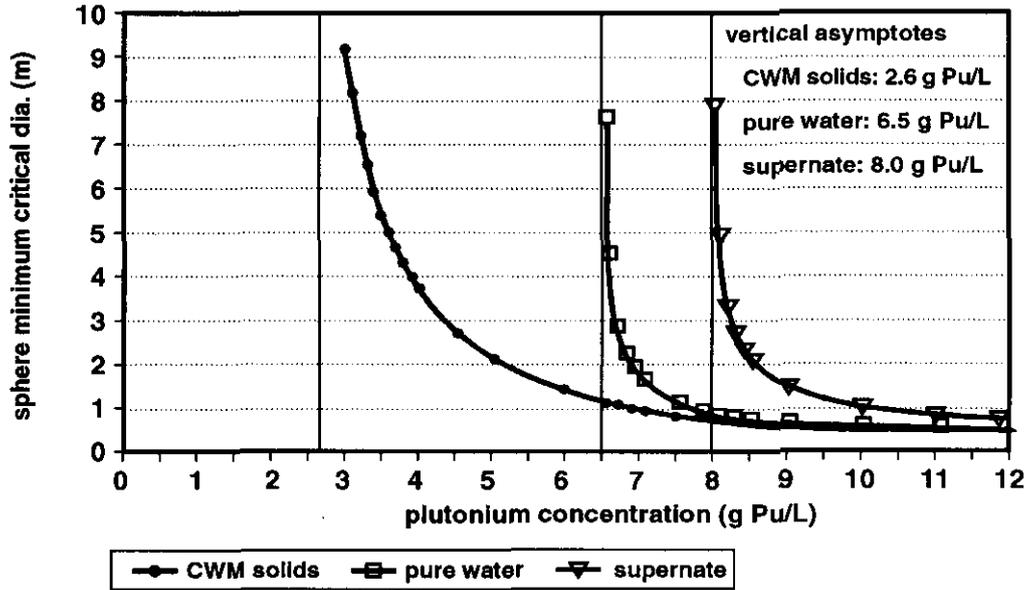
B2.4.3 Slab Minimum Critical Parameters

Figure B-3 shows the slab minimum critical thickness as a function of plutonium concentration. Figure B-4 shows the slab minimum critical mass as a function of plutonium concentration in a 22.9-m (75-ft) diameter tank. The plutonium mass is calculated assuming the waste covers the entire area of a 22.9-m-diameter tank at a uniform depth. A 25-cm-thick water reflector is assumed on top and bottom.

The minimum subcritical mass in a uniform slab is shown conservatively in Figure B-3 to be somewhat less than 1,000 g. When a plutonium minimum critical areal density of 2,582 g/m² is assumed, the minimum critical mass is found to be 1,060 kg. This quantity exceeds the estimated total inventory for all Hanford Site waste storage tanks combined, and is 10 times greater than the largest inventory for any single tank.

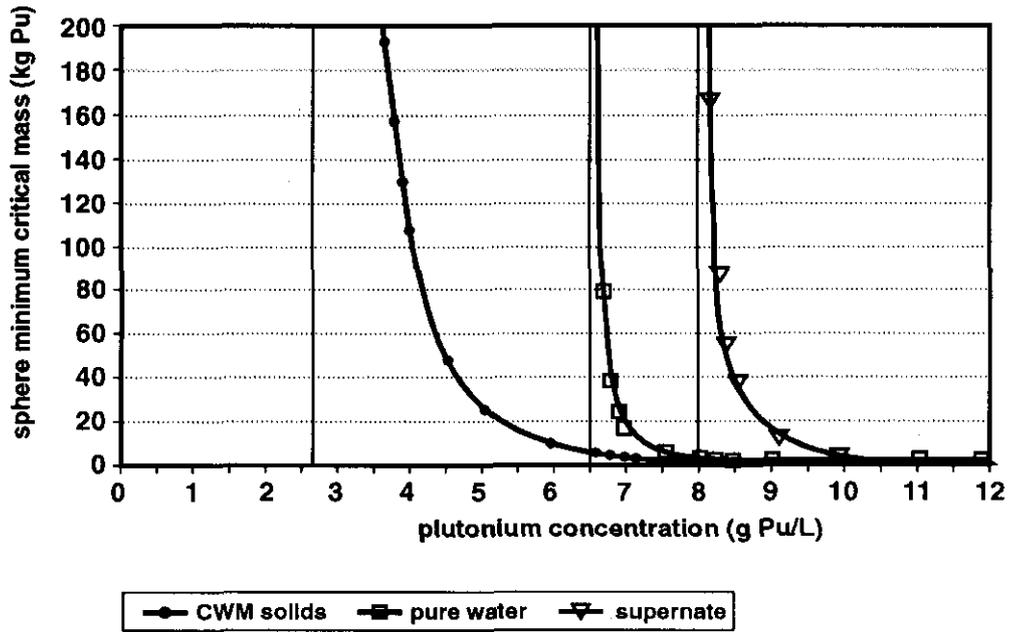
For a waste mixture, the plutonium critical areal density is greater than that for plutonium in pure water. It is conservative to use the minimum critical areal density for plutonium in water for criticality safety evaluation of waste.

Figure B-1 Sphere Minimum Critical Diameter Versus Plutonium Concentration.



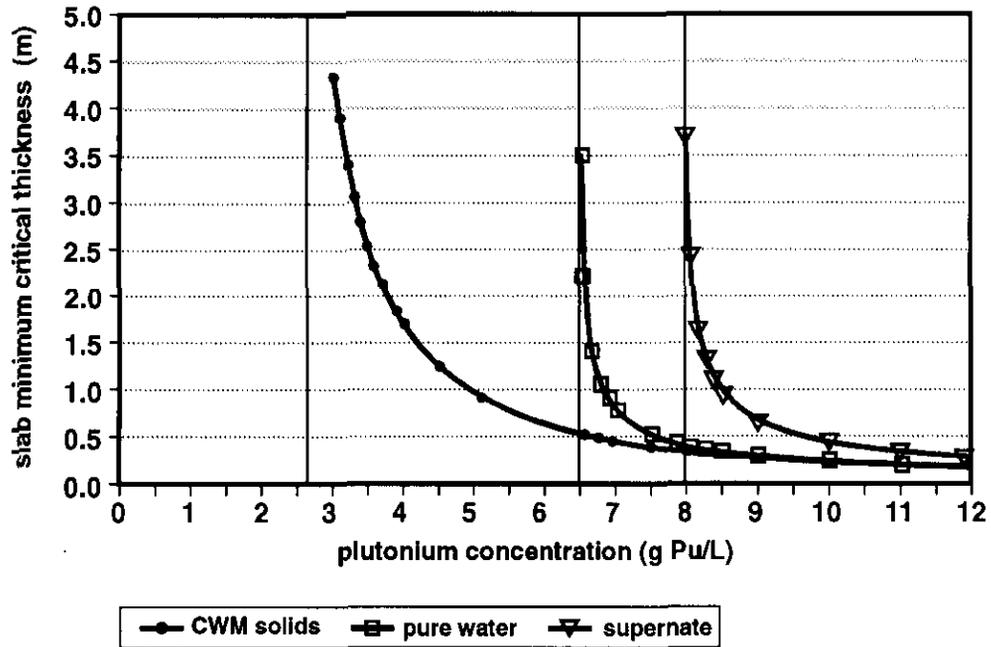
CWM = conservative waste model

Figure B-2. Sphere Minimum Critical Mass Versus Plutonium Concentration.



CWM = conservative waste model

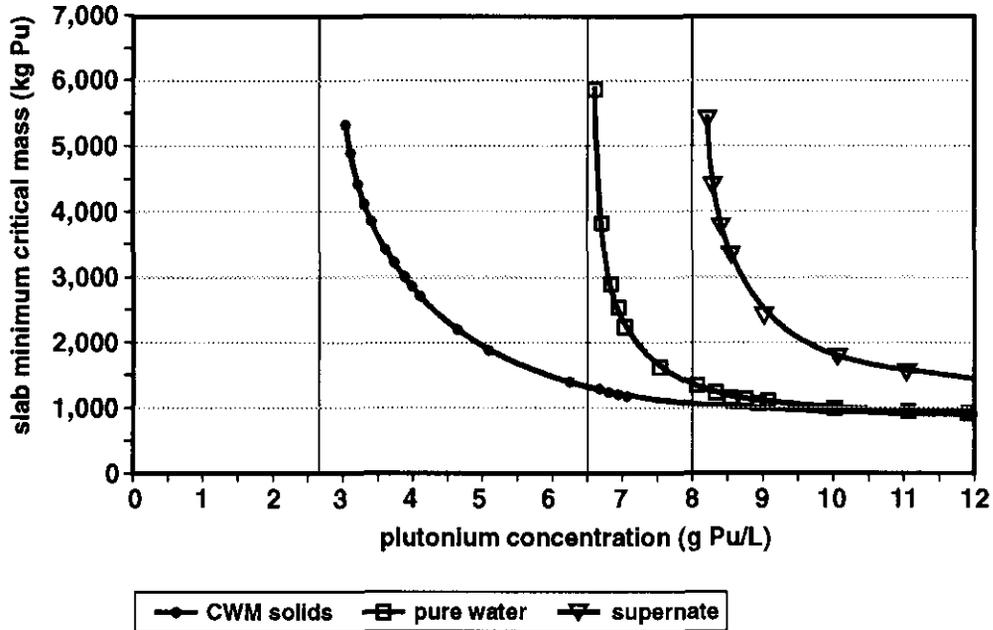
Figure B-3. Slab Minimum Critical Thickness Versus Plutonium Concentration



CWM = conservative waste model

7G96030035.8
R1 SRP-CSD

Figure B-4. Slab Minimum Critical Mass Versus Plutonium Concentration in a 22.9-m- (75-ft) Diameter Tank.



B2.4.4 Plutonium, Uranium, and Iron Mixtures

Rogers (1999) provides a parametric study of K Basin sludge. Calculations of k_{∞} are made for plutonium in mixtures of uranium, iron, and water. Rogers (1999) provides a curve of maximum k_{∞} as a function of the iron/uranium mass ratio for high-burnup N Reactor uranium fuel. See Section B6.0 for an estimation of the maximum achievable k_{∞} based on these calculations.

B3.0 ABSORBER-TO-PLUTONIUM RATIOS

The ability of waste components to absorb neutrons varies greatly. Nevertheless, if the proportion of any component is high enough, subcriticality can be ensured by the neutron absorption of that component. When a subcritical limit mass ratio is met or exceeded, subcriticality is assured for all concentrations of plutonium. This section provides subcritical limit ratios for components selected to be useful in evaluating criticality safety. The convention is adopted to specify ratios with the plutonium in the denominator.

In practice, the quantity of a particular component may not be enough to exceed the subcritical limit mass ratio. In this case, a single mass (or atom) ratio is not sufficient to demonstrate subcriticality, and it is necessary to combine the absorbing capacity of additional waste components to demonstrate subcriticality. Appendix B4.0 provides a method of adding the neutron absorption properties of these various components to assess their combined effect towards ensuring subcriticality.

It should be noted that the absorber-to-plutonium subcritical mass ratio is based on a homogeneous mixture of waste components. Although subcriticality is assured in a homogeneous mixture when the subcritical limit mass ratio is exceeded, the same cannot be said for a heterogeneous mixture unless that subcritical limit mass ratio is exceeded throughout the waste.

B3.1 URANIUM-TO-PLUTONIUM MASS RATIO

Nuclear Safety Guide, TID-7016 Revision 2 (Thomas 1978) states that criticality is not possible for plutonium combined with natural or depleted uranium provided the ratio of plutonium/uranium, also written as $\text{Pu}/(\text{Pu} + \text{U})$, does not exceed 0.0013. Natural uranium contains 0.72 wt% ^{235}U .

For this evaluation, the inverse of the above ratio, or $(\text{Pu} + \text{U})/\text{Pu}$, is used. This inverse ratio places the plutonium in the denominator, where it appears in other ratios used to assess subcriticality. The subcritical limit for the inverse ratio is 770. This is sometimes referred to as the "always safe" limit value for the U/Pu mass ratio. Subcriticality is assured when the mass ratio exceeds this value and the ^{235}U enrichment in the uranium is less than that of natural uranium (0.72 wt%).

Most of the uranium sent to waste storage tanks is depleted with a ^{235}U content less than that of natural uranium. Because most of the waste is a byproduct of the production of plutonium from low-enriched uranium, the uranium/plutonium mass ratio is usually larger than the subcritical limit.

The ratio of $(\text{Pu} + \text{U})/\text{Pu}$ is often simplified to U/Pu . This substitution causes the ratio to decrease slightly and is, therefore, conservative. The error introduced by ignoring the plutonium in the numerator is less than 0.15 %.

B3.2 IRON AND MANGANESE-TO-PLUTONIUM MASS RATIOS

Clemmons and Goslen (1992a, 1992b) calculated the amount of iron and manganese required in a water solution to ensure subcriticality of ^{239}Pu . Criticality is not possible regardless of water moderation if the following mass ratios are exceeded: $\text{Fe}/^{239}\text{Pu} = 160$ or $\text{Mn}/^{239}\text{Pu} = 32$.

B3.3 HYDROGEN-TO-PLUTONIUM ATOM RATIO

Criticality is not possible when the hydrogen-to-plutonium (H/Pu) atom ratio exceeds 3,600.

B3.4 BORON AND CADMIUM

The presence of boron and cadmium in tank waste can be important. Whenever criticality evaluations are made using boron, it is important to recognize that the two primary isotopes have greatly different absorption cross sections and that a proper specification for boron is important. Boron in waste is assumed to be natural boron composed of 20 wt% ^{10}B and 80 wt% ^{11}B .

Cadmium and boron are strong absorbers of moderated neutrons. If the ratio of hydrogen to fissile atoms exceeds 250, subcriticality is assured when either the cadmium/fissile atom ratio exceeds 0.5 or the boron/fissile atom ratio exceeds 2.0. Fissile refers to ^{235}U , ^{233}U , and ^{239}Pu .

The neutron absorption contribution of these elements would dominate the waste model. If they were included in the model, their continued presence in the tank waste would have to be guaranteed. As a practical matter, it would be very difficult, if not impossible, to guarantee the presence of these poisons mixed uniformly throughout the waste.

Still, the large neutron absorption of cadmium and boron means they are potentially important to subcriticality studies. If their presence is independently verified, they provide additional assurance of subcriticality.

B3.5 ADDITIONAL ABSORBER-TO-PLUTONIUM MASS RATIOS

In addition to those previously mentioned, other elements and compounds can be considered for minimum subcritical absorber-to-plutonium (X/Pu) mass ratios. This is because of their high neutron absorption cross sections, or because of their relative abundance in the waste. The minimum subcritical absorber-to-plutonium mass ratios for these are shown in Table B-3 (Rogers et al. 1996). These mass ratios will ensure subcriticality for any degree of water moderation.

Table B-3. Minimum Subcritical Absorber-to-Plutonium Mass Ratios to Ensure Subcriticality¹.

Component	Minimum subcritical X/Pu mass ratio
Calcium	770
Chromium	135
Copper	130
Lanthanum	121
Nickel	105
Nitrogen	61
Nitrate	270
Sodium	360
Thorium	243

Note:

¹Rogers et al. (1996)

For subcritical mass ratios in Tables B-3 and B-4, calculations are provided by Rogers et al. (1996, Appendix A) to verify conservatism. Maximum k_{∞} is found to be no greater than 0.90 at a 95% confidence level over the range of moderation from extremely dry to highly overmoderated.

A subcritical limit k_{∞} of 0.90, rather than 0.95, is used for these mass ratios because no experimental benchmark data was available against which to compare calculations.

B3.6 MATERIALS WITH LOW NEUTRON ABSORPTION CROSS SECTIONS

The CWM assumes that the macroscopic absorption cross section will be greater than 0.01096 cm^{-1} . However, some components in waste, if taken alone, exhibit a smaller macroscopic neutron absorption cross section. Waste components with low neutron absorption cross sections are aluminum, bismuth phosphate, carbon (graphite), silicon dioxide, and zirconium. Minimum subcritical absorber-to-plutonium mass ratios for components with a low neutron absorption are shown in Table B-4, based on Rogers et al. (1996).

Silicon dioxide is a common compound found in sandy soils. Bismuth phosphate comprises a significant portion of some waste streams. Zirconium was used in fuel element cladding and is found in cladding waste. Carbon is a common substance, although in waste it seldom appears in a pure form.

Table B-4. Minimum Subcritical Absorber-to-Plutonium Mass Ratios for Low Neutron Absorbers¹.

Component	Minimum subcritical X/Pu mass ratio
Aluminum	910
Bismuth	48,000
Bismuth phosphate	10,100
Carbon (graphite)	28,000
Silicon	1,400
Silicon dioxide	3,000
Zirconium	4,000

Note:

¹Rogers et al. (1996)

This group of components is important to criticality safety because a hypothetical configuration can be constructed for which the minimum critical plutonium concentration would be less than 2.6 g/L (the CWM value). In practice, however, there are no configurations having a high enough proportion of these components to provide less neutron absorption than the CWM. In addition, even if the proportion of these components were high, criticality would still not be possible at a plutonium concentration less than 2.6 g/L, unless the quantity of plutonium and the volume occupied were large.

B4.0 COMBINING THE EFFECT OF SEVERAL ABSORBERS

B4.1 SUM OF ACTUAL-TO-MINIMUM SUBCRITICAL MASS FRACTIONS

Since waste is made up of many components, it is more realistic to base an evaluation of subcriticality on the combined effects of all the neutron absorbers. For an infinitely large system the neutron multiplication will remain unchanged when the composition of the waste is changed, as long as the macroscopic neutron absorption cross section is unchanged. This fact provides the basis for the following method to take into account the various absorbers. Braun et al. (1994) discusses justification of the method.

Waste is subcritical when the neutron absorption cross section per atom of plutonium is greater than required for criticality. The contribution of a waste component to subcriticality is proportional to its concentration divided by the plutonium concentration; this is the X/Pu. This contribution can be represented as a fraction by dividing by the subcritical limit mass ratio for that component.

The following rules can be used to evaluate a waste composition.

- Calculate the actual X/Pu for each waste component. Divide each actual mass ratio by its corresponding minimum subcritical mass ratio to obtain a fraction. This fraction will be referred to as the actual-to-minimum subcritical mass fraction.
- When the sum of the actual-to-minimum subcritical mass fractions for all individual components is greater than unity, the waste is subcritical.

Therefore,

$$\sum_{j=1}^N \frac{\left(\frac{X_j}{Pu}\right)_{\text{actual}}}{\left(\frac{X_j}{Pu}\right)_{\text{subcritical}}} \geq 1$$

where:

$$\left(\frac{X_j}{Pu}\right)_{\text{actual}} = \text{Actual absorber mass divided by plutonium mass.}$$

$$\left(\frac{X_j}{Pu}\right)_{\text{subcritical}} = \text{Minimum subcritical mass ratio.}$$

When the sum of fractions is greater than 1.0, the total neutron absorption is sufficient to ensure subcriticality in an infinite system of homogeneous waste. The greater the sum of the fractions, the greater the margin of safety.

Caution needs to be exercised in the interpretation of the sum of fractions obtained from characterization data that is incomplete. When the analyzed composition does not include all waste components, the sum of fractions can be too small. Additional evaluation may be required to demonstrate subcriticality.

B4.2 SOLIDS-TO-PLUTONIUM MASS RATIO

If waste density is lowered (assuming a large volume), the minimum critical plutonium concentration is reduced in direct proportion, but the subcritical limit solids/plutonium mass ratio remains unchanged. Therefore, the solids/plutonium mass ratio is a measure of the margin of subcriticality that is independent of solids density. When the plutonium inventory and the total mass of stored waste is known, these parameters may be used to demonstrate subcriticality.

For the CWM, criticality is not possible when the plutonium content is less than 2.1 g/kg solids. The reciprocal of this ratio (i.e., the solids/plutonium mass ratio) is 476. When neutron absorption per unit mass exceeds that of the CWM, subcriticality is assured when the mass ratio exceeds this value, which is expected to be conservative for all actual tank waste. However, if a large fraction of uranium is assumed possible, conservatism requires that the larger subcritical limit mass ratio of 770 that applies to natural uranium be used. Therefore, it is recommended that the subcritical limit mass ratio of 770 be used as a generalized limit for solids. When the total mass of settled solids divided by the total plutonium inventory exceeds 770, and the waste is well mixed, subcriticality is assured for all credible waste compositions.

The solids/plutonium subcritical limit mass ratio provides a method by which the degree of subcriticality of a waste storage tank can be easily estimated. However, it does not remove the need to perform waste characterization analyses to determine the waste composition.

When taken alone, aluminum, bismuth, silicon, and zirconium exhibit a solids/plutonium subcritical limit larger than the 770 value for natural uranium. However, those components are always mixed with a large fraction of other components with a smaller component/plutonium mass ratio, such that the overall mass ratio is less than 770.

B5.0 URANIUM CRITICAL PARAMETERS

In a uranium-water system that is homogenized, criticality is not possible until the enrichment exceeds 1.03 wt% (Clayton and Brown 1965). The *Nuclear Criticality Safety Guide* (Pruvost and Paxton 1996) provides a subcritical limit for uranium mixed homogeneously with water of 0.93 wt% for uranium metal and 0.96 wt% for UO_2 , UO_3 , U_3O_8 , and UO_3F_2 . Although criticality is possible at enrichments as low as 0.72 wt% in precisely defined heterogeneous arrays with low neutron absorbing moderators, such as graphite or heavy water, these configurations would be impossible to achieve in tank waste. Nearly all uranium sent to waste storage tanks is enriched less than 1.03 wt%. There is no chemical or physical means by which the ^{235}U can be separated from the ^{238}U in tank waste.

CPS-T-149-00010, Rev. D-0 (WHC 1988), which has been used in the past to cover operations involving tank waste, has the following limit governing ^{233}U and ^{235}U :

One gram of ^{233}U or ^{235}U in excess of 1 wt% enrichment shall be assumed equivalent to one gram of Pu, unless otherwise restricted by a specification. The amount of ^{235}U less than or equal to the equivalent of 1 wt% enrichment may be excluded in concentration and total mass calculations.

The uranium/plutonium subcritical limit mass ratio of 770 was derived using the assumption that the ^{235}U enrichment is 0.72 wt%. When this mass ratio is used as a measure of subcriticality, assurance must be provided that the uranium is natural or depleted.

Clemmons and Goslen (1992a, 1992b) calculated the amount of iron and manganese required in a water solution to ensure subcriticality of ^{235}U . Criticality is not possible regardless of water moderation if the following mass ratios are exceeded: $\text{Fe}/^{235}\text{U} = 77$ or $\text{Mn}/^{235}\text{U} = 30$.

Rogers (1999) provides a parametric study of uranium combined with iron and containing up to 0.30 wt% plutonium.

B6.0 ESTIMATED NEUTRON MULTIPLICATION CONSTANT

Rogers (1999) provides a parametric study of k_{∞} versus ^{235}U enrichment, plutonium content, and iron content. Although this study was done for K Basin sludge (not stored at tank farms), it provides a means to estimate the highest k_{∞} that is possible without violating the Technical Safety Requirement (TSR) (FDH 1997) (see Section 3.2). The fissionable components of both K Basin sludge and tank waste are derived from N Reactor fuel.

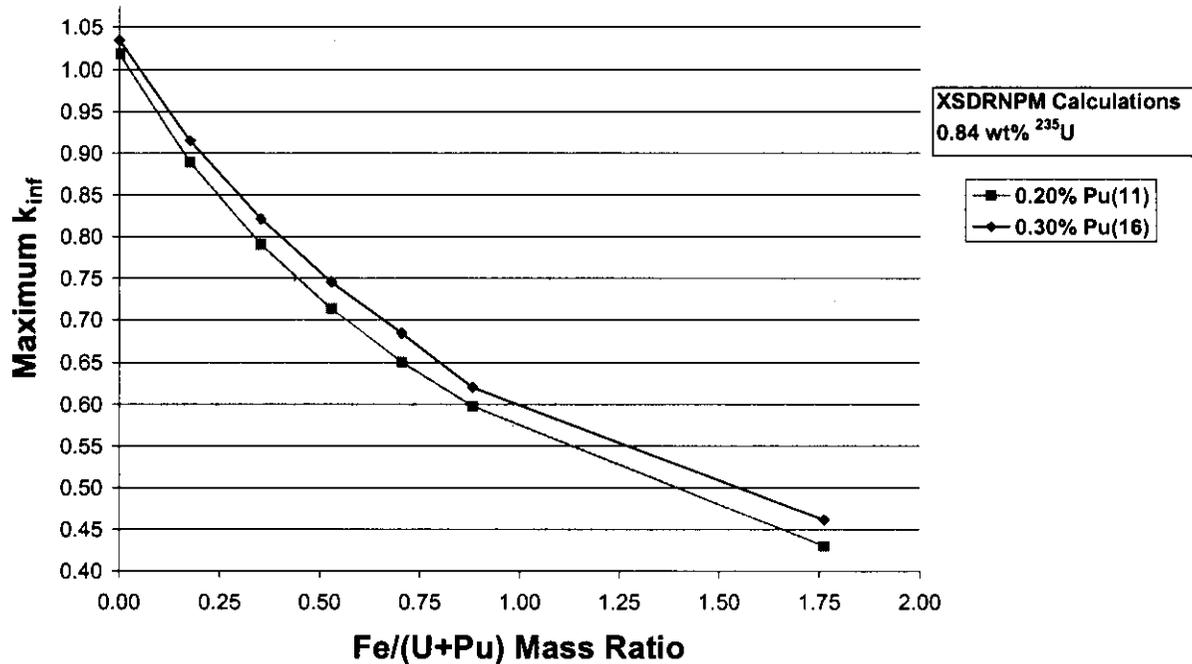
Irradiation of N Reactor fuel is taken into account in the parametric study. Calculations made over the entire range of burnup show that longer irradiation results in higher plutonium content and larger k_{∞} . At greatest burnup, the plutonium content comprises 0.30 wt% of the uranium and contains 16 wt% ^{240}Pu . The average plutonium content in irradiated N Reactor fuel is about 0.20 wt%, and this contains 11 wt% ^{240}Pu . After irradiation, the uranium is depleted with a ^{235}U content less than 0.72 wt%. However, for the calculations, the uranium is assumed to be slightly enriched with a 0.84 wt% ^{235}U content. This composition after burnup is conservative when compared to actual N Reactor fuel.

Figure B-5 shows maximum k_{∞} for homogeneous sludge (as described above) as a function of iron content (Rogers 1999, Figure A-7). To meet the TSR (FDH 1997), the iron/plutonium mass ratio must exceed 160. For this mass ratio calculation, the "plutonium equivalent" inventory is used, and the ^{235}U in excess of 0.72 wt% is treated as if it were plutonium. For 0.84 wt% enrichment, the excess ^{235}U amounts to 0.12 wt% of the uranium, and this is added to the plutonium equivalent inventory.

At maximum burnup, the uranium contains 0.30 wt% plutonium and 0.12 wt% excess ^{235}U , thus providing a "plutonium equivalence" of 0.42 wt%. When multiplied by 160 and divided by the mass of the uranium, the required iron/uranium mass ratio becomes 0.672. At average burnup,

the required iron/uranium mass ratio is found to be 0.512. From Figure B-5 these values correspond to maximum k_{∞} values of 0.72. For unirradiated 0.84 wt% and 1.25 wt% uranium (containing no plutonium), maximum k_{∞} is 0.923 and 1.062, respectively (Rogers 1999, Figure A-2). When iron is added to meet the TSR for the "plutonium equivalence," maximum k_{∞} drops to below 0.83 for both cases.

Figure B-5. Maximum k_{∞} for Homogeneous Sludge as a Function of Iron Content.



In conclusion, it is theoretically possible for incoming sludge to have a k_{∞} as high as 0.83 and yet meet the TSR (FDH 1997). This requires sludge to contain a minimum of neutron absorbing solids and optimal moderation. For irradiated uranium with unseparated plutonium, the highest k_{∞} possible for a configuration that does not violate the TSR is estimated to be 0.72. Normally, the content of neutron absorbing solids is at least several times the minimum subcritical limit, and the maximum k_{∞} is expected to be less than 0.50.

Braun et al. (1994, Appendix A.5.0) evaluates the margin of subcriticality afforded by the composition of tank waste. From several hundred available characterization samples, eight samples were selected on the basis of their apparent low neutron absorber content as providing the most reactive compositions. The MONK6A Monte Carlo criticality code (UKAEA 1988a, 19988b) was used to calculate the neutron multiplication constant, k_{∞} , for infinite systems of these compositions. Seven calculated values were less than 0.12, but the eighth was 0.32. This high k_{∞} appears to result from the plutonium content being reported as 0.1 wt% of the sample weight. This content appears to be an upper limit that over-estimates the plutonium content.

B7.0 REFERENCES

- Braun, D. J., L. D. Muhlestein, T. B. Powers, and M. D. Zentner, 1994, *High-Level Waste Tank Subcriticality Safety Assessment*, WHC-SD-WM-SARR-003, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- Carter, R. D., G.R. Kiel, and K. R. Ridgway, 1969, *Criticality Handbook*, ARH-600, Vol. II, p. III.A.8.100-3, Atlantic Richfield Hanford Company, Richland, Washington.
- Clayton, E. D., and C. L. Brown, 1965, "Criticality and Nuclear Safety of Slightly Enriched Uranium," *Chemical Engineering Progress*, Symposium Series, Vol. 61, No. 60, pp.33-43.
- Clemmons, J. S., and A. Q. Goslen, 1992a, *Minimum Safe Ratios of Fe and Mn to U-235 in an Infinite System (U)*, Nuclear Criticality Safety Analysis, WM-92-3, WER-MER-921143, Westinghouse Savannah River Company, Aiken, South Carolina.
- Clemmons, J. S., and A. Q. Goslen, 1992b, *Minimum Safe Ratios of Fe and Mn to Pu-239 in an Infinite System (U)*, Nuclear Criticality Safety Analysis, WM-92-5, WER-MER-921353, Westinghouse Savannah River Company, Aiken, South Carolina.
- FDH, 1997, *Tank Waste Remediation System Technical Safety Requirements*, HNF-SD-WM-TSR-006, Rev. 0, Section 5.7, "Nuclear Criticality Safety," Fluor Daniel Hanford, Inc., Richland, Washington.
- Paxton, H. C., and N. L. Pruvost, 1987, *Critical Dimensions of Systems Containing ²³⁵U, ²³⁹Pu, and ²³³U*, LA-10860-MS, Los Alamos National Laboratory, Los Alamos, New Mexico.
- Pruvost, N. L., and H. C. Paxton, 1996, *Nuclear Criticality Safety Guide*, LA-12808, Los Alamos National Laboratory, Los Alamos, New Mexico.
- Reynolds, D. A. and D. L. Herting, 1984, *Solubilities of Sodium Nitrate, Sodium Nitrite, and Sodium Aluminate in Simulated Nuclear Waste*, RHO-RE-ST-SWD-024, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- Rogers, C. A., 1993, *CSER 92-009: An Analytical Model For Evaluating Subcritical Limits For Waste in Hanford Site Storage Tanks*, WHC-SD-SQA-CSA-20356, Westinghouse Hanford Company, Richland, Washington.

- Rogers, C. A., 1999, *Criticality Safety Evaluation of Disposing of K Basin Sludge in Double-Shell Tank AW-105*, HNF-3500, Lockheed Martin Hanford Corporation, Richland, Washington.
- Rogers, C. A., K. N. Schwinkendorf, and H. Harris, 1996, *Criticality Parameters for Tank Waste Evaluation*, WHC-SD-SQA-CSA-507, Westinghouse Hanford Company, Richland, Washington.
- Schwinkendorf, K. N., 1994a, *Criticality Analysis of Homogeneous Pu-Water-Waste Systems*. This is Appendix A.6.0 of Braun et al., *High Level Waste Tank Subcriticality Safety Assessment*, WHC-SD-WM-SARR-003, March 1994.
- Schwinkendorf, K. N., 1994b, *Criticality Analysis of Various Idealized Waste Geometries* (Appendix A.7.0 of Braun et al, *High Level Waste Tank Subcriticality Safety Assessment*, WHC-SD-WM-SARR-003, March 1994).
- Thomas, J. T., 1978, *Nuclear Safety Guide, TID-7016, Revision 2*, NUREG/CR-0095, ORNL/NUREG/CSD-6, Oak Ridge National Laboratory, Oak Ridge, Tennessee.
- UKAEA, 1988a, *MONK6 User Guide*, ANSWERS (MONK6) 1, AEEW R2195, Answers Business Center, Winfrith Technology Center, Dorchester, Dorset DT2 8DH, United Kingdom.
- UKAEA, 1988b, *MONK6 A Monte Carlo Code for Criticality Calculations*, ANSWERS (MONK6) 2, AEEW R2248, Answers Business Center, Winfrith Technology Center, Dorchester, Dorset DT2 8DH, United Kingdom.
- WHC, 1988, *Waste Storage Tanks and Associated Equipment*, CPS-T-149-00010, Rev. D-0, Westinghouse Hanford Company, Richland, Washington.

APPENDIX C

CHARACTERIZATION OF TANK CONTENTS

This page intentionally left blank.

C.0 CHARACTERIZATION OF TANK CONTENTS

Sources of characterization data are described, and a summary description of waste streams is provided. Emphasis is given to information and parameters most relevant to demonstrating criticality safety. More detailed descriptions of tank waste can be found in the references.

C1.0 SOURCES OF CHARACTERIZATION DATA

The most detailed information on tank waste composition is provided by the Hanford Defined Waste (HDW) Model and by chemical analyses of waste tank samples. In practice, the available analytical data for a specific storage tank may be limited, making it necessary to use HDW data to characterize the waste.

C1.1 BEST-BASIS INVENTORIES OF CHEMICALS AND RADIONUCLIDES

The Best-Basis Inventory program was established to develop a Best-Basis Inventory (CHG 2000), also called Standard Inventory, of chemicals and radionuclides in waste stored in the 149 SSTs and 28 DSTs. This program develops and maintains estimates of total (global) tank waste inventories, maintains tank specific inventories, and reconciles conflicting information.

Kupfer et al. (1999) estimates the global and tank-by-tank waste component inventories. Kupfer et al. (1999) provides global inventory estimates (based primarily on process history) for 30 nonradioactive components and 46 radionuclides that account for approximately 99% of the total tank waste inventory.

C1.1.1 Tank Waste Information Network System (TWINS)

The Tank Waste Information Network System (TWINS) website on the Hanford intranet provides access to the Tank Characterization Database. This database contains the most current Best-Basis Inventory for each of the storage tanks. Values are provided for each radionuclide in curies. This data can be converted to provide the current best-basis plutonium inventory. This provides the official plutonium inventory for SSTs, which are not included in the DST Plutonium Inventory Database.

Development and maintenance of the Best-Basis Tank Inventories is an on-going effort. As new analytical data or process knowledge become available, the respective Best-Basis Inventories are typically adjusted.

C1.2 PLUTONIUM INVENTORY DATABASE

The plutonium inventory database (LMHC 1998) is the official inventory for DSTs. This database is an Excel spreadsheet that maintains a record of all material transfers in and out of each storage tank and updates the plutonium inventory after each transfer.

No transfers of waste are made to SSTs, and no plutonium inventory database is maintained for them. The plutonium inventory for SSTs is estimated by Whyatt et al. (1996, Table A.4) using data obtained from the Tank Waste Source Term Summary Validation (WHC 1995) and from the Hanford Tank Chemical and Radionuclide Inventories (Agnew 1997).

C1.3 HANFORD DEFINED WASTE MODEL

Agnew (1997) used the HDW Model to estimate the contents of storage tanks in the Hanford tank farms. The basic data used are records of transfers and waste stream compositions compiled into a process and transaction data set. This data was used to construct the Waste Status and Transaction Record Summary (WSTRS) spreadsheet which contains a record of transactions for each tank. The Tank Layer Model (TLM) and the Supernatant Mixing Model (SMM) process data in this spreadsheet. Output from these spreadsheet models is input to the HDW Model to estimate tank inventories. Agnew (1997) provides a printout of these inventories.

Agnew (1997, p. 13) describes the approach used to define the various HDW types (i.e., waste streams) used in his model.

The determination of chemical and radionuclide concentrations for each of the HDW's begin with input of radionuclides and stable chemicals, both of which are used to define the total species in each waste stream as shown in the Block Diagram of HDW Spreadsheet [not reproduced here]. These total species are then separated into two layers, a sludge and a supernatant, that result in different concentrations of species for the two layers.

Each species is precipitated according to a single point solubility. Ions precipitated in more than one salt are simply successively precipitated. Thus, the solids that precipitate are merely representative and are not meant to reflect the actual solids distribution. Because this supernatant is also present in the interstices of the sludge layer, this "supernatant" is included as part of the sludge composition. The solubility of each species is set by a macro that when run on the HDW spreadsheet, adjusts the fraction precipitated parameter so that the supernatant concentration is equal to or less than the target solubility.

Agnew (1997) lists four sources of error in tank inventories. Modeling error is derived from variability in processes from which waste is derived and sent to tank storage. Parameter error results from variation in component solubilities. Transaction error is caused by variability in evaporator blending, by concentrate carryover, and by inaccurate transaction information. Sampling error is primarily the result of waste heterogeneity.

C1.4 ANALYTICAL SAMPLES

Analyses of core samples provide a primary source of waste characterization data. However, the number of core samples which have been analyzed is relatively small compared to the large volume of waste in tank storage. There is considerable variation in component concentrations in different tanks and at different locations in the same tank.

A single core sample might contain material from discrete waste layers. Historically, the contents of the core sample are mixed before being analyzed, so the reported concentrations represent an average value over the core. Such a sample analysis does not represent actual conditions and might instead be a homogenized average of two or more waste types. This mixing of the cored material will not detect layers of high plutonium concentration. A discussion of stratification is provided in Appendix D6.0.

C1.4.1 Data Compiled for Safety Assessment

As part of the *High-Level Waste Tank Subcriticality Safety Assessment* (Braun et al. 1994), a compilation was made of 914 waste sample analyses made between 1974 and 1994. A plutonium concentration was reported for 200 of 312 solids samples and for 306 of 602 liquid samples.

Reported plutonium concentrations are higher for solids samples than for liquid samples. The highest plutonium concentration reported was 0.35 g/L (1.3 g/gal) in a solids sample, but the median value of all solids samples was found to be about 0.01 g/L. The highest plutonium concentration was later found to be in error and was actually smaller than reported. The median plutonium concentration from among all 306 liquid samples is 0.0001 g/L. Two liquid samples are reported with plutonium concentrations greater than 0.01 g/L. It is not known to what extent these values have been influenced by uncertainties associated with the measurements.

In July 1999 gross alpha counts from a sample (core 260, segment 1) taken from SST TX-118 indicated a possible plutonium concentration of 0.89 g/L (Lipke 1999). In making this estimate, the entire measured alpha activity of 36.8 $\mu\text{Ci/g}$ was attributed to ^{239}Pu . Conversion to plutonium concentration was made using a measured sludge specific gravity of 1.5 g/L. This conservative estimate of plutonium concentration provides the highest value that has been obtained from any sample analysis for any tank.

When a plutonium-bearing solution is made alkaline, the plutonium will precipitate along with other solids and settle to the bottom of the tank. A high pH will tend to keep the plutonium combined with solids. Braun et al. (1994) provides a histogram of pH for 310 liquid samples, most of which were taken from SSTs. The lowest pH values found are three values between 7.0 and 8.0 and six values between 8.0 and 9.0. The median pH from all samples is about 12. All sampled waste was found to be alkaline (i.e., with a pH greater than 7).

C1.4.2 Tank Waste Source Term Inventory Validation

During 1994 and 1995 a revised accident analysis was prepared for tank farms that required the determination of source terms (i.e., the highest concentrations) for selected radionuclides and chemicals. The selection process included verification that the sample indicating the highest radionuclide concentration was a valid sample and the laboratory analysis was free of error.

Documentation of source term verification is provided in *Tank Waste Source Term Inventory Validation* (Brevick et al. 1995). Radionuclide tables in Volume II contain measured concentrations grouped according to nuclide.

Cowley (1996) used data from Brevick et al. (1995) to determine the highest plutonium concentrations in DST, SST, and aging waste facility (AWF) liquids and solids. Cowley's values are shown in Table C-1. The highest plutonium concentration found for any waste type was 0.7 g/L, and this came from DST solids. This is an upper bound value, based on the available data, and is expected to be higher than any actual plutonium concentration.

Table C-1. Highest Plutonium Concentrations Determined From Sample Analysis of Each Waste Type¹

Waste Type	Unit Liter Dose Bq/L	Plutonium Concentration g Pu/L
DST Liquids	7.7 E+06	0.0034
DST Solids	1.6 E+09	0.7
SST Liquids	3.6 E+07	0.016
SST Solids	4.4 E+08	0.19
AWF Liquids	1.2 E+06	0.00052
AWF Solids	4.4 E+08	0.19

Note:

AWF = aging waste facility

¹ Cowley (1996)

C1.5 OTHER SOURCES OF INFORMATION

This section lists sources of characterization information that are of lesser importance than those described above.

C1.5.1 TRAC

Track Radioactive Components (TRAC), a software program developed by Jungfleisch (1984) to estimate tank inventories, has been superseded by the HDW Model, Revision 4, and is no longer used to estimate tank waste component inventories.

Records of waste additions and transfers were used to estimate the amounts and kinds of waste in storage tanks. In 1983, the TRAC computer model estimated the total plutonium discharged in waste to be 385 kg (Jungfleisch 1983). The most recent TRAC inventory was generated in 1985, but was not published.

C1.5.2 Estimate Of Total Inventory Based on Historical Records

Roetman et al. (1994) estimated the plutonium in Hanford Site waste tanks based upon historical records. The total plutonium in the tank farms was estimated to be 407 kg, based on annual average estimates. Roetman's conservative bounding estimate was 981 kg, based upon annual maximum estimates.

C2.0 DESCRIPTION OF WASTE STREAMS

Descriptions of waste streams routed to the tank farms can be found in the following references.

- *A History of the 200 Area Tank Farms* (Anderson 1990).
- *The Potential for Criticality in Hanford Tanks Resulting from Retrieval of Tank Waste* (Whyatt et al. 1996).
- *Fluid Dynamics, Particulate Segregation, Chemical Processes, Natural Ore Analog and Tank Inventory Discussions that Relate to the Potential for Criticality in Hanford Tanks* (Serne et al. 1996).
- *Hanford Tank Chemical and Radionuclide Inventories: HDW Model Rev. 4* (Agnew 1997).
- *High-Level Waste Tank Subcriticality Safety Assessment* (Braun et al. 1994).

C2.1 HISTORY OF 200 AREA TANK FARMS

Anderson (1990) provides the following synopsis of waste streams:

Four basic chemical operations were the source of radioactive waste solutions transferred to underground storage tanks since startup of the Hanford Site. These were the bismuth phosphate (BiPO_4) process, the tributyl phosphate (TBP) process, the reduction oxidation (REDOX) process, and the plutonium-uranium extraction (PUREX) process. Three of these were chemical separations programs for the recovery of uranium and plutonium from irradiated reactor fuels. The fourth (TBP) was designed for the recovery of uranium metal waste generated by the BiPO_4 process. In all cases, the aqueous wastes were made alkaline for storage in underground steel tanks.

Anderson (1990) provides the following specific details:

The plutonium and uranium content in the waste usually is specified as a percentage of the quantity processed. Although the waste streams described are the source of nearly all waste stored in the tank farms, the processes generating these wastes are no longer operating. Most waste streams have not sent waste to tank farms for a long time. An estimate of future sources of waste is provided in Appendix E, "Generation of Future Waste."

Bismuth Phosphate (BiPO₄) Process - The bismuth phosphate process generated the following waste types:

1. Alkaline coating removal waste, containing small amounts of fission products, was combined with first-cycle decontamination wastes for storage. About 0.4 percent of the plutonium and about 0.4 percent of the uranium is estimated as having gone to waste.
2. Metal waste from the extraction, containing all of the uranium and about 1 percent of the plutonium.
3. Byproduct cake solution and waste solution from the first decontamination cycle, containing about 1 percent of the plutonium.
4. Second decontamination-cycle waste, containing about 1 percent of the plutonium.

Uranium Recovery Plant - Waste from the Uranium Recovery Plant contained about the same proportion of fission products as did metal waste. This waste is referred to as TBP waste. Anderson (1990) does not specify the content of uranium and plutonium.

REDOX Plant - The REDOX Plant generated the following waste types:

1. Coating waste from the decladding of aluminum-clad fuels, containing about 0.4 percent of the plutonium and 0.4 percent of the uranium.
2. Waste from the decladding of zircaloy-clad fuels. This waste is described as containing about 0.0001 kg of uranium/L (0.001 lb. of uranium/gal). The plutonium content is stated to be equal to the uranium content. This is an obvious inaccuracy because the content of the uranium should be hundreds of times greater than that of plutonium. Perhaps the intent may be to show that the uranium and plutonium are only a small fraction of the original.
3. REDOX waste was the high-level component of the process waste. It contained about 0.05 percent of the original uranium and 0.04 percent of the original plutonium.

PUREX Plant - Waste from the PUREX Plant falls into the following types:

1. Coating waste from the decladding of aluminum-clad fuels. This waste contains about 0.4 percent of the original plutonium and 0.4 percent of the original uranium.
2. Waste from the decladding of zircaloy-clad fuels. This contains the same content of plutonium and uranium as the same type waste from the REDOX Plant.
3. Organic wash waste. This contains about 0.000036 kg of uranium/L (0.0003 lb. of uranium/gal). The plutonium content is very low and is not specified.
4. Neutralized PUREX Plant acid waste. The uranium and plutonium content is very low and is not specified.
5. Waste from the thorium campaigns. The uranium and plutonium content is very low and is not specified.

All of these waste streams are derived from slightly enriched uranium fuel elements that have been burned in a reactor. This fuel has an initial ^{235}U enrichment less than 1.3 wt%, which the burning process reduces to a level below that in natural uranium (i. e., below 0.72 wt%) while at the same time producing plutonium. The plutonium content increases to an average value of approximately 0.10 wt% of the uranium content. The maximum plutonium content will not exceed 0.15 wt% of the uranium.

Plutonium Finishing Plant (PFP) - The *High-Level Waste Tank Subcriticality Safety Assessment* (Braun et al. 1994) provide the following description of waste from the PFP:

The PFP is the only potential source of separated plutonium in the waste tanks. Fuel processing is not performed at the PFP. Its feed source is primarily the PUREX Plant and other process facilities. Waste from the PFP was not sent to the tank farms until 1973. Before that time, PFP waste was discharged to burial drums or cribs.

The PFP consists of three separate operations that generate waste containing plutonium and the Development and Analytical Laboratories. The three operations are the remote mechanical "A" (RMA) oxide line, which converts plutonium nitrate produced at the PUREX Plant and formerly at the REDOX Plant to plutonium oxide powder; the remote mechanical "C" (RMC) metal line, which converts plutonium oxide to plutonium metal via calcium reduction of plutonium fluoride; and the Plutonium Reclamation Facility (PRF), which processes a variety of plutonium scrap materials, such as crucible fragments, to plutonium nitrate to serve as feed to the RMC line.

For additional information on waste streams and present tank contents, see Appendix B of Braun et al. (1994).

C2.2 PROPERTIES OF WASTE RELEVANT TO CRITICALITY HAZARD

Both Chapter 9 of Serne et al. (1996) and Appendix A of Whyatt et al. (1996) are titled *Properties of Waste Relevant to Criticality Hazard*, and they contain the same information. This chapter "characterizes the waste in the underground tanks using historical accounts of flows and compositions of waste stream inputs, waste treatment processes, and analytical results of recent waste samples from the tanks."

The summary and conclusions from Whyatt et al. (1996) provide the following information:

The approach to identifying properties relating to criticality in existing waste storage tanks was to determine the batchwise composition of discharges from the processing facilities to the tank farm storage tanks. Nuclear Materials Control (NMC) records were used for plutonium, uranium, and neptunium transfers because they have been well documented and reviewed for accuracy and because they identified the Hanford Site tanks that received each batch of waste. The NMC records included an accountability volume that was compared, where possible, to plant waste discharge volumes as supportive information. Where possible, the settled sludge concentration of fissile material and insoluble absorbers in tanks that may contain fissile material in concentrations higher than 10 kg was determined on the basis of historical accumulation of process waste streams and by core sample analysis results. These two approaches to estimating inventories were compared and discrepancies discussed.

The results show that the fissile material concentrations in all of the Hanford Site tank waste is likely at subcritical values and that sufficient insoluble absorbers are present to maintain subcritical conditions under safe storage and retrieval operations. Additional sampling of tank waste is recommended for some selected tanks to verify fissile material and insoluble absorber content.

Seven DSTs were estimated to contain more than 10 kg of plutonium: AW-103, AW-105, AW-106, AY-101, AZ-101, AZ-102, and SY-102. Because these tanks are still active, they were evaluated in greater detail. In all, these seven tanks contain five waste types. DST AW-103 waste is similar to DST AW-105 waste and DST AZ-101 waste is similar to DST AZ-102 waste.

C2.3 BASIC WASTE TYPES

For criticality safety, waste is generally categorized into three forms: (1) supernatant, (2) sludge, and (3) saltcake. The settled solids portion of waste is a combination of sludge and saltcake. The chemical and physical characteristics of each type are summarized.

C2.3.1 Supernatant

Supernatant is the liquid portion above the settled sludge. Thermal gradients tend to promote mixing, and the supernatant should be inherently homogeneous. The plutonium concentration in supernatant liquid is very low.

C2.3.2 Sludge

Sludge is a mixture of metal precipitates (with interstitial liquid) which results from the neutralization and settling of chemical separation wastes. Sludge generally consists of metal oxides, hydroxides, carbonates, silicates, phosphates, sulfates, and other insoluble species. Some of these compounds are fairly soluble, but many are insoluble.

Plutonium is associated with sludge in the following ways:

- Sorption onto precipitated metal oxide particles. The sorption bonding of plutonium to strong neutron-absorbing metals, such as iron and manganese, provides direct assurance that neutron absorbers will remain associated with the plutonium.
- Precipitation as plutonium oxide crystals. If pure hydrous plutonium oxide is formed as a crystalline precipitate when acidic waste are made alkaline, it will probably agglomerate or mix with neutron absorbers.
- Formation of solid solutions. Solid solution formation occurs when plutonium co-precipitates with a metal ion in a crystalline lattice.

C2.3.3 Saltcake

Saltcake forms when sodium nitrate and sodium phosphate crystallize during evaporation and cooling of saturated waste solutions. Saltcake is the waste material not evaporated in an evaporator (called evaporator bottoms) that crystallizes after being pumped back to a storage tank and allowed to cool. Saltcake is soluble, and it generally forms the top layer when other solids types are present (i.e., above sludge). Saltcake sometimes forms a crust above the liquid layer.

An important characteristic of saltcake to criticality safety is its solubility. When water is added to saltcake, it dissolves. This allows it to be easily removed from other solids.

Dissolved plutonium remains in solution as saltcake crystallizes and is not found within saltcake.

Only insignificant amounts of plutonium (if any) are sorbed on the surface of saltcake.

Bratzel et al. (1996) provide the following information:

Plutonium concentrations resulting from the evaporation operations were inherently limited by boil-down rates and pumpability considerations. Volume reduction operations resulted in higher concentrations of plutonium in the saltcake than in the original dilute waste solutions. However, most of the concentrated plutonium is in the interstitial liquor

retained by the saltcake crystals. Analyses of the interstitial liquor show that plutonium concentrations are far lower than the maximum solubility limit of 0.2 g Pu/L. Thus, plutonium concentrations in the actual saltcake are even lower than those in the interstitial liquor.

Saltcake may contain some associated sludge. However, the amounts of sludge trapped within the saltcake are much smaller than the sludge layers themselves. Thus, the sludge layer is the only waste type with a realistic potential for an accidental criticality.

Braun et al. (1994, p.14) draws the conclusion that "saltcake waste is highly subcritical because of the very small quantity of plutonium and the very large amount of salt-type neutron absorbers."

C3.0 LIQUID CONTENT

As a rule, waste contains a large percentage of liquid, primarily water. However, to ensure long-term stability, to increase storage volume, and to preclude tank leaks, liquids are sometimes removed from the waste. Since it is often desirable that the liquid content of waste be low, it is important that criticality safety not depend upon the water content.

The high water content of the waste ensures that fissile material is overmoderated. However, there are operations designed to remove water from waste, such as saltwell pumping and passing waste through an evaporator. Although the degree of overmoderation decreases as liquid is removed, the low plutonium concentration ensures that the waste will remain overmoderated after completion of evaporation or pumping. Even if all water were to be removed, the scattering properties of the large fraction of non-fissile material would provide appreciable moderation and absorption.

Most sludge contains at least 40 wt% water. At the low plutonium concentration in sludge this equates to a very high H/Pu atom ratio. Assuming a plutonium concentration of 1 g/L, the H/Pu atom ratio would be about 10,000, as compared to the value of 3,600 which ensures subcriticality. A water content less than 10 wt% would be almost impossible to obtain in practice, and would require heating the waste to drive off water. At the same time, the actual plutonium concentration would be expected to be less than 0.1 g/L.

Criticality safety limits are based on the assumption that the water content (moderation) in waste is optimal. Therefore, the actual volume of liquid in the waste tank does not affect conclusions reached.

When waste is pumped through pipes to tank farms, it must be highly fluid with a large liquid fraction. In the past the plutonium concentration in discharged waste was maintained below 0.013 g/L (0.05 g/gal), and sometimes water was added to meet this limit. This low concentration and high fluidity ensured that the plutonium was spread over a wide area within the storage tank. Within the storage tank a concentration limit for plutonium in settled solids applies, and this concentration is much higher. The solids settle to the bottom and the liquid

(largely water) rises to the surface. The water occupies space and reduces the available storage volume. Pumping supernatant liquid through an evaporator to remove water reclaims this storage space. In this way, water needed to meet the discharge limits is later removed to create storage space.

C4.0 ESTIMATE OF TOTAL PLUTONIUM IN TANK FARMS

Kupfer et al. (1999) provides a best-basis estimate of the total (global) tank farm inventory for each plutonium isotope. These inventories are shown in Table C-2. The best-basis estimate of the total plutonium inventory including all isotopes, is 675.2 kg.

Table C-2. Global Radionuclide Inventory¹

Nuclide	Curies in 177 Tanks (Ci)	Kilograms in 177 Tanks (kg)
²³² U	1.23E+02	0.006
²³³ U	4.76E+02	49.4
²³⁴ U	3.46E+02	55.4
²³⁵ U	1.45E+01	6,730.
²³⁶ U	9.57E+00	149.
²³⁸ U	3.32E+02	958,000.
Total Uranium	---	965,000.
²³⁸ Pu	2.77E+03	0.16
²³⁹ Pu	3.91E+04	633.7
²⁴⁰ Pu	8.93E+03	39.3
²⁴¹ Pu	2.29E+05	2.01
²⁴² Pu	1.16E+00	0.29
Total Plutonium	---	675.2

Note:

¹Data from Kupfer et al. (1999, p.6-19)

Roetman et al. (1994) provides an estimate of the quantity of plutonium discharged as waste based upon a careful review of plutonium production and process efficiencies. In all, an estimated 67.5 metric tons of plutonium was produced at the Hanford Site, of which about 63 metric tons was processed at the Hanford Site. The difference is 4 metric tons of unprocessed plutonium in the K Basins and 528 kg of plutonium sent offsite for processing.

Several methods were used to estimate the mass of plutonium sent to waste. The best estimate for criticality safety evaluation is a conservative estimate based upon the yearly maximum waste loss approximation. Using this method, the total plutonium sent to waste is found to be 950 kg of plutonium from the separations processes and 31 kg from the PFP. This provides an upper limit on the total plutonium at tank farms of 981 kg.

Agnew (1997) estimates the total site inventory of plutonium in waste as 816 kg. The estimated quantity in SSTs is 529 kg and in DSTs is 246 kg. An additional 41.1 kg is estimated to be in cribs and 0.309 kg may have leaked into the ground.

The Plutonium Inventory Database (LMHC 1998) maintains an estimate of the plutonium content in each DST. For November 1999, the sum of the plutonium inventories for all DSTs equals 289.7 kg. This is an average of 10.3 kg of plutonium per DST. The largest plutonium inventory in any DST is 81.29 kg in DST AY-102.

Whyatt et al. (1996, Table A.4) compiles estimates of the plutonium inventory in SSTs obtained from the *Tank Waste Source Term Summary Validation* (WHC 1995), and from the *Hanford Tank Chemical and Radionuclide Inventories* (Agnew 1997). In 1997 SST C-106 had the largest plutonium inventory, but the contents have been transferred to DST AY-102. Now the plutonium inventory of 60.3 kg for SST C-104, based on WHC (1995), is the largest for any SST. This does not include 33 kg of ^{233}U reported by Hess (1991).

C5.0 ESTIMATE OF URANIUM INVENTORY

C5.1 AVERAGE URANIUM-235 ENRICHMENT

Kupfer et al. (1999) gives the best-basis estimate of the total inventory of uranium at tank farms to be 965,000 kg (see Table C-2). Of this 6,730 kg is estimated to be ^{235}U , and the average ^{235}U enrichment is found to be 0.697 wt%. Most uranium in tank waste is depleted.

Agnew (1997), using the HDW Model, Revision 4, estimates the total of uranium at tank farms to be 2,443,749 kg. Agnew places 2,257,185 kg in the 149 SSTs and 186,564 kg in the 28 DSTs. Table C-3 summarizes Agnew's estimates. Considering all uranium, the average ^{235}U enrichment is estimated to be 0.63 wt%, based on Agnew (1997).

Table C-3. Uranium Inventory¹

Location	Uranium kg	²³⁵U wt%
• Location		
NE Quadrant	1,146,765	0.59
SW Quadrant	620,223	0.68
NW Quadrant	489,198	0.67
SE Quadrant	186,564	0.65
Total in Tanks	2,443,749	0.63
Containment Type		
DSTs	186,564	0.65
SSTs	2,257,185	0.63
Cribs	188,469	0.03
Leaks	1,382	0.68
Total Onsite	2,633,600	---

Note:

¹Agnew (1997)

C5.2 WASTE STREAMS WITH ENRICHED URANIUM

Agnew (1997) inputs the composition of 48 waste streams into the HDW Model. The acronyms for 24 waste streams selected for having high uranium content or high ²³⁵U enrichment are listed in Table C-4, and the ²³⁸U and ²³⁵U content in these streams are shown in Table C-5. Nine waste streams contain enriched uranium, with the ²³⁵U content between 0.78 and 0.86 wt%. No waste stream is reported to have enrichment greater than 0.86 wt%.

The best-basis estimate of total uranium provided by Kupfer et al. (1999) is less than half as large as that provided by Agnew (1997). For this reason, Agnew's estimates of uranium content in individual waste streams should be conservative.

The three saltcake streams in Table C-5 show uranium concentrations to only one significant figure, and the uncertainty in the ²³⁵U enrichment is high. Even though the B-Saltcake waste stream indicates a ²³⁵U enrichment of 0.78 wt%, this high enrichment may be the result of the high uncertainty, and the uranium may not actually be enriched.

Table C-4. Acronyms for Waste Streams¹

Acronym	Description of Waste Stream
AR	PUREX waste; "washed" P sludge
B-SltCk	Saltcake from 242-B evaporator, 1951 until 1955
BY-SltCk	Saltcake generated from in-tank solidification in BY Farm, 1965-74
CSR	Waste from cesium recovery from supernatants
CWPI	PUREX cladding waste, 1956-60
CWR1	REDOX waste, aluminum clad fuel, 1952-60
CWR2	REDOX waste, aluminum clad fuel with some Zr fuel, 1961-67
CWZr1	PUREX cladding waste, Zircaloy cladding, 1966-70
CWZr2	REDOX coating waste, Zirconium cladding, 1983-88
MW1	Metal waste from BiPO ₄ and uranium recovery, 1944 to 1951
MW2	Metal waste from BiPO ₄ and uranium recovery, 1952 to 1956
OWW1	PUREX organic wash waste, 1956-62
OWW3	PUREX organic wash waste, 1968-72
P2	PUREX high-level waste, 1963 to 1967
P3	PUREX high-level waste, 1983-88
PfeCN1	Ferrocyanide sludge from in-plant scavenging from uranium recovery
PL2	PUREX low-level waste
R2	REDOX waste, 1958 to 1966
T1-SltCK	Saltcake from 242-T Evaporator, 1951 to 1955
TfeCN	Ferrocyanide sludge from in-tank or in-farm scavenging
UR/TBP	Uranium recovery operation in 222-S; created TBP waste
1C1	First cycle decontamination waste from bismuth phosphate process
1C2	First cycle decontamination waste from bismuth phosphate process
1CfeCN	Ferrocyanide sludge from in-plant scavenging of 1C supernatant waste

Note:

¹Agnew (1997, Appendix F)

Table C-6 lists the nine waste streams shown to contain enriched uranium. The uranium fraction in grams of uranium per gram of solids is used to estimate the quantity of enriched uranium sent to each tank. The total enriched uranium for all waste streams is estimated at 118,000 kg, including the B-Saltcake stream. This is about 4.9% of the total uranium in tank farms. If the B-Saltcake stream is not included, the total enriched uranium is 93,700 kg, or about 3.8% of the total uranium in tank farms. The CWR2 waste stream contains the largest content of enriched uranium and also the largest uranium/sludge mass ratio of 0.0704.

Single-shell tank S-107 has received 16,200 kg of enriched uranium from the CWR2 waste stream. This is the most significant quantity of enriched uranium sent to an individual tank. However, the enrichment of 0.85 wt% is too low for criticality to occur, even in an unlimited quantity with optimal moderation. Criticality is not possible with the uranium in any of the waste streams sent to tank farms, even if it were to be separated from all other solids.

Table C-5. Waste Streams With High Uranium Content or Enrichment¹

Waste Stream	²³⁸ U	²³⁵ U	²³⁸ U	²³⁵ U	Enrichment wt% ²³⁵ U
	Agnew Data in Ci/L		Converted to g/L		
MW1	1.698E-5	7.542E-7	51.0	0.352	0.69
MW2	1.711E-5	7.434E-7	51.4	0.347	0.67
1C1	2.488E-6	1.105E-7	7.47	0.0516	0.69
1C2	2.393E-6	1.040E-7	7.19	0.0486	0.67
UR/TBP	1.21E-7	5.31E-9	0.363	0.00248	0.68
PFeCN1	1.21E-7	5.31E-9	0.363	0.00248	0.68
TfeCN	3.2E-5	5.3E-9	96.1	0.00248	0.00
1CfeCN	2.393E-6	1.04E-7	7.19	0.0486	0.67
R2	1.675E-7	9.057E-9	0.503	0.00423	0.83
CWR1	1.459E-6	6.368E-8	4.38	0.0298	0.67
CWR2	1.434E-6	7.831E-8	4.31	0.0366	0.84
P2	2.314E-6	1.183E-7	6.95	0.0553	0.79
CWP1	1.19E-6	4.96E-8	3.57	0.0232	0.64
CWZr1	9.416E-7	5.066E-8	2.83	0.0237	0.83
OWW1	1.030E-6	4.303E-8	3.09	0.0201	0.65
OWW3	4.54E-7	2.32E-8	1.38	0.0108	0.79
AR	1.87E-7	8.05E-9	0.562	0.00376	0.66
CSR	4.2E-7	1.81E-8	1.26	0.00846	0.67
B-SltCk	1E-6	5E-8	3.00	0.0234	0.77
T1-SltCk	9E-7	4E-8	2.70	0.0187	0.69
BY-SltCk	3E-6	5E-8	9.01	0.0234	0.26
P3	1.015E-6	5.619E-8	3.05	0.0263	0.85
PL2	3.997E-7	2.213E-8	1.20	0.0103	0.85
CWZr2	6.156E-7	3.408E-8	1.85	0.0159	0.85

Note:

¹Data from Agnew (1997, pp B-14 to B-17)

Table C-6. Waste Streams With Enriched Uranium¹

Waste Stream	U/sludge g/g	Enrichment wt% ²³⁵ U	Tank	Volume kL	Uranium ² kg
R2	0.00017	0.84	SX-111	155.2	38
			SX-114	140.0	35
			SX-110	121.1	30
			SX-112	94.6	23
			SX-107	90.8	22
CWR2	0.0704	0.85	S-107	159.0	16,200
			T-101	132.5	13,500
			U-108	98.4	10,000
			C-104	56.8	5,800
			T-105	7.6	770
P2	---	0.80	---	---	---
CWZr1	0.0145	0.84	C-104	102.2	2,150
			S-107	68.1	1,430
			C-101	49.2	1,000
OWW3	0.0410	0.80	C-104	117.3	6,980
B-SltCk³	0.0095	0.78	B-105	492.1	6,800
			B-106	439.1	6,050
			B-101	321.8	4,430
			B-109	318.0	4,380
			B-104	230.9	3,180
P3	0.0363	0.86	AZ-101	102.2	5,380
			AZ-102	49.2	2,600
PL2	0.0111	0.86	AW-104	370.9	5,970
			AW-101	230.9	3,720
			AW-105	94.6	1,520
			AY-102	15.1	240
CWZr2	0.0073	0.86	AW-103	1374.0	14,500
			AW-105	813.7	8,600
			AW-104	18.9	200

Notes:

¹Data from Agnew (1997)²Estimated uranium based on assumed sludge density of 1.45 g/cm³.³Enrichment is uncertain. This uranium may not be enriched.

C6.0 ESTIMATE OF URANIUM-233 INVENTORY

Hess (1991) reviewed historical records to arrive at an estimate of the total quantity of ^{233}U sent to tank farms. Hess summarized his review as follows:

The results of that review indicate that a total of about 45 kg of ^{233}U was discharged out of PUREX to underground tanks in C-Farms during 1966 and 1970 thoria campaigns. Although the total volume of liquid waste transferred was on the order of several million gallons, it is highly probable that the ^{233}U settled in the initial tanks of a cascade of tanks used. This would have resulted in ^{233}U accumulations of 33 kg in tank 104-C (241-C-104) from the 1970 campaign and 12 kg in 102-C from the 1966 campaign. It is recommended that these amounts be added to inventories of 104-C and 102-C, and that future sampling in all of the C-Farm tanks should include tests for thorium and to further track the waste disposition from the thoria campaigns.

Roetman et al. (1994) makes the follow statement concerning ^{233}U :

Another fissionable isotope of criticality concern is ^{233}U . The thorium irradiation process resulted in the direct production of ^{233}U , which was then recovered with the PUREX process. The waste from this recovery was discharged to the waste tanks. Since the recovery of ^{233}U from thoria was much more difficult than the separation of plutonium from uranium, the recovery efficiency was less, approximately 95%. Approximately 45 kg of ^{233}U were discharged to the Hanford waste tanks from the 1966 and 1970 thoria campaigns.

Agnew (1997) estimates the total quantity of ^{233}U to be 994 Ci. When divided by 0.0095 Ci/g, this converts to 104.6 kg, a value twice as large as those given above. The estimated total quantity in SSTs is 87.0 kg and in DSTs is 17.6 kg.

C7.0 TANK-AVERAGED ABSORBER/PLUTONIUM MASS RATIOS

The inventory of a tank can be used to determine the average plutonium concentration and the average solids/plutonium mass ratio for waste in that tank. Likewise, the total inventory of tank farms can be used to determine the average value for these parameters for tank farms as a whole. The average value is also the value "expected" when a new waste sample is analyzed.

Roetman et al. (1994) estimates the total quantity of plutonium discharged to tank storage to have been 981 kg since the beginning of plutonium production. Table C-7 summarizes volumes of various waste types reported by Hanlon (1999).

It is conservative to overestimate the plutonium and underestimate the volume of solids. Supernatant, slurry, and saltcake contain very little plutonium, and their volumes are not included in the estimate of the volume of waste solids. All plutonium is assumed in the slurry and sludge. Using a sludge volume of 49,550 kL (13,090 kgal) and a tank farm plutonium inventory of 981 kg, the average plutonium concentration in sludge is 0.020 g/L. When a low solids density of 1.2 g/cm³ (1,200 g/L) is assumed, the global solids/plutonium mass ratio for tank farms is estimated to be 60,600. Although conservative, this is, nevertheless, estimated to be 76 times greater than required to ensure subcriticality when homogenized.

	SST	DST	Total
Supernatant liquid (kL)			
Total supernatant	5,788	55,250	61,038
Solids (kL)			
Double-shell slurry	0	1,730	1,730
Sludge	44,420	3,400	47,820
Saltcake	79,030	12,150	91,180
Total solids	123,400	17,300	140,700

Note:

¹Hanlon (1999)

C7.1 MASS RATIOS FOR SELECTED TANKS

Solids/plutonium mass ratios are calculated for several storage tanks selected because of having a high plutonium inventory or a low ratio of solids. Double-shell tank SY-102, DST AW-105 and SST C-104 are described to illustrate the quantity of solids expected.

C7.1.1 Mass Ratio for SST C-104

The plutonium inventory for SST C-104 is the largest in tank farms. The inventory, which is conservatively estimated at 89 kg, is based upon an estimated 60.3 kg of plutonium (Whyatt et al. 1996, Table A.4) and 33 kg of ²³³U (Hess 1991). Hanlon (1999) reports that SST C-104 contains 1,117 kL (295 kgal) of sludge. The average plutonium concentration is found to be 0.08 g/L, and the average solids/plutonium mass ratio is 15,055.

C7.1.2 Mass Ratio for DST AW-105

The Plutonium Inventory Database (LMHC 1998) shows the plutonium inventory for DST AW-105 for January 2000 to be 22.95 kg. In addition, the volume of solids is shown to be 1,060 kL (280 kgal), the plutonium concentration to be 0.022 g/L (0.00002 g/cm³) in the solids, and the solids/plutonium mass ratio to be 55,409.

C7.1.3 Mass Ratio for DST AY-102

In January 1999 the Plutonium Inventory Database (LMHC 1998) showed a plutonium inventory for DST AY-102 of 11.55 kg and a solids/plutonium mass ratio of 14,224. Between March and October 1999 nearly the contents of SST C-106 was transferred into this tank, and the plutonium inventory underwent a major increase. In January 2000 the plutonium inventory was listed as 80.18 kg, and the volume of solids was 799 kL (211 kgal). The plutonium concentration in the solids at this time was listed as 0.0966 g/L (0.000097 g/cm³), and the solids/plutonium mass ratio was 13,959.

C7.1.4 Mass Ratio for DST SY-102

The Plutonium Inventory Database (LMHC 1998) shows the DST SY-102 plutonium inventory for January 2000 to be 42.30 kg, the volume of solids to be 269 kL (71 kgal), and the solids density to be 1.56 g/cm³. The plutonium concentration is reported to be 0.15 g/L (0.00015 g/cm³) in the solids, and the solids/plutonium mass ratio is 10,117. This is the smallest proportion of solids for any other tank.

C8.0 UNIQUE ITEMS IN TANK STORAGE

"Unique items" are items disposed of in waste tanks on a one-time-only basis whose physical and chemical characteristics differ greatly from the normal tank contents. Some unique items contain small quantities of fissile material. Unique items disposed of in SSTs are listed in Table C-8. No unique items have been disposed of in DSTs.

No significant reduction in the margin of criticality safety has resulted from unique items in waste storage tanks. Credible configurations of unique items combined with normal tank waste will remain well subcritical.

C8.1 DIATOMACEOUS EARTH

Large quantities of diatomaceous earth containing no fissionable materials were added to SSTs BX-102, SX-113, TX-116, TX-117, TY-106, and U-104. Diatomaceous earth is primarily silicon dioxide, a material with a very low neutron absorption cross section. The subcritical limit plutonium concentration for silicon dioxide is 0.70 g/L, based on a k_{eff} less than 0.95 at a 95% confidence level (Altschuler 1991).

The diatomaceous earth acts as a diluent and decreases the concentrations of waste components, but not their component/plutonium mass ratios. The value of neutron absorption cross section per fissile atom is nearly unchanged by this dilution. There is no mechanism that would selectively combine the plutonium with silicon dioxide. The presence of the diatomaceous earth adds a small amount of neutron absorption. By acting as a diluent, the diatomaceous earth increases the plutonium critical mass and increases the margin of subcriticality.

Table C-8. Unique Contents of Single-Shell Tanks

SST	Contents
BX-101	7 m ³ of ARC-359/organic ion exchange resin.
BX-102	95,000 kg of diatomaceous earth.
BY-105	57,000 kg of Portland cement.
SX-107	41 bottles of neutralized waste from F area. < 1 g of ²³⁹ Pu per bottle.
SX-110	16 plastic bottles (7.6 cm diameter by 137.2 cm long) with: 113 g natural uranium 53 g depleted uranium 6 g ²³⁵ U 204 g ²³⁹ Pu
SX-113	41,000 kg of diatomaceous earth.
TX-116	95,000 kg of diatomaceous earth.
TX-117	41,000 kg of diatomaceous earth.
TY-106	27,000 kg of diatomaceous earth.
U-101	6 casks with experimental fuel elements, shroud tubes, and samarium "poison" ceramic ball containing: 1530 g 4.5% enriched uranium 6 g Pu 180 kCi ⁶⁰ Co 130 Ci mixed fission products Slugs with 130 Ci of ⁶⁰ Co.
U-104	55,000 kg of diatomaceous earth.

C8.2 PORTLAND CEMENT

Portland cement was added to SST BY-105. Portland cement acts as a diluent, but it contains more neutron absorption than does diatomaceous earth. The addition of Portland cement increases the margin of subcriticality.

C8.3 UNIQUE ITEMS CONTAINING FISSILE MATERIAL

"Unique items" containing small quantities of fissile material were added to SSTs SX-101, SX-107, and SX-110. SST SX-107 received less than 42 g of plutonium, and SST U-101 received fuel elements with an estimated 75 g of fissile material.

Single-shell tank SX-110 received 16 plastic bottles holding 204 g ^{239}Pu and 6 g ^{235}U . The most reactive possible configuration for these 16 plastic bottles is a close-packed array with optimal interior water and a close-fitting reflector. A conservative configuration to represent this array is an optimally water-moderated, homogeneous sphere with full water reflection. For ^{239}Pu , the minimum critical mass is 520 g at a concentration of 30 g Pu/L. Assuming all 210 g of fissile material is ^{239}Pu , these 16 containers represent 40% of the smallest possible critical mass.

Each plastic bottle has a diameter of 7.6 cm and a volume of 6.2 L. The average plutonium concentration is 2.1 g/L when confined to the bottles. At optimal concentration of 30 g Pu/L, the plutonium would occupy about 7% of the volume in the bottles. Therefore, if undamaged, the bottles, even if close-packed, would not form an optimal array. In reality, the 16 plastic bottles would likely be spaced apart, and their arrangement in the storage tank would be far from optimal. The most probable arrangements would be expected to represent a very small fraction of a critical mass.

The harsh environment inside a waste tank should rapidly destroy the plastic bottles and disperse their contents. Mixing with waste material would quickly ensure that optimal conditions would not exist.

Waste already in the tank would contain plutonium at a concentration less than 0.10 g Pu/L, a value less than 4% of the minimum critical concentration. The plutonium in the waste, because of its very low concentration, would have only a negligible impact on reactivity. The postulated sphere formed by the unique material would have a fraction of a critical mass of 0.40. When combined with the other waste, the waste would remain well subcritical. Any disturbance or mixing of the waste should further disperse the contents of the plastic bottles and lower the plutonium concentration to a value closer to the tank average concentration.

C9.0 ENHANCED PLUTONIUM PRODUCTION AT SURFACE OF FUEL ELEMENTS

Plutonium is produced during the process of irradiating (burning) uranium fuel in a reactor. However, the plutonium is not produced uniformly over the volume of the fuel, but rather is produced in greater concentration near the outer surface (periphery). Enhanced plutonium production at the surface of uranium fuel elements is a possible source of unaccounted for plutonium in waste streams containing fuel element cladding waste.

Schwinkendorf (1994) examines the possibility that enhanced production near the surface of natural uranium elements might provide a means of increasing the plutonium concentration in nuclear waste. Since fuel near the surface is in intimate contact with the cladding, this surface layer might be removed with the cladding and become part of the waste stream. In other words, fuel in a thin layer next to the cladding not only had a higher content of plutonium, but was discarded as waste in a higher proportion than was fuel from the interior of the element. In addition, its presence would have gone undetected in the laboratory analysis of fuel.

Schwinkendorf (1994) calculated the minimum critical mass for an optimal configuration of material taken from the outer surface of high-exposure, natural uranium fuel elements. For these calculations, the following assumptions were made: fission products (neutron poisons) have been removed, uranium and plutonium stay together, chunks of pure metal of optimum size are formed at an optimum distance from each other, and the interspersed water does not contain neutron poisons. Even for these idealized conditions, the mass required for criticality was in excess of 10 metric tons (10^4 kg) of heavy metal.

When natural uranium is irradiated, plutonium is produced faster than ^{235}U is burned. However, when green fuel with a higher $^{235}\text{U}/^{238}\text{U}$ ratio is irradiated, the ratio of plutonium production to ^{235}U burnup decreases. Schwinkendorf (1997, Appendix G) reaches the following conclusion concerning the radial isotopic evolution during burnup of 0.95 wt% and 1.25 wt% enriched uranium:

In all burnup calculations reported in this appendix, the production of plutonium is enhanced at the outer surfaces of the fuel; there is increased exposure near the surfaces because of self-shielding. However, this effect is at least partially mitigated by the fact that enhanced fissile uranium depletion also occurs near the fuel surfaces. In addition, the increased plutonium production near the surface also has a higher ^{240}Pu content, again, because of the increased exposure at the surface. The number of neutrons produced per fission is higher for ^{239}Pu than ^{235}U , and so the plutonium is worth more than the fissile uranium, but the effect is not large. Scrap material composed of the outer skin of the fuel is more reactive than the average fuel, but not to a significant degree.

For 1.25 wt% enriched uranium, the sum of the ^{235}U and ^{239}Pu concentrations does not exceed that of green fuel at any radial position, regardless of exposure time. In a layer between 0.05 and 0.1 cm thick comprising only a few percent of the total fuel volume, the total fissile concentration remains almost unchanged during exposure. After 52 days exposure of 1.25 wt% uranium elements, the ^{235}U concentration on the surface has decreased by about 0.10 wt% of the uranium, while the ^{239}Pu has increased to about 0.10 wt%. After 192 days exposure, the ^{235}U decrease is about 0.30 wt%, while ^{239}Pu increased to about 0.28 wt%. In other words, in a 0.10 cm deep layer the ^{235}U lost is replaced by an equal quantity of ^{239}Pu , which has a smaller critical mass. When this replacement is taken into account, the outer 0.05-cm thick layer of each element is found to increase in reactivity during irradiation.

Below the surface of the element the total fissile concentration decreases with fuel exposure. After 52 days of exposure, the ^{235}U content has dropped by 0.08 wt% to about 1.17 wt% of the uranium mass, while the ^{239}Pu content has increased to about 0.05 wt%. After 192 days of exposure, the ^{235}U content has dropped by 0.26 wt% to about 0.99 wt%, while the ^{239}Pu content has increased to about 0.14 wt%. Therefore, for the fuel element as a whole, the loss of ^{235}U is 1.6 to 1.8 times greater than the production of ^{239}Pu .

Using 192 days as the upper limit on exposure, the plutonium content of 95% of the 1.25 wt% enriched uranium fuel will not exceed 0.14 wt%, while in the other 5%, the plutonium content will not exceed 0.30 wt%. The plutonium content in the sludge then depends on whether, or not, a higher fraction of the surface uranium becomes sludge than does interior uranium. However, the surface layer is such a small fraction of the total uranium that, even if it became waste at a higher proportion, it would not compose a large fraction of the uranium in any waste stream. In addition, the surface layer would be associated with the cladding, and cladding waste is expected to contain a larger proportion of neutron absorbing cladding components. When the total neutron absorber/plutonium mass ratio in cladding waste streams is taken into account, the margin of subcriticality remains high even with a higher proportion of surface uranium.

C10.0 REFERENCES

- Agnew, S. F., 1997, *Hanford Tank Chemical and Radionuclide Inventories: HDW Model Rev. 4*, LA-UR-96-3860, Los Alamos National Laboratory, Los Alamos, New Mexico.
- Altschuler, S. J., 1991, *CSER 91-011: Minimum Critical Safety Limits for Nuclear Waste*, WHC-SD-SQA-CSA-20339, Westinghouse Hanford Company, Richland, Washington.
- Anderson, J. D., 1990, *A History of the 200 Area Tank Farms*, WHC-MR-0132, Westinghouse Hanford Company, Richland, Washington.
- Bratzel, D. R., W. W. Schulz, R. Vornehm, and A. E. Waltar, 1996, *Tank Farm Nuclear Criticality Review*, WHC-SD-WM-TI-725, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- Braun, D. J., L. D. Muhlestein, T. B. Powers, and M. D. Zentner, 1994, *High-Level Waste Tank Subcriticality Safety Assessment*, WHC-SD-WM-SARR-003, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- Brevick, C. H., L. A. Gaddis, and E. D. Johnson, 1995, *Tank Waste Source Term Inventory Validation*, 3 Volumes, WHC-SD-WM-ER-400, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- CHG, 2000, *Tank Characterization Database*, maintains Best Basis Inventory on Hanford intranet at <http://twins.pnl.gov:8001/TCD/main.html>, CH2M Hill Hanford Group, Inc., Richland, Washington.
- Cowley, W. L., 1996, *Development of Radiological Concentrations and Unit Liter Doses for TWRS FSAR Radiological Consequence Calculations*, WHC-SD-WM-SARR-037, Westinghouse Hanford Company, Richland, Washington.
- Hanlon, B. M., 1999, *Waste Tank Summary Report Summary Report for Month Ending July 31, 1999*, HNF-EP-0182-136, Fluor Daniel Hanford, Inc., Richland Washington.
- Hess, A. L., 1991, *Review of Records for Source of ²³³U in Tank 104-C* (Letter 29210-91-030 to S. D. Godfrey), Westinghouse Hanford Company, Richland, Washington. This is provided in Appendix B of *Criticality Safety of Double-Shell Waste Storage Tanks*, WHC-SD-SQA-CSA-20368 (Rogers 1994).

- Jungfleisch, F. M., 1983, *Supplementary Information to the Preliminary Estimation of the Waste Inventories in Hanford Tanks Through 1980*, SD-WM-TI-058, Rockwell Hanford Operations, Richland, Washington.
- Jungfleisch, F. M., 1984, *Preliminary Estimation of the Waste Inventories in Hanford Tanks Through 1980*, SD-WM-TI-057, Rockwell Hanford Operations, Richland, Washington.
- Kupfer, M. J., A. L. Boldt, K. M. Hodgson, L. W. Shelton, B. C. Simpson, R. A. Watrous; M. D. LeClair; G. L. Borsheim; R. T. Winward; B. A. Higley, R. M. Orme; N. G. Colton, S. L. Lambert, D. E. Place; and W. W. Schultz, 1999, *Standard Inventories of Chemicals and Radionuclides in Hanford Site Tank Wastes*, Lockheed Martin Hanford Corporation, Richland, Washington.
- Lipke, E. J., 1999, *Above Average Sample in 241-TX-118*, (Letter 74F00-EJL-99028 to N. L. Hulse), Lockheed Martin Hanford Corporation, Richland, Washington.
- LMHC, 1998, *Plutonium Inventory Database*, HISI 11350, Lockheed Martin Hanford Company, Richland, Washington.
- Roetman, V. E., S. P. Roblyer, and H. Toffer, 1994, *Estimation of Plutonium in Hanford Site Waste Tanks Based on Historical Records* WHC-EP-0793, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- Rogers, C. A., 1994, *Criticality Safety of Double-Shell Waste Storage Tanks*, WHC-SD-SQA-CSA-20368, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- Schwinkendorf, K. N., 1994, *Analysis of Radial Isotopic Profile for Hanford Site Aluminum-Clad Fuel Elements*. This is Appendix A.8.0 of Braun et al. (1994), *High Level Waste Tank Subcriticality Safety Assessment*.
- Schwinkendorf, K. N., 1997, *Criticality Safety Evaluation for Spent Nuclear Fuel Processing and Storage Facilities*, HNF-SD-SNF-CSER-005, Rev. 3, Fluor Daniel Northwest, Richland, Washington.
- Serne, R. J., G. A. Whyatt, S. V. Mattigod, Y. Onishi, P. M. Doctor, B. N. Bjornstad, M. R. Powell, L. M. Liljegren, J. H. Westsik, Jr., N. J. Aimo, K. P. Recknagle, G. R. Golcar, T. B. Miley, G. R. Holdren, D. W. Jeppson, R. K. Biyani, and G. S. Barney, 1996, *Fluid Dynamics, Particulate Segregation, Chemical Processes, Natural Ore Analog and Tank Inventory Discussions that Relate to the Potential for Criticality in Hanford Tanks*, WHC-SD-WM-TI-757, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- WHC, 1995, *Tank Waste Source Term Inventory Validation*, WHC-SD-WM-ER-400, Westinghouse Hanford Company, Richland, Washington.

Whyatt, G. A., R. J. Serne, S. V. Mattigod, Y. Onishi, M. R. Powell, J. H. Westsik, Jr., L. M. Liljegren, G. R. Golcar, K. P. Recknagle, P. M. Doctor, V. G. Zhirmov, J. Dixon, D. W. Jeppson, and G. S. Barney, 1996, *The Potential for Criticality in Hanford Tanks Resulting from Retrieval of Tank Waste*, PNNL-11304, Pacific Northwest National Laboratory, Richland, Washington.

APPENDIX D

PHYSICAL AND CHEMICAL PROCESSES IN TANK WASTE

This page intentionally left blank.

D.0 PHYSICAL AND CHEMICAL PROCESSES IN TANK WASTE

At the time of discharge, waste is assured of being subcritical because of limits and controls placed upon it by tank farms. Within the waste tank environment, however, physical and chemical processes capable of changing uranium and/or plutonium concentrations are of concern. Waste sent to tank farms in the past was the product of processes in which the uranium and plutonium was first dissolved and then precipitated. This Criticality Safety Evaluation Report (CSER) summarizes published studies of physical and chemical processes of importance to criticality safety, but no new chemistry information is developed.

For criticality to occur, the plutonium concentration must exceed the minimum concentration for which criticality is possible. There are two ways to achieve a high plutonium concentration in waste solids: to receive waste already having a high plutonium concentration or to concentrate plutonium after it reaches the storage tank. Knowledge of tank waste chemistry is required to justify the conclusion that a plutonium concentration high enough to lead to criticality is not credible in the tank waste environment.

Plutonium processing facilities are shut down, and the quantity of plutonium discharged in past operations greatly exceeds that which is likely to be discharged in the future. Although the present or future tense may be used in the following discussion, it applies also to the past.

D1.0 SOURCES OF CHEMISTRY INFORMATION

These documents provide a comprehensive review of the chemistry associated with tank waste. Most of this waste is the byproduct of the extraction of plutonium from reactor fuel.

D1.1 PLUTONIUM CHEMISTRY

Evaluations of plutonium chemistry related to tank waste are documented by:

- *Tank Farm Nuclear Criticality Review* (Bratzel et al. 1996);
- *Fluid Dynamics, Particulate Segregation, Chemical Processes, Natural Ore Analog and Tank Inventory Discussions that Relate to the Potential for Criticality in Hanford Tanks* (Serne et al. 1996);
- *The Potential for Criticality in Hanford Tanks Resulting from Retrieval of Tank Waste* (Whyatt et al. 1996).

Three Russian chemistry studies related to Hanford Site tank waste are documented by:

- *Plutonium(IV) Precipitates Formed in Alkaline Media in the Presence of Various Anions* (Krot et al. 1998);
- *Interaction of Pu(IV,VI) Hydroxides/Oxides with Metal Hydroxides/Oxides in Alkaline Media* (Fedoseev et al. 1998);
- *Purification of Alkaline Solutions and Wastes from Actinides and Technetium by Coprecipitation with Some Carriers Using the Method of Appearing Reagents* (Peretrukin et al. 1998).

Criticality Safety Assessment of Tank 241-C-106 Remediation (Vail 1997b), which documents the evaluation of transferring waste from DST C-106 to DST AY-102, provides a good review of the chemical processes that ensure criticality safety in waste disturbing operations.

D1.2 URANIUM CHEMISTRY

An evaluation of uranium chemistry is found in:

- *Feasibility Report on Criticality Issues Associated with Storage of K Basin Sludge in Tank Farms* (Daling et al. 1997).

D2.0 ALKALINITY

By providing alkaline conditions, the solubility of plutonium is assured of being low, and the plutonium settles to the bottom along with other insoluble waste components. The upper layers of liquid can then be pumped to another tank while leaving most of the plutonium behind. In this way, solids and plutonium can be accumulated in selected tanks.

Based upon published data from chemical experiments with plutonium, the upper limit on the concentration of dissolved plutonium in liquid waste should not exceed 0.0017 g/L (see Appendix D2.0). For uranium, the limiting concentration is 0.017 g/L.

Plutonium can be concentrated by dissolving it and then precipitating it. Dissolution requires a change from alkalinity to acidity, and precipitation requires the reverse. Because chemical compatibility is determined before waste transfer, it is highly unlikely that transfer of an acidic waste would ever be made. If an inadvertent transfer of acidic waste were made, a large quantity would be required to cause dissolution of the plutonium.

Bratzel et al. (1996) provides the following excerpt concerning acidification of waste:

Acidification of wastes in the underground tanks could cause some dissolution of oxyhydroxides or oxides of plutonium and other metals on which the plutonium may be sorbed and thus result in dissolution of some of the plutonium. Of greater consequence would be the large volume of gases generated by reactions with nitrite ions to produce NO_x gases, with carbonate ions to produce CO₂, and with the carbon steel tanks to produce hydrogen gas. Acidification would eventually cause failure of the carbon steel pipelines and steel liner, allowing liquid waste to escape into the environment. Because of these disastrous consequences, accidental acidification of the wastes must be avoided.

D2.1 LIMIT ON pH

The following limits are placed upon the pH for tank waste:

	<u>pH Limit</u>
pH at time of discharge:	8.0 (minimum)
Investigation Levels (for analyzed waste samples)	
For tank with ≤ 30 cm (12.0 in.) depth of supernate:	not limited
For tank containing ≤ 10 kg of plutonium:	not limited
For tank containing > 10 kg of plutonium <u>and</u> with > 30 cm (12.0 in.) depth of supernate:	8.0 (minimum)

A pH between 0 and 7 indicates acidity and between 7 and 14 indicates alkalinity. Water is neutral, neither acidic nor alkaline, with a pH of 7. pH is an indicator of the degree to which material contained in the waste is insoluble. A high pH guarantees a low solubility of plutonium and of neutron absorbers.

The limit on pH is a secondary limit. A low pH does not by itself create conditions that might cause criticality. Requiring the waste to be alkaline (i.e., to have a high pH) forces important components of the waste, including plutonium, to precipitate. This ensures that the plutonium will be combined with a large ratio of neutron absorbing solids. Hence, a limit on pH protects the key assumptions of this evaluation. The pH is important to criticality safety because a high pH increases confidence that the solids/plutonium mass ratio will remain high. When the waste is alkaline, nearly all plutonium entering a tank will remain in that tank, and this permits a more accurate knowledge of tank inventories after waste solutions are transferred between tanks.

More than 1,000 kg of plutonium is required to be spread uniformly across a storage tank to reach the minimum critical areal density. This remains true even if all neutron absorbing solids are removed. The process by which waste is generated and sent to tank storage tends to disperse the plutonium and virtually guarantees a low concentration. It is extremely unlikely that a plutonium concentration even approaching the subcritical limit would be achieved, regardless of the pH.

When the plutonium concentration is less than 7 g/L, criticality is not possible in an unlimited volume of water or in a 50-cm (20-in.) thick waste layer at optimal water content. No matter how acidic, a 30-cm (12-in.) thick layer of supernatant would not be capable of dissolving and redistributing plutonium in any manner capable of even approaching criticality. For this reason, when the liquid level is less than 30 cm thick, no restriction is placed upon the pH.

When the total quantity of plutonium in a tank is less than 10 kg, the average areal density is less than 1.0 % of the minimum required for criticality. For criticality to occur, it would be necessary for a large fraction of the tank contents to come together in a compact volume of less than 500 L. This is not considered credible for two reasons. First, there is no mechanism capable of separating the plutonium from agglomerated solids; and second, there is no mechanism capable of depositing all of the plutonium in a small region of the tank. For these reasons, no restriction is placed upon pH when the plutonium inventory is less than 10 kg.

No restriction is placed on pH when the plutonium inventory is less than, or equal to, 10 kg or the depth of supernatant liquid is less than, or equal to, 30 cm. When the plutonium inventory exceeds 10 kg and the depth of liquid exceeds 30 cm, the pH is required to exceed 8.0.

D2.2 EXPECTED PH

The pH in tank waste is expected to be about 12. In a study by Wodrich et al. (1992), the pH of waste in 139 SSTs was reported as follows: no tanks showed a pH less than 7.0; 5 tanks showed a pH less than 8.0; and 20 tanks showed a pH between 8 and 10. The lowest pH originated from the bismuth phosphate process used at T-Plant between 1944 and 1956 and at B-Plant from 1945 to 1952.

Braun et al. (1994) reported 385 pH measurements, mostly for SSTs. Of these, 9 samples showed a pH below 9.0, but none showed a pH below 7.0. This indicates that about 1 out of every 40 measurements might be expected to show a pH less than 9.0.

Tank farms control the pH of waste to reduce corrosion. The *Operating Specifications for 241-AN, AP, AW, AY, AZ, & SY Tank Farms* (LMHC 1998) specify that tank contents comply with the composition limits provided. Requirements are placed on the nitrite, nitrate, and hydroxide ion (NO_2^- , NO_3^- , and OH^-) concentrations in order to minimize the rate of corrosion and stress corrosion cracking (SCC). The OH^- concentration must exceed 0.01 molar.

D3.0 SOLUBILITY OF PLUTONIUM IN WASTE

The solubility of plutonium in high-level waste is an important concern of criticality safety. By maintaining the waste alkaline, a low solubility for plutonium is assured, and the plutonium will remain combined with solids and be less mobile.

D3.1 CONCENTRATION OF DISSOLVED PLUTONIUM

Hobbs et al. (1993) investigated the influence of hydroxide, nitrate, nitrite, aluminate, sulfate, carbonate, and temperature on the solubility of plutonium and uranium in alkaline salt solutions and reached the following conclusion:

For salt solutions within the range considered by this study, upper 95% tolerance limits for Pu and U solubilities, estimated from analytically measured specie concentrations, are 1.68 mg/L and 17.1 mg/L (6.36 mg/gal and 64.7 mg/gal), respectively. The alkaline salt solutions considered in this study should bound those that will be treated in the in-tank precipitation process.

Hobbs et al. (1993) also concluded that:

Conditions which increase the plutonium solubility do not necessarily increase the uranium solubility. Maximum plutonium solubility is predicted at high nitrate, high hydroxide, high carbonate, and high sulfate concentrations. Maximum uranium solubility is predicted at high hydroxide, high aluminate and high sulfate concentrations and low nitrate, low nitrite, and low carbonate concentrations.

Since the upper bounds of plutonium solubility in an alkaline salt solution is 0.0017 g/L, the concentration in solution is negligible when compared to the minimum critical plutonium concentration of 7.2 g/L in water. For this reason, the tracking of plutonium in supernate satisfies an accountability concern rather than a criticality safety concern.

Hobbs et al. (1993) makes the following observation concerning the effect upon solubility of waste components in a 500 L (132 gal) volume of 10 g Pu/L solution:

The solubilities reported in this document may be higher than those which will be observed in actual waste due to the coprecipitation of the plutonium and the uranium with iron, manganese, and aluminum during neutralization of fresh waste. In a single experiment in which the plutonium and uranium were coprecipitated with iron, the solubility of plutonium was decreased by a factor of about five, and that of uranium increased by about a factor of two.

D3.2 EFFECT OF SODIUM HYDROXIDE ON PLUTONIUM SOLUBILITY

Although the common understanding is that plutonium precipitates in alkaline solution, this simple understanding does not fully describe the complex nature of the chemistry involved in waste. Delegard (1985) shows that the solubility of plutonium increases for increasing NaOH:

The solubility of Pu(IV) hydrous oxide, $\text{PuO}_2 \cdot x\text{H}_2\text{O}$, in air-equilibrated, synthetic, Hanford high-level waste (HLW) solutions was determined as a function of NaOH, $\text{NaAl}(\text{OH})_4$, NaNO_3 , NaNO_2 , and Na_2CO_3 concentrations. The solubility was found to increase with the square of the NaOH chemical activity. The components NaNO_3 and NaNO_2 increased $\text{PuO}_2 \cdot x\text{H}_2\text{O}$ solubility by increasing NaOH activity.

Although the plutonium solubility is found to increase with addition of NaOH, the solubility, nevertheless, remains very low. Delegard (1985) studied the solubility of plutonium in NaOH solutions over the range of NaOH concentration between 1 and 15 molar.

For typical waste, Delegard (1985) states that the NaOH concentration is 4 molar. At this concentration the maximum plutonium concentration is found to be about 0.0005 g/L (0.0019 g/gal or 2×10^{-6} molar). When allowed to set for a long time, the plutonium concentration in solution decreases to a value about 8 times smaller. When the NaOH is increased from 4 molar to 7 molar, the concentration of dissolved plutonium increases by about 10 times, and at 11 molar it is about 100 times greater than at 4 molar. At the very high NaOH concentration of 11 molar, the plutonium concentration in solution should not exceed 0.05 g/L (0.19 g/gal).

Source Term Inventory Validation (WHC 1995) provides a compilation of NaOH concentration measurements taken from about 400 liquid and 150 solids samples from DSTs. The NaOH concentration does not exceed 230 g/L for any of these measurements. This corresponds to a 5.8 molar solution of NaOH. Based on this molarity, the plutonium concentration will not exceed 0.005 g/L in the supernatant liquid in any tank. The same conclusion also applies to an even larger number of measurements taken from SSTs. Although a few measurements were reported with a higher concentration of NaOH, those measurements were all found to be in error or to not apply to the present condition of the waste.

D3.3 EFFECT OF CARBONATES ON PLUTONIUM SOLUBILITY

Bratzel et al. (1996, Figure 3-4) provides a graph of the solubility of plutonium oxyhydride in supernate solutions as a function of carbonate concentrations. Bratzel et al. (1996) describes the impact of carbonate on plutonium solubility as follows:

Dissolution of plutonium oxide to yield solutions containing soluble plutonium concentrations above 0.2 g Pu/L, in carbonate solutions at a pH in the range of 9 to 10, would require oxidation of Pu(IV) to either Pu(V) or Pu(VI). A very strong oxidant such as $\text{Ag}(\text{II})$, boiling nitric acid, chlorine, permanganate ion, etc, is required to effect such oxidation. Even under carefully controlled laboratory conditions, such oxidation of

Pu(IV) is difficult to achieve. Thus, no dissolution mechanism is known to produce a sufficiently high plutonium concentration to be of criticality concern.

The Final Safety Analysis Report (FSAR) (LMHC 1999) provides the following information:

The liquids capable of a 0.2 g Pu/L concentration are those with a large (about 1 molar) carbonate concentration and low pH. Extrapolation of the data indicates that larger concentrations might be supportable if the carbonate concentration exceeds 1 molar. Because experiments show that aging reduces the plutonium concentration in tank liquids by an order of magnitude or more and tank solutions with greater than 1 molar carbonate do not exist, a criticality cannot occur in waste liquids.

In conclusion, although plutonium solubility increases with increasing carbonate concentration and with decreasing pH, the conditions do not exist in tank storage that permit a plutonium concentration in excess of 0.2 g/L in the supernatant liquid, and this concentration is too low to be of concern to criticality safety.

D3.4 UPPER LIMIT ON SOLUBILITY

From the information above, the concentration of dissolved plutonium in high-level waste should never be greater than 0.0017 g/L, even under the most idealized conditions for dissolution. However, the *Source Term Inventory Validation* (WHC 1995) reports a couple of samples with plutonium concentration a little higher than this value. The highest reported concentration is 0.0029 g/L. These values are the plutonium saturation values.

For uranium a higher concentration is possible, and the largest uranium concentration reported for DST liquids is 10.5 g/L. The ^{235}U concentrations would be about 100 times smaller than this value.

D4.0 CHEMICAL SEPARATION AND CONCENTRATION OF COMPONENTS

Bratzel et al. (1996) concludes that "no physical or chemical phenomena or mechanism has been identified that could concentrate fissile material at sufficient quantities or concentrations to result in an accidental nuclear criticality." This report identifies three potential chemical mechanisms that cause plutonium to enter the sludge:

- Sorption onto precipitated hydrated metal oxide sludge particles
- Precipitation as pure plutonium oxide crystals
- Formation of solid-solid solutions with non-neutron absorbers such as Zr(IV).

The importance of these mechanisms is described as follows (Bratzel et al. 1996):

If sorption were the dominant chemical mechanism, bonding of plutonium to strong neutron-absorbing metals such as iron and manganese ensures subcritical conditions. If pure hydrous plutonium oxide formed as a crystalline precipitate when acidic wastes were made alkaline, the plutonium oxide particles would be expected to agglomerate or mix with neutron absorbers. For nuclear criticality to occur for either of the latter cases, large (tens of micrometers in size) plutonium particles free of neutron absorbers must be created and then concentrated by some mechanism. All literature data, as well as calculations performed for modeling operational mechanisms such as saltwell pumping, air lift circulators, and transfer pumps, indicate that creation and separation of such plutonium-bearing particles are not plausible.

Bratzel et al. (1996) states that "because other insoluble oxyhydroxides precipitated in great excess with plutonium oxyhydroxide, co-precipitation of plutonium with the sludge-forming solids occurred." Of several likely co-precipitation mechanisms, the most important is "believed to be sorption of plutonium oxyhydroxide on the solid metal oxyhydroxides. In particular, the oxyhydroxides of iron and aluminum develop large, amorphous surfaces as they are formed that attract transition metal ions (Laitinen 1960)."

Bratzel et al. (1996) describes chemical mechanisms for concentrating the plutonium in a waste tank as follows:

Any chemical mechanism for concentrating the plutonium in a waste tank requires the transport of plutonium from various solid phases to an aqueous liquid phase and then to a compact geometry of a solid phase. Sorbed plutonium must be desorbed into the aqueous phase of the mixture of solid and liquid phases in the waste tanks. However, plutonium sorption on oxyhydroxides may be irreversible under certain chemical environments (DOE 1988; Alberts and Orlandini 1981) unless the solution composition in contact with the plutonium is dramatically altered. Such alteration requires the addition of organic or inorganic plutonium complexants, large changes in the REDOX potential of the solution by addition of oxidants or reductants, or by acidification of the solution.

Potential complexing agents for plutonium are present in all the tanks. Organic complexing agents such as EDTA, HEDTA, nitrilotriacetate, citrate, oxalate, and others are in the various waste mixtures. Even though these compounds are powerful complexants for plutonium in weakly acidic solutions, they are not very effective at the high hydroxide ion concentrations of actual waste supernatant liquids (Delegard et al. 1984). Under alkaline conditions, the organic reagents cannot compete strongly with either hydroxide or carbonate ion bonding to plutonium ions. Anionic complexes of Pu(IV) with hydroxide and carbonate ion have been shown to form under conditions expected in waste tank supernatant liquors. With excess nitrite ions the proposed aqueous species are $\text{Pu}(\text{OH})_2(\text{CO}_3)_2^{(2-)}$ and $\text{Pu}(\text{OH})_4(\text{CO}_3)_2^{(4-)}$. These species have only very low solubility in waste tank supernatant liquids so they are not likely to cause significant plutonium desorption.

The supernatant liquids in the underground tanks contain high concentrations of dissolved nitrite ions, relative to plutonium concentrations, that act as a mild reducing agent, maintaining soluble plutonium almost exclusively as Pu(IV). Because Pu(IV) is sorbed more strongly or is more insoluble than either Pu(V) or Pu(VI), the nitrite ions help drive the plutonium to the solid phase. To change the reduction-oxidation potential of the supernatant liquid requires the addition of huge quantities of oxidizing or reducing agents to the tanks. Such additions are not reasonable in view of the tight control maintained over additions of materials to the tanks.

Dissolution of plutonium oxide to yield solutions containing soluble plutonium concentrations above 0.2 g Pu/L, in carbonate solutions at a pH in the range of 9 to 10, would require oxidation of Pu(IV) to either Pu(V) or Pu(VI). A very strong oxidant such as Ag(II), boiling nitric acid, chlorine, permanganate ion, etc., is required to effect such oxidation. Even under carefully controlled laboratory conditions, such oxidation of Pu(IV) is difficult to achieve. Thus, no dissolution mechanism is known to produce a sufficiently high plutonium concentration to be of criticality concern.

D4.1 CONDITIONS THAT PREVENT PLUTONIUM ACCUMULATION

There are several conditions that tend to prevent the accumulation of a large mass of plutonium in a small, compact volume within a waste storage tank.

- First, alkaline waste permits dissolution of only a very low concentration of plutonium.
- Second, the plutonium would be mixed with a large quantity of solids. These solids would tend to shield the plutonium from the chemicals that would dissolve it.
- Third, because there is no mechanism to force the precipitate to a localized portion of the waste, precipitated plutonium would be expected to spread out over the entire cross section of the tank. This spreading out would ensure a geometry not conducive to criticality.

D4.2 REMOVAL OF NEUTRON ABSORBERS

If chemicals were to dissolve neutron-absorbing components, there are several natural controls preventing criticality.

- The low plutonium areal density would preclude criticality even if all absorbers were removed.

- The variety of chemicals that compose waste would mitigate the impact of removing a specific absorber. Even if one were removed, others should be unaffected. The large solids/plutonium mass ratio would continue to guarantee subcriticality even after some components have been removed.
- The natural tendency of any operation performed on waste is to cause mixing. Components removed chemically would tend to be remixed with the waste.

No chemical process has been identified that might credibly lead to criticality.

D5.0 CHEMICAL SEPARATION OF PLUTONIUM FROM URANIUM

Plutonium is obtained by chemical separation from uranium metal fuel elements that have been irradiated in a reactor. Because the purpose of the separation process is to recover the valuable plutonium, nearly all of it is removed. Consequently, the discarded uranium sent to waste tanks contains only a small fraction of plutonium and less than 70% of the original ²³⁵U. Most plutonium sent to waste disposal is intimately associated with uranium, primarily in the form of oxide.

The possibility of chemically separating plutonium from uranium is discussed in *Properties of Potential Uranium Precipitates as Related to Chemical Processing* (Colvin 1967). Concerning the behavior of plutonium when sodium polyuranate is precipitated, Colvin (1967) provides the following information:

The major constituent of dissolver solution is uranyl nitrate with plutonium and fission products being present at much lower concentrations. When sodium hydroxide is added to dissolver solution, the behavior of plutonium is dependent upon the plutonium valence state.

The plutonium-to-uranium ratio will be higher in the precipitate than in the original solution if plutonium is in the IV valence state. Laboratory experiments, where only a portion of the uranium is precipitated, indicated an immediate 80 percent plutonium enrichment in the precipitate. A prolonged digestion period considerably increases this figure. Plutonium polymer formation is most likely the principle contributor to the observed enrichment. The above experiments were performed with a beginning dissolver solution temperature of 450 °C. Variance in temperature could change the above figure considerably.

The opposite condition exists when plutonium is in the VI valence state. The Pu/U ratio in the precipitate is a factor of 2 less than in the original solution.

Analysis of dissolver solutions indicates that plutonium is normally present as 75-90% Pu(IV).

Colvin (1967) raises the possibility that at least part of the plutonium might separate from the uranium under certain conditions. For the separation process to work, the plutonium and uranium must first be dissolved. However, dissolution is prevented by maintaining a high pH (i.e., the waste is alkaline).

If chemical processes did somehow cause the proportion of plutonium to uranium to increase over a region within the waste, this would not by itself make criticality possible. Before criticality becomes possible, several independent conditions would have to be fulfilled as described below:

- The proportion of good neutron absorbers other than ^{238}U would have to be far less than usual. Even with a total removal of ^{238}U , other absorbers should be more than adequate to ensure subcriticality.
- The plutonium would have to be concentrated many-fold (usually by more than a factor of 100).
- The quantity of plutonium concentrated into a small volume would have to be large enough to go critical.

D6.0 EXCEEDING DISCHARGE LIMITS

A surveillance and monitoring program has been in place since the first waste discharge to detect a high plutonium concentration and to ensure compliance with process limits and controls. Although highly unlikely, it is theoretically possible that at some time in the past an abnormally large quantity of plutonium was discharged, leading to creation of a region of higher than average plutonium concentration.

Waste intended for tank farms is sent first to a holdup tank in the process facility. Holdup tanks do not have a critically safe design. Their diameters are large compared with the minimum size which can be made critical with a highly concentrated plutonium solution. They are recognized as locations with a potential for accidental nuclear criticality. For this reason, these tanks are closely monitored to ensure compliance to criticality safety limits and controls. Audits are made and samples analyzed on a regular basis to identify conditions that might lead to an abnormal accumulation of plutonium. If a violation were to be detected, the waste composition would be adjusted before being discharged to tank farms. After a predetermined volume of waste accumulates, the waste is discharged to the appropriate tank located in the RPP tank farms.

Under routine operations, the quantity of plutonium in a holdup tank is small and is mixed with a much larger quantity of solids. Accumulation of an abnormal quantity of plutonium, followed by its discharge to a storage tank, would require multiple failures of the auditing and sampling program and of nuclear material management procedures.

Because the contents of these tanks are stirred, it is unlikely that plutonium would be

concentrated in a small volume within the larger volume of waste. If highly concentrated plutonium solution were erroneously sent to a holdup tank, it would not likely remain highly concentrated. After leaving the holdup tank, a region of high concentration would be spread over a long length of small diameter transfer piping and the probability of reassembling the plutonium into a compact volume inside of the large storage tank is very small. The most likely final geometry in the large storage tank would be a thin slab of material with an areal density well below the subcritical limit.

Safety factors have been applied to ensure that any credible violation of discharge limits would not lead to a nuclear criticality. In the past the average plutonium concentration in a batch of discharged waste has been maintained at less than 0.013 g/L (0.05 g/gal). In a routine discharge, the plutonium would have to be concentrated more than 200 times above the limit value to reach the minimum critical concentration.

D7.0 SETTLING OF SUSPENDED SOLIDS

Traditionally, the average plutonium concentration for incoming waste was maintained less than 0.013 g/L (0.05 g/gal) (WHC 1988), but the Technical Safety Requirements (TSR) limit is 0.04 g/L (0.15 g/gal) (FDH 1997). For criticality to occur, the plutonium concentration would have to be much greater than this value. Conversely, if a much higher plutonium concentration cannot be achieved, criticality is not possible.

The simplest and perhaps most effective mechanism for concentrating plutonium is settling. Over a period of time settling of particulate and removal of liquid (i.e., by evaporation or pumping) can result in a layer of waste in which the plutonium concentration has increased. The degree by which concentration can increase is determined largely by the volume fraction of solids. After all liquid has been removed, no further concentration increase is possible. To ensure subcriticality, the proportion of solids was required to be large enough that the plutonium concentration would not exceed 1g/L in settled solids.

Different rates of settling for particles of different sizes and compositions may result in a vertical profile in which the plutonium bearing particles are nonuniformly distributed. This process, known as gravity segregation, creates the possibility that the plutonium concentration in a layer of waste might be greater than expected, if conditions are just right. Gravity segregation is, therefore, an important concern for criticality safety.

D7.1 GRAVITY SEGREGATION

If the plutonium does not separate from other solids during settling, the highest possible plutonium concentration would be achieved when all suspendable matter lies at the bottom combined only with interstitial liquid. The remainder of the liquid forms a supernatant layer above the solids. If the settled waste solids are homogenized, subcriticality is assured whenever the solids/plutonium mass ratio exceeds the subcritical limit.

When particles settle through a liquid, the settling velocity varies according to variations in their sizes and densities. Dense particles settle faster than light particles. If the plutonium-bearing particles exhibit a characteristic size and density, then these particles will tend to settle at a characteristic velocity, while other particles will settle at different velocities. This might result in the plutonium-bearing particles being preferentially segregated in layers of higher than average concentration.

Whyatt et al. (1996) reviewed mining literature on gravity separation to see how well techniques of mixing and stirring can be utilized to separate different types of ore. In addition, they used the Transient Energy Momentum and Pressure Equations in Three Dimensions (TEMPEST) code to model fluid dynamics and to estimate the degree by which the plutonium concentration might increase upon settling. They used the TEMPEST computer code to calculate settling velocities for waste mixtures composed of various size particles. For their calculations, they assumed a maximum particle size and a distribution of sizes up to this maximum.

For the calculations, a theoretical mixture of plutonium and iron was constructed. The Fe/Pu mass ratio was set at 580, a value 3 times the subcritical limit of 160. The plutonium and iron were each given the assumed particles size distribution and assumed to be mixed uniformly together. A simulation was then calculated of this mixture being dropped into a water-filled storage tank and being allowed to settle to the bottom. Based upon this simulation, Whyatt et al. (1966) concluded that there are three important ranges for particle size defined in terms of the ability of the plutonium and iron to separate from each other. These ranges are as follows:

1. **Maximum size greater than 100 μm** -- Gravity segregation will occur. Even simple operations, such as stirring, can cause significant segregation of plutonium from iron.
2. **Maximum size between 100 and 10 μm** -- The range of particle size between 100 and 10 μm forms a transition from a size where segregation is likely to a size where it is unlikely. As the maximum particle size is reduced below 100 μm , the ability to segregate according to particle size and density decreases rapidly.
3. **Maximum size less than 10 μm** -- For particle size smaller than 10 μm , significant segregation is very unlikely. Even with specialized equipment, it is difficult to separate particles at this size. Unintentional segregation is not credible.

Using the TEMPEST code to model the behavior of particles following mixing and settling, Whyatt et al. (1966) reached the following conclusions:

- Full 3-dimensional models of solids being pumped out of SST C-106 and into DST AY-102, where they were allowed to settle, and of mixer pump operation in DST SY-102 show no indication of unusual segregation in any area of the tank. The greatest degree of solids enrichment in all simulations was about a factor of 2.5 for the largest solids in the distribution.
- The upper bound on plutonium particle size is below the normal particle size for mineral processing separations by gravity separation. Gravity separation would be difficult even in equipment designed for the purpose and accidental segregation sufficient to create large segregated masses needed to cause criticality does not appear credible.
- Segregation of plutonium by selective flotation of the plutonium appears highly unlikely. If the proper organic molecules were present to allow flotation, they would be expected to float other metal hydroxides along with the plutonium at the pH conditions typical of waste storage tanks. However, even if selective flotation were assumed to occur, the material would spread out in a foam layer across the tank. The 2.6 kg/m² (240 g/ft²) areal density limit for criticality would apply, so that the entire plutonium contents of a waste tank could be concentrated into a foam layer without resulting in a criticality.

D7.2 FORMATION OF LAYERS AS A DISPERSAL MECHANISM

The tendency to form layers is an important waste dispersal mechanism. As waste enters a storage tank, it spreads over a large area, thereby preventing a buildup of waste in a confined volume. An investigation of the waste's tendency to form layers is reported in *Ferrocyanide Waste Tank Hazard Assessment - Interim Report* (Grigsby et al. 1992), as follows:

Evidence for the fluidity of the settled sludge can be seen from the observation of the behavior in the field and in the laboratory. The accumulated sludges in the tanks were pumped between tanks, sometimes for several miles. Settled synthetic In-Plant sludges made in the laboratory were observed to be readily pourable.

The handling of freshly prepared synthetic In-Plant ferrocyanide sludges provides an independent way to predict conical pileup in tanks. Although a carefully controlled and scaled test was not done, incidental observations indicated that sludge poured into transparent plastic containers formed a level surface in a matter of seconds after pouring was terminated. Based on the observation that a level surface was achieved within seconds in a 0.3-m (1-ft)-diameter container, a level surface would be expected within minutes in a 22.9-m (75-ft) diameter tank.

Based on the preliminary calculations presented above, it is concluded that ferrocyanide pumped into storage tanks, for example, would form a horizontal surface within minutes. Fresh sludge would be expected to mix with sludge already in the tank in the immediate vicinity of the outfall. This mixed sludge would spread on top of the preexisting sludge surface. Therefore, while horizontal layers having compositions corresponding to specific batches would be possible, it appears that localized vertical piles of sludge are highly improbable.

An incoming waste stream would spread into a thin layer. Any localized volume of higher plutonium concentration would be dispersed. The spreading process would ensure a relatively uniform areal distribution within the storage tank.

D7.3 CASE STUDY OF STRATIFICATION

After leaving the generator (processing plant), waste is pumped over a considerable distance within a 3-in. diameter pipe before reaching the storage tank. Upon discharge into the large tank this highly fluid liquid-sludge mixture would be dispersed into a layer on top of the existing tank contents. The spreading process would ensure a relatively uniform distribution over a sizeable area within the storage tank. An initial volume of unexpectedly high plutonium concentration would be transformed into a thin layer of higher concentration.

Vail (1997a) examines the possible range of variation in plutonium concentration in sludge in DST AZ-101 formed from discrete discharges of waste from the PUREX Plant. Vail modeled each discharge as forming a thin layer of uniform thickness on top of the existing waste surface. The final waste configuration, based on this model, is one of stratification, and this layering within DST AZ-101 appears to be a reasonable illustration of the nature of layering in all tanks. No attempt has been made to experimentally confirm the detailed structure of thin layers obtained in this model. On a broader scale, however, the existence of layers is well documented.

Since 1981 more than 235 transfers of plutonium-bearing waste have been received into DST AZ-101. The volume of batches ranged from 2,000 L (528 gal) to over 15,000 L (3,963 gal), and the average plutonium concentration ranged from 0 to 0.0242 g Pu/L. The most plutonium reported in a single batch was 242 g.

If gravity segregation is also assumed to occur, the plutonium might have segregated into a layer of higher concentration than would occur by layering caused by lateral dispersal alone. If this happened, however, the remaining solids in a batch would have settled either on top of the plutonium rich layer or below it. Wyatt et al. (1996) concluded that a concentration increase due to gravity segregation of up to a factor of 2.5 would be possible for tank waste. Each plutonium-rich layer would be sandwiched between layers of neutron absorbing solids. The plutonium concentration averaged over the entire layer would be the same as if formed by lateral dispersal alone.

The total plutonium in a layer is the same as in the initial discharge from which the layer was formed. If the plutonium concentration is high, then the thickness of that layer is small. Assuming no increase in plutonium concentration upon settling, Vail (1997a) determined the highest plutonium concentration to be 0.63 g/L in a layer 0.09 cm (0.04 in.) thick. Upon assuming a 3-fold increase in concentration upon settling, Vail (1997a) reaches the conclusion for DST AZ-101 that elevated plutonium concentrations as high as 2 g/L might be possible, but only in layers a fraction of a centimeter thick. When averaged over a thicker slice of waste, the plutonium concentration will be considerably less. The average in a one-inch thick layer would be expected to be less than 0.35 g/L.

D7.4 FLOCCULATION

Flocculation occurs when the attractive forces between particles overcome the electrostatic repulsive forces. Flocculation is a form of agglomeration. When particles flocculate, the unique density and settling velocities of individual particle types are masked by the properties of the flocculate. When rapid flocculation occurs, the plutonium-bearing particles will be effectively bonded to other waste solids and gravity segregation will not occur. A discussion of flocculation of plutonium particles is provided in the tank farms FSAR (LMHC 1999).

The segregation factor of 2.5 is based upon an assumption that there is no cohesion between particles. However, Whyatt et al. (1996) concluded that particles are flocculated under normal tank conditions. This flocculent waste contains agglomerates in which the plutonium is tightly bound with other solids. These agglomerates would counteract any tendency of the plutonium to separate from other solids. Transmission Electron Microscopy (TEM) measurements indicate the most probable primary sludge particle dimension is between 0.010 and 0.050 μm . About 90% of the particles were found to be less than 1 micron. However, these primary particles agglomerate with neutron absorbers, such as iron, into larger particles with dimensions between 10 and several hundred microns. This greatly inhibits the ability of the plutonium to segregate into plutonium-rich layers. For the evaluation of tank waste, it is conservative to ignore the ability of agglomeration to inhibit gravity segregation.

Rapid formation of flocculates will prevent segregation of waste solids provided that the salt concentrations is at least 0.1 molar in the supernatant, the pH exceeds 10, and the mixing intensity is low enough to permit flocculation. These conditions are present in almost all of the tank supernates.

Although, in theory, selective agglomeration of the plutonium-bearing particles is possible, such that these particles bond to each other to form clumps; available evidence supports the conclusion that this does not happen.

D8.0 EVAPORATION

An evaporator is used to remove liquid from waste to create new storage space. From a criticality safety point-of-view, evaporation is similar to settling of solids. After settling, a liquid layer remains above the solids. After evaporation, the liquid has been removed. After either process, the worst-case composition of the solids layer should remain about the same.

Solids deposited after evaporation might achieve a lower water fraction than solids settled below a liquid layer. This evaluation assumes that optimal moderation can be achieved, and this covers the possibility of complete removal of liquids.

D9.0 MIXING

For many waste storage tanks, the only operation performed is the pumping of new waste into the tank. The incoming solids from each batch of waste settle into a layer and the liquid portion mixes with the supernatant liquid.

There are five tanks that have air-lift circulators and two tanks with mixing pumps. The purpose of the air-lift circulators and mixing pumps is to mix the waste. In addition, when waste is transferred from one tank to another, the transfer process will mix the waste. Different waste types will be mixed together after these operations.

When particle size is small, mixing tends to disperse the plutonium. Dispersal tends to create a more homogeneous mixture and to reduce the regions of higher plutonium concentration. When waste types are mixed together, the concentration of each component tends to become closer to an average value. Regions containing high concentrations of plutonium would tend to become smaller. As the mixing continues, the overall mixture becomes increasingly uniform. Mixing is expected to increase the margin of safety by dispersing the plutonium.

When mixer pumps are shut off, the suspended solids will settle and gravity segregation is possible. Gravity segregation is discussed in Appendix D7.1.

D10.0 CONCLUSIONS

A multidisciplinary team of senior technical personnel concluded that the detailed nucleonic-related studies underlying the criticality safety basis of the waste tanks were technically sound and conservative. Their report *Tank Farm Criticality Review* (Bratzel et al. 1996) provides the primary study of chemistry for tank waste. They concluded:

In summary, the phenomena and mechanistic studies performed to evaluate the criticality safety of the waste tanks show that under the existing waste tank conditions, the fissile material cannot become separated and/or concentrated from the neutron absorber oxyhydroxide solids to the extent that it results in a significant reduction of the existing margin of subcriticality (Serne et al. 1996). In addition, the favorable neutronics of the waste tanks support the finding that an accidental criticality is incredible under the existing waste tank conditions.

Note, however, that this finding applies only to existing waste tank conditions. Although the phenomena and mechanistic studies also support forthcoming retrieval operations (i.e., retrieval of waste from tank 241-C-106), future operational activities must be evaluated against the technical-based limits and controls to ensure future subcritical conditions.

A guarantee of subcriticality is provided only when “existing waste tank conditions” are maintained. In the undisturbed waste storage environment these conditions will be maintained indefinitely. However, when operations are performed that transfer waste between tanks or add chemicals, assurance is required that waste conditions remain within “existing tank conditions.” An important requirement for maintaining existing tank conditions is that the pH remain greater than 8.0.

D11.0 REFERENCES

- Alberts, J. J., and K. A. Orlandini, 1981, *Laboratory and Field Studies of the Relative Mobility of ^{239, 240}Pu and ²⁴¹Am From Lake Sediments Under Oxidic and Anoxic Conditions*, *Geochemica et Cosmochimica Acta*, 45, pp. 1933-1938.
- Bratzel, D. R., W. W. Schulz, R. Vornehm, and A. E. Waltar, 1996, *Tank Farm Nuclear Criticality Review*, WHC-SD-WM-TI-725, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- Braun, D. J., L. D. Muhlestein, T. B. Powers, and M. D. Zentner, 1994, *High-Level Waste Tank Subcriticality Safety Assessment*, WHC-SD-WM-SARR-003, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- Colvin, C. A., 1967, *Properties of Potential Uranium Precipitates as Related to Chemical Processing*, ARH-147, Atlantic Richfield Hanford Company, Richland, Washington.
- Daling, P. M., T. S. Vail, G. A. Whyatt, D. I. Kaplan, K. M. Krupka, S. V. Mattigod, J. R. Serne, C. A. Rogers, D. G. Erickson, K. N. Schwinkendorf, and E. J. Lipke, 1997, *Feasibility Report on Criticality Issues Associated With Storage of K Basin Sludge in Tank Farms*, HNF-SD-WM-ES-409, Rev. 0, U.S. Department of Energy, Project Hanford Management Contractor, Richland, Washington.
- Delegard, C. H., G. S. Barney, and S. A. Gallagher, 1984, *Effects of Hanford High-Level Waste Components on the Solubility of Cobalt, Strontium, Neptunium, Plutonium, and Americium*, *Geochemical Behavior of Disposal Radioactive Waste*, G. S. Barney, J. D. Navratil, and W. W. Schultz, Eds., ACS Symposium Series 246, American Chemical Society, p. 95.
- Delegard, C. H., 1985, *Solubility of PuO₂•xH₂O in Alkaline Hanford High-Level Waste Solution*, RHO-RE-SA-75 P, Rockwell International, Richland, Washington.
- DOE, 1988, *Site Characterization Plan, Reference Repository Location, Hanford Site, Washington*, DOE/RW-0164, Vol. 3, U.S. Department of Energy, Washington, DC, pp. 4.1-106.
- FDH, 1997, *Tank Waste Remediation System Technical Requirements*, HNF-SD-WM-TSR-006, Rev. 0, Section 5.7, "Nuclear Criticality Safety," Fluor Daniel Hanford, Inc., Richland, Washington.

- Fedoseev, A. M., N. N. Krot, N. A. Budantseva, A. A. Bessonov, M. V. Nikonov, M. S. Grigoriev, A. Yu. Garnov, V. P. Perminov, and L. N. Astafurova, 1998, *Interaction of Pu (IV,VI) Hydroxides/Oxides with Metal Hydroxides/Oxides in Alkaline Media*, PNNL-11900 (UC-2030), Pacific Northwest National Laboratory, Richland, Washington.
- Grigsby, J. M., D. B. Bechtold, G. L. Borsheim, M. D. Crippen, D. R. Dickinson, G. L. Fox, D. W. Jeppson, M. Kummerer, J. M. McLaren, J. O. McCormack, A. Padilla, B. C. Simpson, and D. D. Stepnewski, 1992, *Ferrocyanide Waste Tank Hazard Assessment - Interim Report*, WHC-SD-WM-RPT-O32, Rev. 1, Westinghouse Hanford Company, Richland, Washington.
- Hobbs, D. T., T. B. Edwards, and S. D. Fleischman, 1993, *Solubility of Plutonium and Uranium in Alkaline Salt Solutions (U)*, WSRC-TR-93-056, Savannah River Technology Center, Westinghouse Savannah River Company, Aiken, South Carolina.
- Krot, N. N., V. P. Shilov, A. B. Yusov, I. G. Tananaev, M. S. Grigoriev, A. Yu. Garnov, V. P. Perminov, and L. N. Astafurova, 1998, *Plutonium(IV) Precipitates Formed in Alkaline Media in the Presence of Various Anions*, PNNL-11901 (UC-2030), Pacific Northwest National Laboratory, Richland, Washington.
- Laitinen, H. A., 1960, *Chemical Analysis*, McGraw-Hill Book Company, Inc., New York, New York, p. 169.
- LMHC, 1998, *Operating Specifications For the 241-AN, AP, AW, AY, AZ & SY Tank Farms*, OSD-T-151-00007, Rev. H-21, Lockheed Martin Hanford Corporation, Richland, Washington.
- LMHC, 1999, *Tank Waste Remediation System Final Safety Analysis Report*, WHC-SD-WM-SAR-067, Rev. 0, Lockheed Martin Hanford Corporation, Richland, Washington.
- Peretrukin, V. F., V. I. Silin, A. V. Kareta, A. V. Gelis, V. P. Shilov, K. E. German, E. V. Firsova, A. G. Maslennikov, and V. E. Trushina, 1998, *Purification of Alkaline Solutions and Wastes from Actinides and Technetium by Coprecipitation with Some Carriers Using the Method of Appearing Reagents*, PNNL-11988, Pacific Northwest National Laboratory, Richland, Washington.
- Serne, R. J., G. A. Whyatt, S. V. Mattigod, Y. Onishi, P. M. Doctor, B. N. Bjornstad, M. R. Powell, L. M. Liljegren, J. H. Westsik, Jr., N. J. Aimo, K. P. Recknagle, G. R. Golcar, T. B. Miley, G. R. Holdren, D. W. Jeppson, R. K. Biyani, and G. S. Barney, 1996, *Fluid Dynamics, Particulate Segregation, Chemical Processes, Natural Ore Analog and Tank Inventory Discussions that Relate to the Potential for Criticality in Hanford Tanks*, WHC-SD-WM-TI-757, Rev. 0, Westinghouse Hanford Company, Richland, Washington.

- Vail, T. S., 1997a, *CSER 96-014: Criticality Safety of Project W-151, 241-AZ-101 Retrieval System Process Test*, WHC-SD-W151-CSA-001, Rev. 1, Duke Engineering & Services, Richland, Washington.
- Vail, T. S., 1997b, *Criticality Safety Assessment of the Tank 241-C-106 Remediation*, WHC-SD-W320-CSA-001, Rev. 1, Duke Engineering & Services, Richland, Washington.
- WHC, 1988, *Waste Storage Tanks and Associated Equipment*, CPS-T-149-00010, Rev. D-0, Westinghouse Hanford Company, Richland, Washington.
- WHC, 1995, *Tank Waste Source Term Inventory Validation*, WHC-SD-WM-ER-400, Westinghouse Hanford Company, Richland, Washington.
- Whyatt, G. A., R. J. Serne, S. V. Mattigod, Y. Onishi, M. R. Powell, J. H. Westsik, Jr., L. M. Liljegren, G. R. Golcar, K. P. Recknagle, P. M. Doctor, V. G. Zhirmov, J. Dixon, D. W. Jeppson, and G. S. Barney, 1996, *The Potential for Criticality in Hanford Tanks Resulting from Retrieval of Tank Waste*, PNNL-11304, Pacific Northwest National Laboratory, Richland, Washington.
- Wodrich, D. D., G. S. Barney, D. L. Becker, G. L. Borsheim, W. C. Carlos, M. J. Klem, R. E. Van der Cook, and J. L. Ryan, 1992, *Summary of Single-Shell Tank Waste Stability*, WHC-EP-0347, Supplement, Westinghouse Hanford Company, Richland, Washington.

This page intentionally left blank.

APPENDIX E
GENERATION OF FUTURE WASTE

This page intentionally left blank.

E.0 GENERATION OF FUTURE WASTE

A survey of anticipated waste sources is provided, based upon estimates from the 1998 revision of *Operation Waste Volume Projection (OWVP)* by Strode and Boyles (1998). Projections of routine waste volumes generated by the various facilities are summarized in Table E-1. In 1998, B-Plant and the PUREX Plant were in terminal clean out (TCO) mode.

Table E-1. Average Monthly Waste Generation Rates¹

Facility	Facility Target Quantity (kgal/month)	Average Quantity 10/96 - 6/97 (kgal/month)
Tank Farms	10.0	2.7
PUREX Plant	0.4	N/A-TCO Mode
UO₃ Plant	Deactivated	0.0
B-Plant	N/A-TCO Mode	N/A-TCO Mode
WESF²	1.7	With B-Plant
PFP	0.4	N/A
S-Plant (222-S Laboratory)	2.1	0.6
T-Plant	1.4	1.4
100 Areas	0.0	0.0
300 Area	4.2	2.2
400 Area	0.2	0.0
WSCF³	0.0	0.0
TOTALS	20.4	6.9

Notes:

¹Based on Strode and Boyles (1998).

²Waste Encapsulation and Storage Facility (WESF)

³Waste Sampling and Characterization Facility (WSCF)

N/A = not applicable)

Although an estimate is made of the future volumes of waste, it is recognized that projected values may change, and unforeseen sources of waste may develop. The information provided in this appendix is descriptive and intended to help understand the needs of the tank farms criticality safety program. Criticality safety limits and controls are not dependent upon this information, and this appendix need not be revised to reflect changes.

In the past, most tank waste was generated from reactor fuel element processing at a rate regulated by plutonium production. However, the mission of the Hanford Site has changed from production to environmental cleanup. In the future, most waste will result from cleanup of older facilities.

E1.0 PUREX PLANT

Deactivation of the PUREX Plant was completed in Fiscal Year (FY) 1997. All fissionable material has been either removed from the facility or stabilized. Stabilized fissionable material is securely fixed in place and does not present any criticality safety hazard. An example of stabilized fissionable material is surface contamination within ducts. Although shut down, PUREX may send approximately 5 kgal of dilute waste once per year to tank farms. The fissionable content of this waste is expected to be extremely low.

E2.0 PLUTONIUM FINISHING PLANT

Operations at the Plutonium Finishing Plant (PFP) include: (1) stabilization of reactive solid residues by muffle furnace calcination; (2) shipping, receiving and storage of special nuclear materials; (3) analytical and development laboratories; and (4) treatment and handling of PFP liquid wastes destined for tank farms and the Effluent Treatment Facility (ETF). According to the *Operation Waste Volume Projection (OWVP)* (Strode and Boyles 1998):

The volume of waste anticipated to be produced for the TPA Compliant Case is developed from the existing waste generation rate at PFP (100 untreated gallons/month), and the anticipated use of a direct denitration vertical calciner coupled with an ion exchange processing system currently being developed and tested by the development laboratories. The vertical calciner is the most promising technology for plutonium residue stabilization and facility clean out. All projection cases projected that PFP stabilization and clean out would generate 27 kgal of additional waste from 1998 through 2006 (Strode and Boyles 1998).

Table E-2 shows the *Operation Waste Volume Projection (OWVP)* (Strobe and Boyles 1998) projections of solids content in PFP waste.

Table E-2. Plutonium Finishing Plant Waste Solids Content¹

Solids in waste (vol%)	
Plutonium Reclamation Facility	3.5
Remote Mechanical C Line	4.4
Laboratory	4.5

Note:

¹Strobe and Boyles (1998)

Plutonium Finishing Plant waste discharged to DCRT 244-TX passes through the storage and treatment holdup tank D5 located in the 241-Z Building. Waste in holdup tank D5 is received from other tanks located in the 241-Z Building: holdup tanks D4, D7, and D8 and process tanks 19, 39, WM-1, and 40. This describes the present discharge pathway, but there is no guarantee it will remain unchanged in the future.

The fissile inventory in holdup tank D5 is determined from analysis of samples, and an independent verification is made of the calculations, before transfer to tank farms. The method used in this determination is described in operating procedures. Adjustments are made to waste in holdup tank D5 before discharge to meet tank farms acceptance criteria. Iron hydroxide is added to ensure compliance with a minimum limit of at least 0.7 vol%.

The transfer data sheet (TDS) must be reviewed and initialed by PFP supervision. It is then transmitted to tank farms for approval before the actual transfer.

E3.0 B-PLANT/WASTE ENCAPSULATION AND STORAGE FACILITY

B-Plant stabilization and shut down was completed in 1998. Just west of B-Plant is the Waste Encapsulation and Storage Facility (WESF) built to support encapsulation and storage of ¹³⁷Cs. WESF is projected to continue to generate 5 kgal/year until 2028. This waste should contain only trace quantities of fissionable material. Nevertheless, verification of compliance to the tank farm Criticality Prevention Specifications (CPSs) is required before transfer.

E4.0 S-PLANT

S-Plant currently provides analytical chemistry services in support of Hanford processing plants and tank characterization. Waste is sent to tank farms from the 222-S Laboratory. Strode and Boyles (1998) describe this waste as follows:

Most of the radioactive liquid waste generated at the laboratory complex originates from analytical activities performed within the 222-S Laboratory in support of tank characterization. Radioactive and radioactive hazardous (mixed) wastes generated by the 222-S Laboratory are discharged to the 219-S Waste Handling Facility. Dilute, non-complexed wastes are currently being transferred via pipeline to Tank 102-SY. Projected S-Plant monthly generations rates were approximately 1.0 to 1.7 kgal/month for FY 1998 through 2028 for all projection cases.

E5.0 T-PLANT

T-Plant's primary mission includes decontamination of radiologically and chemically contaminated equipment. Currently, waste is being sent to tank farms through the 204-AR facility. Strode and Boyles (1998) provide the following information:

T-Plant is currently testing new decontamination techniques (ice blasting and CO₂ decontamination systems) which have reduced liquid waste generations from those reported previously. Dilute, non-complexed wastes collected at T-Plant during decontamination, repackaging, condensate collection, or railcar certification are currently being transported to 204-AR vault via railcar. These wastes contain approximately 5 volume percent solids. Projected T-Plant waste generations were based on a combination of anticipated work loads and actual observed generation rates. The projected volumes supplied by T-Plant engineers ranged from 1.4 kgal/month to 2.7 kgal/month.

E6.0 242-A EVAPORATOR AND LIQUID EFFLUENT RETENTION FACILITY

After a period of shutdown, the 242-A Evaporator was restarted on April 15, 1994. The evaporator is not run continuously, but rather part time in a series of campaigns. The most recent evaporator campaign started in March 1999 to evaporate DSTs AY-102, AP-106, AN-101, and AP-108 containing dilute waste. For the years 1999-2004, it was estimated that 1 to 2 campaigns would be required each year. The volume of waste reduction during a month of evaporator operation will not exceed 5,700 kL (1,500 kgal). During an average month of operation, the processing rate has been about 1,900 kL (500 kgal).

A minimum of two dedicated DSTs is required for operation of the evaporator. Prior to operation, wastes are collected in dilute receiver tanks. Any solids in the initial waste will settle out in the receiver tanks. When these tanks are full, the supernate is transferred to the evaporator feed tank (DST AW-102). Between 4 to 6 months are required for wastes to be sampled and analyzed according to Evaporator DQO requirements before evaporation can be started. After the DST AW-102 is full and DQO requirements are met, supernate is transferred to the 242-A Evaporator for boil-down.

After cycling through the evaporator several times, the concentrated waste is sent to the evaporator receiver tank (DST AW-106). During each pass through the evaporator, the waste concentration increases until it reaches a level consistent with precampaign Process Control Plans. Upon reaching the desired waste volume reduction factor (WVRF), the waste is transferred to DST AW-106. This concentrated waste is referred to as double-shell slurry feed (DSSF).

Evaporator condensate is stored at the Liquid Effluent Retention Facility (LERF), which has a 6.5 million gallon storage capacity.

E7.0 WASTE FROM SINGLE-SHELL TANKS

As part of the Waste Stabilization Program, liquid waste from SSTs is pumped to DCRTs and then to DSTs. The compatibility assessment includes a verification of compliance with the plutonium concentration limits specified in the CPS.

E7.1 SALTWELL LIQUID PUMPING

Liquid removed from a SST is drained through a salt-well screen and is relatively free of solids. Saltwell liquid (SWL) is characterized for chemical compatibility with the DCRTs and DSTs. Saltwell liquid pumping will occur for SSTs that have 50,000 gallons or more of drainable interstitial liquid. In the 200 East Area, the receiver tanks for saltwell liquid are DSTs AN-101, AP-106, and AP-108. In the 200 West Area, DST SY-102 is the saltwell liquid receiver tank.

E7.2 SINGLE-SHELL TANK SOLIDS RETRIEVAL

Strode and Boyles (1998) assumed that retrieval of SST C-106 solids would be completed in June 1999. These solids are stored in DST AY-102.

It is projected that all solids will be removed from all SSTs and that SST site closure will be completed by FY 2024.

E8.0 DOUBLE-SHELL TANKS

Four DSTs in the AY and AZ farms are designated as aging waste tanks to hold high-heat wastes, and the remaining 24 DSTs are used to store non-aging waste that are not high-heat.

E8.1 AGING WASTE DOUBLE-SHELL TANKS

No additional aging waste is expected to be generated by Hanford facilities. Solids retrieved from SST C-106 will be stored in DST AY-102 due to the high heat content. Double-shell tanks for aging waste are equipped with condensers and air-lift circulators.

E8.2 NON-AGING WASTE DOUBLE-SHELL TANKS

Projections for the usage of DSTs include the following (Strode and Boyles 1998):

- Approximately 66 kgal of caustic will be added to DST AN-107 in FY 2000.
- DST AP-106 is currently receiving direct transfers of wastes from B-Plant and rail and truck shipments via the 204-AR vault from S-Plant, T-Plant, 100 Area, 300 Area, and 400 Area.
- In the 200 East Area, DST AP-108 will be the receiver tank for complexed saltwell liquid, and DSTs AN-101 and AP-106 will be the receiver tanks for non-complexed saltwell liquid.

E9.0 WASTE SAMPLING AND CHARACTERIZATION FACILITY

The Waste Sampling and Characterization Facility (WSCF) receives waste from S-Plant. This waste is sent to the Effluent Treatment Facility (ETF) and not to tank farms.

E10.0 EFFLUENT TREATMENT FACILITY

The Effluent Treatment Facility (ETF) started operation in November 1995 to process stored evaporator condensate from the Liquid Effluent Retention Facility (LERF), newly generated evaporator condensate, and aqueous wastewater containing low specific radioactivity. The ETF should not send any streams to DSTs.

E11.0 100 AREAS

Waste from deactivation of the N Basin will not be transferred to DSTs, but instead are to be transferred to the Environmental Restoration Disposal Facility (ERDF).

In 1999 plans to send K Basin sludge to tank farms was discontinued, after the decision was made to store it in a basin at T-Plant in the 200 West area. K Basin sludge contains uranium that has not been processed to remove plutonium. K Basin sludge is an example of a potential waste stream with characteristics different from typical tank waste.

Plans to clean out the 105-F and 105-H Basins are being reviewed.

E12.0 300 AREA

Strode and Boyles (1998) report the following status of waste shipments from the 300 Area in 1998 and the projected future shipments:

Liquid wastes from the various 300 Area Facilities are transferred to the 340 Facility. Liquid wastes collected at the 340 Facility are transferred to the 204-AR vault in 20,000 gallon railroad tank cars (after September 1998, shipments will likely be via a truck tanker due to the pending cessation of rail service). In the future the 340 Facility will be closed and a new facility will be installed for the Pacific Northwest Laboratory to transfer wastes from its 300 Area facilities to the DSTs. Facilities in the 300 Area sent 26 kgal of waste (includes flush) to DSTs (2.2 kgal/month) in FY 1997. All three projections predicted that 2.3 kgal/month of miscellaneous waste would be generated from 300 Area facilities during FY 1998. Projected waste generations for FY 1999 and beyond varied from 0.33 to 1.4 kgal/month.

Rail service has since been discontinued, and shipments are made via tanker truck.

E13.0 400 AREA

The 400 Area contains 3 major facilities: the Fast Flux Test Facility (FFTF), the Maintenance and Storage Facility (MASF), and the Fuel and Material Examination Facility (FMEF). Strode and Boyles (1998) report the following status of waste shipments from the 400 Area prior to 1998 and the projected shipments after 1999:

Shutdown of the FFTF has increased the amount of liquid waste generated by the plant's Sodium Removal System. Approximately 11 kgal of wastes were received from 400 Area in FY 1994-1995 (~0.5 kgal/month). All three projection cases projected a 7 kgal shipment of miscellaneous waste would be generated from the 400 Area facilities every third year starting in FY 1999.

E14.0 REFERENCES

Strode, J. N. and V. C. Boyles, 1998, *Operation Waste Volume Projection*, HNF-SD-WM-ER-029, Rev. 24, Lockheed Martin Hanford Corp., Richland, Washington.

APPENDIX F

“MOST REASONABLE” HYPOTHETICAL ACCIDENT

This page intentionally left blank.

F.0 "MOST REASONABLE" HYPOTHETICAL ACCIDENT

The FSAR (LMHC 1999) addresses the consequences of a postulated criticality. Although no credible sequence of events culminating in a criticality was identified, a scenario was nevertheless constructed to define the "most reasonable" configuration for this hypothetical accident. This scenario illustrates the low probability of a criticality accident.

The initiating waste transfer was chosen to be 500 L (132 gal) of a 10 g Pu/L solution pumped at a rate of 4.7 L/s. This transfer was selected to represent the worst conceivable transfer of waste from PFP following multiple failures of procedures. A higher plutonium concentration would be overly conservative, while a lower concentration would require too large a volume to be credible. The selection of 500 L of 10-g Pu/L solution is based on the following considerations:

- The plutonium concentration must exceed the minimum critical plutonium concentration in water of 7 g/L.
- PFP has a strict control over plutonium inventory, and a discrepancy of hundreds of grams, or more, would be noticed. The quantity of plutonium assumed in this scenario is kept as low as possible to increase the plausibility that the error would not be detected at PFP and the transfer stopped.
- 500 L of a 10 g Pu/L solution is near the point at which criticality would occur at PFP. If a larger volume or a higher concentration of plutonium is assumed, criticality would likely occur at PFP, and a transfer to tank farms would not be made.

In reality, this accident scenario contains enough failures to be considered incredible. A batch containing 5 kg of plutonium would certainly be noticed before transfer. It ignores the many reasons why this solution could not have been accumulated into a single batch without going critical at PFP and then to remain undiluted until reaching tank farms.

The hypothetical accident scenario assumes that a 500-L batch of a 10 g Pu/L solution is transferred intact. Upon entering the DST, the plutonium solution is assumed to form into a compact spherical shape within a deep layer of existing supernatant liquid. The radius of this sphere is assumed to increase until criticality is reached.

A criticality would have an initial burst followed by a rapid shutdown. The release of fission energy would cause thermal expansion and the formation of small bubbles within the liquid. Expansion would reduce the plutonium concentration, and convective currents generated by heat production would expel plutonium from the region of high concentration. The small bubbles would form into larger bubbles leading to the release of steam. Since the tank is far larger than the critical volume, expansion and convection would cause dilution of the plutonium, and there is no mechanism to bring it back together. Even if additional 10 g Pu/L solution enters the tank, the turbulent condition in the waste liquid would be expected to prevent the reassembly of a

critical configuration before the entire 500-L volume is discharged. The postulated criticality was, therefore, modeled as a single burst.

An estimated 89 MJ of energy (equivalent to 2.8×10^{18} fissions) would be released from the postulated DST criticality. Most of this energy would go into heating the liquid within the region of criticality. About 25% of the energy would be released into the headspace in steam.

A criticality accident is considered "beyond extremely unlikely" for several reasons. First, multiple running failures would have to occur without being identified and without interruption by other operations. Second, the operations to which the waste is subjected would naturally result in dilution, and the number of failures required to achieve a critical configuration would be increased. Third, the volume of waste sent to tank farms from PFP has been considerably reduced, and transfers in the future will be infrequent.

F1.0 REFERENCE

LMHC, 1999, *Tank Waste Remediation System Final Safety Analysis Report*, HNF-SD-WM-SAR-067, Rev. 0, Lockheed Martin Hanford Corporation, Richland, Washington.

APPENDIX G

EVALUATION OF PLANNED OPERATIONS

This page intentionally left blank.

G.0 EVALUATION OF PLANNED OPERATIONS

When a new operation is planned, a review will be made to determine if a criticality safety evaluation is needed. If the proposed operations fall within the envelope of existing evaluations, then no additional evaluation is required. For example, operations that do not disturb the waste are already covered. Operations, such as use of airlift circulators and saltwell pumping, that cause movement of the supernatant liquid, but do not disturb settled solids, are also covered. Although some operations that disturb settled solids have been evaluated, this is not true for all such operations. Justification that the operation needs no further evaluation is to be documented in writing.

Activities considered “waste disturbing” include:

- (1) Moving, suspending, mixing, or otherwise disturbing settled solids,
- (2) Combining waste from different tanks,
- (3) Sluicing,
- (4) Operation of mixer or transfer pumps, and
- (5) Changing the basic chemistry.

If these, or similar operations, fall outside of the envelope of permitted operations, a criticality safety valuation must be performed and documented.

When a criticality safety evaluation is performed for an operation previously evaluated in a different tank, the existing CSER may serve as a model, and much of the description and analysis may be copied to the new CSER. To reduce duplication, a simplified approach to evaluating new “waste disturbing” activities is described.

G1.0 SIMPLIFIED APPROACH TO EVALUATIONS

A new evaluation may be written as addendum to this CSER. Information in this CSER need not be copied, but may be included by reference. For example, descriptions of tank farm facilities and of the physical and chemical characteristics of tank waste are provided in this CSER and need not be included in an addendum. Details of “waste disturbing” activities summarized in this CSER need not be included.

A future evaluation should include the following features:

- (1) A description of operations to be performed.

- (2) Reference to this evaluation for facility descriptions and phenomenological descriptions that apply. Significant differences, if there are any, should be described.
- (3) A summary of characterization data for the waste affected and a brief assessment of the margin of subcriticality.
- (4) A brief review of chemistry with a description of any differences from that which has been reviewed in past chemistry studies. If new chemistry concerns are raised, an evaluation of their impact on criticality safety shall be made.
- (5) A statement of conclusions related to criticality safety.

G2.0 BASIS DOCUMENTS ISSUED FOR SPECIFIC OPERATIONS

CSERs that have been issued for specific operations can be placed into three categories:

- (1) waste stabilization (removal of liquid);
- (2) transfer of solids from SST C-106; and,
- (3) evaluation of mixer pump operation.

Waste Stabilization Operations in SSTs

Four CSERs for waste stabilization projects are no longer needed (voided): Rogers (1993a), Rogers (1993b), Rogers (1993c), and Rogers (1994a)

Transfer of Waste Solids from SST C-106 to DST AY-102

Vail, T. S., 1997b, *Criticality Safety Assessment of the Tank 241-C-106 Remediation*, WHC-SD-W320-CSA-001, Rev. 1, Duke Engineering & Services, Richland, Washington. Transfer of SST C-106 waste is completed, and this CSERs is no longer needed (voided).

Evaluation of Mixer Pump Operation

Vail, T. S., 1997a, *CSER 96-014: Criticality Safety of Project W-151, 241-AZ-101 Retrieval System Process Test*, WHC-SD-W151-CSA-001, Rev. 1, Duke Engineering & Services, Richland, Washington.

Vail, T. S., 1996, *CSER 96-004: Criticality Safety of Operability Testing the Mixer Pump in Tank 241-AN-107*, WHC-SD-SQA-CSA-505, Rev. 0, Westinghouse Hanford Company, Richland, Washington.

The important information from these documents has been included in this report. Testing of the mixer pump for DST AN-107 has been put on indefinite hold.

G3.0 THERMAL EFFECTS

The FSAR (LMHC 1999) reports a study of heat convection in a generic tank. The sludge was modeled as wet clay with 50% porosity, a density of 1.4 g/cm^3 , a viscosity of 12.5 cp, and a heat production of 2.66 W/m^3 . Convection cells were found to form near tank walls where cooling occurs. The flow velocity of sludge was found to be $3 \times 10^{-12} \text{ m/s}$ at this rate of heat production. When the heat production was increased to 43.2 W/m^3 , the flow velocity increased to $3 \times 10^{-8} \text{ m/s}$. This flow velocity is very slow when compared to the much greater flow velocity of 0.2 to 2.5 m/s that is required to transport particulate matter. Thermal effects are, therefore, not strong enough to cause migration of plutonium as the result of surface erosion by moving liquid. Therefore, redistribution of plutonium will not occur as a result of thermal effects.

G4.0 SLUICING

The FSAR (LMHC 1999) presents a model of sluicing developed by Serne et al. (1996) as an example of the transfer of solids between storage tanks. Serne et al. (1996) used the TEMPEST code to simulate the sluicing of solids from SST C-106 and the transfer of these solids to DST AY-102. In the sluicing process for SST C-106, supernate was sent through a 4-in., Schedule 40, transfer line to the sluicer nozzles. Each 2.54-cm (1-in.) diameter sluice nozzle directed a stream of water at a pressure of 180 psig and a temperature of 180 °F against the sludge layer to convert it into slurry. This slurry was pumped to DST AY-102 through an encased transfer line. Supernate from DST AY-102 was recirculated back to SST C-106 to be used to sluice more waste.

Solids entering DST AY-102 are assumed to comprise 30 wt% of a slurry moving at a flow rate of 22 L/s (350 gpm) through 2.54-cm (1-in.) nozzles on each of the four legs of a horizontal, cruciform-shaped, distribution header. The particle size distribution measured for SST C-106 solids was applied to the model. Using a particle density of 2.4 g/cm^3 , nine size groups were modeled. The smallest particles ranging from 5 to 10 μm diameter were placed in Group 1, and the largest particles ranging from 50 to 55 μm diameter were placed in Group 9. Two cases were modeled to examine the behavior of plutonium particles. The first case assigned plutonium particles a diameter of 11.5 μm and a density of 11.5 g/cm^3 (i.e., to Group 5), and the second case assigned the plutonium particles a diameter of 2 μm and a density of 6 g/cm^3 (i.e., to Group 1).

The simulation showed the slurry to flow rapidly downward and spread out over the existing settled sludge surface. Group 5 particles after settling were found enriched by a factor of 1.5, while Group 1 particles were not enriched. Group 1 particles (i.e., those of the smallest size) were the last to settle and comprised the top layer. This modeling, specific to SST C-106, indicated that the degree of segregation would not be large.

Literature on separation operations used in mining show that it would be difficult to separate particles of the sizes in tank waste through gravity separation techniques. Small particles are

expected to flocculate, and the settling velocity of plutonium particles would not be different from other sludge. The expectation of flocculation is supported by transmission electron microscopy experiments. Light scattering measurements indicate that sludge particles are agglomerated. Even though segregation might occur between faster-settling and slower-settling agglomerates, the relative plutonium concentrations would not change. In addition, sorption and solid solution mechanisms that operate to bind plutonium to neutron absorbers provide additional barriers preventing segregation.

The source term validation (WHC 1995) provides upper limits on plutonium concentrations of 7.65×10^6 Bq/L (0.0029 g/L) in supernate and 1.57×10^9 Bq/L (~0.60 g/L) in solids. When distributed uniformly over a large volume exceeding several thousand liters, for criticality to occur the plutonium concentration would have to exceed 2.6 g/L in dry sludge or 7 g/L in sludge with a high water content. In a compact volume of several hundred liters, a plutonium concentration in excess of 7 g/L would be required, regardless of the water content. For criticality to actually occur, the segregation factor would have to exceed 10, assuming an initial concentration of 0.6 g/L, the highest concentration reported in the source term validation (WHC 1995). Even assuming a higher initial concentration of 0.89 g/L, as reported in 1999 for SST TX-118 (Lipke 1999), the segregation factor would have to exceed 7.5. In reality, the initial concentration should be far less, and the required segregation factor would be substantially larger. Several kilograms of plutonium would have to be deposited in a relatively small volume. Under credible conditions, the plutonium would deposit in a slab. At 10 g/L of plutonium, a slab must be at least 30-cm (12-in.) thick to go critical, and, if evenly distributed over the entire tank, this requires about 1,200 kg of plutonium.

A multi-disciplinary team of experts from a diverse range of backgrounds studied an exhaustive set of scenarios that might lead to criticality in SST C-106. The team considered whether separation of plutonium from the waste was possible. As part of this team, Vail (1997b) evaluated sluicing operations and provides a good description of the chemical and physical processes at work.

Evidence is provided that the plutonium is chemically bonded (sorbed) onto oxyhydroxides of iron, aluminum, and manganese, and this bonding will remain during the waste aging process. If sorption is the dominant process, then the plutonium and the neutron absorbers to which it is bound will remain in the same ratio, and criticality is precluded. Since it is difficult to prove that sorption is the only precipitation mechanism, the possibility is considered that plutonium might be at least partially separated by hydraulic forces. Even if separation were assumed, the following conditions would also have to occur before criticality becomes possible:

1. Plutonium would have to separate from iron and manganese by factors of 6.7 and 4.7, respectively;
2. Plutonium would need to concentrate by a factor of at least 20;
3. The plutonium rich region would have to form into a compact geometry that would be highly unlikely to occur.

Vail (1997b) concludes "there is no credible mechanism for plutonium to concentrate in sufficient quantities to constitute a nuclear criticality concern." Before being sluiced, SST C-106 had one of the highest plutonium inventories of any Hanford Site waste storage tank, and this conclusion should apply equally as well to sluicing operations for other tanks.

G5.0 SALTWELL PUMPING

Liquid waste enters a saltwell through perforations in the steel wall, and a pump transfers this liquid to another tank. Initially, some sludge and saltcake solids are carried along with the liquid, but the proportion of solids transferred quickly drops as the bulk solids surrounding the saltwell act as a filter to prevent additional solid particles from flowing into the well (Bratzel et al. 1996).

Serne et al. (1996) examined solids transport during saltwell pumping and concluded that the impact for criticality safety is negligible:

Assuming that the initial velocities generated by pumping could mobilize the plutonium-rich particles or the neutron absorber-rich particles near the well, the particles would be pumped out of the tank. This would leave an area in the vicinity of the well with diminished concentrations of either plutonium- or neutron absorber-rich particles. However, because calculated velocities drop markedly as the distance from the well increases, the effect would be very localized. Away from the pump, the velocities are so low that no particle resuspension is plausible and, whatever their size, the sludge particles remain as they were before saltwell pumping.

Simulation results show maximum velocities immediately adjacent to the well of 0.00209 m/s (for a pumping rate of 2 gal/min) during the first minute of pumping. After this time the sludge around the well dries out and flow decreases. The simulations do not account for the liquid already in the well before pumping starts. As discussed in Sections 2 and 4.1, the critical velocities needed to resuspend particles in natural stream beds range from 0.2 to 2.5 m/s. Further, cohesive particles such as fine-grained clays actually require velocities of a few meters/second. The calculations using the STOMP code suggest that the highest velocities will be three orders of magnitude lower than this value and will occur in an area immediately surrounding the pump for very short durations before the flow drops to essentially zero.

In summary it appears that saltwell pumping has not affected particle segregation if the sludge is fine-grained silt and clay-like. If the sludge maintains these characteristics, any future saltwell pumping also will not affect particle segregation. If the tank contains only saltcake, the permeabilities will be higher and flow rates could also be higher. However, saltcake does not contain significant amounts of plutonium, so saltcake particle transport is not an issue for criticality.

In conclusion, saltwell pumping is a benign operation that does not cause significant separation and/or physical concentration of solid materials (Bratzel et al. 1996).

G6.0 MIXER PUMP OPERATION

Serne et al. (1996) modeled mixer pump operations using the TEMPEST computer code. Two cases were modeled: a single rotating jet and two mixer pumps at the center of a DST. The degree of selective relocation of plutonium (includes particle segregation) was found insufficient to cause a concern of criticality. The operation of one or two mixer pumps will not deposit solids in a compact geometry. A single mixer pump would likely deposit solids in an annular ring around the pump.

G6.1 MIXER PUMP IN DOUBLE-SHELL TANK SY-101

A single mixer pump is installed in DST SY-101 to remove the potential for a gas explosion. As part of the justification for this pump, the potential for accidental criticality was reviewed (LANL 1995). Approximately 1 kg of plutonium in this tank is combined with a large amount of iron and other metals. The conclusion was reached that the proposed mixer pump would not lead to a nuclear criticality.

G6.2 MIXER PUMP IN DOUBLE-SHELL TANK AZ-101

Onishi and Recknagle (1997) evaluated operation of a mixer pump in DST AZ-101. A three-dimensional simulation was performed with the time-varying TEMPEST code of the operation of two 300-hp pumps with four rotating 18.3-m/s (60-ft/s) jets. The heterogeneous distribution of the plutonium as a result of 262 sequential discharges to produce a plutonium inventory of 9.8 kg was considered. Based on discharge records, the highest plutonium concentration was assumed to be 1.88 g/L in a thin layer. This value is 30 times greater than the average plutonium concentration. The DST AZ-101 model, with these very conservative initial conditions, predicted a maximum plutonium quantity in the pump housing of 75 g after 10 seconds with an average concentration of 0.6 g/L. After four minutes, the quantity decreased to 10 g and the average concentration decreased to 0.1 g/L. Onishi and Recknagle (1997) concluded that the pump jet mixing operation could not result in criticality.

Vail (1997a) bases his evaluation on Onishi and Recknagle (1997), but provides a greater depth of discussion and assumes a larger plutonium inventory of 23.2 kg. Concerning continued mixing over a long period of time, Vail (1997a) concludes that "additional mixing and settling of all the plutonium in the tank cannot continually increase the concentration above the average concentration allowed by the total mass of plutonium in the tank." If one assumes an extremely high concentration of 8 g/L is achieved, then the total inventory of the tank would form a slab only 0.7 cm (0.26 in.) thick. This slab would be highly subcritical.

G6.3 MIXER PUMP IN DOUBLE-SHELL TANK AN-107

Vail (1996) evaluated the operability testing of the DST AN-107 mixer pump. The plutonium inventory in this tank is estimated to be 3,459 g, with 1,789 g in the supernate and 1,670 g in the

sludge layer. The estimated plutonium concentration in the sludge was 0.002 g/L. For the plutonium areal density to reach the minimum critical value of 2,582 g/m², the entire plutonium content of the solids would have to be contained in an area of only 1.34 m² (14.4 ft²).

In addition, the plutonium concentration would have to increase by a factor of 5,000 to reach the 10 g/L minimum required for criticality with 3,000 g of plutonium. No scenario was found that might credibly result in this degree of concentration. It was concluded that the large margin of safety was sufficient to permit operation of the mixer pump.

G7.0 AIRLIFT CIRCULATOR

An airlift circulator is a vertical steel pipe open at both ends that extends into the supernate layer above the settled waste solids. Air is pumped through the pipe to circulate liquid and to disperse suspended sludge solids. By evening out heat generation, airlift circulators help prevent thermal hot spots. Serne et al. (1996) shows that airlift circulator operation can suspend solids located beneath the circulators, if the particles are small, but this material would be redistributed over the area of the tank. Airlift circulators do not reduce the margin of subcriticality.

G8.0 SLURRY DISTRIBUTOR

A "slurry distributor" is installed in some tanks, such as in DST AY-102, to distribute the incoming slurry over a large area. Distribution of the slurry is accomplished by spraying the slurry from a rotating nozzle to ensure a high degree of dispersal. Dispersal of the incoming slurry reduces the likelihood of creating a region of high plutonium concentration. The incoming waste forms a layer on top of the original waste that is uniform in both thickness and composition. Use of a slurry distributor enhances criticality safety by producing a more uniform distribution of waste components.

G9.0 TRANSFER OF LIQUID WASTE

Stabilization of waste in SSTs is accomplished by pumping drainable liquid out of these tanks and transferring it to newer, more reliable DSTs. Sometimes liquids are also transferred between DSTs. The usual procedure for removing liquid is by pumping from a "saltwell" created by placing a pipe into the waste. Liquid drains into this well through a screen designed to remove particulate.

Plutonium removed with the liquid must either be dissolved or be in the form of fine particulate. The low solubility of plutonium and the low content of particulate puts an upper limit upon the concentration of plutonium in this liquid. A discussion of plutonium solubility is provided in Appendix D. Based upon solubility experiments, the saturation concentration of dissolved

plutonium in waste liquid under the most idealized conditions should not be greater than 0.017 g/L (Hobbs and Edwards 1993, p.1). Depending upon the makeup of the waste, the plutonium saturation concentration might be as low as 0.002 g/L. Solid components in the waste should cause the actual plutonium concentration to be lower than these values.

The *Safety Assessment* (Braun et al. 1994) reports 306 analyzed liquid samples. Of these, two are reported with plutonium concentrations somewhat greater than 0.01 g/L, but the median plutonium concentration is reported at about 0.0001 g/L.

G9.1 PARTICULATE SUSPENDED IN PUMPED LIQUID

Liquid accumulating in a saltwell may contain a small quantity of solids. Since this liquid passes through a fine mesh screen, the size of the particles will be small, and the concentration of particles in the liquid is expected to be small.

The composition of particles suspended in the liquid should be the same as the composition of solids within the waste. The process of becoming suspended should not contribute in any way to increasing the ratio of plutonium to solids mass. The plutonium concentration is very likely to be less than 0.1 g/L and probably will be no greater than 0.01 g/L. The low concentration of particulate suspended in the liquid combined with the low plutonium concentration in solids ensures that the total plutonium in the suspended particulate is small.

When the particulate in the pumped liquid settle, they would form a layer whose composition is the same as the original solids. These particulate would be distributed in a layer of low areal density over the cross sectional area of the receiving tank. They should have a negligible impact upon reactivity within the receiver tank.

G9.2 RECEIVER TANK CONSIDERATIONS

Liquid waste in a SST contains very little suspended solids. When this liquid is transferred to a receiver tank, it forms a layer above the existing solids layer or mixes with supernate already present. The plutonium distribution within this liquid should be nearly uniform, and the plutonium areal density should be nearly uniform.

The liquid volume pumped into the receiver tank would not be expected to form a layer more than several feet thick. Nevertheless, if one makes the conservative assumption that the incoming liquid forms a layer 6.1 m (20 ft) deep, this defines an upper bound on plutonium areal density for the incoming waste. At the same time, if the assumption is made that the plutonium is at saturation concentration, then the plutonium concentration is 0.017 g/L. Using these conservative assumptions, the plutonium areal density is 103 g/m² (9.6 g/ft²), a value only 4% of the minimum critical areal density of 2.6 kg/m² (240 g/ft²). The incoming waste will remain well subcritical, even if the plutonium settles into a layer of any thickness.

If an areal density of 103 g/m² (9.6 g/ft²) were actually achieved, the total plutonium present in a 22.86-m (75-ft) diameter tank would be 42.4 kg. This is greater than the estimated inventory in all but a few of the 177 waste tanks. In practice, dissolved plutonium comprises a small

percentage of the plutonium in a tank. Most plutonium is associated with the solids and would not be removed in pumped liquid. The actual areal density should be at least a factor of 10 less than the value obtained above.

For criticality to occur in liquid waste, the plutonium concentration must exceed 7 g/L. The plutonium saturation concentration of 0.017 g/L is at least 411 times less than required to achieve criticality. Even if a concentration of 7 g/L were actually achieved, criticality would remain unlikely for several reasons. First, this high concentration must be exceeded over a relatively large volume. Second, sufficient plutonium would have to be present. Third, neutron poisons present in the waste would increase the concentration required to achieve criticality.

There are no known mechanisms capable of concentrating the plutonium to a degree even remotely approaching that for which criticality becomes possible. It is concluded that pumping liquid from a SST to a DST cannot remove the waste from a highly subcritical state.

G10.0 TRANSFER OF WASTE SOLIDS

Transfer of waste solids from one tank to another is a waste-disturbing activity requiring special evaluation. Although transfer of waste solids from SST C-106 has been completed, this operation provides a model for similar operations. Criteria are provided against which to judge the acceptability of future waste transfers.

G10.1 TRANSFER OF SOLIDS FROM SINGLE-SHELL TANK C-106 TO DOUBLE-SHELL TANK AY-102

The criticality safety evaluation of the transfer of solids from SST C-106 to DST AY-102 is provided by Vail (1997b). This evaluation supercedes earlier, less comprehensive, evaluation by Rogers (1994a).

Single-shell tank C-106 is a high-heat tank that received periodic additions of water to replace losses due to evaporation. Over a 10 year period about 3,000 kL (800 kgal) of cooling water was added at a rate of about 25 kL (6.6 kgal) each month. This water was required to prevent the waste from drying out and to maintain the temperature within limits. Temperature fluctuations observed on one of the thermocouple trees following addition of cooling water raised concerns for safety. Removal of solids was deemed necessary to remove the need for water addition that caused the fluctuations. Rogers (1994b) discusses the implications of the heat anomaly to criticality safety.

For this discussion, the term "sludge" is synonymous with "solids," and the term "supernate" is synonymous with "liquid."

G10.1.1 Description of Single-Shell Tank C-106

Single-shell tank C-106 is 75 ft (22.86 m) in diameter with a capacity of 2,000 kL (530 kgal). Before removal of solids, it contained 746,000 L (197,000 gal) of sludge stratified in two layers. The top layer consisted of 655,000 L (173,000 gal) of high heat generating sludge (contains ⁹⁰Sr). The bottom layer consisted of 91,000 L (24,000 gal) of hardened coating waste from dissolution of aluminum fuel cladding. Before transfer, the volume of drainable liquid in SST C-106 was 121,000 L (32,000 gal) of supernate and 60,600 L (16,000 gal) of interstitial liquid.

G10.1.2 Characterization of Waste in Single-Shell Tank C-106

Harris (1993) reports eight (8) analytical samples for SST C-106: 3 sludge (solids), 3 supernate (liquid), and 2 combination samples (in which solids and liquids are homogenized). Rogers (1994a) shows concentration of the individual components for the solids and the liquids, based on Harris (1993). Vail (1997b) provides the following description of the contents of SST C-106:

The average concentration of plutonium in the sludge is 0.076 g/L, which is a factor of 34 lower than the minimum critical concentration of 2.6 g/L. The mass ratios for Fe/Pu, Al/Pu, and Mn/Pu are 1,070, 653, and 147, respectively. The Fe/Pu and Mn/Pu mass ratios are much greater than the minimum critical mass ratios for these neutron absorbers (160 and 32, respectively, from Braun et al. (1994)). To reach these minimum criticality limits, the average sludge material must undergo a chemical alteration that concentrates the plutonium 34 times and that separates plutonium from iron and manganese by factors of 6.7 and 4.6, respectively.

When the high plutonium concentration of 0.089 g/kg is multiplied by the high reported solids density of 1.43 kg/L, the plutonium concentration in solids is found to be 0.127 g/L, and the total plutonium content in 746,000 L (197,000 gal) of solids is 94.6 kg. When an estimated 2.9 kg of plutonium in the 182,000 L (48,000 gal) of liquid is added, the upper limit on plutonium inventory becomes 97.5 kg (Rogers 1994a). Agnew (1993) estimated the plutonium inventory of SST C-106 to be 63.7 kg.

G10.1.3 Chemistry Considerations

Prior to transfer of SST C-106 solids, a multi-disciplinary team with expertise in nuclear engineering and waste chemistry was assembled to assess the criticality safety aspects of the transfer operations. The following summarization is taken from Vail (1997b):

There is evidence that the plutonium in the waste tanks will be chemically bonded (sorbed) onto the surface of the metal oxyhydroxides of strong neutron absorbers such as iron, aluminum and manganese (all of which are in large abundance in both tanks). There is also evidence that such bonding will remain during the aging process. If sorption is the dominant precipitation mechanism in these strongly alkaline waste forms, there is absolutely no possibility of criticality -- because any potential concentration mechanism would concentrate the neutron poisons in the same ratio.

The possibility was considered that part of the plutonium might precipitate through a mechanism other than chemical sorption. Even then, under normal conditions, the large amounts of neutron absorbers would always be in close proximity to the plutonium and would ensure subcriticality. In addition, the possibility was considered that hydraulic forces associated with the proposed retrieval process might be capable of concentrating the plutonium to some degree. Nevertheless, there are several layers of protection against the formation of a critical configuration, including the need for near optimal moderation, a large increase in plutonium concentration, and a large quantity of plutonium. For criticality to occur, it would be necessary to increase the plutonium concentration over a large volume by a factor of at least 20 above the maximum measured concentration in DST C-106 of 0.13 g Pu/L.

If all of the plutonium contained in both SST C-106 and DST AY-102 were placed in a single tank, the maximum possible plutonium would not exceed 108 kg. If this plutonium were concentrated to 4 g Pu/L, it would form a sphere about 3.7 m (12 ft) in diameter. The physical possibility of achieving such a compact and highly concentrated sphere of plutonium is extremely unlikely, and this configuration would be subcritical.

Criticality would require the simultaneous occurrence of several low probability phenomena. The multi-disciplinary team was not able to find any scenario that would be capable of causing criticality under credible conditions.

G10.1.4 Sluicing

Before waste was pumped to DST AY-102, it was sluiced into a pumpable liquid using two sluicers installed on opposite sides of SST C-106. Vail (1997b) concludes that there is no mechanism in the process of sluicing capable of concentrating plutonium to the degree necessary to make criticality possible. See Section H4.0 for more information on sluicing.

G10.1.5 Receiver Tank AY-102

Double-shell tank AY-102 was selected to receive waste solids from SST C-106 based upon the similarity and compatibility of the wastes. Double-shell tank AY-102 is a 22.86-m (75-ft) diameter tank containing 2,112 kL (558 kgal) of waste (Hanlon 1999). Of this, 1,999 kL (528 kgal) is supernatant liquid and 114 kL (30 kgal) is sludge. The plutonium inventory is 8.7 kg. A "slurry distributor" is installed in DST AY-102 to spread incoming slurry over a wide area and to provide uniform mixing of the waste. This process reduces the possibility of creating a volume in which the plutonium concentration is much higher than the average.

Vail (1997b) provides the following description of the contents of DST AY-102:

The concentration of plutonium in the sludge is 0.075 g/L, which is a factor of 35 lower than the minimum critical concentration (2.6 g/L)..... If the highest plutonium concentration recorded in tank 241-AY-102 (namely 0.12 g Pu/L) is used, the degree of concentration necessary to approach criticality concern is 22.

The mass ratios for Fe/Pu, Al/Pu, and Mn/Pu are 1,560, 705, and 164, respectively. The Fe/Pu and Mn/Pu mass ratios are much greater than the minimum critical mass ratios for these neutron absorbers (160 and 32, respectively, from Braun et al. (1994)). To reach these minimum criticality limits, the sludge must undergo a chemical alteration that on average concentrates the plutonium 35 times and that separates plutonium from iron and manganese by factors of 9.8 and 5.1, respectively.

The fluidity required for transfer operations ensures a high water content, and neutron absorbers would further increase the minimum concentration of plutonium required for criticality. At the degree of moderation in these waste solids, the plutonium concentration required to achieve criticality is expected to be at least 7.2 g Pu/L. The plutonium concentration in the supernate (liquid) is more than 10,000 times smaller than the minimum critical concentration.

The uranium/plutonium (U/Pu) mass ratio for DST AY-102 waste is found to be about 279 in the solids, or about 36% of that required to ensure subcriticality. The cadmium/plutonium mass ratio of 7.8 converts to an atom ratio of about 16, and the boron/plutonium mass ratio of 53 converts to an atom ratio of about 1,100. The concentrations of both of these elements are far larger than required to ensure subcriticality for the plutonium present.

At the very least, a 50-fold increase in the plutonium concentration would be required for criticality. In reality, the large contents of iron and manganese ensure that a far greater increase would be required. There is no identified mechanism capable of separating plutonium from other components, and the waste is well subcritical in comparison to several components with different chemical properties. For criticality to occur, separation would have to occur over a large volume of solids. No mechanism capable of causing criticality as a result of mixing waste from SST C-106 with that already in DST AY-102 has been identified, and criticality is not considered to be credible.

G10.2 FUTURE TRANSFERS OF SOLIDS

Waste is transferred from one storage tank to another along transfer lines. Most transfer lines have inside diameters less than 4 in. No transfer line has an inside diameter greater than 10 in. Waste that is acceptable for tank storage cannot be made critical in transfer lines. No separate evaluation for criticality safety is required for the transfer lines.

Bratzel et al. (1996) concludes that an accidental criticality is incredible under existing waste tank conditions. Before a transfer of waste is approved assurance is required that conditions in the receiver tank will remain within "existing waste conditions." This requires that waste in the sending and the receiving tanks are chemically similar and compatible. Maintaining "existing tank conditions" requires at minimum a pH greater than 8.0 in both tanks.

Before a transfer of waste is made, a criticality safety review shall be made to document that the parameters are within acceptable limits. Transfer of supernate and saltwell liquid is acceptable when the content of solids is consistent with liquid waste after solids have been allowed to settle out (see Section 9.9). Transfer of mobilized (suspended) solids is acceptable when the following conditions are met: (1) the waste in the receiver tank

remains within "existing waste conditions; and, (2) surveillance requirements are met in both the sending and receiver tanks. If it cannot be shown that these conditions are met, a criticality safety evaluation shall be performed to assess the degree of subcriticality of the receiver tank after transfer in greater detail.

G11.0 REFERENCES

- Agnew, S.F., 1993, *Analysis of the History of the 241-C Farm*, LAUR-93-3605, Los Alamos National Laboratory, Los Alamos, New Mexico.
- Bratzel, D. R., W. W. Schulz, R. Vornehm, and A. E. Waltar, 1996, *Tank Farm Nuclear Criticality Review*, WHC-SD-WM-TI-725, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- Braun, D. J., L. D. Muhlestein, T. B. Powers, and M. D. Zentner, 1994, *High-Level Waste Tank Subcriticality Safety Assessment*, WHC-SD-WM-SARR-003, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- Hanlon, B. M., 1999, *Waste Tank Summary Report for Month Ending July 31, 1999*, HNF-EP-0182-136, Fluor Daniel Hanford, Inc., Richland, Washington.
- Harris, J. P., 1993, *101-AI, 102-AI, & 106-C Data Compendium*, WHC-SD-WM-TI-578, Rev. 0, August 19, 1993.
- Hobbs, D. T., and T. B. Edwards, 1993, *Solubility of Plutonium in Alkaline Salt Solutions (U)*, WSRC-TR-93-131, Savannah River Technology Center, Westinghouse Savannah River Company, Aiken, South Carolina.
- LANL, 1995, *A Safety Assessment for Proposed Pump Mixing Operations to Mitigate Episodic Gas Releases in Tank 241-SY-101: Hanford Site, Richland, Washington*, LA-UR-92-3196, Rev. 14, Appendix I, Los Alamos National Laboratory, Los Alamos, New Mexico.
- Lipke, E. J., 1999, *Above Average Sample in 241-TX-118*, (Letter 74F00-EJL-99028 to N. L. Hulse, July 22), Lockheed Martin Hanford Corporation, Richland, Washington.
- LMHC, 1999, *Tank Waste Remediation System Final Safety Analysis Report*, HNF-SD-WM-SAR-067, Rev. 0, Lockheed Martin Hanford Corporation, Richland, Washington.
- Onishi, Y., and K. Recknagle, 1997, *Tank 241-AZ-101 Criticality Assessment Resulting From Pump Jet Mixing Sludge Mixing Simulations*, PNNL-11486, Pacific Northwest National Laboratory, Richland, Washington.

- Rogers, C. A., 1993a, *CSER 92-008: Waste Stabilization For Single Shell Tank 101-T*, WHC-SD-SQA-CSA-20355, Rev. 1, Westinghouse Hanford Company, Richland, Washington.
- Rogers, C. A., 1993b, *CSER 92-006: Waste Stabilization For Single Shell Tanks 102-BY, 109-BY, 102-C, 107-C, and 110-C*, WHC-SD-SQA-CSA-20352, Rev. 1, Westinghouse Hanford Company, Richland, Washington.
- Rogers, C. A., 1993c, *CSER 93-002: Waste Stabilization For Single Shell Tanks 110-BX and 111-BX*, WHC-SD-SQA-CSA-20359, Rev. 1, Westinghouse Hanford Company, Richland, Washington..
- Rogers, C. A., 1994a, *CSER 94-001: Criticality Safety of Single Shell Waste Storage Tanks*, WHC-SD-SQA-CSA-20363, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- Rogers, C. A., 1994b, *CSER 94-009: Implications of the Heat Anomaly in Tank 106-C to Criticality Safety*, WHC-SD-SQA-CSA-20373, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- Serne, R. J., G. A. Whyatt, S. V. Mattigod, Y. Onishi, P. M. Doctor, B. N. Bjornstad, M. R. Powell, L. M. Liljegren, J. H. Westsik, Jr., N. J. Aimo, K. P. Recknagle, G. R. Golcar, T. B. Miley, G. R. Holdren, D. W. Jeppson, R. K. Biyani, and G. S. Barney, 1996, *Fluid Dynamics, Particulate Segregation, Chemical Processes, Natural Ore Analog and Tank Inventory Discussions that Relate to the Potential for Criticality in Hanford Tanks*, WHC-SD-WM-TI-757, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- Vail, T. S., 1996, *CSER 96-004: Criticality Safety of Operability Testing the Mixer Pump in Tank 241-AN-107*, WHC-SD-SQA-CSA-505, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
- Vail, T. S., 1997a, *CSER 96-014: Criticality Safety of Project W-151, 241-AZ-101 Retrieval System Process Test*, WHC-SD-W151-CSA-001, Rev. 1, Duke Engineering & Services, Richland, Washington.
- Vail, T. S., 1997b, *Criticality Safety Assessment of the Tank 241-C-106 Remediation*, WHC-SD-W320-CSA-001, Rev. 1, Duke Engineering & Services, Richland, Washington.
- WHC, 1995, *Tank Waste Source Term Inventory Validation*, WHC-SD-WM-ER-400, Westinghouse Hanford Company, Richland, Washington.

DISTRIBUTION SHEET

To	From	Page 1 of 1
Distribution	Process Control	Date 02/16/00
Project Title/Work Order		EDT No. EDT-628251
RPP-5296, Rev. 0, "Criticality Safety Evaluation of Hanford Site High-Level Waste Storage Tanks"		ECN No. N/A

Name	MSIN	Text With All Attach.	Text Only	Attach./Appendix Only	EDT/ECN Only
<u>CH2M Hill Hanford Group, Inc.</u>					
C. DeFigh-Price	R2-12	3			
R. A. Dodd	R3-72	X			
L. A. Fort	R2-12	X			
K. D. Fowler	R2-11	X			
C. J. Hopkins	R2-50	X			
W. J. Kennedy	S5-07	X			
N. W. Kirch	R2-11	X			
C. A. Rogers	R2-11	4			
R. G. Stickney	R1-43	X			
L. E. Thomas	S7-86	X			
E. V. Weiss	R2-12	X			
T.C.S.R.C.	R1-10	X			
<u>Fluor Hanford</u>					
T. A. Campbell	T4-56	X			
G. R. Franz	A0-26	X			
T. S. Vail	T4-56	X			
<u>Fluor Federal Services</u>					
C. S. Eberle	B4-44	X			
A. M. Rau	B4-46	X			
H. Toffer	B4-44	X			
<u>Lockheed Martin Services, Inc.</u>					
Central Files	B1-07	X			
<u>Office of River Protection</u>					
W. F. Hendrickson	H6-60	2			
J. D. Voice	H6-60	X			
DOE Reading Room	H2-53	X			
<u>Numatec Hanford Corporation</u>					
A. F. Choho	R3-73	X			
<u>Waste Management Federal Services of Hanford, Inc.</u>					
R. J. Nicklas	T4-56	X			