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Corrosion Considerations for Life Management of Hanford High-Level Waste Tanks

Prepared for the U.S. Department of Energy
Office of Environmental Restoration and
Waste Management

Westinghouse
Hanford Company Richland, Washington

Hanford Operations and Engineering Contractor for the
U.S. Department of Energy under Contract DE-AC06-87RL10930

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CORROSION CONSIDERATIONS FOR LIFE MANAGEMENT OF HANFORD HIGH-LEVEL WASTE TANKS

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ABSTRACT[1]

The potential for corrosion-related aging mechanisms to be active in the Hanford Site waste tanks is frequently questioned and there are related uncertainties. This paper considers surveillance and analyses for evaluating the potential influence of corrosion processes such as stress corrosion cracking, pitting, crevice corrosion, uniform corrosion, microbiologically-induced corrosion, embrittlement, and corrosion of the reinforced concrete steel on the useful life of Hanford radioactive waste tanks.

There are two types of Hanford Site underground reinforced concrete, carbon steel lined waste tank structures. They primarily store caustic nitrate wastes, some at elevated temperatures, from defense reprocessing of spent nuclear fuels. Some of the Hanford waste tanks have leaked radioactive liquid waste to the soil. These leaks are possibly due to nitrate-induced stress corrosion cracking. Major efforts prescribed to avoid nitrate-induced stress corrosion cracking in newer tank designs appear successful. A potential for pitting and crevice corrosion cracking in the carbon steel liners exists. There has been no evidence of significant uniform corrosion of the carbon steel liners and there has been no evidence of waste tank degradation caused by corrosion of the concrete reinforcing steel.

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As much as practical, surveillance for evidence of active corrosion is conducted. This surveillance primarily consists of leak detection and monitoring today. Remote closed-circuit television examinations of one type of Hanford waste tank were recently completed and a remote (robotic) ultrasonic testing (UT) system is under development for quantitative examination of the same type of tanks that received the closed-circuit television examination.

A waste tank life management program is being developed to qualify the Hanford waste tanks for continued safe storage of these wastes. Corrosion evaluations, structural analyses, and surveillance are required to qualify the tanks and to promptly detect evidence of possible distress.

Keywords: aging management, aging mechanisms, life management, Hanford, radioactive waste, high-level waste tanks

INTRODUCTION

The U.S. Department of Energy Hanford Site occupies 570 square miles of sagebrush steppe in Southeastern Washington State. This site currently stores and manages 61 Mgal of radioactive waste resulting from 50 years of weapons production in 177 large underground waste tank structures.

In 1987, the Hanford mission changed from producing special nuclear material for nuclear weapons to environmental cleanup. Even the most aggressive environmental cleanup schedules recognize the need for development of new waste retrieval and disposal technologies and construction of processing facilities to dispose of the Hanford defense wastes. In the interim, as these technologies are developed and processing facilities are constructed, the wastes must be managed in a safe, environmentally responsible manner.

There are two basic types of Hanford underground storage tank structures (Figure 1): Single Shell Tanks (SSTs) (four configurations) and Double Shell Tanks (DSTs) (one configuration). These tanks were originally designed to store radioactive liquid wastes. The SSTs currently store 35 Mgal of sludge and saltcake waste (semi-solid and solid waste forms) and 5 Mgal of pumpable liquids. Of the 149 SSTs, 67 are known or assumed to have leaked radioactive waste to the soil. There are 28 DSTs which currently contain 25 Mgal of waste ranging from semi-solid sludge to liquid supernate[1].

Continued operation of the DSTs and interim storage in the SSTs will be required until the wastes are retrieved and permanently disposed. The radiation and chemical exposure hazards to personnel and equipment, waste tank configurations, limited engineering records, and past operating practices, together with limited interim storage options and long-term operating demands, provide unique challenges to managing the useful life of the Hanford waste tanks. Current capabilities for inspection and major rehabilitation of the Hanford waste tanks are limited by restricted access to the structures.

SYSTEM DESCRIPTION

Hanford High-Level Waste Tanks

Single Shell Tanks were constructed from 1943 to 1964. The SST structures range in capacity from 55 Kgal to 1 Mgal and consist of reinforced concrete shells with a single mild carbon steel plate liner (Figure 1). The existing Double Shell Tanks (DSTs) were constructed from 1967 to 1986 and are all 1 Mgal capacity tanks. The DST structures consist of a totally enclosed, carbon-steel-plate primary tank surrounded by a carbon steel plate lined, reinforced concrete shell (Figure 1). Table 1[2,3] shows Hanford waste tank steel plate specifications along with original construction dates and nominal design lives.

The waste tanks have two primary functions:

Confinement - The tank shells and liners provide confinement barriers for liquid and solid wastes

Structural Stability - The reinforced concrete shells and the primary steel tanks provide acceptable safety margins for continued operation under normal and abnormal loads.

Waste Characteristics

The radioactive wastes stored in Hanford waste tanks originated from chemical reprocessing of defense production reactor irradiated fuels and various plutonium-uranium separations, chemical extraction, and decontamination processes required to support the past weapons production mission. The first fuel separations process (bismuth-phosphate) generated slightly alkaline wastes. Starting in the 1950's, the PUREX (Plutonium-URanium EXtraction) process generated caustic nitrate wastes. Shortly after introduction of the PUREX process caustic nitrate waste, waste tank leaks were discovered at both Hanford and Savannah River. Concentration limits were placed on nitrates, nitrites, and hydroxides to prevent nitrate-induced stress corrosion cracking. Table 2 is an aggregate representation of the major chemical constituents currently stored in Hanford waste tanks.

Waste temperatures have varied from campaign to campaign depending on the concentrations and decay heat of radioactive constituents in wastes transferred to the waste tanks. Previous waste volume reduction efforts in some tanks utilized this decay heat to self-concentrate the waste. Operating specifications have always required temperature limits for the waste contents in order to control the temperatures and thermal gradients in the reinforced concrete structures. Elevated temperatures can overstress and degrade the concrete and accelerate corrosion/aging mechanisms in the waste tank steels. Waste temperatures in Hanford waste tanks currently range from ambient to 88°C (191°F). For most of the waste tanks, waste temperatures have historically been less than 180°C (356°F). Some of the SST wastes are suspected of reaching temperatures up to 315°C (600°F)[5].

CORROSION/AGING CONSIDERATIONS FOR HANFORD HIGH-LEVEL WASTE TANK STEELS

Corrosion/aging of waste tank steels can be manifested in two ways: (1) changes in material properties (e.g., yield strength, ductility, and toughness), or (2) general or localized changes in material geometry (e.g., cracking, thinning, and pitting). Both tend to reduce the ability of the material to perform as designed. Research and operating experience suggest that geometry changes will be more affected by active corrosion/aging mechanisms in Hanford Site waste tank steels than material properties.

Corrosion studies, testing programs, and failure analyses to determine the effect of Hanford wastes on carbon steels used for tank liner plates have been conducted, with few interruptions, since the first SSTs were designed and constructed in 1943-1944. The principal objective of these activities has been to assess the potentials and rates for uniform corrosion, pitting, and nitrate-induced stress corrosion cracking. Test solutions have included synthetic bismuth-phosphate and caustic nitrate wastes. Laboratory testing has been performed in the liquid and vapor phases at various temperatures for periods ranging from 6 months to 6 years[6].

Currently, the scope and requirements are being developed for followup corrosion studies to assess past corrosion trends for the tank liners and estimate future trends. First, a data base of corrosion experiments and results will be organized from Hanford waste tank steel corrosion work, that previously has been organized around specific projects with different needs, schedules, and resources. This data base will be used to support life management decisions.

Stress Corrosion Cracking

Waste chemistries, elevated temperatures, and high residual stresses in the weld heat affected zones are believed to promote stress corrosion cracking in the SST carbon steel liners. Nitrate-induced stress corrosion cracking was recognized as a potentially active mechanism in Hanford SSTs when some tanks storing the PUREX process caustic nitrate waste at the Hanford Site and the Savannah River Site began to leak. Failure analysis of a leaking Savannah River Site waste tank storing similar PUREX caustic nitrate waste confirmed stress corrosion cracking[7].

In order to mitigate future tank leaks that may be caused by nitrate-induced stress corrosion cracking, chemistry specifications were updated to control nitrate, nitrite, and hydroxide concentrations of waste streams entering the waste tanks. In addition to controlling waste chemistry, the DST waste tank design included specifications for a post-weld stress relief anneal of the primary carbon steel tank. There have been no indications of leaks from DSTs that have had both chemistry control and stress-relieved primary tanks.

Pitting and Crevice Corrosion

Pitting of Hanford waste tanks remains a long-term storage issue . Early SST corrosion studies, both in the laboratory and in waste tanks, demonstrated that pitting could occur[6]. Later DST laboratory corrosion studies confirmed pitting could occur in the vapor phase of synthetic PUREX waste[8]. Other conditions that may promote pitting in Hanford waste tanks have been postulated. Carbon dioxide in the headspace may cause a hydroxide depletion in the meniscus which can lower the pH enough to initiate pitting at the waterline. In addition, previous raw water and dilute waste additions may

create a dilute layer of nitrite solution with a lower pH than the bulk solution, and this layer may be aggressive enough to initiate pitting. Neither of these postulated conditions has been studied enough to confirm that they exist.

The physical properties of the waste stored in the SSTs are conducive to formation of crevices between the steel liners and accumulated saltcake where stagnate solutions may cause significant attack. It has also been suggested that condensation running down the side walls of the tanks may be depleted of any corrosion inhibiting constituents and may be trapped in a crevice between a saltcake surface and the carbon steel liner of SSTs.

Uniform Corrosion

Although a uniform corrosion rate of 0.5 mil-per-year is the reference life-limiting condition for design life of the Hanford DSTs, corrosion studies on mild carbon steels with simulated Hanford wastes and examinations of mild carbon steel equipment removed from waste tanks have shown no significant uniform corrosion. The practice of maintaining the waste chemistry in highly alkaline solutions (pH > 10) is expected to keep uniform corrosion rates low.

Microbiologically-Induced Corrosion

Microbiologically-Induced Corrosion (MIC) may be of concern in the annular spaces of the DSTs if stagnate rainwater or raw water could accumulate. This scenario is unlikely because leak detection systems in the annular spaces continuously monitor for moisture. In the event stagnate solutions did accumulate in the annular spaces of DSTs, the high radiation fields inside these annular spaces would be expected to suppress microbial action.

Embrittlement

There are no significant concerns for embrittlement of the Hanford waste tank steels. The operating temperatures are sufficiently high to avoid loss of ductility, hydrogen is not generated in sufficient concentrations to cause hydrogen embrittlement, and the stored wastes do not produce neutron (embrittling) radiation.

Corrosion of Concrete Reinforcing Steel

A number of chemical constituents contained in Hanford defense wastes are capable of attacking the concrete and possibly the reinforcing steel. The carbon steel liners in SSTs were designed to keep the aggressive waste constituents away from the concrete. If the structures functioned as designed, the permanently dry concrete would not support any form of electrolytic corrosion of the reinforcing steel. However, leaks in some of the SST carbon steel liners have allowed limited, or localized contact. The structural consequences of this potential contact depend largely on: (1) the size and location of the contact area (e.g., locations with low stresses near the center of the concrete base slab would have the least structural effects), (2) whether the concrete is significantly cracked allowing penetration, and (3) the rate of attack on the concrete[9].

In the early 1980s, testing was conducted to experimentally estimate the relative durability of reinforced concrete exposed to simulated waste[10]. Reinforced concrete specimens, which represented a section of a SST with the steel liner, were preloaded to prescribed stress levels and tested. The simulated waste solution (with various

concentrations of potentially aggressive sodium compounds) was leaked through the liner onto the concrete. The testing was performed at 82°C (180°F), 85% relative humidity. At selected intervals, petrographic examinations were made to evaluate changes in the concrete durability. Also, reinforcing steel from the specimens was examined for corrosion and tested for reductions in yield strength and ductility.

After 6 months of exposure to the simulated waste, the reinforcing steel showed no evidence of corrosion or disruption of the initial mill scale. Physical testing did not indicate steel strength or ductility reductions (Figure 3), and petrographic examination of the concrete showed no adverse reactions with the simulated waste solutions.

In DSTs, the reinforced concrete is separated from the wastes by the primary carbon steel tank and a secondary carbon steel liner. Photographic evidence of exposed reinforced concrete in the SST domes indicates no significant degradation (i.e., no spalling, cracking, or discoloration).

LIFE MANAGEMENT OF HANFORD HIGH-LEVEL WASTE TANKS

A waste tank life extension program is under development to qualify the existing waste tanks for continued storage of Hanford radioactive wastes by establishing the minimum safe useful life of existing SSTs and DSTs under current operational, postulated upset, and natural phenomena hazard loads; and remain able to withstand future retrieval loads. The Tank Farm Life Extension Program is expected to:

- Establish the present structural condition or status of the waste tanks
- Develop comprehensive structural and corrosion acceptance criteria for SSTs and DST to maintain confinement and structural stability
- Determine and conduct the structural analyses and corrosion evaluations necessary to predict tank performance and changes in condition caused by continued operation
- Plan and implement the SST and DST structural and corrosion surveillance required to support life extension predictions and promptly detect indications of possible structural distress
- Determine and account for the dependency of SST and DST operating lives on tank farm support systems including ventilation, electrical distribution, instrumentation, and waste transfer
- Develop options for SST and DST operating life extensions.

In summary, ongoing corrosion and material evaluations, structural analyses for long-term operating demands, and surveillance will be conducted to qualify the waste tank structures and predict their minimum safe remaining useful life.

WASTE TANK STRUCTURAL SURVEILLANCE

Waste tank structural surveillance or measurements and observations are required to confirm: (1) confinement of the liquid waste, and (2) acceptable structural conditions. As much as practical, surveillance should detect any evidence of active corrosion in the steel tanks. The surveillance should also be able to promptly detect evidence of possible structural distress. The potential release of waste, radiation exposure to workers and equipment, and other accessibility problems for the underground tanks restrict this surveillance and have made comprehensive condition surveys for the tanks impractical. The types of steel tank corrosion-related surveillance currently used and planned in the near future are described below and summarized in Table 3.

The level of the contents has always been measured and monitored in the Hanford Site SSTs and DSTs. Level measurements may indicate potential leaks in the steel tanks, and also provide an indication of active corrosion mechanisms.

The DST annular spaces are continuously and directly monitored for leakage from the primary steel tanks. Conductivity probes are used to detect free liquids and continuous air monitors are used to detect radiation. Both will activate alarms at manned locations. This monitoring, to date, confirms that the Hanford Site DSTs are not leaking, and indicates little active corrosion in the primary tanks.

A remote operated closed-circuit television camera was recently deployed in the DST annular spaces via top risers which extend to grade. Visual inspections of portions of the primary tank exterior and secondary liner interior wall surfaces were conducted. The images were recorded on video tape with the ability to detect, or confirm the absence of corrosion on these surfaces. The video tape provides a baseline for subsequent inspections. However, an active corrosion mechanism on the primary tank interior wall in contact with the waste would have to be quite advanced to be detected by this type of surveillance.

A Hanford DST UT inspection system is being developed. The UT transducers will be mounted to a robotic carriage to be deployed in the annular spaces. This system will be able to conduct UT scans on the primary and secondary steel tank walls and have a limited ability to conduct UT examinations on the primary tank bottom via the ventilation air slots in the concrete slab underneath the primary tank. The robotic carriage will be able to precisely return UT transducers to locations in the DST annular space previously examined and conduct repeat UT examinations. This allows the UT examinations to be used for baseline comparisons with sequential UT examinations. Also, followup confirmation examinations can be conducted as necessary. The UT examinations are expected to measure wall plate thicknesses, detect and measure the depth of pitting, and detect and measure the length and depth of cracks initiating from the surface (stress corrosion cracks). The UT examinations are expected to be sufficiently extensive and comprehensive to determine whether significant pitting, crevice corrosion, uniform corrosion, or stress corrosion cracking is occurring.

STRUCTURAL ANALYSES

Structural analyses of the Hanford waste tanks are conducted for long-term performance demands including normal, abnormal, and future retrieval loads. These analyses are detailed and predict continuous structural response of the reinforced concrete shell and the internal steel plate tanks during service. The basic objectives of the structural analyses are:

Confirm that the structure can withstand the anticipated long-term loadings with acceptable stable response.

Assess the structural safety margins for normal, abnormal, and future retrieval loads.

These structural analyses have been performed for the design of Hanford Site DSTs. They are conducted to obtain updated structural evaluations for both the SSTs and DSTs, to provide the technical base for operating limits, and to support waste tank life management tasks. The corrosion-related considerations included in the structural analyses for the internal steel tanks are uniform corrosion allowances and stress limits (below tensile yield) to preclude stress corrosion cracking. These considerations apply mainly to the primary steel tanks within the DSTs.

SUMMARY AND CONCLUSIONS

An effective life management program for the Hanford Site waste storage tank structures is necessary to make decisions regarding continued management of waste in existing tanks and the need for new storage capacity. The information gained from an effective life management program for the Hanford Site waste tanks is also necessary to determine time constraints placed on development of retrieval and disposal technologies and construction of processing facilities.

Conclusions

- The stress relief anneal and waste chemistry controls placed on the DSTs appear to have removed stress corrosion cracking as a major corrosion/aging mechanism for the DSTs. Many of the past leaks from SSTs (non-stress relieved, less chemistry controls) may have been the result of stress corrosion cracking. Stress corrosion cracking has enough significant degradation potential to warrant further studies and surveillance in both SSTs and DSTs. However, the feasibility of SST examinations (limited access) should be studied carefully before such commitments are made.

- Pitting and crevice corrosion remain long-term storage issues. Both of these mechanisms have enough significant degradation potential for DSTs and SSTs to warrant further studies and surveillance. The planned DST UT examinations will be able to detect and measure pitting or crevice corrosion if these forms of degradation are widely present.

- Uniform corrosion is adequately controlled with existing practices. This mechanism does not require additional studies or surveillance; however, ongoing surveillance to monitor for signs of degradation should continue. The DST UT examinations are expected to provide further confirmation.

- MIC is not a significant threat to continued operation of DSTs or SSTs. This mechanism does not require additional studies or surveillance.

- Corrosion of the reinforcing steel embedded in the concrete tank structures is believed to be insignificant. There likely has been limited contact between the SST concrete structure and leaking waste contents. However, surveillance, laboratory testing, and analyses to date indicate that this is not seriously detrimental to either the concrete or the reinforcing steel. In the DSTs, there has not been, nor is it likely that there will be, waste in contact with the

concrete or the reinforcing steel. However, the seriousness of possible effects in the event of concrete reinforcing steel corrosion in Hanford waste tanks warrant confirmatory surveillance.

- The Hanford Site waste tank Life Management Program should include corrosion and structural surveillance to ensure the waste tanks are qualified for continued safe use with acceptable confidence levels.

ACKNOWLEDGEMENTS

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NOTICE

The table below has been revised to correct errors published in the orignal NACE paper.

TABLE 1: HANFORD WASTE TANK STEELS

Tank Farm	Type	Steel Specification	Tanks	Year Constructed
Single Shell Tanks (SST)				
241-B	55 Kgal SST	AWWA A7-39	4	1943-44
	550 Kgal SST	AWWA A7-39	12	1943-44
241-C	55 Kgal SST	AWWA A7-39	4	1943-44
	550 Kgal SST	AWWA A7-39	12	1943-44
241-T	55 Kgal SST	AWWA A7-39	4	1943-44
	550 Kgal SST	AWWA A7-39	12	1943-44
241-U	55 Kgal SST	AWWA A7-39	4	1943-44
	550 Kgal SST	AWWA A7-39	12	1943-44
241-BX	550 Kgal SST	AWWA A7-39	12	1946-47
241-TX	750 Kgal SST	AWWA A283-46	18	1947-48
241-BY	750 Kgal SST	NRA A283-46T	12	1948-49
241-S	750 Kgal SST	NRA A283-46T	12	1950-51
241-TY	750 Kgal SST	NRA A283-49T	6	1951-52
241-SX	1 Mgal SST	NRA A283-52T	15	1953-54
241-A	1 Mgal SST	ASME A283-52T	6	1954-55
241-AX	1 Mgal SST	ASME A201-61T	4	1963-64
Double Shell Tanks (DST)				
241-AY	1 Mgal DST	ASTM A515 GRADE 60	2	1968-70
241-AZ	1 Mgal DST	ASTM A515 GRADE 60	2	1971-70
241-SY	1 Mgal DST	ASTM A515 GRADE 65	3	1974-77
241-AN	1 Mgal DST	ASTM A537 CLASS 1	7	1977-79
241-AW	1 Mgal DST	ASTM A537 CLASS 1	6	1978-80
241-AP	1 Mgal DST	ASTM A537 CLASS 1	8	1983-86

TABLE 2: AGGREGATE CHEMICAL COMPOSITION OF CURRENT HIGH-LEVEL WASTE INVENTORY IN HANFORD WASTE TANKS[4]

COMPONENT	SINGLE SHELL TANKS			DOUBLE SHELL TANKS
	LIQUID (WT%)	SLUDGE (WT%)	SALT CAKE (WT%)	SLURRY (WT%)
$NaNO_3$	20.8	25.3	81.5	14.8
$NaNO_2$	15.8	3.8	1.7	5.6
Na_2CO_3	0.6	2.2	0.5	1.0
NaOH	6.2	5.3	1.5	7.0
$NaAlO_2$	12.5	1.2	1.4	6.0
NaF	-	-	-	0.4
Na_2SO_4	-	1.0	1.3	0.3
Na_3PO_4	2.3	15.8	1.6	0.8
KF	-	-	-	0.4
FeO(OH)	-	1.3	-	0.2
Organic Carbon	0.17	-	-	1.2
NH_4^+	-	-	-	0.08
$Al(OH)_3$	-	2.9	-	4.9
$SrO \cdot H_2O$	-	0.1	-	-
Na_2CrO_4	1.3	-	-	-
$Cr(OH)_3$	-	0.2	-	0.02
$Cd(OH)_2$	-	0.1	-	-
$Ni(OH)_2$	-	-	-	<0.1
$BiPO_4$	-	0.5	-	-
Cl^-	-	0.1	-	-
$Ni_2Fe(CN)_6$	-	0.6	-	-
$P_2O_5 \cdot 24WO_2 \cdot \rightarrow 44H_2O$	-	<0.1	-	-
$ZrO_2 \cdot 2H_2O$	-	0.50.5	-	0.2

TABLE 2: AGGREGATE CHEMICAL COMPOSITION OF CURRENT HIGH-LEVEL WASTE INVENTORY IN HANFORD WASTE TANKS[4]

COMPONENT	SINGLE SHELL TANKS			DOUBLE SHELL TANKS
	LIQUID (WT%)	SLUDGE (WT%)	SALT CAKE (WT%)	SLURRY (WT%)
Fission Products	-	--	-	<0.01
H_2O	40.2	33.33.66	10.5	56.2
Other	<0.1	5.55.5	-	<0.01
Hg^+	-	0.12 ppm	-	-
Total	100.0	100.0	100.0	100.0
Density, g/ml	1.6	1.7	1.4	1.3

TABLE 3: SUMMARY OF POTENTIAL SURVEILLANCE AND ANALYSES

Potential Actions	Stress Corrosion Cracking	Pitting	Crevice Corrosion	Uniform Corrosion	MIC	Concrete Reinforcing Steel
Stress Analysis	DST & SST	N/A	N/A	N/A	N/A	SST & DST
Waste Chemistry	DST & SST	DST & SST	DST & SST	DST & SST	N/A	SST
Waste Physical Properties	N/A	N/A	DST & SST	N/A	N/A	N/A
Thermal History	DST & SST	DST & SST	DST & SST	DST & SST	N/A	SST & DST
Load History	DST & SST	N/A	N/A	N/A	N/A	SST & DST
Remote UT - Welds	DST	N/A	N/A	N/A	N/A	N/A
Remote UT - Bulk	DST	DST	DST	DST	N/A	N/A
Remote UT - Interface	DST	DST	DST	DST	N/A	N/A
Still Photography	DST & SST	DST & SST	DST & SST	DST & SST	N/A	SST
CCTV/Video	DST	DST	DST	DST	DST	N/A
Stereo Photo/Video	Potential	Potential	Potential	Potential	Potential	N/A
Leak Detection	DST & SST	DST & SST	DST & SST	DST & SST	DST & SST	SST
Dome Elevation Surveys	N/A	N/A	N/A	N/A	N/A	SST & DST
Electrochemical Probes	Potential	Potential	Potential	Potential	N/A	N/A
Resistance Probes	N/A	Potential	Potential	Potential	N/A	N/A
Coupons	Potential	Potential	Potential	Potential	N/A	N/A

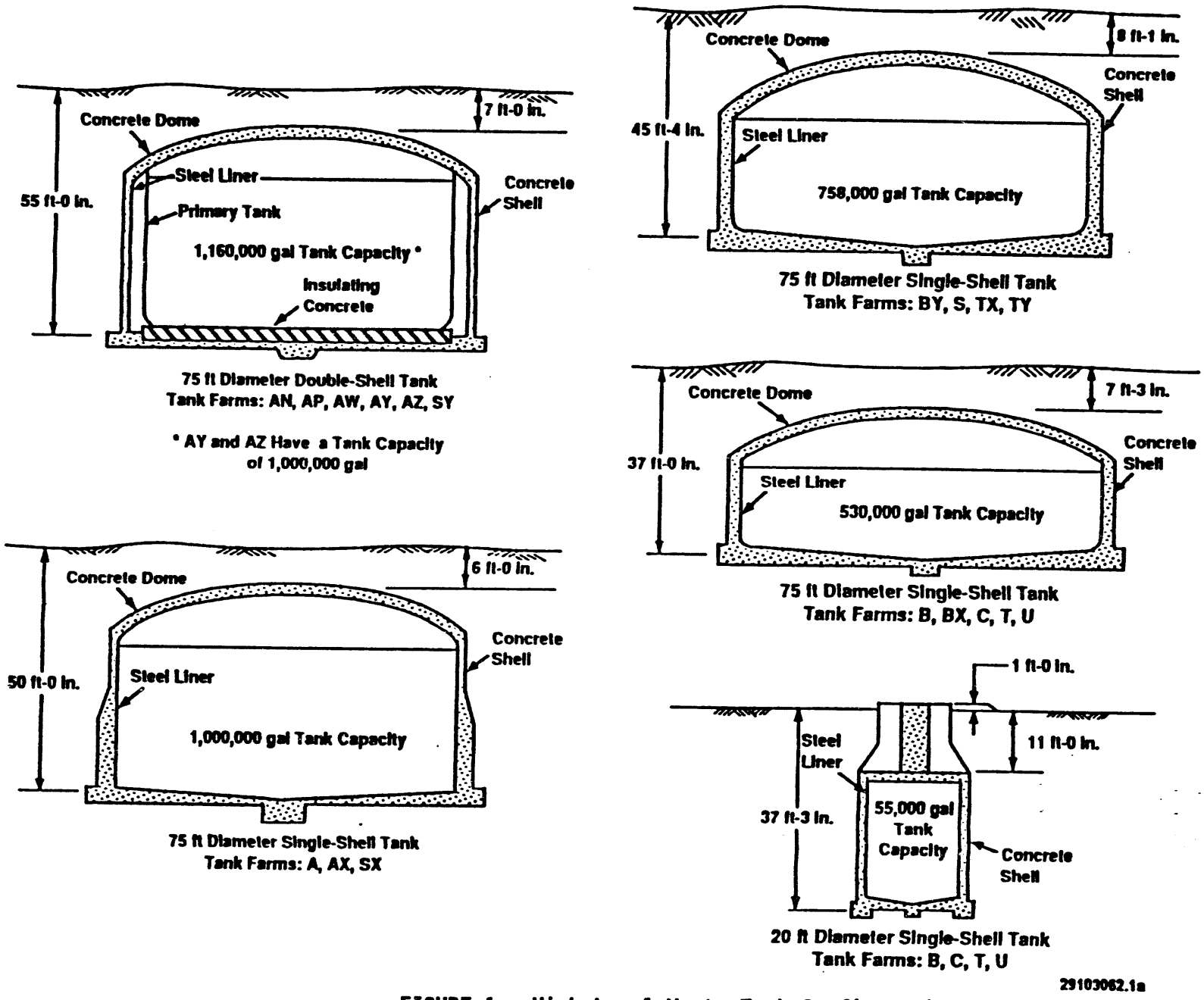

FIGURE 1 - High-Level Waste Tank Configuration

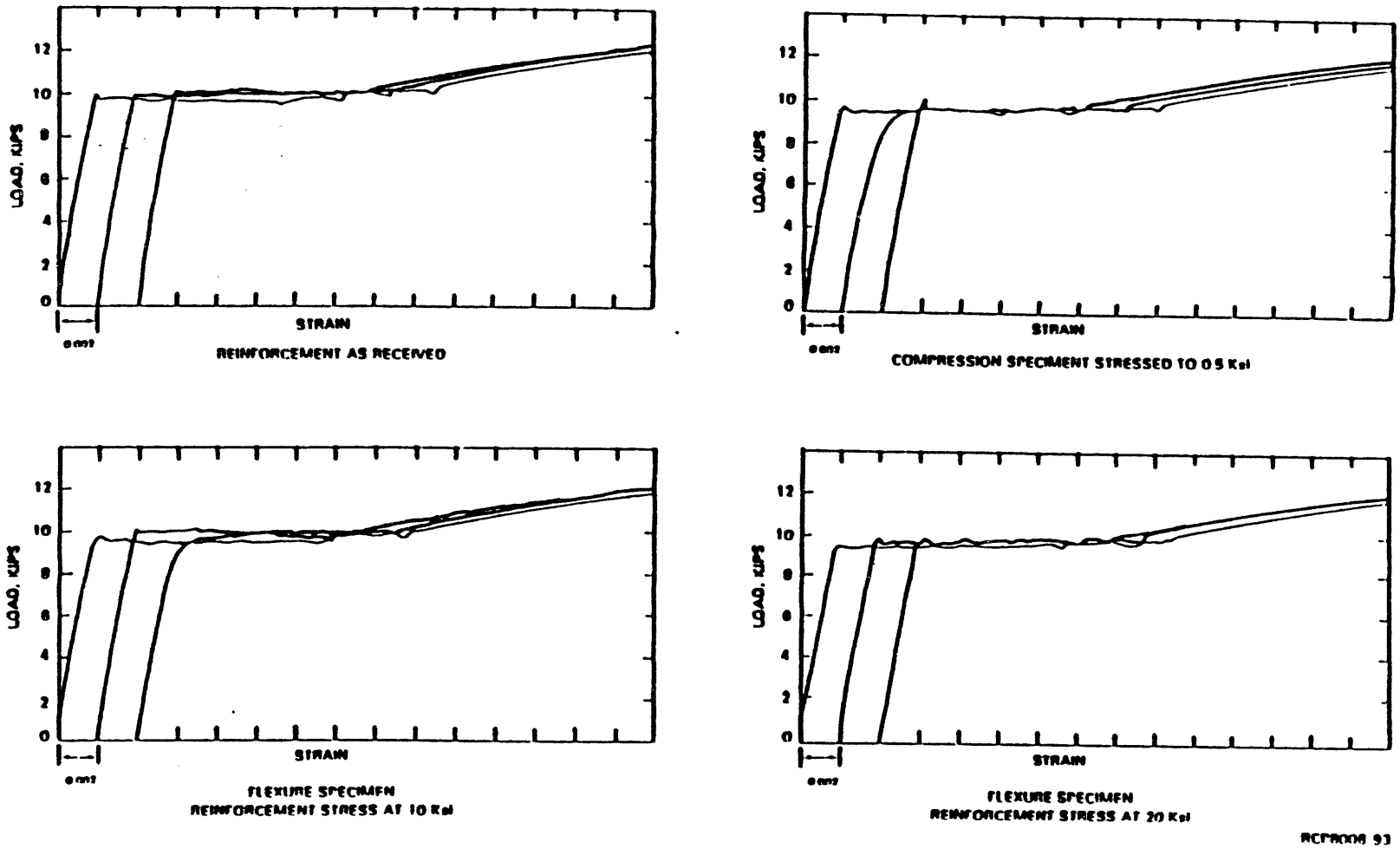

FIGURE 2 - Load Strain Relation of Reinforcement from Concrete Specimens Exposed 6 Months to Simulated Waste Solutions

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