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Nuclear Criticality Safety Assessment for Tank 38H Salt Dissolution

by

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NUCLEAR CRITICALITY SAFETY ASSESSMENT FOR TANK 38H SALT DISSOLUTION (U)

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1. Summary

This assessment report of sample results of the accumulating insoluble solids from Tank 38H demonstrates that an inherent subcritical condition for nuclear criticality safety exists during saltcake dissolution. This report also defines criteria for future sampling of Tank 38H for continued verification of the inherent subcritical condition as saltcake dissolution proceeds. Current Tank 38H sample results demonstrate subcritical conditions based on neutron poisons to fissile ratios. These ratios are summarized below,[1,2,3,4]

Single Element Neutron Poison

Sample No.	Na	Cr	Fe	Mn
Safe Ratio	150	66	76	30
HTF-003	3,700	310	630	1,100

2. Introduction

Distributed throughout the saltcake are insoluble solids containing fissile material. As the saltcake dissolves the insoluble solids can accumulate. Thus, dissolution of saltcake requires verification of subcritical conditions. The goal of the criticality safety program in HLW is to demonstrate an inherent subcritical condition exists to allow bulk saltcake dissolution to occur without the need to implement criticality safety controls. It is a requirement to demonstrate that the accumulating insoluble solids containing fissile material are inherently subcritical for all process conditions or implement criticality safety controls to prevent a criticality.[3]

Dissolution of saltcake occurs whenever saltcake is contacted by solution that is unsaturated in salts. Nitrite and nitrate compounds are the main saltcake constituents; therefore, if the solution contacting the saltcake is unsaturated in these salts, salt dissolution occurs. Salt dissolution from small volume additions (e.g., rain water additions, etc.) have considerable less potential than bulk salt dissolution to result in the accumulation of insoluble solids that were distributed throughout the saltcake.[5]

Bulk saltcake dissolution may occur due to the large volume of unsaturated salt solution such as Defense Waste Processing Facility (DWPF) recycle waste being fed into the 242-16H evaporator system. This system includes Tank 38H that is the focus of this assessment. Salt dissolution has intentionally been performed in Tank 41H, previously the 242-16H concentrate receipt tank, by a waste removal project. Data from insoluble material obtained during the initial Tank 41H salt dissolution has supported the inherent subcritical parameters for Tank 38H dissolution.[6]

Saltcake that has been formed as a result of operating the 242-16H evaporator is a concern for criticality safety for a number of reasons: 1) high enriched uranium and plutonium discards were received in the H-Area tank farm, 2) during evaporation the soluble uranium in solution can reach saturation conditions and crystallize from solution, and 3) large volume reductions of solution occurred in this evaporator system increasing the potential for uranium to have exceeded saturation. Plutonium is also present as fissile material and is accounted for as equivalent uranium.

It is also recognized that the insoluble material found in saltcake will be inherently subcritical for the following reasons: 1) uranium is not the only element that crystallizes from evaporate concentrate solution as an insoluble solid, 2) sludge solids are expected due to the fact sludge settling is not a requirement of the 242-16H evaporator system, and 3) most insoluble solids are neutron poisons or diluents.

This assessment report is provided to demonstrate the inherent criticality safety of Tank 38H solids that will accumulate during saltcake dissolution. The sample results continue to confirm that sufficient neutron poisons are associated with the fissile material contained in insoluble solids which accumulate during saltcake dissolution. This report also provides guidance for verification of subcritical conditions for continued dissolution of saltcake in Tank 38H.

3. Discussion

3.1 Tank 38H Salt Dissolution

Since the receipt of DWPF recycle, a very dilute salt solution, the feed to the 242-16H evaporator system has been lower in salt content. Therefore, the evaporator concentrate can be unsaturated and has the capacity to dissolve salt.

Initial saltcake dissolution in Tank 38H occurred when unsaturated solution contacted the saltcake that had formed on the cooling coils. Further saltcake dissolution will be hindered by the saturated salt solution above the saltcake. This is due to the fact that typical saturated salt solution has a higher specific gravity than the unsaturated salt solution. Photographs taken after decanting the concentrate from Tank 38H (Figure 1 and 2) demonstrate the changing conditions of the saltcake.

In Tank 38H, salt dissolution is limited by a number of factors: 1) salt dissolution occurs only if the unsaturated solution contacts the saltcake, 2) salts dissolution kinetics, and 3) required heat of salt dissolution (i.e., endothermic reaction).

3.2 Sampling Tank 38H Insoluble Solids

Sampling of insoluble solids in Tank 38H has been accomplished by use of a variable depth sampler (VDS) and a saltcake coring device. The variable depth sampler has an aluminum foil covering over the sample bottle mouth that dissolves away in the alkaline tank environment. To obtain a sample of the accumulating insoluble solids using the variable depth sampler, the sampler must be lowered to the desired elevation prior to the foil dissolving. The saltcake core sampler is a hollow tube designed to retain the saltcake when extracted.

The following is a discussion of each individual sample obtained from Tank 38H. Two VDS samples were obtained and the washed insoluble solids results are expected in the near future for use in this report. These two VDS samples represent in-situ conditions of solids accumulating above the saltcake. Current results of these samples indicate more than ample neutron poisons are present.[7] Additional washing of the solids is required to remove soluble compounds before this sample data is used in this report. Ample amounts of neutron poisons are expected to remain after washing.

Tank 38H VDS Sample HTF-001

Date: October 4, 1996 Riser: H Depth: 6" above saltcake

Current sample results represent the in-situ conditions in the tank where the insoluble solids are mixed with solids that may dissolve when contacted with unsaturated solution. Interstitial liquid has been removed from the solids but dissolution with inhibited water has not been completed. Once the washed insoluble solids results are complete, this report will be revised to include these samples.

Tank 38H Salt Core Sample HTF-003

Date: October 10, 1996 Riser: H Depth: Salt/Supernate Interface

The sampler was driven into the saltcake approximately 12 inches to obtain a sample of saltcake for analysis.[3,7] This sample has been repeatedly washed with inhibited water to remove soluble constituents.

Tank 38H VDS Sample HTF-004

Date: October 10, 1996 Riser: H Depth: Salt/Supernate Interface

Current sample results represent the in-situ conditions in the tank where the insoluble solids are mixed with solids that may dissolve when contacted with unsaturated solution. Interstitial liquid has been removed from the solids but dissolution with inhibited water has not been completed. Once the washed insoluble solids results are complete, this report will be revised to include these samples.

3.3 Tank 38H Insoluble Solids Analysis

The analysis of Tank 38H insoluble solids is performed by the Savannah River Technology Center (SRTC). The samples are highly radioactive and are delivered to the SRTC Shielded Cells in shielded doorstops. Within the Shielded Cells facility the samples are prepared for analysis. Preparation is required to: 1) prepare a subsample of the total "as received" sample, 2) wash soluble solids from the insoluble solids, and 3) prepare a subsample of the insoluble solids for analysis.

Additional details of the SRTC sample preparation and analysis effort are defined in a Test Plan[8] requested by a HLWE Technical Task Request.[9]

3.4 Tank 38H Insoluble Solids Analysis Results

Table 1 below lists the sample analysis results for the insoluble solids that will be used to demonstrate subcritical conditions exist in Tank 38H. Additional results may be found in the SRTC Technical Report describing the sample analyses and results.[7]

Table 1

Tank 38H Insoluble Solids Sample Results (wt.%)

Sample No.	Na	Cr	Fe	Mn	U_{eq}^{235}
HTF-003	17	1.4	2.9	5.0	0.0045

3.5 Insoluble Solids Sampling Frequency

Salt sample(s) (e.g., VDS, salt core sampler) will be required prior to the next decant from Tank 38H to confirm that adequate neutron poisons are present. Minimal salt dissolution is expected from the addition of supernate to Tank 38H because of the layering effect. The duration between Tank 38H decants is expected to be between 1 to 2 months depending on the variable evaporator concentrate drop rate and the scheduled direct receipt of DWPF recycle into Tank 38H.

Minimal salt dissolution is expected in Tank 38H with the present fixed jet configuration (i.e., 248 inches) and the supernate stratified layering behavior. All additions to Tank 38H are made through inlets at or above the operating fill limit of 362 inches. Depending on the specific gravity of the solutions and the current Tank 38H supernate specific gravity profile, supernate (or concentrate) additions will stratify and form a layer in the tank with respect to their specific gravity. The layering occurs because Tank 38H has no mechanical mixing. The only solution mixing that occurs within the tank is from diffusion, thermal gradients (e.g., hot concentrate), and the agitation from supernate addition and extraction.

Therefore, the supernate behavior can be described in Tank 38H based on the influent supernate characteristics. This behavior has been observed during the gradient density testing at SRTC.[10]

(1) when supernate additions have a low specific gravity and supernate is covering the saltcake, the saltcake will not be contacted by the low specific gravity supernate. Therefore, if the supernate addition is unsaturated, saltcake dissolution is expected to occur by two mechanisms: diffusion (slow) and thermal mixing.

(2) when supernate additions have a high specific gravity, the saltcake will be contacted by displacing lower specific gravity supernate. If the supernate addition is unsaturated, salt dissolution will occur.

(3) supernate additions will cause localized salt mining due to the contact with the salt layer if the supernate is unsaturated. This effect is significantly mitigated as the supernate level increases since the supernate layer prevents contact with the salt layer.

Additionally, salt dissolution will be impeded due to the current Tank 38H jet suction elevation (248 inches) and the maximum salt cake elevation (~254 inches) from video inspection. Initially after each decant, the saltcake will be exposed to the influent supernate additions. Salt dissolution will occur if the supernate is unsaturated. Unsaturated supernate will become saturated which will cause an increase in specific gravity. As additional supernate is added, it will follow the supernate behavior described above.

Eventually Tank 38H salt elevation will approach 248 inches and a saturated supernate layer is expected to remain over the salt cake after each decant. Salt dissolution below 248 inches is expected at a slow rate since the salt dissolution will be occurring from diffusion and thermal mixing. As mentioned previously, localized salt dissolution will occur below the drop point, so some salt dissolution may occur below the 248 inch elevation.

4.0 Subcritical Parameter Assessment

4.1 Single Element Neutron Poison Ratios

Listed below in Table 2 are the weight ratios of single element neutron poisons to fissile material identified in the insoluble material that accumulates in Tank 38H. The safe weight ratios[1] were established in the referenced Nuclear Criticality Safety Evaluations (NCSEs).[2,3,4] The nuclear criticality "safe ratio" is the amount of inherent neutron poisons to uranium that will maintain a mixture subcritical under all conditions; therefore, all weight ratios greater than the safe ratio are considered subcritical and safe.

Table 2

Tank 38H Single Element Weight Ratios to Fissile Material

Sample No.	$Na:U_{eq}$	$Cr:U_{eq}$	$Fe:U_{eq}$	$Mn:U_{eq}$
Safe Ratio	150	66	76	30
HTF-003	3700:1	310:1	630:1	1100:1

4.2 Combined Element Neutron Poison Ratios

Listed in Table 3 is the weight ratio of combined neutron poisons to fissile material identified in the insoluble material accumulating in Tank 38H. The safe weight ratio was established in the referenced Nuclear Criticality Safety Evaluation (NCSE).[4] The nuclear criticality "combined safe ratio" is the ratio of inherent neutron poisons to each other as compared to uranium. The combined ratio will maintain a mixture subcritical under all conditions; therefore, all weight ratios greater than the combined safe ratio are considered subcritical and safe.

Table 3

Tank 38H Combined Element Weight Ratio to Fissile Material

Sample No.	$Fe:Mn:U_{eq}$
Safe Ratio	20:20:1
HTF-003	630:1100:1

4.3 Density of Fissile Material

The maximum fissile material density is the weight percent fissile material of the insoluble solids times the maximum settled insoluble solids density. If the maximum settled solids density is not determined, then it is conservative to use the maximum crystalline density of the insoluble solids. Even the average crystalline density of the insoluble solids represents a conservative assumption. This is a conservative assumption for conditions in HLW because to achieve the crystalline densities, the insoluble solids would need to be dewatered and lattice voids filled.

The effective density of the initial salt dissolution cycle for Tank 38H is calculated by multiplying the total fissile material weight percent by the uranium density (3.93 g/cc) of uranium precipitate solids.[11] Thus, if the Tank 38H insoluble solids on the salt surface have an average density of 3.93 g/cc, then the uranium concentration could be derived by applying the effective weight percent of the uranium solids to the precipitated solids layer.

$$U_{eq}\ \text{Density}^{Tk\,38H} = 0.0045\ \text{wt\%}\ U_{eq} \cdot \frac{1\ \text{wt. fraction}}{100\ \text{wt.\%}} \cdot \frac{3.93\ \text{g}}{\text{cc}} \cdot \frac{1{,}000\ \text{cc}}{\text{liter}} = 0.18\frac{\text{g}}{\text{L}}U_{eq}$$

Thus it is shown that the density of Tank 38H precipitated solids (i.e., slurry) is below the aqueous solution ANSI/ANS 8.1 single parameter limit of 11.6 g/L. The factor of safety is the ratio of 11.6/0.18 or 64. This implies not only would the uranium bearing solids be required to settle to 3.93 g/cc, but the slurry would force the lattice void spaces to be filled 64 times yielding an incredible mixture density of 251 g/cc. This fissile mass density calculation is given strictly for comparison to the safety factor for a hydrogen-poisoned liquid system. Tank 38H is heavily poisoned by inherent neutron absorbers common to the saltcake formation process; therefore, the aqueous solution concentration of 11.6 g/L is conservative because of the inherent elemental (e.g., nitrogen) replacement of the aqueous hydrogen component. The subcritical reactivity of the tank under all scenarios is demonstrated by the neutron poison to fissile material weight ratios.[3]

5.0 Criteria for Future Salt Dissolution in Tank 38H

This section contains a discussion of expected conditions in Tank 38H and criteria for future sampling. The HLWE Criticality Safety Engineer is responsible for monitoring Tank 38H conditions. The HLW Criticality Safety Engineer has "Stop Work" authority if the technical basis cannot be verified.

October 23, 1996 Sampling Criteria

The next planned decant transfer has been evaluated and determined to be within the safety basis by sample HTF-003. The subsequent planned decant transfer (e.g., December 1996) will be the next assessment point. At the next assessment point, the HLWE Criticality Safety Engineer will determine specific actions that include sampling Tank 38H. Additional actions may be requested such as salt soundings and alternative sampling methods.

At this time, saltcake dissolution in Tank 38H is limited due to the current supernate level of 330 inches which limits the contact of potentially unsaturated salt solution with the saltcake (~254 inches).[12] However, the continued recycle of supernate out of Tank 38H and the continued receipt of fresh evaporator concentrate may dissolve saltcake.

Figure 1: Tank 38H Salt Profile on April 3, 1996

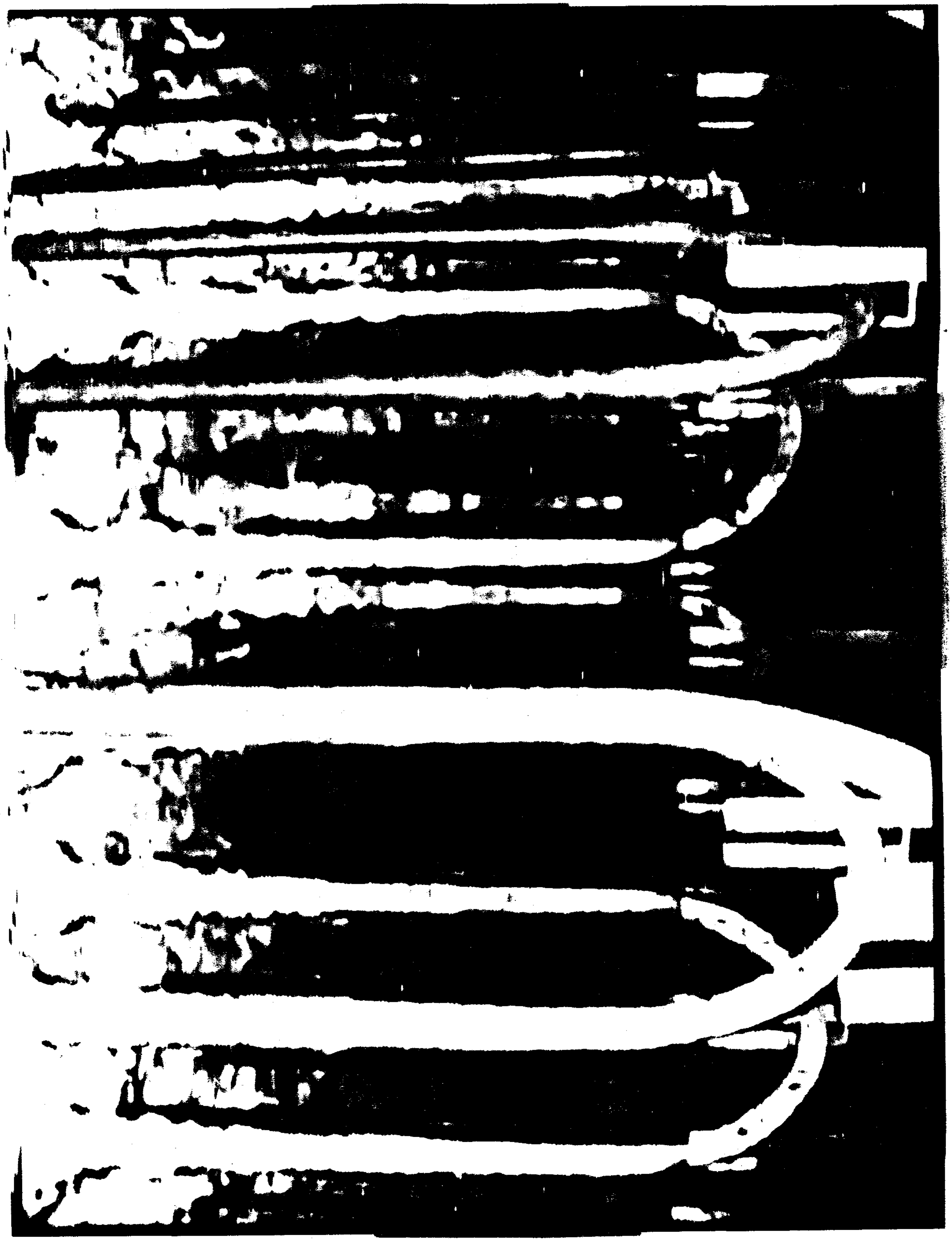

Figure 2: Tank 38H Salt Profile on August 6, 1996

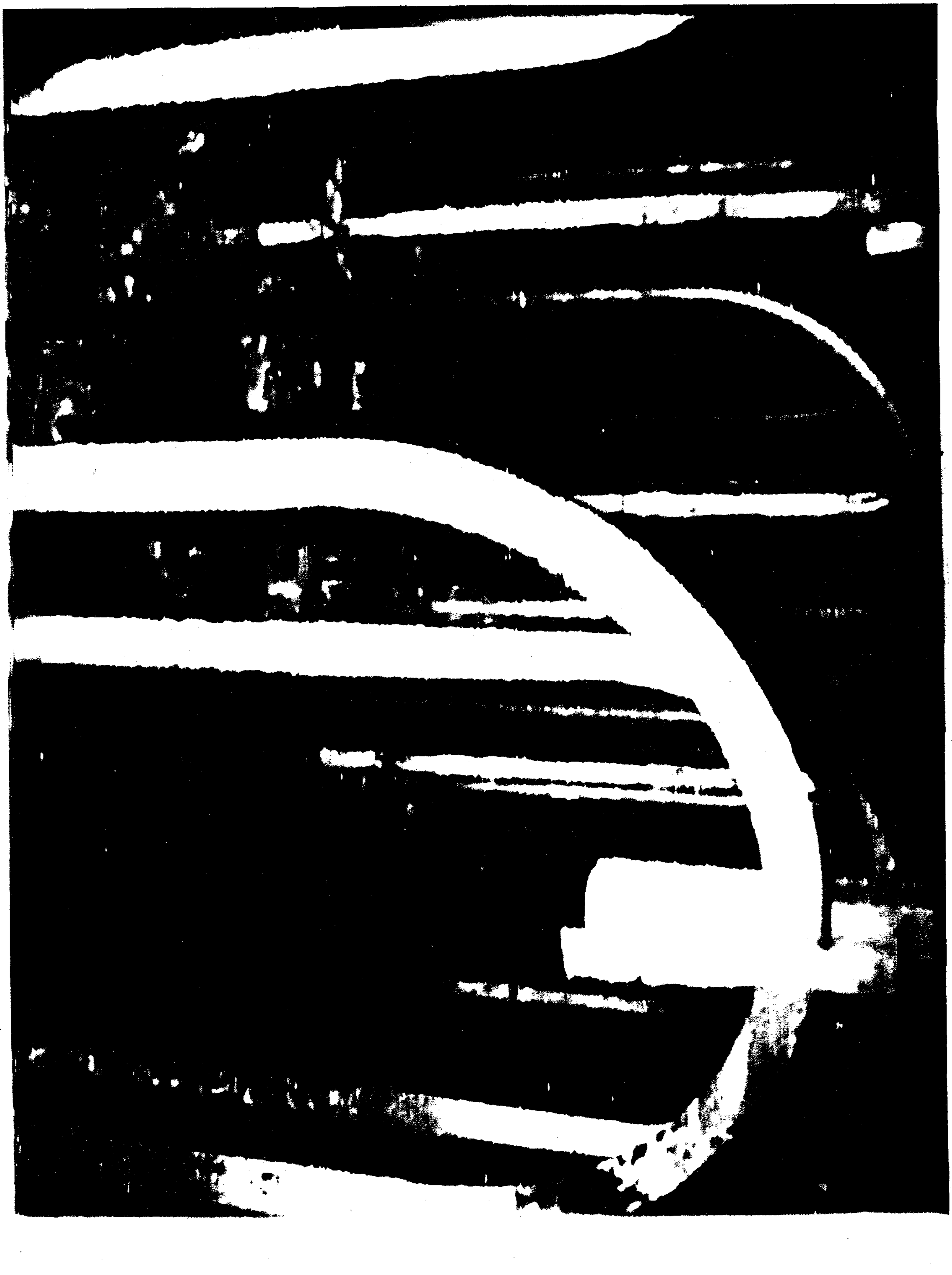

Figure 3: Tank 38H Salt Formation Close-up on August 6, 1996

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[1] WSRC-RP-96-0544, "Preliminary Analytical Results for Tank Farm Samples in Support of Salt Dissolution Evaluations (U)," D. T. Hobbs, Rev. 0, 10/21/96.

[2] EPD-CTG-960046, "Metal Poisons in Waste Tanks," T.G. Williamson, 7/31/96.

[3] N-NCS-H-00034, Nuclear Criticality Safety Evaluation - "Minimum Safe Ratios of Inherent Saltcake Metals to Uranium," R.H. Ross, 10/14/96.

[4] WER-WME-92-1143, Nuclear Criticality Safety Analysis WM-92-3 "Minimum Safe Ratios of Fe and Mn to U-235 in an Infinite System (U)," J. S. Clemmons, 9/18/92.

[5] WSRC-TR-93-0081, "Evaluation of Potential Accumulation of Uranium and/or Plutonium in the HLW Evaporator Systems (U)," M. C. Chandler, Rev. 0, 2/1/93.

[6] WSRC-TR-94-0057, "Final Report: Analysis of Tank 41H Saltcake Samples #2 and Comparison to Sample #1," D.T. Hobbs, 1/26/94.

[7] WSRC-RP-96-0544, "Preliminary Analytical Results for Tank Farm Samples in Support of Salt Dissolution Evaluations (U)," D. T. Hobbs, Rev. 0, 10/21/96.

[8] WSRC-RP-96-0526, "Technical Task Plan for the Analysis of Tank Farm Samples in Support of Salt Dissolution Evaluations (U)," D. T. Hobbs, Rev. 0, 10/13/96.

[9] HLE-TTR-97-003, "Tank 38 Salt Analysis to Determine Neutron Poisons," 10/9/96.

[10] WSRC-TR-96-0160, "An Investigation of Density Driven Salt Dissolution Techniques (U)," B. J. Weirsma, Rev. 0, 8/96.

[11] WSRC-RP-94-1213, "Characterization of the Tank 41H Saltcake Insoluble Solids," D. T. Hobbs, 10/31/94.

[12] HLW Morning Report, 10/22/96.

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