EPD-CTG-94-0037

Nuclear Criticality Safety Evaluation–DWPF Late Wash Facility, Salt Process Cell and Chemical Process Cell (U)

by

T. G. Williamson

Westinghouse Savannah River Company
Savannah River Site
Aiken, South Carolina 29808

DOE Contract No. **DE-AC09-89SR18035**

This paper was prepared in connection with work done under the above contract number with the U. S. Department of Energy. By acceptance of this paper, the publisher and/or recipient acknowledges the U. S. Government's right to retain a nonexclusive, royalty-free license in and to any copyright covering this paper, along with the right to reproduce and to authorize others to reproduce all or part of the copyrighted paper.

DISCLAIMER

DISCLAIMER

SAFETY ENGINEERING DEPARTMENT

CRITICALITY AND FIRE ANALYSIS SECTION

CRITICALITY TECHNOLOGY GROUP **EPD-CTG-940037**

October 17,1994

UNCLASSIFIED
DOES NOT CONTAIN UNCLASSIFIED
CONTROLLED NUCLEAR INFORMATION

KEYWORDS
Criticality
Defense Waste
DWPF

ADC &
REVIEWING
OFFICIAL JR Chandler
~~C. E. Apperson~~ JR Chandler

RETENTION
Permanent

Nuclear Criticality Safety Evaluation - DWPF Late Wash Facility, Salt Process Cell and Chemical Process Cell (U)

APPROVAL

Author CTG	T. G. Williamson — T. G. Williamson	10/17/94 Date
Technical Reviewer CTG	R. W. Rathbun — R. W. Rathbun	10/17/94 Date
Manager CTG	JR Chandler for CE Apperson — C. E. Apperson	10/24/94 Date

Distribution:

F. Beranek	730-B	A. F. McFarlane	730-B
C. E. Apperson	773-24A	J. F. Mincey	773-24A
R. W. Rathbun	773-786-1A	R. L. Reed	773-42A
B. C. Ha	773-A	L. M. Papouchado	773-A
E. F. Duhn	773-A	M. J. Plodinec	773-A
L. F. Landon	704-T	D. A. Crowley	704-1T
K. R. Yates	992W-2	H. H. Elder	704-S
A. G. Sarrack	992-1W	N. H. Kuehn	704-T
W. T. Goldsten	704-S	B. Mangiante	992-1W
J. R. Chandler	773-24A	J. F. Ortaldo	704-S
J. R. Harbor	773-43A	E. W. Holtzscheiter	773-A
C. T. Randall	704-T	S. A. Budenstein	704-1T
J. R. Fowler	704-Z	R. A. Jacobs	704-T
D. A. Sharp	992W-1	M. C. Chandler	703-H
J. W. Ray	704-35S	Tech.Information Man.	703-43A
CTG Files	773-24A		

Table of Contents

List of Figures

List of Tables

1.0 INTRODUCTION

The Savannah River Site (SRS) High Level Nuclear Waste will be vitrified in the Defense Waste Processing Facility (DWPF) for long term storage and disposal. This is a nuclear criticality safety evaluation for the Late Wash Facility (LWF), the Salt Processing Cell (SPC) and the Chemical Processing Cell (CPC) of the DWPF.

2.0 DESCRIPTION

Waste salt solution is processed in the Tank Farm In-Tank Precipitation (ITP) process and is then further washed in the DWPF Late Wash Facility (LWF) before it is fed to the DWPF Salt Processing Cell. In the Salt Processing Cell the precipitate slurry is processed in the Precipitate Reactor (PR) and the resultant Precipitate Hydrolysis Aqueous (PHA) product is combined with the sludge feed and frit in the DWPF Chemical Process Cell to produce a melter feed. The waste is finally immobilized in the Melt Cell. A simplified flow diagram is shown in figure 1. The processes included in this NCSE are enclosed by the dashed lines in figure 1.

Material in the Tank Farm and in the ITP and ESP processes have been shown to be safe against a nuclear criticality by others. The precipitate slurry feed from ITP and the first six batches of sludge feed are safe against a nuclear criticality and this evaluation demonstrates that the processes in the LWF, the SPC and the CPC do not alter the characteristics of the materials to compromise safety.

2.1 Late Wash Facility

In the Late Wash Facility (1) a batch of precipitate slurry is transferred from the Tank Farm into the Late Wash Precipitate Tank (LWPT). While the contents of LWPT are agitated, sufficient sodium tetraphenylborate (NaTPB) is added to the tank to reprecipitate the soluble cesium, potassium and ammonium and provide an excess of NaTPB. The slurry is circulated through the cross-flow filter to increase the content of insoluble solids while wash water containing NaTPB is added at a rate equivalent to the filtration rate. The filtrate is accumulated in the Late Wash Hold Tank (LWHT). The filtrate in the LWHT will be sparged of benzene, chemically adjusted, and returned to the H-Area Tank Farm. The washed precipitate slurry is

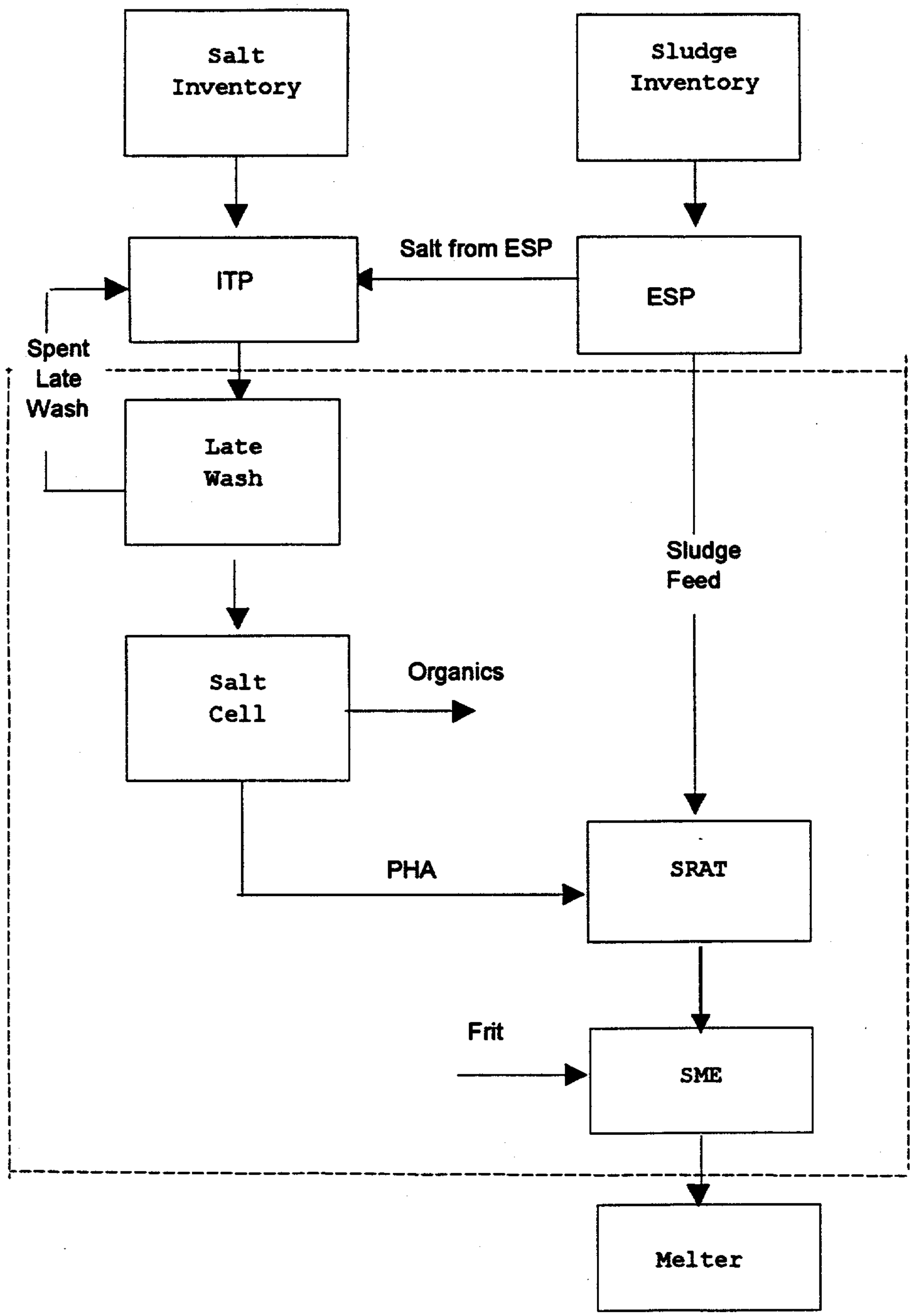

Figure 1

DWPF Processes

transferred to the Precipitate Reactor Feed Tank (PRFT) in the DWPF Salt Processing Cell (SPC).

The LWF contains two 6160-gallon stainless steel tanks, the Precipitate Tank (LWPT) and the Hold Tank (LWHT) each located in a concrete cell (2). In a separate cell is the Cross-Flow Filter which has a 30-gallon capacity (including piping) on the tube side and 120-gallon capacity on the shell side. Also there is a 550-gallon Surge Tank which contains sludge.

In the In Tank Precipitation process sodium tetraphenylborate (NaTPB) and monosodium titanate (MST), $NaTi_2O_5H$ are added for the removal of radioactive cesium and strontium. In this process the MST will also adsorb uranium and plutonium and mix in with the entrained sludge as insoluble solids. The fissile materials will remain adsorbed on the MST through the late wash process and through the Salt Processing Cell.

2.2 Salt Processing Cell

In the Salt Processing Cell the precipitate slurry from the LWF is first stored in the Precipitate Reactor Feed Tank (PRFT) and then processed in the Precipitate Reactor (PR). In the PR, tetraphenylborate is decomposed by thermal and copper-catalyzed acid hydrolysis reactions to produce benzene and an aqueous phase containing soluble sodium, potassium and cesium. The Precipitate Hydrolysis Aqueous (PHA) product is then fed to the Chemical Process Cell (CPC) where it is combined with the sludge feed.

2.3 Chemical Processing Cell

Alkaline sludge slurry is transferred from the Tank Farm Extended Sludge Processing (ESP) operation to the Sludge Receipt Tank (SRAT) in the Chemical Processing Cell. The sludge slurry is treated with nitric acid in the SRAT to control the flow properties of the slurry and to react with several of the components in the slurry, primarily nitrates and carbonates. The nitric acid requirement is calculated to achieve the final pH of 4.5-5.5 with the contribution of formic from the PHA.

To treat the slurry in the SRAT with acid, the slurry temperature is first raised to 85-95 °C. If required, the slurry volume is then reduced by evaporating water from the slurry. After the slurry volume has been adjusted by the addition of nitric acid, the temperature is raised to the boiling point (99-102°C) and PHA from the Precipitate Reactor Bottoms Tank (PRBT) is added to the acidified sludge at a controlled volumetric rate that equals the rate of condensate production. This method of feed control maximizes the free vapor volume within the SRAT and minimizes entrainment of solids into the condensate.

After addition of the PHA, the SRAT contents may be boiled at total reflux for a sufficient period to steam-strip mercury from the slurry. From the SRAT the material is sent to the Slurry Mix Evaporator (SME) where glass frit is added in preparation for the Melter.

3.0 REQUIREMENTS DOCUMENTATION

There are no specific documents which apply uniquely to this evaluation.

4.0 METHODOLOGY

This evaluation is based primarily on approved Nuclear Criticality Evaluations and national standards. Some computations were made with the HRXN and ANISN modules of the Joshua J70 system (3). HRXN computes the atom densities for mixtures and combines them with the Hansen-Roach 16-group nuclear cross sections to calculate infinite system multiplication factors and to prepare cross sections for transport codes. The ANISN module is a one-dimensional discrete-ordinates transport code. The Joshua J70 system and Hansen-Roach cross sections in HRXN are well accepted for criticality safety analyses.

5.0 CONTINGENCY ANALYSIS

Material for the Late Wash Facility, the Salt Processing Cell and the Chemical Processing Cell comes from the Savannah River Site Tank farms and is first processed through the In Tank Precipitation process and Extended Sludge Process. The ITP and ESP have been shown to be safe against a nuclear criticality and to have wide margins of safety. The primary nuclear criticality defense in these processes is the low concentration of fissile materials and the high abundance of neutron absorbers in the salt solution and the sludge.

The basic characteristics of the precipitate slurry and the sludge feeds which provide margins of safety are not altered in the LWF, the Salt Processing Cell or the Chemical Processing Cell. The fissile materials either remain with the insoluble neutron absorbers or are dissolved into solution. In either case the ratio of fissile material to absorber (MST, iron or manganese) in the insoluble portion or the ratio of hydrogen-to-fissile material in the soluble portion far exceeds safe values so that criticality cannot occur.

A group of persons knowledgeable in the DWPF processes identified several upset conditions which might lead to criticality problems. In this assessment these upset conditions are shown to be safe against a nuclear criticality with a safety factor of at least five for all materials expected in the first six batches of material.

Because the feed materials to the LWF, the Salt Processing Cell and the Chemical Processing Cell have been shown to be safe with a wide margin of safety and that criticality is deemed incredible for these materials, and because the processes in these facilities do not significantly reduce the margins of safety, criticality is also incredible in the LWF, the Salt Processing Cell and the Chemical Processing Cell. Thus a double contingency analysis is not required.

6.0 EVALUATION AND RESULTS

The processes considered in this evaluation (4) are those enclosed in the dashed line in Figure 1, that is the LWF, the Salt Processing Cell and the Chemical Processing Cell (SRAT and SME). The precipitate slurry from ITP is shown to be safe against a nuclear criticality by others with a wide range of safety because of the neutron absorbing properties of monosodium titanate and the low concentration of entrained sludge. In this evaluation it is shown that these margins of safety are not compromised in the LWF and Salt Processing Cell.

The first six batches of sludge feed from the ESP are safe against a nuclear criticality because of the neutron absorbing properties of iron and manganese and because of the low concentration of fissile materials and low enrichment. In this evaluation it is demonstrated that these margins of safety are not compromised in the Chemical Processing Cell.

A group of persons knowledgeable in the DWPF processes has considered possible criticality problems in the DWPF (5). The assumptions in the assessment were:

> The slurry feeds are inherently safe thus, unless a mechanism exists to separate fissile materials from bulk solids (precipitate and sludge), nuclear criticality is impossible. Process areas in which chemical and physical interactions might separate fissile material from the solids are considered.
>
> No oxalic acid will be used in the DWPF process cells. If oxalic acid is introduced, its effects must be documented and analyzed.
>
> There is no isotopic separation mechanism in the DWPF processes.

The group concluded the following regarding the individual components of the DWPF:

For the Late Wash Facility an assessment will be made of the effect of filtration on the separation and accumulation of fissile materials.

For the Salt Processing Cell the group concluded that experimental data is needed to establish if uranium and plutonium is dissolved or remains with the insoluble solids during the hydrolysis reactions. If the uranium and plutonium are in solution, then there is no nuclear safety concern because the concentrations are very low. The following upset conditions which could occur if the fissile materials desorb from the MST are considered:

> Fouling of the cooling coils by tars and accumulation of fissile materials on the coils. For fouling analysis the total amount of fissile material in four batches will be assumed. This value is based on previous experience.
>
> Accumulation of fissile material due to loss of agitation. In this case fissile material separates from MST and preferentially settles. The analysis is to be based on the processing of two batches as loss of agitation is an upset which would be detected before processing of more than two batches is completed.

Accumulation of fissile material in the sump, the condenser/decanter or the organic evaporator. Two years worth of precipitate hydrolysis feed which is approximately equivalent to a sludge batch will be used as an upper bound. These accumulations are of concern for those fissile materials which become soluble.

Two nuclear safety issues identified by the review group for the Chemical Process Cell are:

Plutonium oxide may be dissolved in mercury and accumulate in the SRAT mercury trap.

There may be fouling and accumulation of fissile materials on the cooling coils of the SRAT.

These issues are addressed in the evaluation. Other issues related to the melter are addressed in a separate NCSE (6).

Sludge from the Tank Farms which is expected in the first six batches (approximately 20 years of DWPF operation) is discussed in Appendix C. In this evaluation material balance tables generated by the integrated High Level Waste flowsheet model are used to estimate material contents for the various processes. The flowsheet model includes uranium and plutonium but does not distinguish the isotopic content. In some cases 5% of the uranium content is used to estimate the ^{235}U content and a Pu/U ratio of 0.1 to estimate the ^{239}Pu content. This is done to impart a conservative factor of at least five to the analysis.

6.1 Late Wash and Salt Processing Cell

Feed to the LWF comes from the ITP process where sodium tetraphenylborate (NaTPB) and monosodium titanate (MST), $NaTi_2O_5H$, are added for the removal of radioactive cesium and strontium. In this process the MST will adsorb strontium, uranium and plutonium and remain as an insoluble solid. This precipitate slurry is washed, filtered and transferred to the LWF.

There are two streams of fissile material entering the process in the precipitate feeds. These are 1. fissile material adsorbed on the monosodium titanate (MST), and fissile material in the entrained insoluble sludge solids of the salt solution. The entrained insoluble solids are limited to 400 mg/l by the process requirement of the In-

Tank Precipitation process, because of consideration of the cross flow filters operating characteristics (7).

Goslen (8) and Goslen and Bess (9,10) have analyzed uranium and plutonium mixed with MST and have determined ratios for which the bias-adjusted multiplication factor for an optimally moderated water-reflected sphere remains less than 0.95. In these analyses they used the 16-group Hansen Roach cross-section set with the Joshua criticality modules HRXN and ANISN(3).

Chandler (11) demonstrated the safety of fissile materials adsorbed on MST based on minimum safe-weight ratios of MST to fissionable materials computed by Goslen and Bess and the maximum adsorption capacity of MST measured by Hobbs (12). Bess (13) combines these conditions with canyon facility data to evaluate the potential for criticality in the ITP process due to the adsorption of fissile material from solution. He used as a bounding condition only the waste from the H area High Heat Waste Process, which involved plutonium and enriched uranium. This report further demonstrates that the potential for criticality in the ITP process due to adsorption of fissionable materials by MST is not credible.

In the Late Wash Facility sodium tetraphenylborate (NaTPB) is added to reprecipitate the soluble cesium, potassium and ammonium and provide an excess of NaTPB. The slurry is circulated through the cross-flow filter to increase the content of insoluble solids while wash water containing NaTPB is added. The filtrate is accumulated in the Late Wash Hold Tank (LWHT) where it is sparged of benzene, chemically adjusted, and returned to the H-Area Tank Farm. The washed precipitate slurry is transferred to the Precipitate Reactor Feed Tank (PRFT) in the DWPF Salt Processing Cell (SPC). There are no chemical mechanisms in the LWF processes to separate the fissile material from the MST (14) so the system remains safe. If the fissile materials were to separate from the insoluble portion and go into solution, the hydrogen-to-fissile ratio is high enough to maintain criticality safety.

The cross flow filter is a sintered metal filter rated at nominal pore size 0.5 micron and fabricated from stainless steel (2). There is no physical mechanism in this process to separate the fissile materials from the nonfissile ones. The filter system includes backpulsing capability which is used to mitigate the accumulation of solids in the filter.

In appendix A the processes in the LWF and SPC are analyzed for material with characteristics expected in the first salt solution feed. The material balance tables used in this analysis are generated by the integrated HLW flowsheet model (15,16) which incorporates the chemical and physical processes in the DWPF. In addition to the MST-to-fissile ratio, there are other parameters which provide a margin of safety. These include the high hydrogen-to-fissile ratio, the low fissile mass which will be in any of the tanks, and the low enrichment. In Appendix A it is shown that this material is safe because of the H/F ratio and the fissile mass with a factor of at least five margin. Another factor, which provides an additional margin but which is not included in the analysis, is the presence of boron which is a good neutron absorber.

It is demonstrated in Appendix A that fissile material accumulating on the heating and cooling coils cannot lead to an unsafe situation. It is also demonstrated that, if 35% of the fissile material were to separate from the MST and accumulate in an optimum condition for criticality, the uranium from a single batch would not exceed a multiplication factor of 0.92. In these considerations an enrichment of 5% is used to estimate the fissile mass. This provides a safety margin of a factor of five compared to the materials which will be processed in the DWPF.

In summary, material processed in the LWF and the Salt Processing Cell will remain safe from a nuclear criticality because the fissile materials remain bound to the MST in a safe ratio. If the fissile nuclides were to separate from the insoluble materials and become soluble the high hydrogen-to-fissile ratio will keep the mixture safe. Also for the materials to be processed in the DWPF the fissile material content is low enough that a critical mass will not accumulate in tanks of the LWF or the SPC. Other neutron absorbers which are prevalent will also mitigate against a nuclear criticality.

6.2 Chemical Processing Cell

Alkaline sludge slurry is transferred from the Tank Farm Extended Sludge Processing (ESP) operation to the Sludge Receipt Tank (SRAT) in the CPC. The sludge slurry is treated with acid in the SRAT by adjusting the slurry volume and raising the temperature to the boiling point (99-102°C). PHA from the Precipitate Reactor Bottoms Tank (PRBT) is added to the acidified sludge at a controlled volumetric rate that equals the rate of condensate production. After addition of the PHA, the SRAT contents may be boiled at total reflux for a sufficient period to steam-strip mercury from the slurry.

In the Slurry Mix Evaporator (SME) the sludge feed is dewatered and frit is added to make glass for feed to the melter. Clark (17) has evaluated the SME and has shown it to be safe for some upset conditions.

Selected materials for the first six sludge batches are listed in Appendix C.

The feed from the Extended Sludge Process to the SRAT is critically safe because of the safe weight ratios of iron and manganese to fissile materials. Clemmons and Goslen (18, 19) analyzed systems containing wet and dry mixtures of iron and manganese with the fissile nuclides ^{235}U and ^{239}Pu and determined mass ratios for which the computed value of the infinite multiplication factor did not exceed a bias adjusted value of 0.95. In these analyses they used the 16-energy-group Hansen Roach cross-section set with the Joshua criticality module HRXN (3). The most reactive system is the dry system and the addition of small amounts of water (H/F <10) drives the multiplication factor to lower values.

In the SRAT the sludge is acidified and heated to make the sludge properties more suitable for vitrification. Coleman, et al. (20) have measured the effects of adding formic and nitric acids to radioactive SRS sludge under conditions that simulated acid and temperature conditions of the SRAT. As the pH decreases from 7, iron shows no measurable solubility until the pH reaches 3.8. Less than 5% of the iron will dissolve when the SRAT is operated in the pH range 4.5-5.5. Manganese exhibits characteristics intermediate between sparingly soluble iron and soluble sodium. About 40% of the manganese in the sludge is dissolved in the pH range 4.5-5.5 and this solubility does not increase when pH 3.0 is reached. Thus, sludge that is safe in the ESP because of the iron-to-fissile ratio is also safe in the Chemical Processing Cell. Sludge that is safe in the tank farm because of the manganese-to-fissile ratio is safe if the safety margin is greater than two. As shown in Appendix C this includes the material from eight tanks. In addition there are 24 tanks for which the contents are safe by a combination of iron and manganese even if both the iron and manganese contents are reduced by a factor of two.

In appendix B the processes in the SRAT are analyzed for material with characteristic expected in the first batch. The material balance tables used in this analysis are generated by the integrated HLW flowsheet model (15,16) which incorporates the chemical and physical processes in the DWPF. The material in this flowsheet is characteristic of the material in Batch One feed, which is a mixture of material from Tanks 42 and 51, and contains uranium enriched to less than 1% ^{235}U.

In summary the material in the Chemical Processing Cell is safe because the neutron absorbers, iron and manganese remain in the insoluble fraction. If the fissile materials remain insoluble, the ratios of iron and manganese to fissile nuclides is sufficient to maintain safety. If the fissile materials do not remain with the insoluble fraction then the hydrogen-to-fissile ratio is adequate to maintain criticality safety. Also it is shown that caking of the fissile material on the cooling and heating coils cannot lead to a criticality incident.

7.0 DESIGN FEATURES AND ADMINISTRATIVELY CONTROLLED LIMITS AND REQUIREMENTS

No specific design requirements or administratively controlled limits are required.

8.0 SUMMARY AND CONCLUSIONS

Material for the Late Wash Facility, the Salt Processing Cell and the Chemical Processing Cell comes from the Savannah River Site Tank farms and is first processed through the In Tank Precipitation process and Extended Sludge Process. The ITP and ESP have been shown to be safe against a nuclear criticality and to have wide margins of safety. The primary nuclear criticality defense in these processes is the low concentration of fissile materials and the high abundance of neutron absorbers in the salt solution and the sludge.

The basic characteristics of the precipitate slurry and the sludge feeds which provide margins of safety are not altered in the LWF, the Salt Processing Cell or the Chemical Processing Cell. The fissile materials either remain with the insoluble neutron absorbers or are dissolved into solution. In either case the ratio of fissile material to absorber (MST, iron or manganese) in the insoluble portion or the ratio of hydrogen-to-fissile material in the soluble portion far exceeds safe values so that criticality cannot occur.

Several upset conditions are shown to be safe against a nuclear criticality with a safety factor of at least five for all materials expected in the first six batches of material.

9.0 REFERENCES

1. WSRC-RP-92-793 (Rev. 4) "Technical Bases -DWPF Late Washing Facility (U)".

2. S. A. Edwards, M. H. Bailey, "Hazards Assessment Document, Defense Waste Processing Facility, Late Wash Facility", WSRC-TR-93-649 (Rev. 1), July, 1994.

3. H. K. Clark, "Joshua Nuclear Criticality Safety Modules" DPSTM-86-700-3, 1986.

4. T. G. Williamson, "Criticality Safety Analysis - Late Wash Facility, Salt Processing Cell and Chemical Processing Cell", Calc. Note N-CLC-A-0024, 1994.

5. B. C. Ha, "Assessment of Nuclear Safety and Nuclear Criticality Potential in the Defense Waste Processing Facility (U)", WSRC-RP-93-692, Rev. 0, 1993.

6. T. G. Williamson, "Nuclear Criticality Safety Evaluation DWPF Melter - Batch 1(U), SRT-CMA-930039 (1993).

7. D. D. Walker, "Process Requirements for the In-Tank Precipitation", SRT-LWP-92-142 (1992).

8. A. Q. Goslen, "Nuclear Criticality Safety Analysis WM-92-1, Maximum Safe Loading of U-235 Monosodium Dititanate, Rev.1 (U)", WMR-WMT-920788, 1992.

9. A. Q. Goslen and C. E. Bess, "Nuclear Criticality Safety Analysis WM-92-2, Maximum Safe Masses at Various U-235 Loadings on Monosodium Dititanate, Rev.1 (U)", WMR-WMT-920979, 1992.

10. A. Q. Goslen and C. E. Bess, "Nuclear Criticality Safety Analysis WM-93-1, Determination of the Safe Accumulated MST Mass Additions in the In Tank Precipitation Process (U)", WER-HLE-930558, 1993.

11. M. C. Chandler, "Nuclear Criticality Safety Bounding Analysis for the In-Tank-Precipitation (ITP) Process (U)", WSRC-TR-93-171, 1993.

12. D. T. Hobbs, "Fissile Solubility and Monosodium Titanate Loading Tests (U)" WSRC-RP-92-1273, 1993.

13. C. E. Bess, "Nuclear Criticality Safety, Bounding Analysis for the In-Tank-Precipitation (ITP) Process, Impacted by Fissile Isotopic Weight Fractions (U), WSRC-TR-94-004 Rev. 0, 1994.

14. B. C. Ha, "Formic Acid Boiling Experiments of Monosodium Titanate Containing Adsorbed Depleted Uranium", WSRC-RP-94-1133, 1994.

15. A. S. Choi, J. R. Fowler, J. R. Edwards Jr., and C. T. Randall, "High Level Nuclear Waste Treatment in the Defense Waste Processing Facility - Overview and Integrated Flowsheet Model (U)", WSRC-MS-91-401, 1991.

16. A. S. Choi, "Material Balance Tables for the DWPF Coupled Feed Flowsheet with Batch One Sludge (U)", WSRC-TR-93-203 Rev. 0, 1993.

17. H. K. Clark, "Defense Waste Processing Facility Nuclear Safety Assessment", DPST-82-965, Nov. 1982.

18. J. S. Clemmons and A. Q. Goslen, "Nuclear Criticality Safety Analysis WM-92-3, Minimum Safe Ratios of Fe and Mn To U-235 In An Infinite System (U)", WER-WME-921143, 1992.

19. J. S. Clemmons and A. Q. Goslen, "Nuclear Criticality Safety Analysis WM-92-5, Minimum Safe Ratios of Fe and Mn To Pu-239 In An Infinite System (U)", WER-HLE-921353, 1992.

20. C. J. Coleman, N. E. Bibler, D. M. Ferrara and S. F. Siegwald, "Reactions of Formic and Nitric Acids with Savannah River Site Radioactive HLW Sludges in the DWPF Pretreatment Steps" WM'94, Proc. of the Symposium on Waste Management, 1994.

21. ANSI/ANS 8.1 1983, "American National Standard for Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors", 1983.

22. Drawing W 754253 Rev 2 "Process Vessels - PR & PRBT Equipment Requirement Drawing"

23. WSRC-TR-92-340-2 (Rev.5), "Functional Performance Requirement, DWPF Late Wash Facility", July 1994.

24. R. D. Carter, G. R. Keil and K. R. Ridgway, "Criticality Handbook, ARH-600, Atlantic Richfield Hanford Co., 1969.

25. Drawing W 752193, Rev 5 "Tank Assembly - SME/SRAT/MFT/DWG Equipment Requirement Drawing", 1985.

26. Drawing W 752197, Rev 1 "Coil Assembly - SME/SRAT Equipment Requirement Drawing". 1984.

27. B. C. Ha, N. E. Bibler, D. M. Ferrara, "Plutonium in the Mercury of the SRAT Product", SRT-GTC-93-0061, 1993.

28. WSRC-TR-94-0191, "Estimated Sludge Inventory For Individual Tanks", May 1994.

29. HLW-OVP-94-0077 "HLW System Plan Revision 3 (U), May 1994.

30. J. S. Clemmons, "Assessment of Criticality Safety Parameters Based on Estimated Waste Tank Sludge Contents" HLW-HLE-94-0388, 1994.

APPENDIX A LATE WASH FACILITY AND SALT PROCESSING CELL

A-1 REPRESENTATIVE FEED

The primary defense against criticality in the LWF and the SPC is the fact that the fissile materials will remain with the insoluble portion of the slurry and the combination of fissile materials and MST cannot become critical. In this appendix it is demonstrated that there are other safety factors which mitigate against a nuclear criticality. These factors are the high hydrogen-to-fissile atomic ratios, the low fissile mass, and the abundance of poisons, such as boron and iron. The question of accumulation of fissile materials on the PR cooling coils and the question of settling and agitation of fissile materials will also be addressed.

There are two streams of fissile material entering the process in the precipitate feeds. These are 1. fissile material adsorbed on the monosodium titanate (MST), and 2. fissile material in the entrained insoluble sludge solids of the salt solution. The entrained insoluble solids are limited to 400 mg/l by the process requirement of the In-Tank Precipitation process, because of consideration of the cross flow filters operating characteristics. (7)

The material balance tables for LWF feed (1) are used in this analysis as a representative material balance for the DWPF coupled feed processes. These balance tables are generated by the integrated HLW flowsheet model (13) and have been reported by Choi (16). The data used in this analysis from reference 1 have higher uranium and plutonium contents than Choi's data.

Material destined for DWPF can be divided into two classes, material with larger amounts of uranium but with low ^{235}U content (< 1.0 wt% ^{235}U) and material with lesser amounts of fissile material but higher enrichment (see Appendix C). This analysis is used to indicate margins of safety for several parameters. In many cases an enrichment is used to compute an equivalent ^{235}U content to demonstrate a margin of safety.

The contents of streams in the LWF containing appreciable fissile materials are summarized in Table A-1. Stream 418 is the feed to the LWF and is composed of the soluble and insoluble components shown in streams 655 and 656. Stream 201 is the material passed from the LWF to the Salt

Processing Cell and streams 661 and 419 are representative of the LWF internal heel and slurry contents.

Table A-1
LWF Material Contents

	Stream 655	Stream 656	Stream 418	Stream 201	Stream 661	Stream 419
	Soluble Feed	Insoluble Feed	Sol. and Insol.	From LWF to SPC	Heel	Slurry
Stream lb/hr	372.2	37.73	409.9	463.8	60.31	517.1
Water lb/hr	357.5	0	357.5	415.5	54.03	451.2
Water Hydrate	0	0.1982	0.1982	0.1982	0.02578	0.224
Al_2O_3 lb/hr	0	0.131	0.131	0.131	0.01704	0.148
CsTPB lb/hr	0	0.4199	0.4199	0.4826	0.06275	0.5453
Fe_2O_3 lb/hr	0	0.3702	0.3702	0.3702	0.04813	0.4183
KTPB lb/hr	0	33.26	33.26	38.22	4.97	43.19
MnO_3 lb/hr	0	0.08603	0.08603	0.08603	0.01119	0.09722
NH_4TPB lb/hr	0	0	0	3.623	0.4711	4.094
Na_2CO_3 lb/hr	5.226	0	5.226	0.4232	0.05504	4.345
$NaNO_2$ lb/hr	3.55	0	3.55	0.2875	0.03738	2.951
$NaNO_3$ lb/hr	0.06895	0	0.06895	0.005584	0.0007262	0.05733
$NaTi_2O_5H$ lb/hr	0	0.8493	0.8493	0.8493	0.1104	0.9597
PuO_2 lb/hr	0	3.781E-04	3.781E-04	3.781E-04	4.917E-05	4.273E-04
$PuO_2(NaTi_2O_5H)_2$	0	2.262E-04	2.262E-04	2.262E-04	2.942E-05	2.556E-04
U_3O_8 lb/hr	0	0.2153	0.2153	0.2153	0.028	0.2433
Sum lb/hr	366.3	35.5	401.9	460.4	59.9	508.5
% of Stream	98.4	94.2	98.0	99.3	99.3	98.3
Density lb/ft^3	63.84	70.11	64.37	63.16	63.16	63.96
Density g/cc	1.024	1.124	1.032	1.013	1.013	1.025

The row showing the percentage of the stream is the percentage of the material in the stream which is represented by these selected compounds. With the exception of the insoluble components, more than 98% of the mass is included with these compounds. The missing compounds in the insoluble stream are primarily organic salts.

These processes show that the uranium and plutonium remains in the insoluble portion with the monosodium titanate (MST), the potassium tetraphenylborate (KTPB), and with the iron and manganese oxides. The mass ratios of uranium and plutonium to one another and to hydrogen, iron, manganese, boron and MST, all of which are effective neutron poisons, are in Table A-2.

Table A-2

Mass Ratios

	Stream 656	Stream 418	Stream 201	Stream 661	Stream 419
	Insoluble	Sol+Insol	From LWF	Heel	Slurry
Uranium lb/hr	0.1822	0.1822	0.1822	0.0237	0.2059
Plutonium lb/hr	0.0004	0.0004	0.0004	0.0001	0.0005
Iron lb/hr	0.2589	0.2589	0.2589	0.0337	0.2926
Manganese lb/hr	0.0544	0.0544	0.0544	0.0071	0.0614
Boron lb/hr	1.0035	1.0035	1.1531	0.1499	1.3031
MST lb/hr	0.8493	0.8493	0.8493	0.1104	0.9597
Uranium wt frac	4.83E-03	4.45E-04	3.93E-04	3.93E-04	3.98E-04
Plutonium wt frac	1.10E-05	1.01E-06	8.93E-07	8.93E-07	9.05E-07
MST wt frac	0.0225	0.0021	0.0018	0.0018	0.0019
Pu/U	0.0023	0.0023	0.0023	0.0023	0.0023
Fe/U	1.42	1.42	1.42	1.42	1.42
Mn/U	0.30	0.30	0.30	0.30	0.30
Fe/Pu	625	625	625	625	625
Mn/Pu	131	131	131	131	131
B/U	6	6	6	6	6
B/Pu	2423	2423	2785	2784	2784
U/MST	0.2146	0.2146	0.2146	0.2147	0.2146
Pu/MST	0.0005	0.0005	0.0005	0.0005	0.0005
H/F	28	51107	59394	59388	57076

The data in this table further indicate that the ratios of the fissile materials to one another and to the neutron poisons, MST, Fe and Mn, are not changed in the LWF processes and the boron ratios are not changed significantly. For this table only boron with the KTPB is included and the boron in CsTPB, NH_3TPB and NaTPB is ignored.

For this system the uranium-to-MST mass ratio is 21.46%. If the enrichment of ^{235}U is less than 5% (less than 2.2 X 10^{-5} weight fraction ^{235}U in the feed) the ratio ^{235}U:MST is 1.07%. Goslen and Bess (10) have demonstrated an equivalency between uranium and plutonium on MST which is proportional to the MST mass and plutonium weight fraction. In this case the plutonium adds 0.05% to the uranium loading making the equivalent ^{235}U:MST 1.12%. The safe weight loading for an infinite system is 1.96 wt% ^{235}U on MST. Thus for a feed stream similar to Batch One with less than 5% ^{235}U in uranium the MST is a sufficient poison to maintain the system subcritical for this feed material. This confirms that the feed material is safe because of the neutron absorption properties of MST with a safety factor of at least 5.

In the LWF feed stream the hydrogen-to-uranium ratio exceeds 50,000. The safe hydrogen-to-fissile ratio for ^{235}U is 2250 and for ^{239}Pu is 3630 (21). Thus, if some or all of the fissile materials were to leave the insoluble MST and form soluble compounds there would no criticality problem because of the high hydrogen-to-fissile ratio.

The LWF is capable of supplying washed precipitate at a rate of one batch per each 61 hours (1). The nominal batch size is about 3,266 gallons, which consists of 2,883 gallons precipitate transferred from Tank 49H, 383 gallons of sodium tetraphenylborate wash and 650 gallons heel. The feed and the heel contain fissile materials. For these calculations a 3000-gallon feed batch will be combined with a 650 gallon heel.

In the Salt Processing Cell the PR tank is a large tank with an internal agitator and sets of heating and cooling coils. Dimensions and volumes are listed in Table A-3 (22)

Table A-3

PR Tank Dimensions

	Inch	Cm
Diameter to inner coil center line	97	246
Diameter to outer coil center line	114.63	291
Clearance from outer coil to tank wall	1.5	3.81
High liquid level	160	406

Tank Volumes	Gallons	Liters
Inside coils	5118	19370
Tank wall	7835	29660

For this analysis 7800 gallons, the approximate volume to the tank wall was used with no account taken for the reduced volume caused by the agitator and the coils.

In the PR, tetraphenylborate is decomposed by thermal and copper-catalyzed acid hydrolysis reactions to produce benzene and an aqueous phase containing soluble sodium, potassium and cesium. These processes do not remove the fissile materials from the MST (14). Even if the fissile materials were removed from the insoluble components and became soluble, they would be in solution and be in a safe condition because of the large amount of hydrogen in the tank.

The volumes and contents of a 3000 gallon batch, of the heel, of both the shell and tube sides of the filter, of the insoluble part of each batch, of a full LWPT Tank (23), and or the PR Tank are listed in Table A-4. In this table it assumed that the filter is completely plugged with the insoluble part of the mixture.

Table A-4

Single Batch Content

	One Batch	Heel	Filter	Insoluble	LWF	PR Tank
Vol. gal	3000	650	150	254	6160	7800
Vol. liters	11355	2460	568	960	23318	29523
Density g/cc	1.032	1.013	1.121	1.121	1.0321	1.0127
Mass kg	11719	2491	638	1079	24063	29897
Uranium g	5210	979	3082	5210	10697	11746
Plutonium g	11.84	2.22	7.00	11.84	24.31	26.69
Uranium g/l	0.4588	0.3979	5.4290	5.4290	0.4588	0.3979
Plutonium g/l	0.0010	0.0009	0.0123	0.0123	0.0010	0.0009

If the uranium enrichment is 5% ^{235}U or less (less than 2.2 X 10^{-5} weight fraction ^{235}U in the feed), then the ^{235}U mass in one batch and in the heel is 309 g for a 3000-gallon batch. If the LWPT could be filled to its 6160-gallon capacity the ^{235}U content in the tank is 534 g and the full PR tank content 587 g. These values are below the single parameter limit for aqueous solutions of 700 g ^{235}U (21). Similarly the plutonium contents are more than a factor of ten less than the 450 g single parameter limit for ^{239}Pu. The rule-of-fractions, based on 700 g ^{235}U and 450 g ^{239}Pu, allows 72 g ^{239}Pu to be mixed with 587 g ^{235}U. At 5% ^{235}U the filter contains 154 g ^{235}U and 7 g Pu. Thus, if the fissile material were to separate from the solids and concentrate in the filter the total material in this batch would be safe. This analysis is done to demonstrate that one batch of feed with characteristics similar to Batch One will be safe based on the total fissile mass in the LWF and SPC with a factor of 5 margin in fissile material content. This factor of five allows for such upsets such as double batching.

The portion of entrained insoluble solids separate from the MST is limited to 400 mg/l. These insoluble solids will contain 1.93 X 10^{-3} g/l uranium and 4.4 X 10^{-6} g/l plutonium. In one batch, 11355 liters, the mass of uranium in the entrained insoluble solids is 22 g and the mass of plutonium is 0.05 g. In the event that these solids were to separate from the MST and accumulate in some manner from several batches, they pose no problem because of the small fissile masses involved.

In addition to the MST poison, the low fissile mass and the high hydrogen-to-fissile atomic ratio there are copious amounts of neutron poisons which further mitigate against a nuclear criticality. These poisons include primarily boron, as well as iron and manganese which are present to a lesser extent.

In summary material processed in the LWF and the Salt Processing Cell will remain safe from a nuclear criticality because the fissile materials remain bound to the MST in a critically safe ratio. If the fissile nuclides were to separate from the insoluble materials and become soluble the high hydrogen-to-fissile ratio will keep the mixture safe. Also for the materials to be processed in the DWPF the fissile material content is low enough that a critical mass will not accumulate in tanks of the LWF or the CPC. Other neutron absorbers which are prevalent will also mitigate against a nuclear criticality.

A-2 FISSILE MATERIAL CAKING ON PR TANK COILS

The PR tank has both cooling and heating coils in the lower part of the tank. These coils are schedule 80S pipe at a 3.5 inch pitch and arrayed in two concentric rings, (22) the inner ring at a diameter of 97 inches to the coil centerline and the outer ring at a coil diameter of 114.63 inches. The first eight coils from the bottom are heating coils and the next five are cooling coils.

The HRXN code was used to generate cross sections for an homogenized cell 3.5 inches square containing concentric rings (from the center) of water, steel, fissile material (gunk), and water. The thickness of the fissile material region was calculated based on a uranium concentration of 150 g uranium/l (this concentration is close to the optimum hydrogen-to-fissile ratio) at 5% enrichment spread uniformly over all of the cooling coils. A total of 25 kg U (1250 g

^{235}U) and 3125 g ^{239}Pu was equally divided between the inner and outer coils. The details of this calculation follow:

Sch 80 pipe:	OD 2.375 inches: radius 3.016 cm	
	ID 1.939 inches: radius 2.462 cm	

Inner coils

Coil location:	Dia. 97 in. Radius 123 cm
Surface area per pipe	1.46×10^4 cm²
Surface area 5 pipes	7.32×10^4 cm²

Outer coils

Coil location:	Dia. 114.63 in. Radius 145 cm
Surface area per pipe	1.73×10^4 cm²
Surface area 5 pipes	8.63×10^4 cm²

These homogenized mixtures were used in cylindrical ANISN computations with seven regions as follows:

1	U/Pu solution	δr = 118 cm
2	Inner coils	δr = 8.89 cm
3	U/Pu solution	δr = 14.1 cm
4	Outer coils	δr = 8.89
5	U/Pu solution	δr = 2.5 cm
6	Steel wall	δr = 1.3 cm
7	Water reflector	δr = 30 cm

where the U/Pu solution is a solution representing the PR tank feed uranium and plutonium only, 4.96×10^{-2} g/l U_3O_8 and 5.95×10^{-3} g/l PuO_2. (These analyses were done with an earlier flow sheet and which are different from later concentrations. Because of the dilute fissile content the results are not significantly altered with later feed compositions). Computations were done with the fissile material (1250 g ^{235}U and 3125 g ^{239}Pu) on the coils, without taking credit for the iron in the coil walls, and without ^{238}U. The results of these ANISN runs are in Table A-5

Table A-5
ANISN Results - Material on Cooling Coils

Condition	K_{eff}
25 kg uranium uniformly distributed over all coils (Job 6710)	0.357
25 kg uranium uniformly distributed over all coils, No iron in coils walls (Job 9547)	0.520
25 kg uranium uniformly distributed over all coils, Full iron, No U-238 (Job 9577)	0.362

For the homogenized cell with low enriched uranium there might be cell effects because of lumping of the material. The last case was run with no U-238, however with the same ^{235}U, Plutonium and iron as the second case. The cross sections were homogenized as solutions with U-238 replaced with water solvent and the H/F ratio increased from 2800 to 3200 with the replacement. The change in reactivity for the cylindrical core is 0.005 between these two cases.

These computations were done with the one-dimensional cylindrical ANISN model with no account taken for axial leakage, which would be significant for these short coils.

For these loadings on the coils the tanks remain safe. This conclusion is not a surprise when it is considered that these coils are far from the center of the tank in a geometry which is not favorable for criticality. Further, if this amount of uranium were to deposit on the cooling coils, the coils would be covered by a layer approximately 1.0-cm thick, which would certainly effect the heat transfer characteristics of the coils and be detected by the performance characteristics.

A-3 ACCUMULATION OF FISSILE MATERIAL DUE TO LOSS OF AGITATION

One of the upset conditions postulated by the review team was the accumulation of fissile material due to loss of agitation. In this case it is postulated that some fissile material separates from MST and preferentially settles. The analysis is to be based on the processing of two batches as loss of agitation is an upset which would be detected before processing of more than two batches is completed. A single batch of feed contains 5210 grams of uranium, Table A-4. Ha (14) has shown that as much as 35% of the uranium can be separated from MST under the extreme conditions of boiling in formic acid and that no measurable plutonium is separated under these conditions. In this case the uranium which would be available to resettle is 1825 grams.

A set of HRXN calculations was done to determine the hydrogen-to-fissile atom ratio which is most reactive for the base case uranium to plutonium mixture. For these cases the Pu to U ratio was maintained at 0.125 and the uranium was included as U_3O_8 with 100% ^{235}U and the plutonium as PuO_2 with 100% ^{239}Pu. The results for varying the uranium concentration are shown in Table A-6.

The data in Table A-6 indicate that this mixture of uranium and plutonium is most reactive with a hydrogen-to-fissile ratio of about 110 and a concentration of about 200 g-U/l. It also indicates that the multiplication factor is not very sensitive to the uranium concentration over a broad range of concentrations.

Table A-6

HRXN - U-Pu Solutions
JO 8964, 8969

U conc g/l	H/F	K inf
500	42.8	1.835
400	54.4	1.846
300	73.7	1.856
200	112.4	1.858
100	228.4	1.813
90	254.2	1.800
80	286.4	1.783
70	327.8	1.760
60	383.0	1.729
50	460.3	1.685
40	576.3	1.622
30	799.6	1.524
20	1156	1.356
10	2319	1.016

The materials in Table A-6 were included in a series of ANISN computations in which a search was done for the size of a critical sphere with a large water reflector with a K eff = 0.92. These results are in table A-7.

Table A-7

Water Reflected Spheres
K eff = 0.92: 100% ^{235}U
JO 8964, 8969

Uranium Conc g/l	Sphere Vol. l	Uranium Content g	Fissile Content g
500	4.84	2420	2723
400	4.88	1952	2196
300	4.99	1497	1684
200	5.31	1062	1194
100	6.59	659	742
90	6.92	623	701
80	7.35	588	662
70	7.94	556	625
60	8.78	527	592
50	10.08	504	567
40	12.35	494	556
30	17.16	515	579
20	33.02	660	743
10	500.0	5000	5625

The data in Table A-7 indicates that at 100% ^{235}U the minimum mass is 494 g. These calculations were done with a ^{239}Pu-to-^{235}U ratio of 0.125.

The effect of changing the uranium enrichment was studied with a series of HRXN-ANISN computations in which the ^{235}U enrichment was varied for fixed uranium and plutonium concentrations. The ANISN cases were done for a spherical-geometry solution core with 30-cm water reflector and a core radius search for 0.92 K eff. For each sphere the uranium loading and the ratio of the uranium loading to a batch containing 1825 g uranium was calculated. These data are shown in Tables A-8 through A-12.

Table A-8

Uranium Concentration 300 g/l (JO 9034)

Enrich. %	Volume Liters	Uranium g	^{235}U g	^{239}Pu g	#Batches @ 1825 g
35	6.97	2092	732	261	1.1
30	7.28	2185	655	273	1.2
25	7.65	2296	574	287	1.3
20	8.12	2438	487	305	1.3
15	8.74	2623	393	328	1.4
10	9.59	2878	288	360	1.6
5	10.84	3252	163	406	1.8
2	11.94	3582	72	448	2.0

Table A-9

Uranium Concentration 200 g/l (JO 9038)

Enrich. %	Volume Liters	Uranium g	^{235}U g	^{239}Pu g	#Batches @ 1825 g
35	7.82	1565	548	196	0.9
30	8.27	1654	492	207	0.9
25	8.83	1765	441	221	1.0
20	9.55	1910	382	239	1.0
15	10.52	2105	316	263	1.2
10	11.92	2384	238	298	1.3
5	14.08	2817	141	352	1.5
2	16.07	3214	64	402	1.8

Table A-10

Uranium Concentration 150 g/l (JO 3275)

Enrich. %	Volume Liters	Uranium g	^{235}U g	^{239}Pu g	#Batches @ 1825 g
35	8.93	1340	469	167	0.7
30	9.56	1434	430	179	0.8
25	10.36	1554	388	194	0.9
20	11.42	1713	343	214	0.9
15	12.89	1933	290	242	1.1
10	15.07	2260	226	283	1.2
5	18.64	2796	140	350	1.5
2	22.11	3316	66	414	1.8

Table A-11

Uranium Concentration 100 g/l (JO 9041)

Enrich. %	Volume Liters	Uranium g	^{235}U g	^{239}Pu g	#Batches @ 1825 g
35	11.76	1167	408	146	0.6
30	12.90	1290	387	161	0.7
25	14.41	1441	360	180	0.8
20	16.50	1650	330	206	0.9
15	19.57	1957	293	245	1.1
10	24.51	2451	245	306	1.3
5	33.55	3355	168	419	1.8
2	43.59	4359	87	545	2.4

Table A-12

Uranium Concentration 50 g/l (JO 3416)

Enrich. %	Volume Liters	Uranium g	^{235}U g	^{239}Pu g	#Batches @ 1825 g
35	26.87	1343	470	168	0.7
30	32.01	1600	480	200	0.9
25	39.76	1988	497	248	1.1
20	52.50	2625	525	328	1.4
15	76.42	3821	574	478	2.1
10	134.0	6700	670	837	3.7
5	365.0	18250	912	2281	10.0

Table A-13

Number of Feed Batches at 1825 g U per Batch not on MST

Enrich. %	300 g/l	200 g/l	150 g/l	100 g/l	50 g/l
35	1.1	0.9	0.7	0.6	0.7
30	1.2	0.9	0.8	0.7	0.9
25	1.3	1.0	0.9	0.8	1.1
20	1.3	1.0	0.9	0.9	1.4
15	1.4	1.2	1.1	1.1	2.1
10	1.6	1.3	1.2	1.3	3.7
5	1.8	1.5	1.5	1.8	10.0
2	2.0	1.8	1.8	2.4	

Table A-13 shows the number of feed batches at 1825 g uranium per batch which would be required to have a spherical configuration with a k_{eff} of 0.92. The data indicates that at 2% ^{235}U enrichment, a mixture which includes the fissile materials from as much as 1.8 batches of feed material would be safe (K_{eff} less than 0.92) if it were to be formed into a sphere of aqueous material at any concentration and be fully water reflected. This computation assumes that 35% of the uranium and all of the plutonium separates from the MST, then recombines in an aqueous mixture at optimum H/F ratio in spherical geometry. The computation neglects other neutron absorbers. Although Table A-13 does not show that all of the uranium from two batches does not maintain K_{eff} less than 0.92, it does indicate that the material from one batch is safe and that it requires a extraordinary set of circumstances for two batches for enrichments of 2%.

To this point the analysis has been based on the fissile material collecting in an aqueous sphere in a reactive condition. If the fissile material settled on the bottom of the tank in a flat layer it would not be expected to be in a more reactive geometry. This can be demonstrated by considering the case of the material from Table A-10, 150 g/l uranium enriched to 5% ^{235}U in a sphere radius 16.41 cm for which K_{eff} is 0.92. If this material were spread uniformly over the bottom of the tank in a circle with diameter 97 inches, the space inside the inner set of coils, the solution thickness would be 0.4 cm or about 0.15 inch. For a 100% enriched uranium solution in water, a fully water reflected slab has a critical thickness of 2.5 inches at a uranium concentration of 150 g/l and for plutonium at the same concentration the critical thickness is 2.4 inches (24). The solution in the PR tank is less than one-tenth of these critical thickness values and contains lower enriched uranium and a much lower concentration of plutonium. Thus settling of the material on the tank bottom does not present a criticality problem.

APPENDIX B - CHEMICAL PROCESSING CELL

B-1 Sludge Receipt Tank (SRAT)

Feed material for the SRAT comes as washed sludge from the Tank Farm and Precipitate Hydrolysis Aqueous (PHA) Solution from the Precipitate Reactor.

Tank Farm material destined for DWPF can be divided into two classes, material with larger amounts of uranium but with low ^{235}U content (< 1.0 wt% ^{235}U) and material with lesser amounts of fissile material but higher enrichment (see Appendix C). For example, Batch One sludge contains uranium with less than 1.0 wt% ^{235}U. This analysis with this feed is used to indicate margins of safety for several parameters. In many cases an enrichment is used to compute an equivalent ^{235}U content to demonstrate a margin of safety.

Selected contents of these feed streams for a representative sludge are listed in Table B-1 (16). The first column in Table B-1 has the composition of the sludge, the second column the feed from the PR tank and the third column the material which will be processed in the Slurry Mix Evaporator (SME), which is the next step in the process. The first column represents the material which initially enters the tank and the third column represents the material which remains after processing in the SRAT. These two materials will be considered in the criticality analysis. This analysis with these feed streams is used to indicate margins of safety for several parameters. In several cases an enrichment is used to compute an equivalent ^{235}U content to demonstrate a margin of safety.

The row labeled % Flow indicates the percentage of the material represented by these selected compounds. Missing are materials containing other neutron absorbers, primarily sodium.

Table B-1
Chemical Processing Cell Streams

	Sludge to DWPF		Salt Cell to SRAT		SRAT to SME	
	Lb/hr	Wt Frac.	Lb/hr	Wt Frac.	Lb/hr	Wt Frac.
Stream	465.8		671.2		457.8	
Water	395.9	8.50E-01	638.2	9.51E-01	355.2	7.76E-01
Water hydrate	6.426	1.38E-02	0.34	5.07E-04	6.327	1.38E-02
Al_2O_3	9.819	2.11E-02	0.3105	4.63E-04	10.28	2.25E-02
FE_2O_3	23.25	4.99E-02	0.4769	7.11E-04	23.73	5.18E-02
H_3BO_3	0		8.076	1.20E-02	8.076	1.76E-02
MgO	1.427	3.06E-03	0.000754	1.12E-06	0.2856	6.24E-04
MnO_2	3.338	7.17E-03	0.03838	5.72E-05	2.026	4.43E-03
$Mn(COOH)_2$	0		0.09662	1.44E-04	2.349	5.13E-03
NiO	0.2809	6.03E-04	0.02289	3.41E-05	0.2887	6.31E-04
PuO_2	8.45E-03	1.81E-05	9.45E-05	1.41E-07	8.55E-03	1.87E-05
PuO_2 (MST)			2.39E-04	3.56E-07	2.39E-04	5.22E-07
U_3O_8	2.308	4.95E-03	0.02	2.98E-05	2.328	5.09E-03
Sum	442.8	0.95	647.6	0.96	410.9	0.90
% Flow	95.05%		96.48%		89.76%	
Density lb/hr	69.36		63.29		71.29	
Density g/cc	1.1120		1.0147		1.1430	

The SRAT is a large tank with an internal stirrer and sets of heating and cooling coils. Dimensions, full tank volume and nominal operational volume are listed in Table B-2 (24, 25)

Table B-2

SRAT Tank Dimensions

		Inch	Meters
Inner Diameter		144	3.66
Maximum Liquid Height		154	3.91
Nominal Liquid Height		88	2.24
Tank Volume	1.09×10^4	gallons	
	4.11×10^4	liters	
Nominal volume	6.23×10^3	gallons	
	2.36×10^4	liters	

For the maximum volume, in which the tank is filled to the maximum height, and the nominal volume and for the concentrations of materials listed in Table B-1 the uranium and plutonium contents of the SRAT are listed in Table B-3.

Table B-3

Uranium and Plutonium Content of SRAT

Sludge Feed	Maximum	Nominal
U_3O_8	227 kg	130 kg
Uranium	192 kg	110 kg
PuO_2	830 g	475 g
Plutonium	732 g	419 g
SRAT to SME		
U_3O_8	239 kg	137 kg
Uranium	202 kg	116 kg
PuO_2	888 g	508 g
Plutonium	783 g	448 g

It is noted that the SRAT is a large tank which can contain significant amounts of fissile materials. Characteristics and ratios of materials in the SRAT are listed in Table B-4.

Table B-4
Material Characteristics

		Sludge to DWPF	Salt Cell to SRAT	SRAT to SME
Uranium	lb/hr	1.953	0.017	1.970
Plutonium	lb/hr	0.007453	0.000169	0.007621
Iron	lb/hr	23.95	1.18	24.43
Manganese	lb/hr	2.109	0.024	1.280
Boron	lb/hr	0.000	1.412	1.412
Uranium	wt. frac.	4.19E-03	2.52E-05	4.30E-03
Plutonium	wt. frac.	1.60E-05	2.51E-07	1.66E-05
Water	wt. frac.	0.8637	0.9513	0.7897
Hydrogen	atm/b-cm	6.42E-02	6.45E-02	6.03E-02
Uranium	atm/b-cm	1.19E-05	6.56E-08	1.26E-05
Plutonium	atm/b-cm	4.48E-08	6.42E-10	4.79E-08
H/F		5354	974783	4770
Pu/U		0.0038	0.0100	0.0039
Fe/U		12.26	69.49	12.40
Mn/U		1.080	1.433	0.650
Fe/Pu		3213	6980	3205
Mn/Pu		283	144	168
B/U		0	83.42	0.72
B/Pu		0	8378	185

The uranium ratios and H/F ratios in this table are for total uranium. For uranium at 5% ^{235}U enrichment the lowest Fe:^{235}U ratio, that in the sludge feed becomes 245:1. The safe Fe:^{235}U weight ratio is 77:1. The safe weight Mn:^{235}U ratio is 30:1. For uranium at 2% ^{235}U enrichment the manganese ratio after processing is 32:1. The margin of safety for the iron ratio is greater than five but the margin of safety for manganese is a factor of two.

The H/F ratios in Table B-4 are ratios for hydrogen to uranium and ^{239}Pu. For 5% enriched uranium, the lowest H/F ratio, that in the SME feed becomes 95,000, well in excess of the safe value. Either the fissile material remains with the metal oxides of iron and manganese, in which case the system is safe because of the poison effect, or the fissile material goes into solution, in which case the system is safe because of the high hydrogen-to-fissile ratio.

In summary the material in the Chemical Processing Cell is safe because the neutron absorbers, iron and manganese remain in the insoluble fraction. If the fissile materials remain insoluble, the ratios of iron and manganese to fissile nuclides is sufficient to maintain safety. If the fissile materials do not remain with the insoluble fraction then the hydrogen-to-fissile ratio is adequate to maintain criticality safety.

B-2 FISSILE MATERIAL CAKING ON SRAT COILS

The SRAT tank has both cooling and heating coils in the lower half of the tank. These coils are 2 nps schedule 40S pipe at a 3.5 inch vertical pitch and arrayed in three concentric rings at coil centerline diameters 46.5, 52.5 and 58.5 inches (26). The coils are staggered so they are arranged in an isosceles triangle with a base 3.5" (8.89 cm) and height 3.0" (7.62 cm). There are two sets of coils, each with 15 pipes (five at each radial location), separated 29 inches vertically.

The HRXN code was used to generate cross sections for a cell 8.89 X 7.62 cm containing concentric rings (from the center) of water, steel, fissile material, and water. The coil characteristics are listed below.

2 inch nps:	OD 2.375 inches:	radius 3.016 cm
Sch 40 pipe:	ID 2.067 inches:	radius 2.625 cm
Coil radius:	Dia. 46.5 inches:	radius 59.0 cm
Coil radius:	Dia. 52.5 inches:	radius 66.7 cm
Coil radius:	Dia. 58.5 inches:	radius 74.3 cm
Surface area per pipe		
First radial site:	7031 cm^2	
Second radial site:	7938 cm^2	
Third radial site:	8847 cm^2	
Surface area 15 pipes:	1.19×10^5 cm^2	
Surface area 30 pipes:	2.38×10^5 cm^2	

The pipe sizes are nominal dimensions and the actual pipe thickness may be as much as 12.5% less than the difference between the outer and inner radii. To account for thinner pipes cases were done with the wall thickness reduced to 75% of the nominal value and to zero by reducing the outer diameter.

Two cases were considered: A dry mixture containing the oxides U_3O_8, Al_2O_3, Fe_2O_3, and MnO_2 and a mixture with all of the U_3O_8 deposited on the coil surfaces. The latter case represents a situation in which all of the fissile material is separated from other oxides and deposits on the coils.

Homogenized mixtures of the fissile material, the cooling pipes and water in the pipe flow area were used in cylindrical ANISN computations with five regions as follows:

1	U/Pu solution	δr = 55.24 cm
2	Homogenized coils	δr = 22.86 cm
3	U/Pu solution	δr = 105 cm
4	Tank wall	δr = 1.27 cm
5	Water reflector	δr = 30 cm

where the U/Pu solution is a solution representing the SRAT tank feed. The homogenized coils region contains the mixture of fissile material, metal oxides, iron and water representing the heating and cooling coils with deposited material.

A material containing U_3O_8, Al_2O_3, Fe_2O_3, and MnO_2 was spread over the coils. This fuel mixture has a density 4.33 g/cc and contains 6.9 weight percent U_3O_8. If this mixture completely fills the free space around the coils 147 kg of U_3O_8, or 25 kg U-235 at 20% enrichment, would be in this volume. For this case the cylindrical ANISN gives a multiplication factor of 0.458 (JO 8389). With the pipe walls reduced to 75% of the nominal thickness and to zero the multiplication factors are 0.481 and 0.546 (JO 8869) respectively. This mixture is safe for both pipe thicknesses.

A full uranium content 250,000 g U_3O_8 at 8.39 g/cc spread over the area of all of the pipes produces a fissile material thickness 0.125 cm and a nominal uranium content of 130,000 g produces a material thickness of 0.065 cm. These uranium contents are higher than that in the feed as the values were rounded up since plutonium is not included explicitly in this calculation. Homogenized materials composed of water in the pipe flow area, iron in the pipe walls, fissile material with several thicknesses, and water in the balance of the cell were constructed. These materials were homogenized over a rectangular cell 8.89 X 7.62 cm and included in the cylindrical ANISN. The results from several cases are listed on Table B-5.

Table B-5

ANISN Results - Material on Cooling and Heating Coils (JO 7941, 7991, 8391, 8860, 8867)

	Fissile Thickness	Wall Thickness	U-235 Enrichment	K eff
Case 1	0.125	Full	15%	0.915
Case 2	0.125	0.75	15%	0.955
Case 3	0.125	Zero	10%	0.955
Case 4	0.125	Zero	5%	0.688
Case 5	0.065	Full	20%	0.793
Case 6	0.065	Full	15%	0.682
Case 7	0.065	0.75	20%	0.834
Case 8	0.065	0.75	15%	0.721
Case 9	0.065	Zero	15%	0.875
Case 10	0.065	Zero	10%	0.714

In these calculations no account has been taken for transverse leakage. Case 2 was repeated with a transverse leakage corresponding to a cylinder height 99.34 cm ($B^2 = 10^{-3}$ cm^{-2}). Each set of coils is 45 cm high so this approximation squeezes the two sets of coils together and ignores the water gap between them. With the transverse leakage the multiplication factor is reduced from 0.955 to 0.929 (JOB 8391).

For a worst case it is assumed that 250 kg U_3O_8 at 15% ^{235}U enrichment and separated from the other metal oxides becomes caked on the SRAT coils and that the iron in the coils had been partially dissolved away. For this case the computed multiplication factor is 0.929. This provides a margin of at least 15 between the case considered and the lower enrichments or lower uranium contents of expected sludges.

B-3 SUMP AND MERCURY TRAP

The Chemical Process Cell contains the SRAT tank. The tank is large enough so that no assumptions have been made about finite geometry. Thus if the material is safe without finite geometry corrections, it would also be safe if the tank were to leak and its contents fall to the floor or into a sump. This is true as long as the material which has leaked to the sump does not change, i.e. there are no processes, such as evaporation, which will concentrate the fissile materials.

The SRAT tank has a mercury trap, which is an indentation in the tank bottom 13.25" deep and 12.0" diameter. A postulated accident has mercury with plutonium collecting in this trap. This possibility is no longer considered a problem as the amount of plutonium that might be in the mercury is less than 6 X 10^{-6} gram of plutonium per gram of mercury (27). At this concentration only about 2 grams of plutonium, a nonsignificant amount, would accumulate in the trap.

APPENDIX C - SLUDGE CHARACTERISTICS

Analyses based on historical records provide information about materials in the Tank Farms which will may eventually be processed through DWPF. A tank inventory summary (28) of sludge fissile material contents is in Table C-1. Also listed are the materials expected in the first 6 batches. Tanks 23 and 24 are not listed in Table C-1 as they may not be included in Batch 6. These batches include the mixed contents of individual tanks as follows (29):

Batch 1 - Tanks 42, 51;
Batch 2 - Tanks 8, 11, 15;
Batch 3 - Tanks 4, 7, 12, 14, 47;
Batch 4 - Tanks 5, 6, 9, 10, 13, 26, 35;
Batch 5 - Tanks 1, 2, 3, 32, 33, 34, 39, 43;
Batch 6 - Tanks 17, 18, 19, 21, 22, 23, 24.

Equivalent ^{235}U is defined two ways for this table. In the column headed Equiv. ^{235}U the quantity is computed as ^{235}U + (700/400) ^{233}U, where 700 and 400 are the acceptable safe masses in grams of ^{235}U and ^{233}U respectively in aqueous solutions (18). This definition does not account for the plutonium. Enrichment in the eighth column and Pu/U in the ninth column are computed from this definition of equivalent ^{235}U. In the last column, headed ^{238}U:Eq. ^{235}U, the quantity is defined as ^{235}U + 1.92(^{233}U + ^{239}Pu + ^{241}Pu) (29). This is called equivalent fissile mass, FM, in the section following Table C-1. The table illustrates the fact that the feed material to DWPF contains either uranium with a low ^{235}U enrichment or with a small content of uranium and the combinations mixed to make the first six batches of sludge all contain low enriched uranium and a low Pu:U ratio. In several places in the analysis for this NCSE an enrichment of 5% and a Pu/U ratio of 0.1 is used to estimate margins of safety. The uncertainty in these fissile content values is not available. For an assumed uncertainty of a factor of 5 in ^{235}U content, all 6 batches have ^{235}U enrichment less than 1% and Pu/U ratio less than 0.1.

Table C-1
Sludge Inventory
Fissile Contents

Tank Number	Uranium kg	^{235}U g	^{233}U g	^{239}Pu g	^{241}Pu g	Equiv. ^{235}U	Enrich.	Pu/U	^{238}U:Eq. ^{235}U
1	1323	8167	0	2139	3	8167	0.62%	0.0016	161
2	61	376	0	318	0	376	0.62%	0.0052	161
3	152	939	0	383	0	939	0.62%	0.0025	161
4	10016	35951	0	9616	18	35951	0.36%	0.0010	278
5	7474	48448	0	7755	13	48448	0.65%	0.0010	153
6	7425	30763	0	4095	16	30763	0.41%	0.0006	240
7	27414	175553	0	58978	102	175553	0.64%	0.0022	155
8	20771	69757	0	31858	85	69757	0.34%	0.0015	297
9	89	546	0	100	0	546	0.61%	0.0011	162
10	17	106	0	51	0	106	0.62%	0.0030	159
11	194	14631	211	23942	868	15000	7.69%	0.1279	12
12	2092	31211	3955	37262	527	38132	1.76%	0.0181	56
13	5606	72487	4194	23452	172	79826	1.40%	0.0042	71
14	239	1920	39	972	2	1988	0.83%	0.0041	120
15	40	28308	1433	19588	199	30815	75.79%	0.4947	0
17	251	625	0	792	3	625	0.25%	0.0032	401
18	466	1454	0	1550	4	1454	0.31%	0.0033	319
19	55	94	0	174	0	94	0.17%	0.0032	584
21	8	1091	18	18	0	1122	13.95%	0.0023	6
22	32	2734	111	0	0	2928	9.03%	0.0000	10
26	6034	10241	0	38507	189	10241	0.17%	0.0064	588
30	0	150	0	493	37	150	NA	3.5333	NA
32	102	18362	0	56814	2917	18362	18.00%	0.5856	5
33	25726	43659	0	16612	79	43659	0.17%	0.0006	588
34	24056	52761	0	20924	98	52761	0.22%	0.0009	455
35	139	21852	0	52966	3174	21852	15.72%	0.4039	5
36	0	65	0	77	4	65	NA	1.2462	NA
39	71	23438	0	82302	5589	23438	33.01%	1.2379	2
40	4838	18611	148	15960	46	18870	0.39%	0.0033	256
41	0	66	0	0	0	66	NA	0.0000	NA
42	3477	33336	777	20631	128	34696	0.99%	0.0060	100
43	42	13713	0	1696	96	13713	32.65%	0.0427	2
47	4523	8629	0	36412	160	8629	0.19%	0.0081	523
51	6887	24640	79	22864	66	24778	0.36%	0.0033	277
Batch 1	10364	57976	856	43495	194	59174	0.57%	0.0042	174
Batch 2	21005	112696	1644	75388	1152	114997	0.55%	0.0036	182
Batch 3	44284	253264	3994	143240	809	258855	0.58%	0.0033	170
Batch 4	26784	184443	4194	126926	3564	190315	0.71%	0.0049	140
Batch 5	51533	161415	0	181188	8782	161415	0.31%	0.0037	318
Batch 6	812	5998	129	2534	7	6224	0.77%	0.0031	129

Clemmons (30) has analyzed the content of these tanks for iron, manganese and ^{238}U:Eq. ^{235}U Ratios and demonstrated that 27 of the tanks are safe by the iron ratio only, 25 safe by manganese only, 26 safe by combination of iron and manganese, and 11 by ^{238}U only. The safe tank numbers are listed below:

Safe by iron only: (27 tanks) 1, 2, 3, 4, 5, 7, 8, 9, 10, 11, 12, 13, 14, 15, 17, 18, 19, 21, 22, 26, 32, 35, 40, 42, 43, 47, and 51.

Safe by manganese only: (25 tanks) 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 17, 18, 19, 26, 32, 35, 40, 42, 47, and 51.

Safe by iron and manganese: (26 tanks) 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 17, 18, 19, 26, 32, 35, 39, 40, 42, 47, and 51.

Safe by ^{238}U only: (11 tanks) 1, 4, 5, 6, 8, 9, 17, 18, 19, 33, and 34.

The combined tanks making Batches 1 through 6 have the following iron, manganese and ^{238}U to equivalent fissile mass ratios:

	Fe/FM	Mn/FM	Mn/2FM	$^{238}U/^{235}U$
Batch 1	1092	58	29	174
Batch 2	365	57	28	182
Batch 3	374	104	52	170
Batch 4	551	87	44	140
Batch 5	206	25	13	318
Batch 6	388	37	19	430

All batches have Fe/FM in excess of the safe weight ratio of 160:1. Three batches, #3, #4 and #6 have Mn/FM ratio in excess of the safe weight ratio 32:1. All batches have $^{238}U/Eq.^{235}U$ in excess of the safe ratio of 103:1. All batches are safe by the Fe:Mn:FM ratios.

Material in the SRAT is critically safe because of the ratios of iron to fissile nuclides in the sludge and the ratios of Fe:Mn:FM. The iron ratio is not changed by the processes in the SRAT but the manganese ratio can be reduced up to 50%.