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SEP 16 1994

ENGINEERING DATA TRANSMITTAL

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1	1	Cog. Eng. CA Rogers	<i>CA Rogers</i>	8/15/94	-01						
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18. Signature of EDT Originator <i>CA Rogers</i> 8/15/94	19. Authorized Representative Date for Receiving Organization <i>TS Van</i> 8/16/94	20. Cognizant/Project Engineer's Manager <i>JW Daughtry</i> 8/17/94	21. DOE APPROVAL (if required) Ltr. No. ☐ Approved ☐ Approved w/comments ☐ Disapproved w/comments
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This document was reviewed following the
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7. Abstract

Water is periodically added to Tank C-106 to cool its waste. In March 1994 addition of water was temporarily discontinued to determine if the tank could be adequately cooled at a lower water level. Following an addition of water, a temperature fluctuation was observed on one of the thermocouple trees. This Criticality Safety Evaluation Report (CSER) explains why the anomalous temperature measurements could not have been caused by nuclear criticality.

Waste in Tank C-106 was discharged from processing facilities under controls designed to ensure that the contents of the tank would remain well subcritical under all credible conditions. The observed temperature profile does not fit the profile expected from a criticality event. In addition, there has been no indication of any significant increase in the rate of water evaporation.

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CSER 94-009

Title: IMPLICATIONS OF THE HEAT ANOMALY IN TANK C-106
TO CRITICALITY SAFETY

Prepared by: CA Rogers Date: 8/15/94
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Approved by: M. N. Islam Date: 8/18/94
M. N. Islam
Manager, Waste Tank Safety Assurance

1.0 SUMMARY AND CONCLUSIONS

Following anomalous temperature readings in Tank C-106, a request was made to evaluate the possibility that the observed temperature fluctuations may have been caused by a nuclear criticality. This Criticality Safety Evaluation Report (CSER) explains why a nuclear criticality could not have caused the observed temperature changes.

Consideration of the amount and concentration of plutonium in Tank C-106 leads to the conclusion that criticality is extremely unlikely, if not impossible. The mass ratio of solids to plutonium is estimated to be at least 23 times as great as required to assure subcriticality in a uniform mixture. The measured plutonium concentration is at least 31 times smaller than the minimum required to achieve criticality under conditions of optimal moderation. Waste was sent to tank storage well mixed, and there are no known processes which might credibly have separated plutonium from the other components and concentrated it to the level required to achieve criticality.

These conclusions are independent of the tank farms investigation that reached the conclusion that the anomalous temperature readings in riser 14 were caused by a shifting of waste. This shifting brought the waste into direct contact with the thermocouple located near the center of the tank and caused the recorded temperature to increase (see Appendix A).

The observed temperature does not fit the profile which would be expected from a criticality event.

2.0 BACKGROUND

Waste stored in Tank C-106 generates considerable heat (estimated at 120,000 Btu/hr). A ventilation system and periodic addition of cooling water are used to remove this heat. In March 1994 the decision was made to temporarily discontinue the addition of water to lower the level of the waste in the tank. From this time until water was again added on June 17 the temperature of the waste remained stable. After the addition of water was resumed, the temperature of the waste underwent a steady rise. Water was then added with the objective of stabilizing the temperature. However, the temperature then continued to increase and began to oscillate after more additions were made.

A detailed description of Tank C-106, its contents, and recent water additions is provided in Appendix B. Included is a description of the fluctuation in water level and a schedule of water additions. Appendix C contains a description of the observed temperature profile.

3.0 DISCUSSION

Under normal conditions the heat generated from fission is far too small to influence the temperature of the waste. The heat from fission would become sufficient to raise the temperature only if a self-sustaining neutron chain reaction were to occur. This would cause a rise in energy (heat) output, followed by a rise in temperature and a change in solution density. After a short time the criticality would cease, and the energy output would drop rapidly.

Under certain conditions a solution containing plutonium might fluctuate between supercritical and subcritical and the energy output might oscillate (pulsate). It has been suggested that the temperature curve observed in Tank C-106 (see Appendix C) might be the result of such oscillations. This CSER reviews the potential for criticality and compares the observed temperature profile to that which might result from a criticality.

Waste in Tank C-106 is described as a sludge below a liquid surface. This configuration is very different from those for which energy oscillations have been observed. The exact nature of the energy release which would result from a criticality and its effect on the waste is uncertain.

3.1 Criticality Safety Evaluation

Waste in Tank C-106 was discharged from processing facilities under controls designed to ensure that it remain well subcritical under all credible conditions. The probability of achieving a concentration even approaching the minimum critical concentration is very low. Not only does the plutonium concentration have to be high enough, but the quantity of plutonium within a limited volume must be large enough. A detailed discussion of criticality safety parameters is found in Appendix D.

The minimum concentration of plutonium which can be made critical in conservatively defined waste was calculated to be 2.6 g/L (Rogers 1993). The waste layer in Tank C-106 is less than 7-ft thick, and the minimum plutonium concentration required for criticality in a slab of this thickness is at least 4.0 g/L. A conservative estimate of the plutonium concentration in Tank C-106 solids based upon 3 analyzed waste samples is 0.127 g/L. Therefore, the measured plutonium concentration is at least 31 times less than the minimum value which can be made critical under conditions of optimal moderation.

The overall solids/plutonium mass ratio for Tank C-106 is at least 11,020, a value 23 times greater than the subcritical limit of 476 (Rogers 1993). The Fe/Pu mass ratio is measured to be 653, a value 4 times larger than the subcritical limit of 160. The Mn/Pu mass ratio is measured to be 89, a value almost 3 times larger than the subcritical limit of 32. These ratios show that the margin of subcriticality is large.

Waste was received well mixed, and conditions within tank storage would tend to maintain it well mixed. Even if one postulates a chemical reaction capable of removing a specific waste component (or even several components) from a region of the tank, there would still remain far more absorption in the remaining components than required to ensure subcriticality. In addition the

plutonium itself would have to be greatly concentrated before criticality is possible.

All available information indicates that the waste is highly subcritical and will remain subcritical under all conditions which might credibly develop.

3.2 Qualitative Discussion of Temperature Oscillations

A major difficulty with discussing a hypothetical criticality in high-level waste is the lack of experimental data for critical systems which are similar to this waste. Discussions are greatly complicated by the complexity and by the lack of accurate characterization of the waste.

The uncertainties make it possible to develop a wide range of hypothetical scenarios with a wide range of possible consequences. In theory, it appears possible to design a critical system to imitate almost any observed data. However, it is unlikely that any consensus would be reached by independent reviewers as to its applicability. It would not be possible to demonstrate that the theoretical model even remotely resembles the actual waste configuration. For these reasons, conclusions drawn from hypothetical scenarios must be considered as qualitative only and used with caution.

Between March 10 and June 17 when no water was added to Tank C-106, the temperature was observed to be stable. Water was added on June 17 to resume normal operations. Following this water addition, there was at first a rapid rise in temperature followed by a period of oscillation as more water was added. A graph of the observed temperature is provided in Appendix C.

Criticality would not be expected to occur following the addition of water, for the following reason. Water had earlier been removed from the waste through evaporation. Since the water evaporated from the surface of the waste, the degree of moderation inside the waste would not have changed. Since the layer of water above the sludge would have reflected neutrons back into the waste, its removal would have reduced the potential for criticality. Removal of the upper water layer would be expected to increase the margin of safety.

For a criticality to occur, the sludge must initially be overmoderated, which it very likely was. Water would then have to be removed from the sludge to lower the degree of moderation. For this to happen, the entire layer of water above the sludge would have to be removed (evaporate). If criticality were to occur, it would occur while water was being removed from the sludge. As soon as water was added, the degree of moderation would return to an earlier level. Since criticality had not occurred earlier, it would not occur now. This observation strongly suggests that the rapid temperature rise which came after the water was added was not a result of criticality.

Criticality always produces energy. Temperature oscillations caused by criticality would be expected to begin with a sudden rise in temperature, and the criticality would be expected to terminate after a short time. Following criticality the waste would be expected to cool at a rate more gradual than the temperature rise. Before beginning oscillation on June 26 the temperature was rising slowly then fell swiftly, and during the subsequent oscillations the temperature rose and fell at about the same rate. This is not the

expected temperature profile following a criticality. Again, the observation suggests (but does not prove) that the temperature change was not a result of criticality.

When regular oscillations began, these oscillations began with a relatively rapid drop in temperature. This is characteristic of a sudden cooling process. Oscillations caused by criticality should begin with a rise in temperature. The fact that the oscillations began with a drop in temperature strongly suggests that they were not driven by criticality.

A nuclear criticality produces activated fission products. Identification of these fission products is a sure way to demonstrate that criticality has occurred. Demonstration that no fission products were produced provides verification that a criticality has not occurred. A problem associated with the identification of fission products is that many are short lived and detection of their presence must be made soon after their release.

Dose rates are measured three times each week on the C farm exhaust stack High Efficiency Particulate Air (HEPA) filters. During the period that the temperature cycles were occurring, the measured dose rate did not detect any activity levels which could be attributed to short-lived fission products. The measured dose rates remained normal and do not provide any reason to believe that a criticality occurred.

3.3 Pulsating Criticality Scenarios

For a critical system to pulsate, it must be fluid. Mechanisms which cause a system to pulsate are associated with solutions. Three scenarios generally associated with a pulsating criticality and a brief discussion of their applicability to Tank C-106 are as follows:

1. A criticality occurs in a container (or confined region) of solution of unsafe geometry. Liquid is thrown from the container. The ejected liquid drains back into the container until it causes another excursion and repeats the process.

At the time of the temperature oscillations waste in Tank C-106 was probably best described as a sludge with a wet surface. There is no evidence that liquid was ejected out of the surface of the waste.

2. A fissile solution flows slowly into an unsafe container, until a criticality occurs which throws solution from the container. The input of new fissile material eventually leads to another criticality, and the process is repeated.

No new waste had been sent to Tank C-106, except for periodic additions of cooling water, for over fifteen years. In addition, there are no mechanisms capable of creating a slow, continuous flow of fissile solution from within the tank into a limited volume, and there is no mechanism which might credibly deposit the plutonium in one region of the tank. The large fraction of solids in the waste effectively limits the highest plutonium concentration which can credibly be achieved.

3. A postulated criticality forms bubbles of radiolytic gas. The solution expands until the neutron chain reaction stops. The bubbles dissipate and the solution shrinks causing another criticality. In this scenario the average solution density is first reduced and then restored.

This type of oscillating criticality would be expected to require very precise conditions to create. The viscous nature of the waste should preclude this possibility. However, it is impossible to estimate with any certainty the exact nature of the energy release and its effect on the waste. No change in the surface of the waste was observed.

4.0 REFERENCES

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APPENDIX A
STATUS REPORTS

The following CC:MAIL information messages were sent to all Westinghouse-Hanford Company and BCSR employees to keep them informed of the status of the investigation of Tank C-106.

PRIORITY MESSAGE
July 25, 1994

Many of you are aware that we have been investigating temperature measurement irregularities in Tank C-106. The thermocouple temperature readings in riser 14 started increasing June 17 following completion of a process test. This was classified as an off-normal occurrence because the readings did not match the historical pattern for this thermocouple. We've been investigating and have determined that the thermocouple temperature readings in riser 14 have been increasing because the waste in the tank has shifted, not because the tank's behavior has changed. For the first time, the thermocouple located near the center of the tank has come in direct contact with the waste.

When 30,000 gallons of water were added following a process test that lowered the water level in the tank, the sludge was redistributed and the hole around the thermocouple was filled in. The thermocouple is now giving accurate readings and should continue to do so in the future. As of today, the temperature was recorded at 209.1° Fahrenheit and should stabilize between 210 and 230° F. The temperatures in the tank are well below the safety limit of 350° F and pose no threat to public or worker safety.

Based on thermodynamic modeling of the tank, the riser 14 thermocouple should have been recording temperatures of about 210° F. Instead, its reading has been around 140°, much lower than expected. Due to these low readings, the measurements made by this thermocouple have been considered inaccurate. Therefore, the readings from the thermocouple in riser 8 and the thermodynamic model have been used in the safety analysis. The riser 8 thermocouple readings have remained stable during this time.

An in-tank video tape from last month has shown that a water envelope has been surrounding the thermocouple in riser 14 so that it was measuring the temperature of the water instead of the temperature of the waste. Evaluations by both Pacific Northwest Laboratory and Los Alamos National Laboratory are in agreement with our analysis. The news media was briefed on this subject Friday.

John L. Lee
Director, TWRS Plant

GENERAL DELIVERY MESSAGE
August 1, 1994

I would like to update you on the status of Tank C-106 following the temperature reading increases reported in a priority message July 21, 1994. Even though we have brought in an outside group of experts in heat transfer waste chemistry and tank safety to review Tank C-106 data, I can assure you there has been no change in the tank's safety status. It is being managed in a safe manner and is not a threat to employees, the public or the environment.

The group is looking at historical and current trends in C-106 so that we can thoroughly understand this tank's behavior. They will review our conclusions and action plans as well as advise us on any additional actions that should be taken. Meanwhile, the U.S. Department of Energy is closely reviewing our work.

Among the conclusions that our review has provided is that there is no indication of increased radiation readings in or around the tank, on the HEPA filters, or on the ventilation system's continuous air monitor. Furthermore, we have confirmed that the evaporative water cooling of the tank waste is consistent with historical levels. This supports the belief that the tank is not trapping heat and heating up.

The tank's data continues to be analyzed and contingency plans are being evaluated to ensure continued safe storage of waste. An in-tank video camera is being installed to provide continuous visual monitoring of the tank's interior. Temperature and liquid levels are being monitored closely and the ventilation flow rate has been increased.

The temperature readings from near the center of the tank began increasing June 17, 1994, after water was added to the tank following a process test during which the water was allowed to evaporate. The temperature measurements are still rising, but at a slower rate. As of this morning, the temperature measured by the thermocouple in riser 14 was 215° F. The increase does not signify a change in the waste's behavior, but is occurring because the waste was redistributed and is coming in direct contact with the temperature sensors. A statistical evaluation of the actual temperature data indicates that the temperature will stabilize between 210 and 230° F. The thermodynamic model supports the statistical evaluation.

The increase in temperature measurements has been upgraded from an "off-normal" occurrence to an "unusual" occurrence by plant management to give increased visibility to our investigation. This category change does not reflect a difference in the tank's safety. It is an indication of the level of interest in the temperature measurement rise.

We will continue to keep you informed.

John L. Lee
Director, TWRS Plant

GENERAL DELIVERY MESSAGE
August 5, 1994

As you all know, C-106 has taken center stage both at TWRS and in the public media. I wanted to update you on the status of the C-106 effort.

We put together several teams to examine the issues and to plan for contingencies. These teams include a C-106 heat transfer modeling team, a C-106 monitoring team, a contingency planning team, a radiation readings team, and a senior management team to evaluate lessons learned from the process test.

We brought in outside heat transfer experts from PNL, LANL, and industry to ensure an impartial review. Overall, the conclusions are:

- The tank is not experiencing a bulk heatup, but rather the one thermocouple is now immersed in sludge and reaching equilibrium temperatures. For the past three days the statistical evaluation shows the temperatures are stable, but a few more days will be needed to confirm C-106 has leveled off.
- A steam bump is unlikely and the radiation readings are stable.

Each of these conclusions are undergoing an independent review to ensure we have a consensus by all the technical experts.

As you may have heard, we put in place a number of contingency actions, including:

- Respirators required for all work in C Farm;
- Restricted work in C Farm and each activity evaluated by a special review team;
- Operational trigger points and actions defined for temperature/level;
- Special emergency plans developed for C-106;
- Spare equipment, including an exhauster, identified and package planning ongoing;
- Special monitoring (radiation, video, etc.) identified and implemented; and
- Plans developed for providing additional cooling.

The next steps are completing the independent review of the technical conclusions and reaching agreement on criteria for when we will resume normal operations in C Farms. We have also initiated a special independent review team to examine the process test and identify what we need to do differently in the future. I will keep you abreast of these actions as they are completed.

John Lee, Director
TWRS Plant

APPENDIX B

DESCRIPTION OF TANK C-106 AND CONTENTS

VOLUME OF WASTE

Bailey (1993) describes Tank C-106 in *Functional Design Criteria - Tank 241-C-106 Waste Retrieval Project W-320* as a 75-ft diameter single-shell tank with a capacity of 530,000 gal. It contains 197,000 gal (746,000 L) of sludge stratified in two layers, as follows:

- * The top layer consists of 173,000 gal (655,000 L) of high heat generating sludge. The heat generation is caused by ^{90}Sr , but neither the heat nor the ^{90}Sr significantly influence the margin of criticality safety.
- * The bottom layer consists of 24,000 gal (91,000 L) of hardened coating waste from dissolution of aluminum fuel cladding.

Hanlon (1992) lists the volume of drainable liquid in Tank C-106 at 48,000 gal (183,000 L). Of this, 32,000 gal is supernate and 16,000 gal is interstitial liquid.

CHARACTERIZATION OF WASTE

Harris (1993) compiled characterization data dating back to 1975 for Tanks AY-101, AY-102, and C-106. Eight (8) analytical samples are reported for Tank C-106: 3 sludge (solids), 3 supernate (liquid), and 2 combination samples. Harris lists all of these samples and shows the high, low, and average value of each component concentration. Table 1 shows the high, low, and average concentration of the individual components for the solids and the liquids in Tank C-106.

For this discussion the term "sludge" is synonymous with "solids" and the term supernate is synonymous with "liquid."

A combination sample represents the configuration in which the solids and the liquids are homogenized within the analyzed sample. The average, high, and low values reported by Harris for the total waste are derived directly from the two combination samples. They do not represent a volume averaging obtained from independent sludge and supernate samples.

The concentration for solids is provided in units of grams of solids per kilogram of waste (g/kg). This can be converted to units of g/L by multiplying by the waste density. Harris reports the average value of the solids density to be 1.40 g/cm^3 , with the high and low reported values being 1.43 and 1.37 g/cm^3 . For this evaluation the high value of 1.43 g/cm^3 is used. The density of solids refers to the density of the settled solids, including the liquid portion.

The high estimate of the plutonium concentration in solids is given as 0.089 g/kg . When multiplied by the density of 1.43 kg/L , the converted plutonium concentration becomes 0.127 g/L .

Table 1. Composition of Solids and Liquids in Tank C-106.
(Some components less important to criticality safety are not included.)

Component	Concentration in Solids, g/kg			Concentration in Liquid, g/L		
	High	Low	Average	High	Low	Average
Plutonium	0.089	0.025	0.054	0.016	0.000	0.0036
Aluminum	40.90	30.00	36.45	0.76	0.03	0.27
Barium	4.89	4.89	4.89	0.005	0.005	0.005
Boron	0.019	0.019	0.019	0.010	0.010	0.010
Cadmium	0.210	0.001	0.111	0.026	0.026	0.026
Calcium	11.90	11.90	11.90	0.01	0.01	0.01
Chromium	1.35	0.98	1.17	0.33	0.01	0.17
Iron	64.10	52.10	58.10	0.01	0.01	0.01
Lead	1.06	1.06	1.06	1.68	0.08	0.64
Magnesium	6.56	0.46	3.51	0.01	0.01	0.01
Manganese	14.10	1.84	7.97	0.20	0.20	0.20
Mercury	---	---	---	0.02	0.02	0.02
Nickel	0.97	0.68	0.82	0.89	0.07	0.48
Phosphorus	9.21	9.21	9.21	0.34	0.34	0.34
Potassium	1.47	1.47	1.47	0.42	0.42	0.42
Silicon	71.00	20.60	45.80	2.58	0.11	1.34
Silver	0.53	0.53	0.53	0.01	0.01	0.01
Sodium	117.00	36.80	76.40	127.42	73.83	91.09
Uranium	0.41	0.00	0.20	0.96	0.16	0.56
Zirconium	2.17	0.74	1.45	0.29	0.29	0.29
TOC	4.62	4.62	4.62	20.00	2.52	11.26
Chlorine	---	---	---	0.80	0.15	0.25
CO ₂	---	---	---	91.90	18.60	44.86
NO ₂	---	---	---	13.25	2.99	9.75
NO ₃	1.33	0.93	1.13	112.20	1.40	67.15
PO ₄	93.70	93.70	93.70	11.10	0.85	4.04
SO ₄	4.85	0.94	2.89	6.47	3.52	4.99

An estimate of the total plutonium content is obtained by multiplying the volume of waste, either solids or liquid, by the corresponding maximum concentrations. The total plutonium content in 197,000 gal (746,000 L) of solids is found by multiplying by a concentration of 0.127 g/L to be 94.6 kg. For 48,000 gal of liquid at a plutonium concentration of 0.016 g/L, the total plutonium content is found to be 2.9 kg. The total plutonium for Tank C-106 is estimated to be 97.5 kg. This value is based upon the largest measured density and the largest measured plutonium concentration and is therefore considered to be an upper limit on the expected total plutonium content.

Agnew (1993) estimates the plutonium inventory of Tank C-106 as 63.7 kg.

LIQUID LEVEL AND WATER ADDITION

The following information describing the addition of water and the liquid level was provided by J. S. Schofield.

Over a period of years the liquid level over the surface of waste in Tank C-106 has been maintained between 75 and 79 in. Due to heat generated within the waste there is a continual evaporation of water causing the liquid level to slowly drop at a rate of about 2.2 in. per month. This means that about 198 gallons of water evaporate per day. When the level drops to 74.5 in., water is added to raise it back to approximately 79 in.

The liquid level is measured by a conductivity probe manufactured by the Food Instrument Corporation (FIC) and referred to as the FIC level indicator. The measured liquid level is referred to as the FIC level.

In March, 1994, addition of water was discontinued to allow the liquid level to drop. On May 20, 1994, the sludge level was measured at 69.5 in. Although water continued to evaporate after this date, the indicated level of the waste surface remained at 69.5 in. due to the FIC plummet touching a piece of pipe in the tank.

On June 16, 1994, the liquid level was estimated at 67.5 in., assuming a sludge porosity of 100%. The actual level of the surface was likely between 66.5 and 67.0 in. Beginning June 17 water additions were again made. A summary of water additions and FIC levels is provided in Table 2.

Table 2. Water Additions

Date	Water Added Gallons	FIC Level Inches
6/17/94	0	69.5
6/17/94	2347	69.5
6/22/94	1749	70.5
6/23/94	2273	71.3
6/24/94	2122	72.0
6/28/94	2706	72.7
6/29/94	3075	73.8
7/3/94	14425	78.8

Close observation of the liquid level on July 11 indicated a random cyclic variation between 78.3 and 78.7 in. The reason for this fluctuation appears to be rippling of the surface by rising bubbles.

APPENDIX C
TEMPERATURE MEASUREMENTS

THERMOCOUPLE LOCATIONS

Thermocouples are located in Tank C-106 at two locations to measure temperatures. One location is under riser 8 near the outer wall of the tank, and the other is under riser 14 near the center of the tank. At each location thermocouples are placed at three different depths within the waste (top, middle, and bottom) to obtain a temperature profile. In riser 8 there are 3 thermocouples in the waste and 3 in the vapor space above the waste. In riser 14 there are 3 thermocouples in the waste and 4 in the vapor space.

Thermocouples are designated as TC-1, TC-2, etc., beginning at the lowest level. Thermocouples numbered TC-4 and higher are located in the vapor space. In riser 14 there is no TC-7, but there is a TC-8.

MEASURED TEMPERATURES

Typical measured temperatures prior to cessation of water addition in March, 1994, are shown in Table 3.

Table 3. Historical Temperature Measurements

Riser	Measurement location above bottom, inches	Temperature (°F)
8	62 (TC-3)	115-120
8	38 (TC-2)	135-140
8	15 (TC-1)	150-155
14	64 (TC-3)	85-90
14	40 (TC-2)	95-100
14	16 (TC-1)	120-125

Temperatures for riser 8 were stable within historical seasonal variations around 155°F.

Temperatures for riser 14 showed a much greater change than those for riser 8 and were seen to oscillate. A graph of measured temperatures is shown in Figure 1.

The temperature profile for TC-1 located at the bottom of riser 14 is described as follows:

1. Immediately following water addition on June 17 the temperature increased from 123 to 135° F in about 1 hour (approximately 12° F/hr). This was followed by 2.5 days of much slower temperature rise to about 142° F (about 0.12° F/hr).
2. On June 19 the temperature dropped at a steady rate of about 1° F/hr until it reached 127° F. At this point it remained stable for several hours.

3. On June 21 the temperature began a steady rise lasting 6 days to 172° F (about 0.29° F/hr).
4. On June 26 the temperature dropped about 15° F in about 12 hours (about 1.3° F/hr). An important consideration to criticality safety is that the temperature began an abrupt drop indicating that heat is being removed and not generated.
5. On June 27 the temperature abruptly changed from decreasing to increasing and continued to increase at 1.3° F/hr until it reached about 170° F.
6. After this, for about 12 hours the temperature increased at a much slower rate of about 0.1° F/hr.
7. The temperature profile approximates steps 5, 6, and 7 above in a series of 6 oscillations.
8. After cessation of oscillations, the temperature continued a slow rise of about 0.05° F/hr.

The period of a cycle was seen to increase with each oscillation. At the same time the peak temperature slowly increased with each cycle.

C-106, Riser 14

APPENDIX D
CRITICALITY SAFETY ANALYSIS

PLUTONIUM CRITICAL PARAMETERS

To evaluate criticality safety it is necessary to know the value of certain important parameters which indicate whether or not criticality is possible. Before being used to justify criticality safety, the range over which the parameter can be varied while the system remains subcritical must be known. A parameter used to show that a system is critical is referred to as a critical parameter, and The value at which the system becomes critical is referred to as its critical value.

For purposes of criticality safety a subcritical limit is defined for each critical parameter. The subcritical limit is the largest (or sometimes the smallest) value of a parameter which will assure that a system is subcritical, after taking into account the uncertainties in determining it. In this discussion a "critical" value generally is taken to mean the subcritical limit value and will therefore describe a system that is actually subcritical. The neutron multiplication constant (either k_{eff} or k_{inf}) for subcritical limits used for this evaluation will not exceed 0.95 with a 95% confidence level.

^{240}Pu is fissionable, but not fissile. The presence of even a small quantity of moderating material increases the critical mass of this isotope very rapidly until criticality is not possible with it alone. In tank waste ^{240}Pu acts as a mild poison. Its presence increases the quantity of ^{239}Pu required to achieve criticality. Therefore, it is conservative either to ignore the presence of ^{240}Pu or to assume all plutonium is ^{239}Pu (i.e., to replace ^{240}Pu with ^{239}Pu).

A model of waste, known as the Conservative Waste Model, is described in *CSER 92-009: An Analytical Model For Evaluating Subcritical Limits For Waste in Hanford Site Storage Tanks* (Rogers 1993). Subcritical limits provided in that report are the basis for this evaluation. The primary goal of this model was to select a waste composition known to have a larger macroscopic absorption cross section than that of any real waste. Critical parameters are calculated using the assumption of optimal water moderation. In this way, subcritical limits calculated using the conservative waste model are conservative relative to the actual critical values for any real waste.

PLUTONIUM CONCENTRATION

Criticality is not possible for plutonium in a plutonium-water mixture unless the plutonium concentration exceeds 7.2 g/L. When plutonium is mixed with other materials in addition to water, criticality can become possible at lower concentrations. A lower critical concentration occurs when the total macroscopic absorption cross section per plutonium atom is less than for water. For criticality to be possible in tank waste according to the Conservative Waste Model, it is necessary for the plutonium concentration to be at least 2.6 g/L over a large volume. Alternately, criticality is possible with a higher plutonium concentration over a smaller volume.

For a sphere of waste having a plutonium concentration of 4.0 g/L to achieve criticality requires that the sphere be 15.4-ft in diameter. This sphere would have a volume of 53,750 L and contain 215 kg of plutonium.

For a slab of this waste to go critical it would have to be almost 7-ft. thick. The waste layer in Tank C-106 is less than 7-ft thick, and the minimum plutonium concentration required for criticality is therefore at least 4.0 g/L.

At 10 g Pu/L the critical mass in tank waste is approximately 3.0 kg of plutonium in a volume of 300 liters.

When the ^{235}U enrichment is less than that of natural uranium (0.72 wt%), the "always safe" limit value for the U/Pu mass ratio is 770. This is the reciprocal of 0.0013 for the Pu/(Pu + U) mass ratio provided by the *Nuclear Safety Guide* (Thomas 1978).

URANIUM

In a homogenized mixture of uranium in water, criticality is not possible until the enrichment exceeds 1.03 wt%. Criticality is possible at enrichments as low as 0.72 wt% for precisely defined heterogeneous arrays in moderators with low neutron absorption cross-sections (i.e. graphite, heavy water, beryllium), but such a system would be impossible to achieve in tank waste. Nearly all of the uranium sent to waste storage tanks has an enrichment less than 1.03 wt%. There is no practical chemical or physical means by which the ^{235}U can be separated from the ^{238}U . Because of the low enrichment criticality is not possible as a result of a chain reaction in the uranium.

CONDITIONS REQUIRED FOR CRITICALITY

Plutonium has been discharged to waste storage tanks at a concentration of less than 0.05 g/gal, or 0.013 g/L. Plutonium in the incoming waste would have to be concentrated more than 200 times before criticality becomes possible.

The probability of achieving a concentration even approaching the minimum critical concentration is very low. For criticality to become possible this concentration would have to be achieved over a large volume of waste. Not only does the plutonium concentration have to be high enough, but the quantity of plutonium within a limited volume must be large enough.

For criticality to occur a large quantity of plutonium would have to accumulate in a compact volume. In other words, a large proportion of the available plutonium would have to accumulate in a very small fraction of the waste volume. This configuration of plutonium would be extremely unlikely, if not impossible, to achieve, based upon the known mode of discharge into the tanks and the large ratio of solids to plutonium.

For a criticality to occur with 100 kg of plutonium requires that the plutonium concentration be greater than 4.0 g/L. A conservative estimate of the plutonium concentration in Tank C-106 solids based upon 3 samples is 0.127 g/L. For criticality to be possible in Tank C-106 the plutonium concentration would have to be increased by more than 31 times the highest measured value.

MODERATION

Criticality is not possible when the hydrogen to plutonium atom ratio exceeds 3660. When the water content is so large as to ensure subcriticality, a system containing fissile material is said to be overmoderated.

For criticality to occur with the removal of water, the waste would have to initially be overmoderated. Allowing the waste to dry out, as it would eventually if no water were to be added, would actually increase the reactivity. However, the waste is so far subcritical that the increase would have no significance to safety. Even with complete drying, the waste would remain well subcritical.

NEUTRON ABSORBERS

A compilation of component/plutonium mass ratios for waste in Tank C-106 is shown in Table 4. The subcritical limit for mass ratios for selected components is shown in Table 5 (Rogers 1993).

A comparison of the actual mass ratio to the subcritical limit for that mass ratio is a measure of ability of that component to ensure subcriticality. In a homogeneous mixture subcriticality is assured when the fraction obtained for this ratio exceeds 1.0. By adding the fractions for various components one obtains a measure of the degree of subcriticality for the waste taken as a whole.

Table 4. Mass Ratios of Solids and Liquids in Tank C-106.
(Based on average and high concentrations from Table 1.)

Component	Solids Mass Ratio Component/Plutonium		Liquids Mass Ratio Component/Plutonium	
	Average/High	Avg./Average	Average/High	Avg./Average
Boron	0.213	0.352	0.625	2.78
Cadmium	0.529	2.06	1.63	7.22
Iron	652.8	1076.	0.625	2.77
Manganese	89.5	147.6	12.5	55.5
Silicon	514.6	848.2	83.7	372.2
Sodium	858.4	1,415.	5,693.	25,300.
Uranium	2.25	3.70	35.0	155.5
NO ₂	---	---	609.4	2,708.
NO ₃	12.7	20.9	4,197.	19,650.

Table 5. Mass Ratio Subcritical Limit

Component	Absorber/plutonium mass ratio limit
Aluminum (Al)	910
Chromium (Cr)	135
Iron (Fe)	160
Nickel (N)	105
Nitrogen (N)	61
Nitrate (NO ₃)	270
Manganese (Mn)	32
Sodium (Na)	360
Silicon (Si)	1400
Uranium (U)	770
Water	150

The subcritical limit of the Fe/Pu mass ratio is 160, and the measured mass ratio is 653. The measured mass ratio is four times as large as the subcritical limit. Therefore, the concentration of iron in the solids is far more than required to ensure subcriticality. Iron is an important component to criticality safety and tends to stay associated with plutonium.

The subcritical limit of the Mn/Pu mass ratio is 32, and the measured mass ratio is 89, almost three times as large. Therefore, the concentration of manganese in the solids is far more than required to ensure subcriticality.

The sum of the fractions of the actual to limiting mass ratios for iron, manganese, sodium, and silicon is about 10. These four components alone therefore provide 10 times as much neutron absorption as is required to assure subcriticality in a homogeneous mixture.

Harris (1993) reports that the average solids density is 1.4 g/cm³ (1,400 g/L). Using the highest plutonium concentration of 0.127 g/L, the solids/plutonium mass ratio is found to be 11,020. This is 23 times greater than the subcritical limit for conservative model waste of 476.

It can be seen from the above information that the margin of subcriticality for Tank C-106 waste is large. At the very least a 31-fold increase in the plutonium concentration would be required for criticality to reach the minimum critical concentration of at least 4.0 g/L. However, the large contents of iron and manganese ensure that a greater increase would be required.