

# Nuclear Waste Management and Criticality Safety

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## NUCLEAR WASTE MANAGEMENT AND CRITICALITY SAFETY

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### ABSTRACT

Since 1945 waste streams containing radioactive isotopes have been discharged to underground storage tanks on the Hanford Site in Washington State. At least 49 different waste streams containing low concentrations of  $^{235}\text{U}$  and  $^{239}\text{Pu}$  have been transferred to tank farm facilities. Optimizing tank space, combined with the variability of operations, results in a complex waste matrix that is difficult to characterize. Characterizing tank waste is difficult because of its relative inaccessibility inside the storage tanks and because of the large degree of uncertainty in the composition and distribution of components. Nuclear criticality safety controls are based on precise configurations that are conservative when used to represent the waste environment. However, the safety philosophy governing waste storage requires that the waste be controlled and monitored, and the margin of safety be quantified.

### INTRODUCTION

Between 1944 and 1987, nine plutonium production reactors operated on the Hanford Site, converting a small fraction of the  $^{238}\text{U}$  in the uranium metal fuel into plutonium. The process of separating plutonium from the fuel was conducted in four chemical separations plants using three different processes: (1) bismuth phosphate, (2) REDOX and (3) PUREX. These processes were not 100% efficient, so each waste stream sent to underground storage contained plutonium in small concentrations. Although  $^{235}\text{U}$  is present, it appears almost entirely in depleted uranium for which criticality is not possible. For this reason, the primary fissile component of concern to criticality safety is plutonium. Limits applied to waste are commonly expressed in terms of the plutonium content.

Early separation processes discharged large quantities of waste to the tank farms. To reduce waste volumes, uranium was removed from waste with tributyl phosphate. In this process waste was pumped from the storage tanks into a process building where the uranium was removed. It was pumped back to tank farms into a different tank. This extensive amount of intra-tank pumping increased the difficulty of keeping track of the plutonium inventory in tanks.

Because of the potentially serious consequences, preventing criticality was considered a fundamental priority from the first transfer of waste to storage. Criticality safety concerns in the processing plants are well documented. Information provided in historical documents report the always safe ratio of plutonium-to-uranium at 0.0018 as a basis for criticality control. Specifications based on this subcritical limit were set at a maximum ratio of 0.0017, or 1500 g Pu/ton of U [1]. A major difficulty in verifying today that this limit was always met is finding data sheets that identify the plutonium content of waste streams discharged to the underground storage tanks.

Although information in historic documents allude to their existence, criticality safety evaluations and calculations before 1979 about waste storage have not been found. The 1979 Criticality Safety Analysis Report (CSAR) [2] reviewed both critical slabs and spheres of waste. Based on samples from four tanks, the CSAR concluded that a criticality in waste was impossible at concentrations less than 3.0 g Pu/L of solids. Conservative specifications established the limit on plutonium in waste streams at 0.05 g Pu/gal of waste. At that time a limit of 125 kg was placed on the total mass of

plutonium permitted in a waste tank, and the concentration of plutonium was limited to 1 g Pu/L in settled solids. This CSAR did not provide a comprehensive analysis of ways that plutonium might become more concentrated within the waste matrix. Since then, a more in-depth evaluation of concentrating mechanisms has been made.

Criticality safety analysts are sometimes asked to quantify the confidence that an adequate margin of safety exists. However, to determine the conditions by which plutonium will become chain reacting requires that the composition of the waste and the spatial distribution of components be known. This is a seemingly impossible task because empirical data on all locations in all 177 waste tanks do not exist and historical data is of limited use.

A casual observer might see the safety of tank storage was demonstrated by the 50 years it was stored without criticality. The practitioner, however, understands that 50 years of accumulation, combined with future plans for retrieval and in-tank pretreatment, require an aggressive program to establish margins of safety.

## DISCUSSION

In 1992, declaration of an Unreviewed Safety Question (USQ) related to criticality safety centered on the lack of defined confidence in the Hanford Site waste tank fissile material inventory and the lack of data on the distribution of fissile material in the waste tanks. The possibility of a localized region of higher concentration exceeding the concentration limits could not be discounted. The USQ was closed in 1994 after hundreds of sample analyses were reviewed and the conclusion was reached that a distribution of plutonium capable of criticality could not be achieved under normal operations. Even after closure of the USQ, however, concerns regarding the distribution of fissile material in the waste have continued. Accordingly, the basis for criticality safety control in the tank farms was reviewed and updated.

The first step taken to upgrade the Criticality Safety Program was to reevaluate the minimum critical parameters applied to tank waste. Rogers [3] developed a conservative model based on chemical analyses of 28 waste samples. The sole purpose of this model was to provide a basis for the calculation of conservative critical parameters. Conservatism was assured by selecting the maximum concentrations of components that are good neutron scatters and poor neutron absorbers, and minimum concentrations of components that are good neutron absorbers. The primary goal is to provide a waste composition known to have a smaller macroscopic absorption cross section than does any real waste.

Based on the conservative waste model, the minimum critical plutonium concentration in an infinite system is 2.6 g/L in settled solids. Subcritical parameters were calculated using optimal water moderation and are therefore independent of the actual water content. The density of the solids was conservatively assumed to be 1200 g/L, a value smaller than expected to occur. If a plutonium concentration of 2.6 g/L is not exceeded at any location, criticality is not possible, even if the waste is not homogeneous.

The plutonium content in relationship to the solids content is an important parameter for criticality safety, and it is perhaps the easiest parameter to control to ensure subcriticality. For this reason, limits used in the criticality prevention specifications (CPS) are based on the ratio of solids to plutonium.

The alkalinity of the waste keeps the solubility of plutonium low. Hobbs et al. [4] reports an upper limit on plutonium solubility in alkaline salt solutions representative of tank waste to be 0.017 g/L. The CPS for the waste tanks limits the concentration of plutonium in discharged waste to not more than 0.033 g/L (0.125 g/gal) in the waste. At the same time the solids-to-plutonium mass ratio in a batch before discharge must be at least 1000. In reality, the solids content averaged over many discharges is expected to be much greater than this minimum permitted value. When the plutonium inventory in a tank exceeds 10 kg, the tank averaged solids-to-plutonium mass ratio must be greater than 5000.

The limit on total plutonium mass has been eliminated. A plutonium tracking spreadsheet has been developed, which provides a continuous inventory, including the solids-to-plutonium mass ratio, for all double-shell waste receiver tanks.

Resolving concerns about the plutonium inventory and distribution require determining how much plutonium is in the storage tanks. Two estimates were made for the total plutonium sent to tank farms. The first by Roetman et al. [5], based on the average exposure of monthly fuel discharges in the reactors versus the chemical process efficiencies, estimates 981 kg. The second, by Tusler [6], based on samples and flow sheet compositions, estimates 1078 kg. The results of these two independent activities are remarkably close. Quantified confidence in the accuracy of the plutonium inventories, however, has not been established. In addition, there are still questions about the distribution of fissile material that have not been addressed.

Characterization of tank waste is best accomplished by obtaining full-depth core samples and completing chemical and radiological analyses on them. Caution must be exercised in evaluating core sample data packages for plutonium inventory purposes. It is important to take into account the method used in the analysis when interpreting the results. The plutonium content of solids that have been centrifuged and also the plutonium content when the solids are dissolved in the waste have been found to be higher than for samples of settled solids. We think that the process used to obtain the data might show a higher plutonium content than would ordinarily occur in solids allowed to settle. For example, the highest plutonium concentration found in settled solids is 0.35 g/L, whereas the highest concentration found in centrifuged solids is 0.697 g/L.

Tank averaged solids-to-plutonium mass ratios provide added assurance for the margin of safety for a waste tank. However, the total quantity of solids in the tanks or waste discharge streams is not always known. Nevertheless, it is also possible to develop subcritical limits based on the mass ratio for specific components of the waste. Rogers [7] reports the subcritical absorber-to-plutonium ratios for several individual elements. Some important ones are: U/Pu = 770, Fe/Pu = 160, Mn/Pu = 32, Al/Pu = 910, and Cr/Pu = 135. These minimum subcritical ratios may be used to verify compliance with storage and discharge limits. Regardless of the plutonium concentration, a criticality can not occur if the ratio of the concentration of specific neutron absorbers to the concentration of plutonium is maintained larger than the subcritical limit.

Although data from samples is an important tool, the ability of obtain this data is limited by the number of access ports to the tanks. An effort is underway to identify an alternative means of measuring component inventories (specifically plutonium) and distributions in the tanks. Measurements have been made using passive foil, passive detector scans, active detector scans and gas sampling, but no conclusions have been reached as to the utility for using these methods. The estimated total quantity of plutonium in the waste storage tanks provides an inventory boundary for continuing analysis; however, the distribution of waste components remains undefined.

An effort has been made to identify mechanisms that could preferentially concentrate plutonium in localized volumes of a waste tank. The impact of exceeding discharge limits, settling of plutonium, and evaporation and chemical separation on plutonium concentration was evaluated by Rogers [7]. The waste will remain subcritical, unless the plutonium concentration increases above the minimum for which criticality is possible. The plutonium concentration is

capable of increasing above the discharge limits primarily through the settling of the plutonium bearing solids. However, criticality is not possible unless the plutonium areal density exceeds 2.6 kg/m<sup>2</sup> (240 g/ft<sup>2</sup>). To achieve this average areal density over the large area of a storage tank requires more than 1,000 kg of plutonium. There is no current data to suggest any tank contains anywhere near this amount of plutonium. Rogers concludes that a criticality cannot occur, because no credible concentrating mechanisms capable of achieving a minimum critical concentration over a sufficiently large volume to achieve criticality can be identified.

An assessment of the margin of safety in the tanks is documented in SARR-003 [8]. Analytical data provided in this report was taken from nearly all of the tanks. The 960 samples are arranged according to waste type (sludge, saltcake or supernate) and origin (e.g. PUREX, Plutonium Finishing Plant (PFP)). The following conclusions were drawn from this data.

- The highest plutonium concentration is found in the sludge. The highest individual concentration found is 7 times less than the minimum concentration for which a criticality is possible.
- The water content is generally higher than that which results in the minimum critical plutonium concentration in the waste model. In other words, tank waste is usually overmoderated.
- The solids content in waste contains sufficient absorbers to ensure that criticality cannot occur.

The data strongly supports the supposition that localized concentrations do not exceed the minimum for which criticality is possible. Even though complete characterization of tanks is not available and the waste is generally heterogenous, the hundreds of samples that are now available do provide convincing evidence that the waste is well subcritical and will remain so in the future.

## RESULTS

The increased attention given to the safe storage of high-level tank waste has resulted in a review of tank limits and controls. The 149 single-shell tanks are inactive, highly subcritical, and will never receive more fissionable material. For these reasons, no limits are required on plutonium concentration and/or total mass in these tanks. The only control placed on single-shell tanks is the requirement that activities that may compromise the form and distribution of the fissile material (e.g. dewatering, sampling, contamination

control) be evaluated and approved before they are carried out. A list has been compiled of permitted operations. Any future operation that is not on the list must be evaluated before the activity can be initiated.

For the double-shell tanks, no limit is placed on the plutonium inventory. This is acceptable because the tank inventory is obtained directly from the plutonium concentration, and there is no acceptable way to verify compliance to a mass limit independent of measurements of plutonium concentration. It is redundant to place a limit on both parameters. Limits have been established on pH, on plutonium concentration and on the solids-to-plutonium mass ratio.

A program of surveillance and monitoring of waste in the processing plants before discharge and also in storage tanks has been in place since fuel processing at the Hanford Site began. Periodic audits and appraisals ensure operations are conducted according to requirements and identify nonconforming conditions. A process for investigation and recovery from any nonconforming condition is in place.

Concerns about the quantified accuracy of the margins of safety will continue to be addressed. Characterization data will play an important role in waste retrieval, pretreatment and disposal activities. Increased concern for the criticality safety of tank waste containing low concentrations of fissile material will ensure that adequate knowledge is available to address questions that arise as future environmental restoration operations or processes are undertaken.

#### REFERENCES

1. Clayton, E. D. and W. A. Reardon, 1961, *Nuclear Safety and Criticality Of Plutonium*, HW-71666, General Electric, Richland, Washington.
2. Carter, R. D., 1989, *CSAR 79-007: Underground Waste Storage Tanks and Associated Equipment*, SD-SQA-CSA-20108, Westinghouse Hanford Company, Richland, Washington.
3. Rogers, C. A., 1993, *CSER 92-009: An Analytical Model For Evaluating Subcritical Limits For Waste in Hanford Site Storage Tanks*, WHC-SD-SQA-CSA-20356, Westinghouse Hanford Company, Richland, Washington.
4. Hobbs, D. T., T. B. Edwards, and S. D. Fleishman, 1993, *Solubility of Plutonium and Uranium in Alkaline Salt Solutions (U)*, WSRC-TR-93-056, Savannah River Technology Center, Westinghouse Savannah River Company, Aiken, South Carolina.
5. Roetman, V. E., S. P. Roblyer, and H. Toffer, 1994, *Estimation of Plutonium in Hanford Site Waste Tanks Based on Historical Records*, WHC-EP-0793, Rev. 0, Westinghouse Hanford Company, Richland, Washington.
6. Tusler, L. A., 1995, *Double Shell Tank Plutonium Inventory Assessment*, WHC-SD-WM-TI-640, Westinghouse Hanford Company, Richland, Washington.
7. Rogers, C. A., 1994, *CSER 94-004: Criticality Safety Of Double Shell Waste Storage Tanks*, WHC-SD-SQA-CSA-20368, Westinghouse Hanford Company, Richland, Washington.
8. Braun, D. J., L. D. Muhlestein, T. B. Powers, and M. D. Zentner, 1994, *High Level Waste Tank Subcriticality Safety Assessment*, WHC-SD-WM-SARR-003, Rev. 0, Westinghouse Hanford Company, Richland, Washington.