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Incredibility of a Postulated Criticality Event for Enriched Uranyl Nitrate Storage

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INTRODUCTION

At the H-Canyon outside facilities, highly enriched uranyl nitrate solutions collected from leaks or during deliberate maintenance activities is collected into 5-gallon polyethylene bottles. The current criticality safety evaluation (Ref. 1) for the facility postulates that a criticality event is possible if the solutions are allowed to concentrate through evaporation, freezing, storing multiple bottles in a geometrically unfavorable array, or any combination of these conditions. Using chemical data on uranyl nitrate freezing from Savannah River National Laboratory (Ref. 2 - 6), protected limits on concentration and enrichment, and normal operational restrictions, criticality was shown to be incredible under all normal and credible abnormal conditions.

A bounding normal solution is analyzed under normal conditions and shown to be substantially subcritical. A bounding credible abnormal solution is defined as 9.2 g U-235/L and 73 wt. % enriched and 0 M excess nitric acid. The concentration and enrichment are limitations already protected by the facility for other processes. Because acidity is not specifically restricted, the most reactive lower bound is selected. The credible abnormal worst placement is in a concrete basin reflected on three sides. There the bottle(s) would be subject in the worst cases to flooding (full water reflection) and postulated concentration through evaporation or freezing.

Computational modeling will be performed using KENO-VI in the SCALE 6.1 code package.

DESCRIPTION OF THE WORK (HEADING A)

Modeling of the 5-gallon Storage Bottle

The 5-gallon storage container is a commercially available polyethylene bottle with a specially printed circumferential label indicating ¼, ½, and ¾ full with ¾ full indicated as the “Haz Fill Line”. The outer dimensions of the bottle are nominally 15” tall and 11” diameter; however it is not a right circular cylinder with the lower 2.25” tapered. There is an indentation on the bottom approximately 0.5” deep to accommodate the molded head of a second container to allow for stacking. The head also includes a molded pour spout which sits below the top of the bottle thus ensuring an air space to allow for volume expansion of the contents during freezing. Measured volume of the container was 5.13 gallons using room temperature tap water and calculated to be 5.19 gallons. Conserving volume, the geometry nuances are accounted for and the bottle is modeled as a right circular cylinder retaining the same outer diameter and adjusting the height to 14.34” to maintain the container volume.

Normal Conditions

The normal solution that may be collected and temporarily stored in the container is 7 g U-235/L, ≤73 wt. % enrichment, and ≤0.4 M excess nitric acid. There is no requirement on the acidity and the lower extreme of 0 M excess nitric acid is the most reactive condition.

This analysis also considered real operational restrictions. The normal operation is to fill the container no further than the labeled ¾ full line. This maintains the 35 pound lifting limit for the worker and allows a single operator to transport the container by hand and pour it into an existing storage tank. Once the container is filled it is stored while the solution is characterized, usually 72 hours but procedurally required to be less than 28 days. There is no requirement on storage location but it is usually on a concrete pad out of direct exposure to weather. The most reactive place available to store the bottle is outside in a basin where it would have concrete reflection on three sides. Finally, procedures limit the operation to a maximum of 9 containers, though in practice no more than 4 have ever been needed at one time for any evolution.

The bounding normal conditions modeled for the bottle are ¾ full of a 7 g U-235/L, 73 wt.% enriched (9.589 g total U/L), 0 M excess nitric acid uranyl nitrate solution. This is stored on a concrete pad surrounded on three sides by 24 inches of concrete with concrete boundary conditions applied beyond that and on three sides by 24 inches of 300 K air with vacuum boundary conditions applied beyond that. The 24 inches is the length extended beyond the outermost edge of the bottle or array. Analyses are performed using a single bottle and 4 bottles modeled in the closest fit array (11” triangular pitch).

Credible Abnormal Conditions

The credible abnormal solution is 9.2 g U-235/L at 73 wt. % enrichment and both of those parameters are protected by other limits for facility operation. The limiting acidity is 0 M excess nitric acid. There are two credible abnormal arrangements: a single container that has been overfilled past the ¾ limit to the top of the pour spout or an array of more than 9 normally filled units. Multiple overfilled containers are not considered a credible condition. Thus the credible abnormal outside conditions are the containers, in the basin, being flooded (complete water reflection on three sides), or being exposed to air and allowed to concentrate through evaporation or freezing.

Flooding and Evaporation

For the flooding scenarios, only two cases were necessary. First a single overfilled container is placed in a flooded basin. Second, a 3x3x1 closest fit array of normally filled containers is placed in a flooded basin. The basin is modeled as a concrete pad having three sides of 24 inches of concrete with concrete boundary conditions applied beyond that and three sides of 24 inches of 300 K water with water reflection boundary conditions applied beyond that.

For the evaporation scenarios, the single overfilled bottle or array of normally filled bottles are placed on a concrete pad surrounded on three sides by 24 inches of concrete with concrete boundary conditions applied beyond that and on three sides by 24 inches of 300 K air with vacuum boundary conditions applied beyond that. Evaporation is modeled by removing discrete amounts of water (fraction of original volume) leaving behind a more concentrated uranyl nitrate solution with the uranium mass preserved. The solution retains the same diameter but has an increasingly shorter height as evaporation occurs. The arrays modeled are a 3x3x1, 3x3x2, 5x5x1, 10x10x2, 14x14x3, and an infinite array. The infinite array is modeled as a container with periodic boundary conditions on the top and bottom faces and mirror boundary conditions on the axial surface.

Freezing with Uranium Entrainment in Ice

Rather than assuming an increasingly more concentrated solution in the freezing scenario, chemical data from Savannah River National Laboratory was used to account for uranium entrainment in the ice. The SRNL data (Ref. 2 – 6) spanned the range of 0.363 to 0.955 volume fraction of the original solution frozen. Freezing took on the order of hours to days for volumes of less than 400 mL and indicated a freezing point of ~26 F, making the scenario itself unlikely due to the South Carolina environment. Fig. 1 presents the accumulated data from these reports showing a fraction of the uranium is entrained in the ice during freezing. The SRNL data also provided two other important conclusions. First, freezing occurs radially uniform and mostly top down due to those being the exposed surfaces. This physically restricts the geometries possible and helps to isolate the more concentrated solutions into the lower center of the containers in a freezing array. Second, the relative fraction of uranium entrained in a given volume fraction of ice does not vary with the initial volume or initial concentration of solution provided that solution is homogenously mixed to begin with. Therefore the SRNL data can be used to estimate the relative fraction of the total uranium entrained in the ice during freezing for the solution being examined. This chemical process segregates a fraction of the fissile material from the solution into a lower density region of the container.

For the freezing scenarios, the single overfilled bottle or array of normally filled bottles are placed on a concrete pad surrounded on three sides by 24 inches of concrete with concrete boundary conditions applied beyond that and on three sides by 24 inches of 270 K air with vacuum boundary conditions applied beyond that. Freezing is modeled by selecting a fixed volume fraction of the original solution to freeze. The fraction of the uranium entrained in that volume is interpolated from the SRNL data. The liquid portion is modeled as a cylinder of concentrated solution with the same height to diameter ratio as the original unfrozen solution and in contact with the bottom of the container for conservatism (additional reflection). The solid is modeled as an annulus around the liquid and a disk above it, and its volume expanded due to the difference in density between water and ice. Expansion is accommodated completely by the head space in the container for all cases.

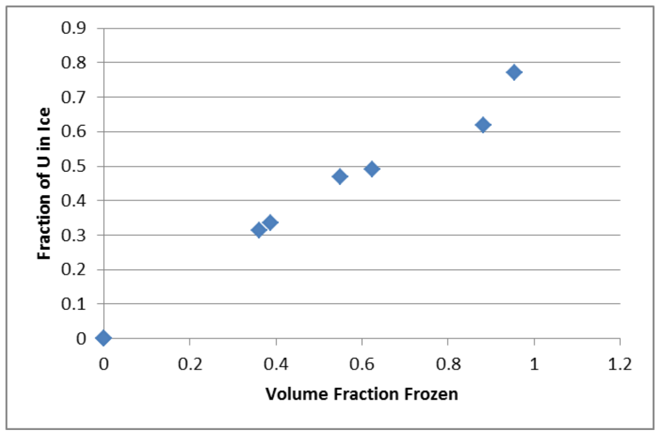

Fig. 1. Faction of solution uranium entrained in ice per the volume fraction of the initial solution frozen.

RESULTS

All multiplication factor values discussed below are the calculated eigenvalue of the system (k-calc) plus two times the Monte Carlo uncertainty (sigma). Models were run with sufficient histories to drive Monte Carlo uncertainties below 0.0005 k for all cases.

The normal conditions had multiplication factor of 0.343 for the single bottle and 0.428 for the 4 bottles. The credible abnormal flooded conditions were not much higher at 0.490 for the single container and 0.604 for the 3x3x1 array of bottles.

Evaporation for a single overfilled bottle and for finite sized arrays of normally filled bottles increased multiplication only slightly with evaporation up to approximately 50 volume % evaporated and then fell off rapidly (Fig.2). While procedurally limited to 9 bottles, conservative larger arrays of 18 (3x3x2) and 25 (5x5x1) bottles were also modeled in the event procedures are changed in the future. The 18 bottle case also accounted for 2 level stacking. Only with an infinite array, where the areal density of the concentrated solution disks became very large, did the system diverge to supercritical states with evaporation. Arrays of 200 (10x10x2) and 588 (14x14x3) bottles were modeled and only with the larger array did the finite system begin to show characteristics of the infinite array. It is beyond extremely unlikely that 588 or even 200 bottles would be used.

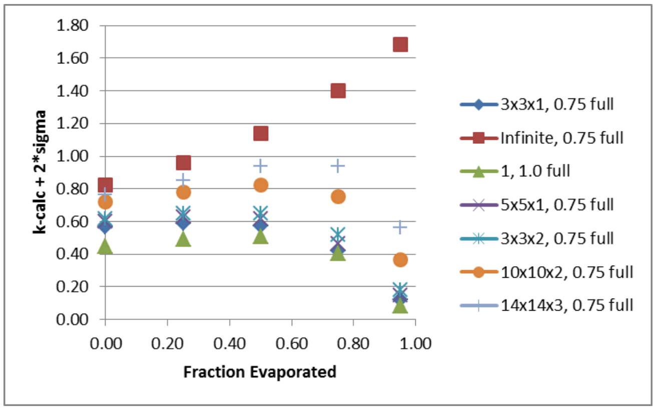

Fig. 2. Multiplication factor of the system subject to evaporation per the fraction of the initial volume evaporated.

Multiplication factor due to freezing conditions for the infinite array varied little over whole range of freezing examined. For the single overfilled bottle and the 3x3x1 array of bottles, there was a slight increase in multiplication factor at 75 volume % frozen which is attributed to a more reactive combination of reflection, moderation and concentration, which did not persist as freezing continued. No larger finite arrays were evaluated since the infinite array multiplication factor was less than 0.83 (Fig. 3).

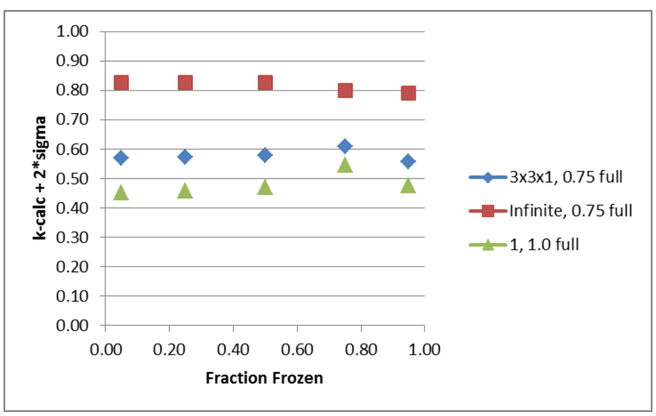

Fig. 2. Multiplication factor of the system subject to evaporation per the fraction of the initial volume evaporated.

Since the safe value of the multiplication factor (with some margin applied) for highly enriched uranium solution systems would be 0.956 (Ref. 7), no normal or credible abnormal condition challenges criticality. These results show the postulated criticality event is not credible which contributes to simplification of the facility safety basis.

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