REFERENCE 180

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5. Critical Experiments with Arrays of Three-Litre Bottles Filled with $Pu(2.8)(NO_3)_4$, *B. M. Durst, E. D. Clayton, J. H. Smith (PNL)*

The safe storage of nuclear fuel is a prime concern of the nuclear fuel cycle worldwide. In the reprocessing of nuclear fuels, such as in the Purex Facility at Hanford, plutonium nitrate solutions are commonly stored in stainless steel or polyethylene cylindrical bottles, and yet to date, an experiment has never been performed determining the effect on criticality of spacing or moderation between bottles.

To address this problem, Rockwell Hanford Operations recently initiated a series of experiments at the Battelle Critical Mass Laboratory with arrays of bottles containing plutonium nitrate solution. The experiments were performed with 3-ℓ polyethylene bottles containing plutonium with a ^{240}Pu content of 2.8 wt%. The objective of these experiments was to provide benchmark data to check calculational codes used in the criticality safety program at the Purex Facility. The experiments addressed a number of factors affecting criticality, namely, (a) the critical air gap between bottles in an array of a fixed number of bottles, (b) the number of bottles required for criticality if the bottles are touching, and (c) the effect on critical array spacing and critical bottle number due to the insertion of a hydrogenous substance into the air gap between bottles.

The experiments were performed with sixteen 3-ℓ polyethylene bottles filled with $Pu(NO_3)_4$. The fissile solution had an average concentration of ~117 g/ℓ plutonium, with the ^{240}Pu content being 2.8 wt% at a free acid molarity (H^+) of ~6.7. The solution specific gravity was 1.421. The bottles were Plexiglas reflected on all sides and bottom, as shown in Fig. 1. The top was left unreflected. Bottles were positioned with the help of an aluminum framework that could be moved accurately in both the x and y directions. Four tiny aluminum pins securely held each bottle in place to prevent tipping and bottle movement.

Criticality was approached by loading about half of the critical array on each side of a split table device. The fuel loading adjustment was made by hand, and, after each subsequent change to the system, the table was closed remotely, completing the array geometry desired. The critical approach method was used to measure the desired critical spacing or critical bottle number, depending on the experiment.

After the initial series of experiments was performed with bottles separated by air gaps, Plexiglas shells of varying thicknesses were placed around each bottle to investigate how moderation between bottles affects both the number of bottles required for criticality and the critical spacing between each bottle.

The experiment results are graphically summarized in Fig. 2. The minimum number of bottles required for criticality was found to be 10.9 bottles, occurring for a square array where bottles were touching one another. As the bottles are spaced apart, more bottles were needed to remain critical. For 16 bottles in a square array, the critical separation between bottle surfaces in both x and y directions is 0.96 cm. The addition of Plexiglas around each bottle decreased the critical bottle number, compared to those separated only by air, but the critical bottle number, even with interstitial plastic in place, was always >10.9 bottles. The most reactive configuration was a tightly packed array of bottles with no intervening material.

As a result of these experiments, calculational benchmarks now exist for criticality analysis of bottle arrays of plutonium nitrate solutions. The experiments illustrate that subcritical mass in an array can be substantially increased if bottles are sufficiently spaced apart. However, if spacing is to be used to ensure criticality safety, the effects of any intervening hydrogenous material must be carefully considered, since this will reduce the margin of safety gained by spacing.

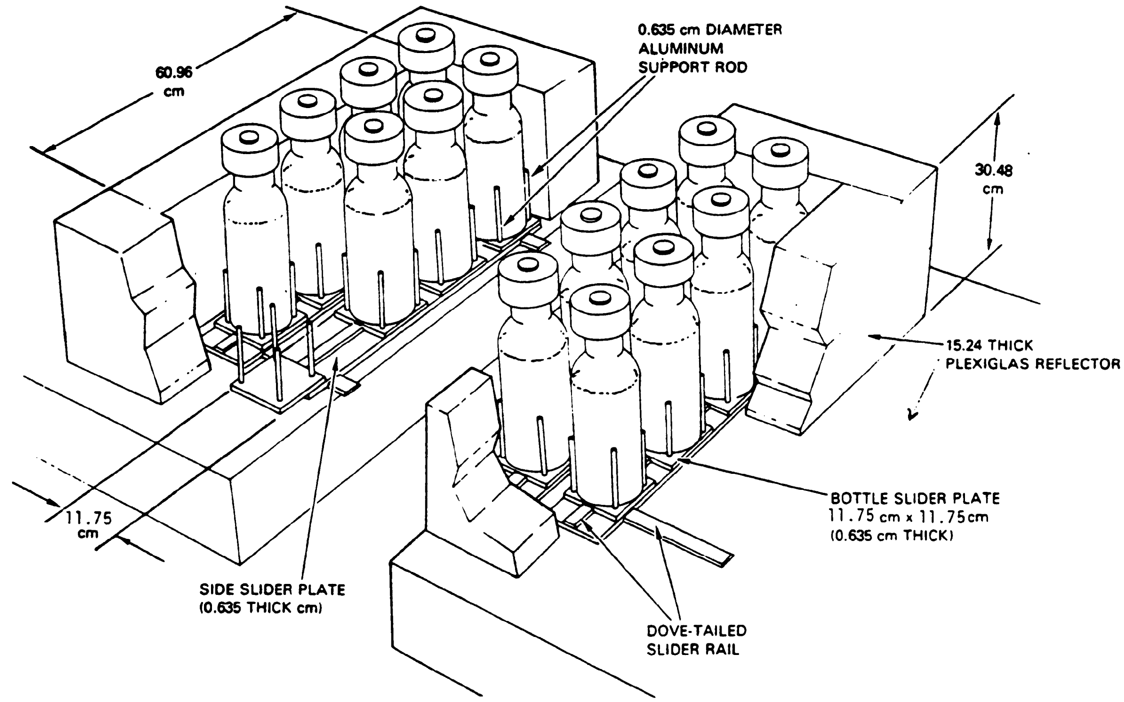

Fig. 1. Isometric view of interacting 3-ℓ bottles.

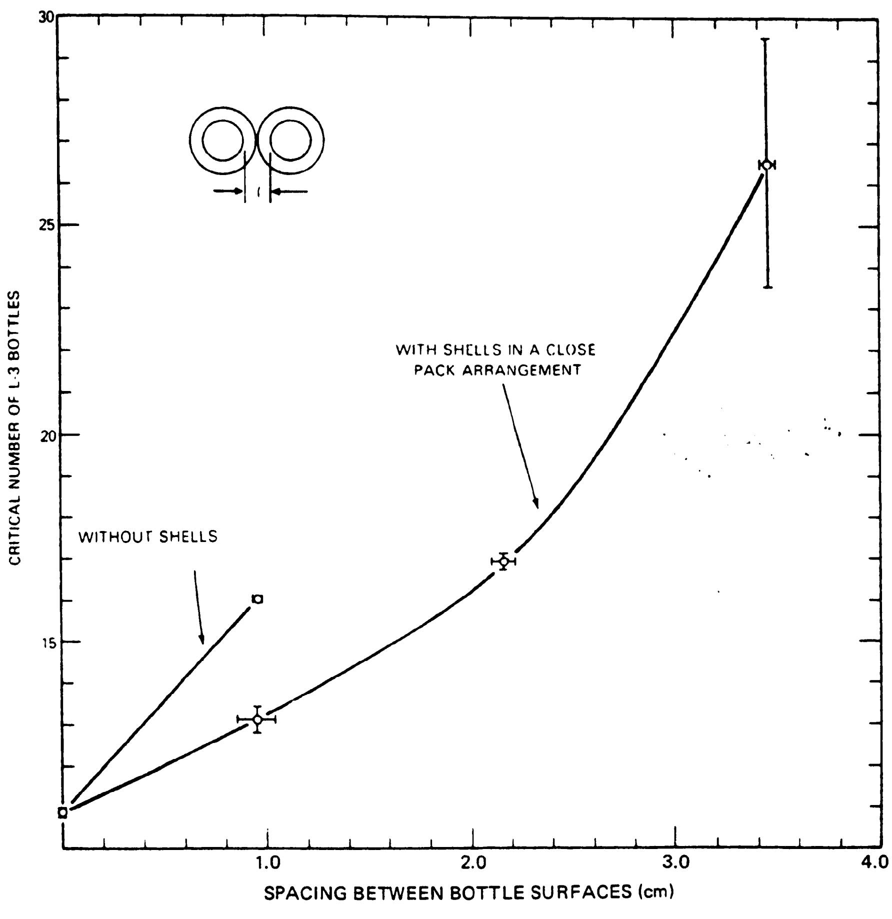

Fig. 2. The effect of Plexiglas shells on critical number of 3-ℓ bottles.