Modernization at the Y-12 National Security Complex: A Case for Additional Experimental Benchmarks

M. L. Thornbury, Consolidated Nuclear Security, Y-12 National Security Complex
C. Juarez, Consolidated Nuclear Security, Y-12 National Security Complex
A. W. Krass, C. S. Engineering, Inc., subcontractor to CNS, LLC at the Y-12 National Security Complex

INTRODUCTION

The form and composition of enriched uranium at intermediate stages of the un-irradiated nuclear fuel cycle strongly influence the technical basis for nuclear criticality safety of each unit operation. For example, the uranium enrichment of gaseous UF_6 gives rise in the late 1940s to an interest in the nuclear properties of $^{235}UO_2F_2$ in aqueous solution [ref. 1]. Similarly, the purification of fissile materials by solvent extraction gives rise in the early 1950s to an interest in the nuclear properties of $^{235}UO_2(NO_3)_2$ in aqueous solution [ref. 2]. The evolution of these, and related, technologies on an industrial scale sustains an experimental criticality program in the United States well into the 1980s.

Efforts are underway at the Y-12 National Security Complex (Y-12) to modernize the recovery, purification, and consolidation of un-irradiated, highly enriched uranium metal. Successful integration of advanced technology such as Electrorefining (ER) eliminates many of the intermediate chemistry systems and processes that are the current and historical basis of the nuclear fuel cycle at Y-12. The cost of operations, the inventory of hazardous chemicals, and the volume of waste are significantly reduced by ER. It also introduces unique material forms and compositions related to the chemistry of chloride salts for further consideration in safety analysis and engineering. The work herein briefly describes recent investigations of nuclear criticality for $^{235}UO_2Cl_2$ (uranyl chloride) and $^{6}LiCl$ (lithium chloride) in aqueous solution. Of particular interest is the minimum critical mass of highly enriched uranium as a function of the molar ratio of ^{6}Li to ^{235}U. The work herein also briefly describes recent investigations of nuclear criticality for ^{235}U metal reflected by salt mixtures of $^{6}LiCl$ or $^{7}LiCl$ (lithium chloride), KCl (potassium chloride), and $^{235}UCl_3$ or $^{238}UCl_3$ (uranium tri-chloride). Computational methods for analysis of nuclear criticality safety and published nuclear data are employed in the absence of directly relevant experimental criticality benchmarks.

Computer code validations in current use at Y-12 for highly enriched uranium cover a wide variety of materials, geometry, and neutron spectra. However, the current lack of experimental criticality benchmarks that involve chlorides as the primary constituents necessitates reliance on a narrowly defined area of applicability and gross conservatism in the derivation of nuclear criticality safety limits. The consequence is often complex or awkward arrangements of fissile material and equipment whose design inhibits innovation and efficiency. Although the current state of the art in neutron transport codes is more advanced than previous eras, the need persists for additional experimental criticality benchmarks that are relevant to the current and future technology of a modernized Y-12.

CALCULATIONAL WORK

The chemical form, mass, distribution, and nuclear properties of ^{6}Li and Cl play a vital role in the design of equipment and safety analysis of the ER system. Neutron scatter and absorption by these nuclei over broad energy spectra greatly influence nuclear criticality in both moderated and unmoderated systems of ^{235}U. The critical radii (k_{eff} = 1.0) of simple material distributions in spherical geometry are estimated with discrete-ordinate computer code calculations (XSDRN) to yield basic criticality safety information. Each estimate is confirmed to produce a calculated $k_{eff} = 1.0$ with two additional Monte Carlo computer code calculations (MCNP and KENO V.a) for identical specifications of material, geometry, and dimension. Due to the lack of experimental data associated with ^{6}Li as a dissolved absorber in critical systems of aqueous solutions, a novel corollary with dissolved ^{10}B is also briefly explored.

RESULTS

The calculated critical mass and volume relationships of uranium in aqueous solutions of $^{235}UO_2Cl_2$ and $^{6}LiCl$ with spherical geometry and full water reflection for $^{6}Li/^{235}U$ ratios of 0, 0.1, 0.2, 0.3, and 0.4 are shown in Figure A-1. Their similarities and differences are made clear by comparison to the available critical data for homogeneous mixtures of U(93.2)[†] metal in water and aqueous solutions of $U(93.2)O_2F_2$ (uranyl fluoride) [Figures 6 and 7 of ref. 3], for non-uniform distributions of U(93.2) metal in water [ref. 4], and for homogeneous mixtures of $U(93.2)O_2$ in water [Table 4 of ref. 5]. Such materials provide useful context because they are the computational and experimental basis for many of the subcritical ^{235}U limits in common use from the ANSI/ANS-8 standards.

Each curve for a given $^{6}Li/^{235}U$ ratio in Figure A-1 reveals the familiar characteristic that the spherical critical mass and the spherical critical volume achieve absolute minimum values. These minimum values increase significantly with small increases in the ^{6}Li content. The mass relationship is further quantified in Figure A-2 along with the

---

[†] The notation U(93.2) signifies un-irradiated uranium enriched to 93.2 percent ^{235}U, by weight.

results for the "equivalent" solutions that arise from a substitution of one-fourth as many ^{10}B atoms for every ^{6}Li atom. Even in the absence of ^{6}Li in a uranyl chloride solution (i.e., $^{6}Li/^{235}U$ ratio = 0), the effect on the critical dimensions of a system is significant. For example, it is seen in Figure A-1 that the minimum critical volume of an aqueous uranyl chloride solution (~9.6 liters) is about 50% larger than that of an aqueous uranyl fluoride solution (~6.4 liters). This is partly due to the effect of mild neutron absorption in chlorine and partly due to the effect of a lower uranium density in the structure of the uranyl chloride complex.

The calculated total critical mass of uranium in systems of ^{235}U metal (ρ=18.81 g/cm^3) reflected by various materials in spherical geometry is shown in Figure A-3. The various materials represent the principal constituents of the ER salt mixtures. Their similarities and differences are made clear by comparison to the available critical data for un-moderated U(93.5) metal with water or graphite reflection [Figure 42 and Table 28 of ref. 6]. Each curve for a given material in Figure A-3 reveals the familiar characteristic that the spherical critical mass of ^{235}U metal decreases as the amount of neutron reflection is increased until a "saturation" thickness, or mass, is achieved. The curves for $^{235}UCl_3$ and $^{238}UCl_3$ diverge upward because the vertical axis represents the total mass of uranium to include both the ^{235}U metal in the core plus the ^{235}U or ^{238}U content of the reflector. In the case of $^{238}UCl_3$, the additional reflector material beyond saturation continues to increase the total mass of uranium in the system, but no longer decreases the critical mass of ^{235}U metal in the core. In the case of $^{235}UCl_3$, the additional reflector material continues to decrease the critical mass of ^{235}U metal in the core, but only at the expense of a much larger mass with a lower density of ^{235}U in the reflector (i.e., the reflector material has a much larger spherical critical mass of its own without need of a ^{235}U metal core). The results suggest that the constituents of the ER salt mixtures are very inefficient neutron reflector materials, and that the effect is significant. For example, it is seen in Figure A-3 that the critical mass of a system with 40 to 50 kg of salt reflection (~36 to ~70 kg total U) increases by about 50% to about 190% larger than that of a system with 40 to 50 kg of water or graphite reflection (~24 kg total U). Furthermore, none of the salt constituents decrease the total critical mass to a value below that of metal reflected with ordinary water.

CONCLUSIONS

The investigations of this work represent unique material compositions and distributions of great importance to the technical basis of nuclear criticality safety of a modernized Y-12. Sufficient evidence exists to support the conclusion that available criticality data, methods, and standards that are based on past practices and experimental benchmarks remain useful. However, gross conservatism inherent in their application to the design and deployment of advanced technologies such as Electrorefining severely limits the potential benefits to operational safety and efficiency. The consequence of unnecessarily restrictive component dimensions and mass limits include: frequent periods of shutdown for recovery and disposition of fissile material by-products; an increased frequency of unit operations, some of which are hazardous (e.g., heavy lifts, hot or electrically energized equipment manipulations, fissile material movements, and the like); an increased number and variety of individual fissile material process containers; a need for increased fissile material storage capacity; etc. The opportunities for process improvement are constrained without additional experimental criticality benchmarks that are relevant to the current and future nuclear fuel cycle of un-irradiated, highly enriched uranium.

REFERENCES

1. C. K. Beck, A. D. Callihan, J. W. Morfitt, and R. L. Murray, "Critical Mass Studies, Part III," **K-343**, Carbide and Carbon Chemicals Company, K-25 Plant, Oak Ridge, Tennessee, April 19, 1949.

2. Dixon Callihan, D. F. Cronin, J. K. Fox, and J. W. Morfitt, "Critical Mass Studies, Part V," **K-646**, Carbide and Carbon Chemicals Company, K-25 Plant, Oak Ridge, Tennessee, June 30, 1950.

3. Norman L. Pruvost and Hugh C. Paxton, eds., "Nuclear Criticality Safety Guide," **LA-12808**, Los Alamos National Laboratory, September 1996.

4. C. M. Hopper, "Eight-Inch Water Reflected Critical Parameters of U(93.2) Metal Spheres within Uranium Metal and Water Solutions of Various H/U-235 Ratios," **CCG-184**, Oak Ridge Y-12 Plant, January 28, 1980.

5. D. W. Magnuson, "Critical Experiments with Enriched Uranium Dioxide," **Y-DR-120**, Oak Ridge Y-12 Plant, November 30, 1973.

6. H. C. Paxton and N. L. Pruvost, "Critical Dimensions of Systems Containing ^{235}U, ^{239}Pu, and ^{233}U, 1986 Revision" **LA-10860-MS**, Los Alamos National Laboratory, July 1987.

APPENDIX A

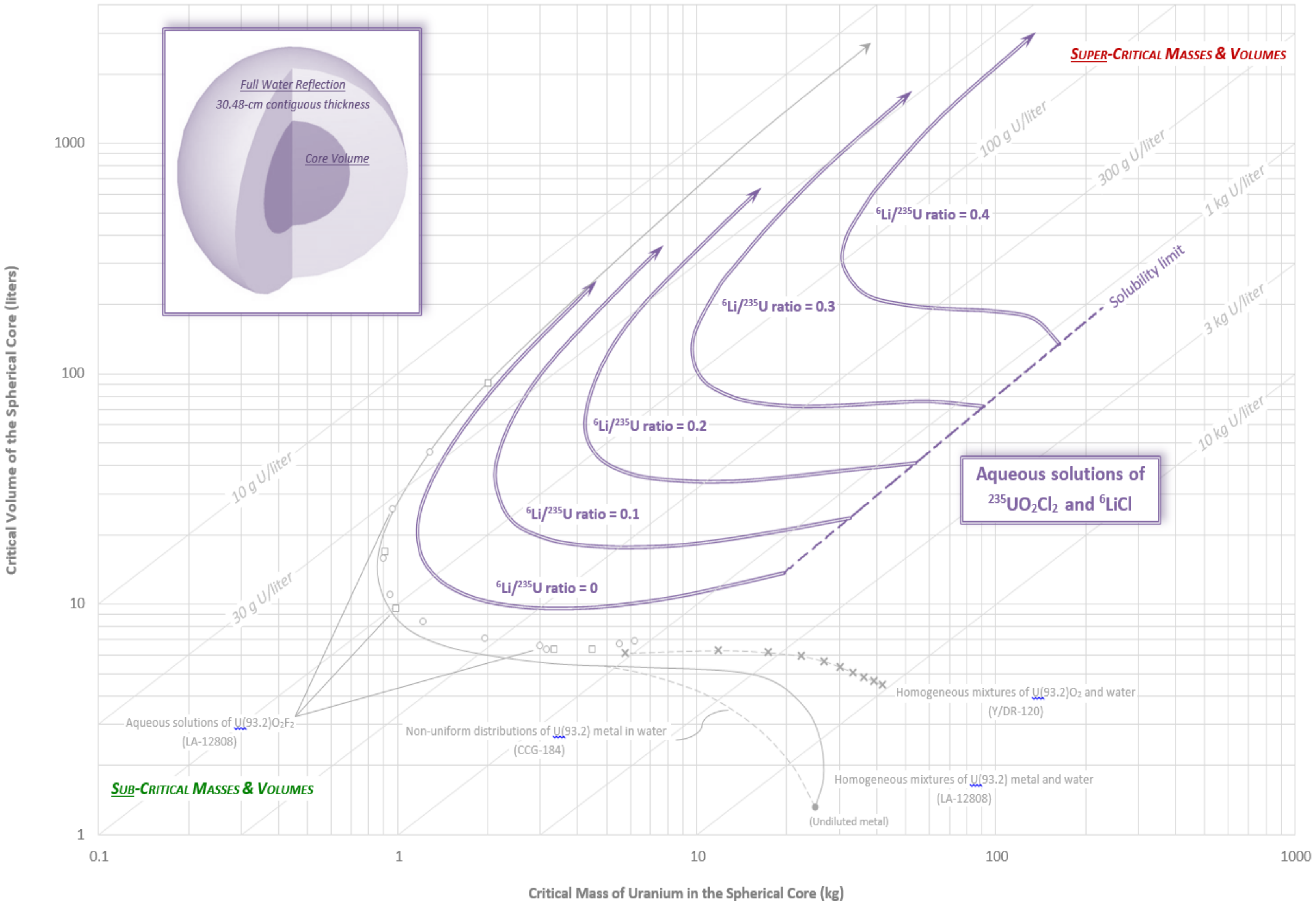

Figure A-1: Critical Masses and Volumes for Aqueous Solutions of $^{235}UO_2Cl_2$ and ^{6}LiCl in Spherical Geometry with Full Water Reflection

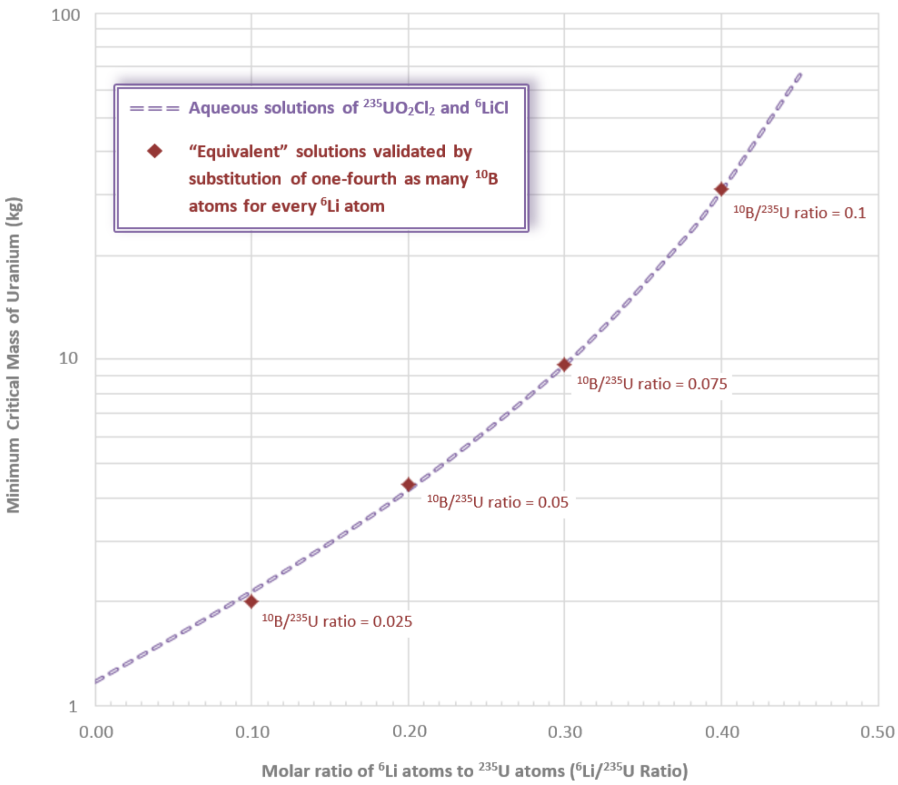

Figure A-2: The Minimum Critical Mass of Uranium for Aqueous Solutions of $^{235}UO_2Cl_2$ and $^{6}LiCl$ in Spherical Geometry with Full Water Reflection as a Function of the $^{6}Li/^{235}U$ Ratio

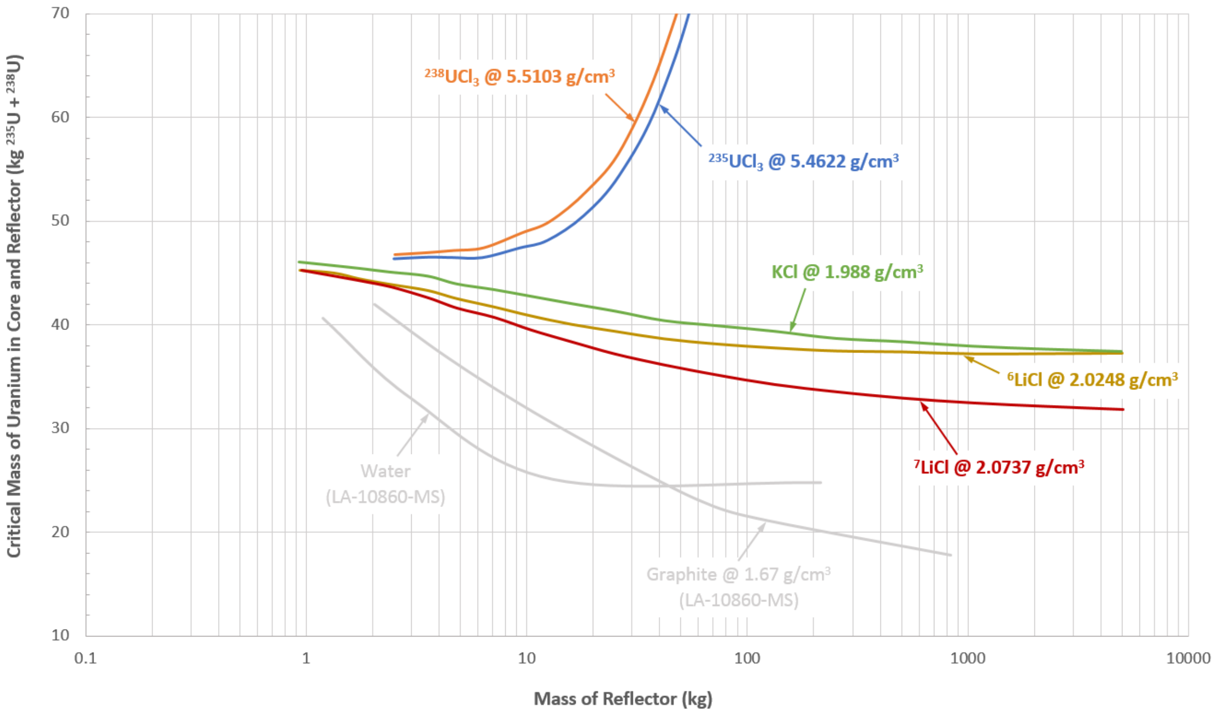

Figure A-3: Total Critical Mass of Uranium in Systems of ^{235}U Metal (ρ=18.81 g/cm^3) Reflected by Various Materials in Spherical Geometry