Crediting a Soluble Neutron Absorber at Y-12 Under ANSI/ANS-8.14-2004

V. B. Lollar, 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 Y-12 National Security Complex (Y-12) is performing work to modernize the recovery, purification, and consolidation of un-irradiated, highly enriched uranium metal. The development of advanced technology such as Electrorefining (ER) eliminates many of the intermediate chemistry systems and processes that are the current and historical basis of uranium processing at Y-12. The ER system processes impure broken uranium metal pieces into a high-purity uranium metal disk. The overall system consists of an ER cell, salt vaporization furnace (SVF), and uranium consolidation furnace (UCF). The ER vessel is comprised of a large cylindrical furnace that contains a eutectic electrolyte mixture and is loaded with uranium metal in an anode basket. The eutectic mixture consists of LiCl, KCl, and UCl_3 at specific weight percentages and is heated above its molten temperature (nominally 500°C). A basket of metal is lowered into the cell and an electric potential is applied between the anode basket and cathode collection rods. Uranium metal ions within the anode basket is oxidized to U^{3+} ions which diffuse into the surrounding electrolyte while at the same time, the uranium ions (U^{3+}) dissolved in the bulk electrolyte are reduced at the cathode rods to form U^0 metal crystals. Once enough charge has been passed through the ER circuit for uranium metal crystals to accumulate at the cathode, the material is collected into a product collection basket (Fig. 1). The product at this point consists of both high-purity uranium metal and substantial electrolyte. The remaining electrolyte is removed by the SVF and UCF.

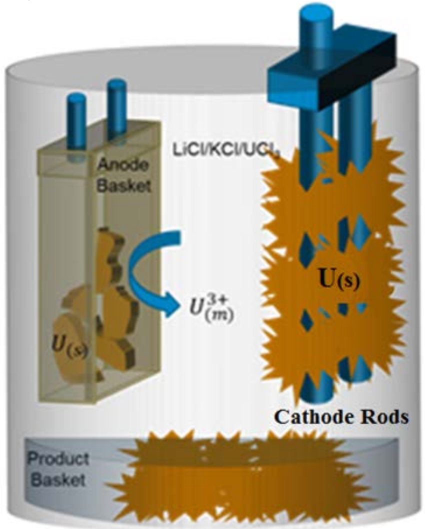

Figure 1. Schematic showing the loading of uranium metal in the Y-12 electrorefiner, dissolution, deposition, and collection of metal in the product basket

The ER cell is an unfavorable geometry cylindrical vessel mounted beneath the floor of a process-dedicated glovebox. The fissile masses required for efficient production are higher than what would normally be allowed per common nuclear criticality safety practices implemented at Y-12. Therefore, a new solution is required to ensure subcriticality under normal and credible abnormal conditions for this process. The proposed solution is to credit a soluble neutron absorber in the ER cell to control reactivity. ANSI/ANS-8.14-2004 provides guidance for the use of soluble neutron absorbers for process and handling operations in which solutions of neutron absorbers are used for criticality control. The standard addresses neutron absorber selection, system design and modifications, safety evaluations, quality control programs, and facility operations with soluble absorbers. The guidance and requirements provided by ANSI/ANS-8.14-2004 present unique challenges for Y-12 and its new electrorefining process.

DISCUSSION

The neutron absorber selected for use in the Y-12 electrorefining process is ^{6}Li. The lithium in the LiCl salt component of the mixture is enriched to 95 atom percent ^{6}Li, which has a thermal neutron absorption cross section of ~940 barns.

As with any parameter controlled for criticality safety, and particularly important with soluble neutron absorbers, one must ensure that the controlled parameter is maintained within the range that has been shown by experiment or evaluation to maintain subcriticality. The following sections address the requirements for crediting a soluble neutron absorber and how Y-12 intends to meet them.

Neutron Absorber Selection

The chemical compatibility of the neutron absorber with the process must be evaluated per ANSI/ANS-8.14-2004. ^{6}Li is not a new chemical added to an aqueous solution, but is simply added by enriching the ^{6}Li content in the LiCl that is already required by the electrorefining process. The enrichment of lithium does not affect the chemical behavior of the mixture or alter the chemical compatibility or solubility of the neutron absorber.

ANSI/ANS-8.14-2004 requires evaluation of the radiation effects such as depletion by absorption or radiolysis on the absorber over its operating life. The primary sources of neutrons in the cell include spontaneous fission of uranium isotopes, $^{7}Li(\alpha,n)^{10}B$ reactions, and $^{37}Cl(\alpha,n)^{40}K$ reactions. Y/DA-9986, *Tritium Production in the Y-12 Electrorefining Cell*, was performed to determine the highest rate of tritium production from the reaction $^{6}Li + n = \alpha + {}^{3}H$. Consequently, this is also the maximum depletion rate of ^{6}Li through neutron absorption. The report performed simulations to conservatively find the highest rate of neutron absorption in ^{6}Li (absorber depletion). The maximum neutron flux was found to be ~7 x10^{-4} neutrons/s-cm^{3} in a homogenous mixture of LiCl, KCl, and UCl_3 with a total of 10 kg of uranium metal loaded into the cell. If this maximum neutron flux occurs for 20 years, the expected operating life of the cell, and every neutron produced is absorbed by the ^{6}Li, only ~2 x 10^{-13}g of ^{6}Li would be depleted. A criticality event is also evaluated for depletion of the neutron absorber. According to LA-12808, *Nuclear Criticality Safety Guide*, studied several criticality accidents and found that the maximum number of fissions was ~3 x 10^{18} fissions. Conservatively, 1 x 10^{20} fissions is assumed with an average of 2.54 neutrons/fission to determine the neutron absorber depletion from a criticality event. If every neutron from 1000 simultaneous criticality events was immediately absorbed by the ^{6}Li in the ER vessel, only 2.54 g of ^{6}Li would be depleted. These scenarios show that absorber depletion from radiation effects is negligible over operating life.

Radiation effects do not significantly deplete the neutron absorber; however, as part of the normal electrorefining process, ^{6}Li is removed and added to the ER cell. Uranium metal crystals are removed from the ER cell for further processing and have some amount of salt mixture adhered to them. Additional ^{6}Li is added to the cell when salt eutectic mixture is added to maintain the required salt height in the cell. In order to ensure the amount of ^{6}Li in the ER cell is maintained per its requirements, the mass of salt must be tracked throughout the process. A robust mass tracking program is in development to track the amount of ^{6}Li in the ER cell from additions and subtractions of salt.

Moderation and reflection can alter the effectiveness of the neutron absorber and must also be evaluated. Lithium salts are soluble in water and moderation is controlled in the process. Moderation is controlled for the ER cell by performing operations in a glovebox and limiting the sources of external liquids. In addition, small amounts of liquid that might enter the ER cell during operation would flash to steam due to the operating temperatures of ~500°C. In the event of a moderation upset, water entry into the cell actually improves the effectiveness of the neutron absorber as neutrons would be slowed down by the increased moderation and more readily absorbed by the ^{6}Li.

System Design and Modifications

The ER cell design must provide features to prevent inadvertent concentration of fissile solution and removal, dilution, or degradation of the neutron absorber. Dilution and removal of the absorber is controlled by preventing liquids from entering the ER cell. Operations are performed within a glovebox with installed drains to limit the amount of liquid that could enter the cell and dilute the absorber. The gloveboxes are also positive pressure and sealed to provide further protection from in-leakage of liquids. Finally, no overhead fissile solution lines are present in the vicinity of the equipment. Additionally, the ER cell is designed to accommodate more than double the amount of soluble neutron absorber required for criticality safety purposes and that amount is maintained for efficient electrorefining operation.

ANSI/ANS-8.14-2004 requires that the system is designed to allow for inspection, sampling, and verification of the neutron absorber prior to use and during operation of the system. Y-12 has robust programs in place to ensure the requirements of the salt mixture meets operational and criticality safety requirements prior to and during operation. The system design allows for easy sampling of the eutectic salt mixture in the cell and is considered part of normal operating conditions.

Criticality Safety Evaluation

ANSI/ANS-8.14-2004 requires criticality safety evaluations to include allowances for uncertainties in the concentration, distribution, and neutronics properties as well as account for potential neutron degradation or non-uniform distributions. A Criticality Safety Process Study (CSPS) was performed to evaluate the safety of the ER cell operation under normal and credible abnormal conditions. The minimum mass of ^{6}Li required for safe operation of the cell was found from computer simulations and the resulting minimum salt mass was calculated. Safety factors were then applied to the minimum salt mass to ensure subcriticality. A safety factor of 10 was applied to account for chemical sampling and analysis errors such as well as uncertainties in the ^{6}Li concentration and distribution. An additional factor of 2 was applied to account for any uncertainties in the ^{6}Li mass.

Criticality safety evaluations must be based on data from applicable experiments or validated calculations. Currently there is a lack of experimental criticality benchmark data that involves chlorides as the primary constituents and necessitates reliance on a narrowly defined area of applicability and gross conservatism in the derivation of nuclear criticality safety limits. Critical experiments are needed for further validation of their nuclear properties and behavior. Experiments will be requested through the DOE Nuclear Criticality Safety Program (NCSP) Integral Experiment Request (IER) process. However, in the absence of critical experiment data for ^{6}Li, it is possible to compare the behavior of ^{6}Li with critical experiment data from aqueous solutions of ^{10}B. Critical mass studies of ^{235}U with ^{6}LiCl were performed in RP YAREA-F-0559, *Critical Mass Studies: Aqueous Solutions of* $^{235}UO_2$ *and* $^{6}LiCl$ *in Spherical Geometry*. This report uses validated computational methods for analysis of nuclear criticality safety data and published nuclear data in the absence of directly relevant experimental critical benchmarks. Validated data from critical experiments with ^{10}B, which behaves similarly to ^{6}Li in its ability to absorb neutrons in a 1/E fashion, was compared to nuclear data of ^{6}Li in the ENDF/D-VII database. The report proved useful to the narrowly defined interest of the minimum critical mass of uranium in a spherical geometry in the absence of directly comparable critical experiments.

Quality Control Program

Y-12 has an established quality control program to ensure proper neutron absorber acquisition, storage, preparation, and usage. Inspection, sampling, and testing intervals will be established to confirm the mass and isotopic concentration of ^{6}Li during electrorefining operations. The Y-12 Quality program will be used to comply with ANSI/ANS-8/14-2004.

Facility Operation with Soluble Absorbers

ANSI/ANS-8/14-2004 requires that system design parameters important to criticality safety are verified to conform to specifications and that proper mixing and concentration of the neutron absorber are verified before use. Y-12 implements a Configuration Management Program that ensures physical design features important to criticality safety are maintained. The facility is required to verify the proper mixing and concentration is verified before use. The proper mixture weight percentages and isotopic concentrations are required for both efficient electrorefining and criticality safety. The characteristics of the eutectic salt mixture are confirmed before operation and on periodic intervals during operation. Additionally, a mass tracking program is required to track the amount of ^{6}Li in the ER cell at all times for material accountability and criticality safety purposes.

CONCLUSION

Y-12 has plans in place to meet the requirements in ANSI/ ANS-8/14-2004 and credit soluble neutron absorbers in its ER process. The selection of ^{6}Li as a soluble neutron absorber does not present any additional chemical hazards for the Y-12 ER process because lithium is already a constituent of the eutectic mixture and only the isotopic concentration of lithium is increased. The addition and removal pathways of the neutron absorber during normal operations have been evaluated and the amount of ^{6}Li will be tracked with a mass tracking program and periodic sampling of the eutectic salt mixture will occur throughout the process. These processes ensure the required amount of ^{6}Li for criticality safety is present in the ER cell at all times. A CSPS has been developed to show the ER cell remains subcritcal under normal and credible abnormal conditions. However, benchmark critical experiment data with chlorides as the primary constituents of a eutectic mixture are needed to validate the calculations to support criticality safety. Finally, robust quality and operational programs are already in place at Y-12 and are relied upon to meet the operational requirements of ANSI/ANS-8/14-2004.

REFERENCES

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