LA-UR-17-22805
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Intended for: American Nuclear Society Nuclear Criticality Safety Division Topical 2017, 2017-09-10/2017-09-15 (Carlsbad, New Mexico, United States)

Issued: 2017-04-06

Investigations into Validation of Plutonium Solutions for Criticality Safety Analysis

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EXTENDED ABSTRACT

Plutonium solution chemistry is among the most complex of all the elements in the periodic table. Under acidic conditions up to four different oxidation states can be present in significant amounts in a single solution [1,2,3]. From the perspective of nuclear criticality safety, accurate information regarding the concentration of plutonium and other constituents in solution must be well-known in order to correctly model plutonium solution systems for analysis. This requires knowledge of the solvent, plutonium oxidation state, chemical speciation, and density of solution.

ANSI/ANS-8.24-2007 *Validation of Neutron Transport Methods for Nuclear Criticality Safety Calculations* [4] defines validation as the process of quantifying the suitability of a computer code system for use in nuclear criticality safety analysis. New tools have been developed to support sensitivity-uncertainty based nuclear criticality safety validation [5].

The majority of available critical experiment benchmark information for plutonium solution systems pertains to plutonium (IV) nitrate solutions [6]. This paper will discuss the validation of plutonium solution systems for nuclear criticality safety analysis and discuss how one new sensitivity-uncertainty tool for validation of MCNP6, Whisper, can provide information to support validation of such systems. Case study results for various solution systems will be presented.

In aqueous acid solutions plutonium can exist simultaneously in four oxidation states; Pu(III), Pu(IV), Pu(V), and Pu(VI) . The oxidation state is largely responsible for chemical reactions in solution. Recovery, separation and purification processes depend on maintaining, adjusting, and controlling the oxidation state. Criticality safety analysis requires an understanding of the chemistry of solutions and the ability to make accurate and conservative assumptions and predictions of the chemical and physical properties of the system.

In order to analyze such systems using computational methods and establish subcriticality, the understanding of the system must go further and encompass both how process conditions affect k-effective of the system, and for validation how process conditions affect the upper subcritical lim-

it (USL) as well. The majority of critical experiment data available for plutonium solutions is for plutonium (IV) nitrate solutions. Of the forty experiments presented in the ICSBEP Handbook, thirty-six of them are plutonium nitrate experiments. Sensitivity-uncertainty analysis of the system allows a deeper look into validation information and may even be useful to establish an upper sub-critical limit for solution systems in other solvent mediums in addition to nitrate. A recent response issued by the DOE Criticality Safety Support Group (CSSG) provides guidance on validation of criticality safety codes when only limited critical or exponential experiment data are available [7]. If sensitivity studies indicate that the nuclides with limited data are unimportant to the validation, the computational method can be validated for the application using traditional methodologies. Further '*for situations where a nuclide is determined to be important to the validation and limited data exist, validation may still be possible'*. An additional margin, separate from and in addition to any margin needed for extending the validation applicability, should be used to compensate for the limited data. Sensitivity and uncertainty tools may be used as part of the technical basis for determining the magnitude of the margin. Discussion will include potential examples of solution systems that may be relevant to such guidance.

ACKNOWLEDGMENTS

This work was supported by the US DOE/NNSA Nuclear Criticality Safety Program.

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