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Rapid, Non-Radioactive Methods for Prediction and Quantification of Radiolytic Radical Decomposition Products in Nuclear Separations

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ABSTRACT:

Monoamide complexants have been explored as potential replacements for tributyl phosphate (TBP) in PUREX, THOREX, and other back-end waste separations processes, since they have greater U and Th selectivity, are composed of readily incinerable C, H, O, and N elements, and their degradation products hinder separations less than TBP degradation products. Monoamides also can have a variety of alkyl substituents, allowing solubility tuning of the extracted metal species to increase extraction efficiency. For over 60 years, monoamide complexants have been examined for their promising actinide separation abilities, but their radiolytic stability data is limited, preventing selection of the most stable monoamides and hindering separations scale-up for industrial use. γ-Radiolysis studies are low-throughput, so reports of monoamide radiolytic stability include only one or a few complexants tested under varied conditions, preventing stability comparisons over a wide range of complexants and conditions. These issues hinder attempts to understand processes that govern monoamide radiolytic stability and the ability to rapidly identify the most promising monoamides for investigation and scale-up. This project aims to establish correspondence between high-throughput, non-radioactive radical assays and classical γ-radiolysis methods to determine monoamide complexant stability and degradation products.

Radicals generated during γ-radiolysis (hydroxyl radical, organic radical cations, superoxide, etc.) are the same as damaging radicals generated in biological systems. Non-radioactive biochemical radical assays were developed to generate these radicals to determine their ability to damage biological components, but this technology has not been applied in nuclear separations. We propose that these high-throughput, non-radioactive radical assays can be used as an initial screening tool to determine radiolytic stability of monoamide complexants for nuclear separations. These assays would quickly identify promising complexants for classical γ-radiolysis studies and process development for industrial scale-up.

For this project, we will: 1) synthesize monoamide complexants with various alkyl substituents and examine their radiolytic stability and degradation products using γ-radiolysis techniques; 2) perform high-throughput, non-radioactive radical assays on the same monoamide complexants to determine their degradation products and relative stabilities when exposed to hydroxyl radical, organic radical cations, superoxide, and peroxynitrite; and 3) correlate γ-radiolysis degradation and non-radioactive radical assay data to determine if γ-radiolytic decomposition is approximated by any or all of the non-radioactive radical assays, as well as similarities of monoamide degradation products from γ-radiolysis and the non-radioactive radical assays. Our work will develop high-throughput, non-radioactive methods to provide a screening tool for determining potential monoamide radiolytic stability prior to traditional but low-throughput γ-radiolysis studies. These methods, which could also be extendable to other classes of complexants, will accelerate DOE work toward single-cycle solvent extraction processes, aid in translating laboratory-scale separations to industrial scales, and facilitate fuel cycle separations research.