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Advanced Screening Approaches for Accelerating Development of Separations Technologies

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ABSTRACT:

Traditionally, aqueous used nuclear fuel reprocessing has primarily focused on separating uranium (U) and plutonium (Pu) from the minor actinides (MA) and fission products. The vast majority of reprocessing facilities operating worldwide use the Plutonium Uranium Reduction Extraction (PUREX) process or a derivative thereof to recycle U and Pu. However, high level waste, such as PUREX raffinate, generates much heat and remains highly radiotoxic for millions of years, mainly due to long-lived fission products and MA. Advanced reprocessing concepts have therefore aimed at separating elements with long-lived, heat-generating radioisotopes to reduce the long-term radiotoxicity of the waste for storage. In addition, while U and Pu are the key actors for fuels, other valuable resources present in used nuclear fuel can be harvested as valuable resources to support societal needs.

Unfortunately, the potential for significant technological advancements in reprocessing is often stifled by the rate of metal chelator design and evaluation from benchtop to process scale. Overall, current practices aimed at overcoming physiochemical property detriments such as solubility, poor selectivity, kinetics, etc. utilize a stepwise evolutionary process to develop structure function relationships that enable solving hard technological problems. While such a development process has been shown to work, ultimately it results in a prolonged path to application, which comes with a substantial financial burden. A unified approach to screening would be highly beneficial, allowing for the early down selection of materials with suitable physiochemical properties spanning thermodynamics, radiation and radiochemistry, process performance (loading capacity, kinetics, phase separations, chemical stability etc.), and disposal.

The goal of this project is to establish a unified selection criterion for chelating molecular structures that can be implemented into a software package to more efficiently address ligand applicability to metal ion separation problems for current and future nuclear fuel cycles. By establishing this criterion, we seek to enable the accelerated, cost-effective discovery of new separation workflows, as well as their implementation beyond early radiotracer experiments.

We propose to move beyond simple solution thermodynamics and incremental modifications of existing ligand structures from obvious molecular families such as the polyaminocarboxylates, amide derivatives, or hydroxamic and phosphonic acids, by demonstrating the need for early incorporation of process parameters in the discovery pipeline. Through the combination of literature data survey, additional physical property measurements and proof-of-concept screening experiments we will provide a clear phenomenological understanding of the potential of new ligand architectures as new reagent for used nuclear fuel aqueous reprocessing. Results from this project will therefore enable a wide-cast screen of molecular architectures from large existing libraries of ligands, combined with batch simulations of various conditions for multistage countercurrent liquid-liquid extraction, that would allow for the identification of new separations methodologies based on a series of well-defined critical criteria stemming from the entire extraction process on industrial scale.