Developing Preorganized Chelators for Separating Nuclear Fission Products

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v1:
Concentrations of nuclear fission products are statistically related to the identity of the source nuclear fuel material that is used. Being able to selectively chelate these products as opposed to their contaminant counterparts helps to increase the accuracy of the concentrations of the nuclear fission products, leading to more accurate predictions in the source material. Typical contaminants to nuclear fission products are those that have the same properties when being characterized by the same analytical methods. Developing preorganized chelators for these species allows for their selective chelation. Preorganizing chelators involves studying and understanding current chelators and other functional groups that bind well to the ions of interest. This work focuses on the use of 3,4,3-LI(1,2-HOPO) as a chelator for nuclear fission products, and exploring possible derivatives generated from HostDesigner when replacing the 4 carbon linker. These generated structures are then evaluated using a soon-to-be-published prediction model from the Critical Materials Institude at Ames Lab, which is able to predict stability constants for the structures using their respective SMILEs format. These structures are then evaluated using DFT to ensure that the trends that are seen for the predictive model are retained in the ab initio perspective. Additional post-processing can be done to evaluate the properties that are most responsible for increased selectivity.

v2:
The concentrations of nuclear fission products are statistically linked to the identity of their source nuclear fuel. Accurate quantification of these products, however, is often hindered by the presence of contaminants with similar chemical properties that complicate standard analytical methods. Selective chelation of fission products offers a pathway to improving the precision of source material identification. This work explores the development of preorganized chelators designed for selective binding of key fission products over their contaminants. Specifically, we focus on modifying the well-established chelator 3,4,3-LI(1,2-HOPO) by varying its four-carbon linker using HostDesigner to generate potential derivatives. These candidate structures are evaluated using a predictive stability constant model developed by the Critical Materials Institute at Ames Laboratory, which estimates binding strengths from molecular SMILES representations. Promising candidates are further validated using density functional theory (DFT) calculations to assess whether predicted binding trends are maintained at the ab initio level. Post-analysis investigates structural features contributing most significantly to selectivity enhancement, guiding future chelator design.