Nuclear Fuel Remediation: A Computational Study

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Introduction

Increased global attention on sustainable energy initiatives sparked renewed, global interest in nuclear energy. However, a significant pitfall of nuclear energy is the production of high-level nuclear waste (HLW) with an extensive half-life. HLW waste storage poses a significant environmental and security risk. Current methods of nuclear fuel post-processing often involve long-term burial of waste in remote locations, such as the Swedish SKB Clab facility, shown in figure 1. Methods of remediating SNF using chemical extraction agents may reduce the danger of HLW long-term storage and provide a means of recycling waste into reactor-grade fuel. This computational research project aimed to study four chemical extraction agents to understand if they possess properties suitable for use in extraction procedures.

Key Words: Basis set, density functional theory, spent nuclear fuel (SNF), high level waste (HLW), Reprocessing fuel cycle (RFC).

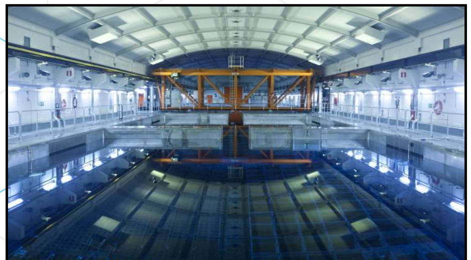
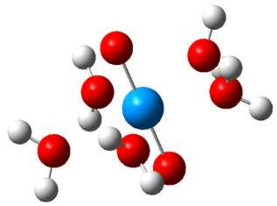


Figure 1: *HLW storage in remote cooling ponds.*

Test Methods

The nitrogen and sulfur-based chelation agents studied included $C_8H_{17}N_3O_4$ (TNDA), $C_6H_{12}N_2O_4$ (EDDA), $C_4H_{10}OS_2$ (Mercapto). A variation of Mercapto, labeled 2-Mercapto, was also studied. These ligands were selected based upon their use as chelation agents for heavy metals or radioactive isotopes. Actinide dioxide compounds PuO_2, UO_2, and NpO2 were modeled in pentaaquo structures, as shown in figure 2. Pentaaquo structures are pentagonal structures where H_2O surrounds an element; these structures model SNF submerged in a cooling pond at a HLW storage facility.



IR frequency peak harmonic animation

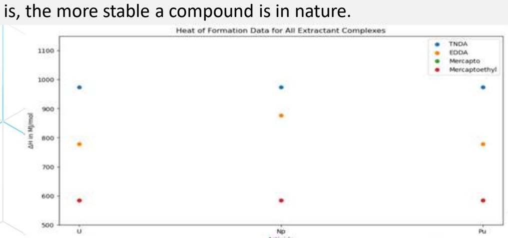
Figure 2: *A typical actinide pentaaquo structure.*

Ligands, pentaaquo structures, and actinide ligand complexes were simulated separately using the 6-31G(d) basis set for atoms S, O, C, and N. The MWB60 basis set was used for actinides Np, Pu, and U; all molecules were simulated in both free space and solvent conditions. Geometry optimization and frequency calculations were performed for all molecules. Figures 3, 4, and 5 show the 3D models of TNDA, modified BAL, and EDDA complexed with an actinide. Finally, the research team modeled the collected data using the standard heat of formation equation. The equation modeled the change in enthalpy during the formation of 1 mole of a desired compound. Equation 1 shows the standard formula.

$$\Delta H^o_{reaction} = \sum \Delta H^o_f(products) - \sum \Delta H^o_f(Reactants)$$

Discussion & Results

Graph 1 shows a comprehensive bar graph of the data collected for each simulation. The more negative ΔH (measured in MJ/mol) is, the more stable a compound is in nature.



Graph 1: *Calculated* ΔH in keV.

TNDA

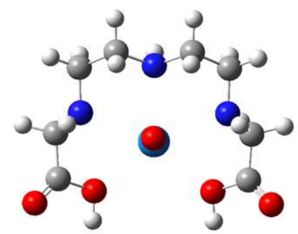


Figure 3: *TNDA actinide complex.*

Figure 3 shows the optimized geometry of TNDA when bound to AnO_2.

MERCAPTO

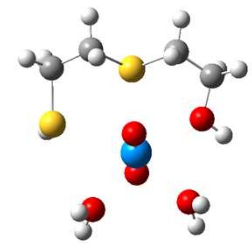


Figure 4: *Mod. BAL actinide complex.*

Figure 4 shows the Mercapto complex with optimized geometry.

EDDA

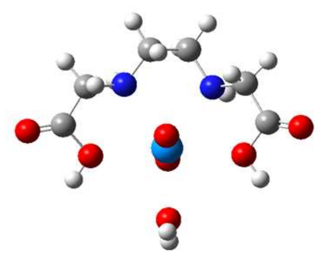


Figure 5: *EDDA actinide complex.*

Figure 5 shows the optimized geometry of EDDA. Note that one water molecule is bound to the AnO_2.

Conclusion

Experimental results revealed unique properties for each of the extraction agents studied. The Mercapto and 2-Mercapto agents did not have a significant difference between actinides on graph 1, indicating a lack of selectivity for the actinides studied. EDDA was marginally selective for Np. Finally, TNDA did not display any significant selectivity for any of the actinides studied. Future work on this subject may include a study of additional chelation agents or actinide elements. Future studies of these extracting agents that expand upon the whole range of actinide elements will provide a better understanding of how these extraction agents may be used to remediate SNF from HLW storage facilities.

References:

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