**BACKGROUND**

- 16 nuclear weapons production sites from the Cold War still contain materials and waste contaminated with radiation (300 million liters of highly radioactive waste stored in tanks at 3 sites alone).
- A number of tanks have leaked, exposing the environment to some of the most dangerous and complicated materials known to exist on Earth.
- Need to extract and separate the radioactive materials from the remainder of the waste.
- New separation materials must be designed that can withstand intense levels of radiation, acidity, alkalinity, and temperature.
- New materials need to have high selectivity for the radioactive species (actinides).
- Necessary to have a fundamental understanding of interactions between materials used for separation and radioactive species (host-guest interactions).

**METHODS**

1. **High-Throughput Screening Algorithm:** HostDesigner (HDS) code
   - Software that uses molecular design algorithms that are capable of identifying optimal integrated ligands for a specified metal at the molecular mechanics level of theory.
   - Algorithm: OVERLAY – a linking molecule from the LIBRARY forms two bonds with one input fragment defined by the user.
   - Two output files: one with structures sorted by geometric parameters (rmsd), other sorted by estimated conformational energy parameters (hdoc).

2. **Density Functional Theory (DFT): ADF** code
   - Amsterdam Density Functional (ADF2016) used for QM density functional theory calculations.
   - Functional: Hybrid B3LYP-D3/B for Uranium, D2P for all other elements.
   - Snap (single point) binding energy calculation.
   - SCF convergence criterion = 1e-6 Hartree.

**RESULTS**

Uranium with four individual, unlinked ligands

-uranium + 4L \rightarrow [UL]_{4}^{1-} \ldots \ldots \Delta E

<table>
<thead>
<tr>
<th>Ligand</th>
<th>ΔE (kcal/mol)</th>
<th>ADF Ranking</th>
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</table>

Two ligands connected by a link from HostDesigner: 6 structures sorted by geometric parameters (rmsd)

-uranium + 2L + [L(hdce)] \rightarrow [UL_{2}(hdce)]^{1-}

<table>
<thead>
<tr>
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</table>

Two ligands connected by a link from HostDesigner: 6 structures sorted by conformational energy parameters (hdoc)

-uranium + 2L + [L(hdce)] \rightarrow [UL_{2}(hdce)]^{1-}

**DISCUSSION**

The difference in binding energy (134.4 kcal/mol on average) between the structures with linked ligands and those without suggests that linked ligands provide more stability than individual ligands.

When this technique is extended to other actinides, the trend in binding energy remains the same. These preliminary results shown to the left will be examined in more detail, and could lead to the discovery of a ligand with the ability to separate actinides.

**CONCLUSIONS**

- The original Uranium catecholate complex is stabilized when HostDesigner is used to connect two of the ligands.
- Use of HostDesigner for design of potential new ligands expedites the process of ranking ligands in order of binding affinity for the actinide elements.

**FUTURE WORK**

- Replace Uranium with other actinide elements and compare the binding energies.
- Connect the ligands through the aromatic sections rather than the amide methyl groups.
- Run further calculations on the most stable linked ligand complexes (geometry optimization, vibrational frequency).
- Identify other experimental structures to run through HostDesigner.

**BIBLIOGRAPHY**


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