DOE Award Number: DE-SC0021420

Recipient Name: University of Iowa

Street Address/City/State/Zip: 2 Gilmore Hall, Iowa City, IA 52242

PI: Tori Z. Forbes

Business Mailing Address of PI: W374 Chemistry Building, Iowa City, IA 52242

Telephone Number of PI: 319-384-1320

Email of PI: tori-forbes@uiowa.edu

Title: Impact of intermolecular interactions on the spectroscopic signals, energetics, and redox behavior of high valent ^{237}Np.

Project Period: 09/01/2020 – 3/14/2024

Report Submission Date: 07/03/2024

Reporting Period Start and End Date: 09/01/2020 – 3/14/2024 (Final report)

1. Introduction and Research Aims

The objective of this proposal was to apply a system level approach towards the understanding of neptunyl behavior in aqueous solution and solid-state phases to fully interpret vibrational spectra, understand the energy landscape, and control redox behavior. Neptunium chemistry in aqueous systems is dominated by the high valent neptunyl ($Np(V/VI)O_2^{+}/^{2+}$) cation, but our fundamental understanding of this moiety is lacking and impedes our ability to control its chemical behavior. This neptunyl unit is unique because it has two strong bonds to oxo groups, and the bond strength and reactivity of the moiety can be varied by the electronic structure of the Np and the ligands in the equatorial plane. With a strong neptunyl bond, the oxo group is essentially passivated and does not participate in additional engagement with ions or molecules in the second coordination sphere. Weakening this bond allows for additional oxo interactions to take place (Fig. 1) and can lead to activation of vibrational modes, changes in the energetics of the system, and unexpected electron transfer reactions. These fundamental chemical processes hamper the interpretation of spectral data, change the energetics of the system, and result in unpredictable redox behavior that influences radiochemical separations and materials processing. A systems chemistry approach that incorporates the influence of second sphere interactions is critical to delineating the specific ions and molecules that control the behavior of neptunium in aqueous solutions.

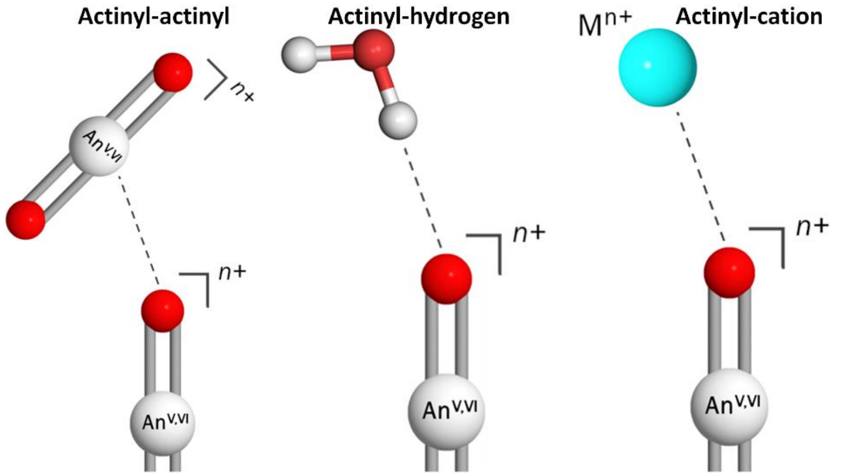


Figure 1. Impacts of actinyl-actinyl (AAI), actinyl-hydrogen (AHI) and actinyl-cation (ACI) interactions on Np chemistry will be explored in this proposal.

Our overall hypothesis was that intermolecular interactions (e.g. electrostatic, H bonding) controlled the resulting spectroscopic signals, thermodynamic stability, and redox behavior of Np. Furthermore, we hypothesized that large effects occured with strong oxo interactions (e.g. neighboring charge dense cations, cooperative hydrogen bonding networks, and diamond-type actinyl-actinyl interactions). These hypotheses were tested through evaluation of neptunyl complexes in aqueous systems using spectroscopic, diffraction, and computational techniques. The following research objectives were identified to test our hypotheses:

Objective 1: Identify conditions that result in perturbations of the spectral features of the neptunyl cation and evaluate the changes in the electronic structure.

Objective 2: Evaluate the energetic changes because of intermolecular interactions with the neptunyl oxo group.

Objective 3: Explore the impact of intermolecular interactions to influence the stability of Np(V), Np(VI), and Np(VII) in aqueous solutions.

2. Significant findings and products from the funded work.

2.1. Prediction of spectral signatures associated with diamond and T-shaped actinyl-actinyl

interactions within Np(V) solids and solutions (Objective 1).

We initially evaluated simple Np(V) salt solutions (perchlorate, chloride, and nitrate) to identify the nature of the neptunyl-neptunyl interactions. Overall, we saw an increase in the number of vibrational bands as we increased the concentration of the Np(V) solution (Pyrch *et al.*, 2022a). Vibrational modes for molecular clusters that included actinyl-actinyl interactions, including T-shaped dimers, trimers, and tetramers were calculated using DFT methodologies to develop a better understanding of the spectral signatures. From the computational studies, we determined that GGA functionals (PBE) were better at predicting geometric and vibrational energies for monomeric system, but the use of hybrid functionals (B3LYP) was needed to properly describe the larger oligomeric units. DFT experiments also indicated that there is activation of multiple vibrational modes involving the neptunyl stretch that is beyond the typical symmetric and asymmetric stretching modes. We were able to link the prominent modes within the 600-700 cm^{-1} window to the symmetric stretching features of the trimeric and tetrameric units. These building units were also present in the solids crystallized from evaporation of the solutions, indicating that at high Np(V) concentrations, the presence of multiple bands within the Raman spectroscopy are caused by the formation of oligiomeric units that contain neptunyl-neptunyl interactions.

In the case of simple salts, we noted only T-shaped neptunyl-neptunyl interactions could be identified in the solution, but additional DFT calculations indicated very little differences in the energetics between T-shaped and diamond-shaped interactions. We noted in the literature that diamond-shaped neptunyl-neptunyl interactions were prominent in the presence of carboxylate ligands and we hypothesized that the ability of the ligand to bridge the metal centers may help drive the formation of the diamond-shaped interaction. We explored this hypothesis through the experimental and computational analysis of a series of carboxylate ligands containing different aromatic and aliphatic functional groups to drive different steric conformations (Augustine *et al.*, 2023a). The major outcome of this study was that the presence of carboxylates drove the formation of actinyl-actinyl interactions at much lower Np(V) concentrations than what is observed in simple salt solution. The spectral analysis is much more complicated due to the presence of ligand bands and combination modes, but the results are supported by DFT vibrational analysis. In addition, we saw no significant impact in the identity of the functional group associated with the carboxylate ligand, suggesting that there were limited effects caused by the presence of electron donating or withdrawing groups or steric hindrance in this case.

Summary: Our studies have led to definitive spectral assignments for neptunyl-neptunyl interactions in aqueous solutions that will be valuable for predicting Np(V) speciation in solution.

2.2. Actinyl-hydrogen and Actinyl-cations interactions influence spectral signatures of high valent actinides (Objective 1) and are a driver for formation energetics (Objective 2).

We began studying U(VI) systems to evaluate spectral changes based upon uranyl hydrogen and uranyl-cation interactions. For the spectral evaluation of the actinyl-hydrogen system, we explored a set of cobalt hexamine uranyl tricarbonate compounds using X-ray diffraction, Raman spectroscopy, and DFT calculations (Pyrch et al. 2022b). The major findings are that (1) asymmetry in the hydrogen bonding network can results in activation of the v_3 asymmetric stretching mode associated with the uranyl cation and (2) this activation is the result of subtle differences in the bond distance of the uranyl moiety. In addition, we collaborated with the Blakemore group (U Kansas) to evaluate the uranyl-cation interactions with vibrational spectroscopy because their macrocyclic ligand offers the opportunity to co-encapsulate the UO_2^{2+} and alkali (Cs^+, Rb^+, K^+, Na^+, and Li^+) and alkali-earth cations (Ca^{2+}, Sr^{2+}). From this work, we also linked the dependence of the U=O symmetric stretching frequency on the Lewis acidity (pK_a) of the metal cation aqua complex (Kumar et al., *in preparation*).

From these experimental results, we utilized DFT calculations to systematically evaluated how changes in the bond asymmetry impacted the v_1 and v_3 bands and developed DFT methodology that could be utilized to evaluate energetics of these systems. Benchmarking studies of different exchange–correlation functionals and dispersion corrections were carried out on actinyl-cation ($K_2[UO_2Cl_4]\cdot 2H_2O$, $Rb_2[UO_2Cl_4]\cdot 2H_2O$, and $Cs_2[UO_2Cl_4]$) and actinyl-hydrogen ($(C_{10}H_{10}N_2)[UO_2Cl_4]$, $(C_{12}H_{12}N_2)[UO_2Cl_4]$, and $(C_{12}H_{14}N_2)[UO_2Cl_4]$) model compounds. We found that the PBE+U+D3BJ methodology reproduced geometric and electronic structure parameters most accurately (Augustine *et al.*, 2023b). The addition of a Hubbard U correction was also the most important parameter for accurately describing the bonding interactions in the hybrid structures. Additionally, the electronic and vibrational properties of the uranyl were studied through PDOS and phonon analysis. Changing the intermolecular interactions through differing N-heterocyclic cations had little effect on the electronic and vibrational energy levels of these systems. The vibrational calculations did show the appearance of combination modes between uranyl and the N-heterocyclic cation in both Raman and IR spectroscopy, which is an important consideration when assigning vibrational bands within these systems. A DFT + thermodynamics (DFT + T) framework was adapted to actinide elements to calculate the formation enthalpies of each structure. Using this approach, we were able to predict values of ΔH_f to within ~21 kJ/mol for H-bonding systems and within 4 kJ/mol for $Cs_2[UO_2Cl_4]$. More importantly, the method was able to accurately capture the experimental trends in ΔH_f observed between each system, contrary to referencing each system back to its standard state.

From these benchmarking studies, we completed a more comprehensive evaluation of actinyl hydrogen bonding interactions and developed predictors that could be used to determine energetics of the hydrogen interactions (Rajapaksha *et al.*, 2023a). We synthesized and characterized 14 compounds containing the $[UO_2Cl_4]^-$ synthon with interactions to hydrogen donors and determined their formation enthalpy (ΔH_f) through isothermal acid calorimetry. The DFT + T approach showed a high level of agreement (largest absolute error = 11 kJ/mol) to the experimental values. Three predictors (packing efficiency, cation protonation enthalpy, and hydrogen bond enthalpy) were explored with the strongest correlation to the hydrogen bond enthalpy, highlighting the importance of the hydrogen bond network in the relative stability of the compound. In addition, we found that the strength of the hydrogen bond network could be tied to a red-shift of the symmetric uranyl stretching mode. This work provided a definitive bridge between thermodynamic energies and spectroscopic features and the molecular-level details of the geometry and electronic structure.

From the U(VI) work, we applied this theoretical framework to interactions between $[Np(V)O_2Cl_4]^{3-}$ and $[Np(VI)O_2Cl_4]^{2-}$ and hydrogen bond donors or alkali cations (Rajapaska *et al.*, 2023b). Theoretical studies were again benchmarked against calorimetric and spectroscopic data, but only two neptunium phases, $[C_5NH_6]_2[Np(VI)O_2Cl_4]$ and $[C_4N_2H_{12}][Np(VI)O_2Cl_4]$, were used due to challenges in performing these studies with transuranic elements. This highlights the importance of accuracy within the DFT calculations for the Np system, as we were able to computationally assess seven Np phases. The DFT+T methodology was validated against the experimental data and the neptunyl hydrogen interaction energies or the electrostatic attraction energies for the alkali cations were found to be the major descriptor for the energetic trends. The DFT calculations also showed a strong correlation between the degree of NCIs by axial oxygen and a red-shift of Np=O symmetrical stretch (v_1) wavenumbers and quantitatively demonstrated that NCIs can weaken the Np=O bond.

Summary: We have definitively demonstrated how actinyl-hydrogen and actinyl-cation interactions can influence spectral features for uranyl and neptunyl system. In addition, we have developed DFT methodologies to predict enthalpies of formation for uranyl and neptunyl systems and linked stability to chemical descriptors (hydrogen interaction energies or the electrostatic attraction energies).

2.3 *Oxidation of Np(VI) to Np(VII) is influenced by the identity of the alkali cation in solution and takes place through interactions with the $^{\bullet}OH$ radical (Objective 3).*

We explored the oxidation of Np(V) to the $[Np(VII)O_4(OH)_2]^{3-}$ anion in LiOH, NaOH, and KOH solutions via Raman and UV-Vis-NIR spectroscopy (Kravchuk *et al.*, 2024). Upon addition of ozone to the Np(V) alkaline media, a one-electron, step-wise oxidation of Np(V)→Np(VI)→Np(VII) was observed. The rate of oxidation was influenced by the identity of the alkali cation, as KOH media resulted in rapid oxidation to Np(VII) compared to LiOH and NaOH. We attributed this observation to the outer-sphere interactions of the alkali metals with differing charge-density having an effect on the redox behavior of neptunium in solution. Additionally, the initial oxidation of Np(V) resulted in the presence of intermediate Np(VI) solid-state phases that were identified by powder X-ray diffraction to be mainly composed of hexavalent neptunate phases. We collaborated with Dr. Richard Wilson at Argonne National Laboratory, who had previously isolated and structurally characterized these Np(VI) intermediate phases, to further analyze and identify their spectral signals (Hutchinson *et al*., 2023) These solid-state Np(VI) neptunates undergo further oxidative dissolution to form the soluble $[Np(VII)O_4(OH)_2]^{3-}$ anion, but the observed rates displayed an opposite effect, as the KOH media exhibited slow dissolution compared to LiOH and NaOH.

DFT calculations were further used to explore the Np(V) oxidation to the Np(VII) through reactions with the hydroxide and superoxide radicals that are formed when O_3 reacts in a basic solution (Kravchuk *et al.*, 2024). Computational results suggested that all interactions with $HO^{\bullet}$ resulted in a favorable oxidation of both the Np(V) and Np(VI) metal centers, while it was unfavorable to oxidize the neptunium center with $O_2^{\bullet-}$. Overall, two unique oxidation pathways were found in the generation of the $[NpO_4(OH)_2]^{3-}$ anion. The most likely and favorable path consisted of an initial inner- or outer-sphere electron transfer of $[NpO_2(OH)_4]^{3-}$ with the hydroxide radical to form $[NpO_2(OH)_4]^{2-}$. From here, the Np(VI) species could undergo further oxidization with $HO^{\bullet}$ through hydrogen transfer to generate the trioxo $[NpO_3(OH)_3]^{2-}$ structure, which readily transforms into the $[NpO_4(OH)_2]^{3-}$ anion. The second pathway saw an initial hydrogen transfer of $[NpO_2(OH)_4]^{3-}$ to create the trioxo Np(VI) species, which could undergo another oxidation through hydrogen transfer to generate the Np(VII) tetraoxo anion. The formation of the Np(VI) trioxo intermediate was found to be a reasonable intermediate structure based on the coordination environments observed in the Np(VI) solid-state species formed during the oxidation experiments. Identifying the importance of the $HO^{\bullet}$ in the oxidation state of Np also inspired our future work in understanding the role of free radicals in neptunium chemistry.

Summary: The counterion influenced the rate of Np(V) oxidation to Np(VI) neptunates and $[Np(VII)O_4OH]^{3-}$ complexes. Oxidation to Np(VII) during ozonolysis takes place through the presence of hydroxyl radicals.

2.5. *U(VI) and Np(V) hydrolysis in chloride media*

During our work with uranyl and neptunyl tetrachlorides, we also synthesized novel hydrated forms of these complexes that led to interesting insights into the hydrolysis of the actinide cation. In the uranyl system, we were able to synthesize the uranyl aqua chloro complex ($(C_4H_{12}N_2)_2[UO_2Cl_4(H_2O)]Cl_2$) using piperazinium as a charge-balancing ligand, and the structure was determined using single-crystal X-ray diffraction (Rajapaksha et al., 2023c). DFT calculations in this study demonstrated that changing the primary coordination sphere from $[UO_2Cl_4]^{2-}$ to $[UO_2Cl_4(H_2O)]^{2-}$ significantly strengthens the U═O bonds due to the stabilization of uranyl σ_g, σ_u, π_g, and π_u orbitals. Stronger U═O bonding is also reflected in minimal hydrogen bonding with the oxo group and manifests in blue-shifting of the uranyl stretching features in the vibrational spectroscopy. With a stronger U═O bond, there is a reduction in the Lewis basicity of the axial oxygen atom, which limits additional hydrogen bonding interactions. While actinyl tetrachloro complexes tends to form discrete molecular units, Np(V) chloro and aquachloro complexes have been observed as 1-D chain topologies. We were able to synthesize, structural characterize, and vibrational analyze two 1-D Np(V) chain compounds: $(CH_6N_3)_2[NpO_2Cl_3]$ and $Rb[NpO_2Cl_2(H_2O)]$ (Rajapaksha *et al.*, 2024). Structural analysis of the solid phases revealed the presence of 1-D chain topologies built from repeating $Np(V)O_2^+$ units linked by bridging Cl^- anions. The Np=O bond lengths within these chains are like those observed for the isolated $[NpO_2Cl_4]^{3-}$ coordination complex, however bond order analysis indicates that the neptunyl bond is weakened by 6.5% compared to the molecular unit. Raman spectroscopy showed that there was a red-shift in the neptunyl symmetric stretch and weakening of the Np=O bond associated with the guanadinium neptunyl chloride compound compared to rubidium form. DFT calculations were also performed to evaluate the formation the 1-D chain from the molecular precursors, and the thermodynamically favored pathway suggest that propagation of the molecular units occur via dissociative ligand substitution reactions.

2.6 *Workforce training and recruitment of students into the field*

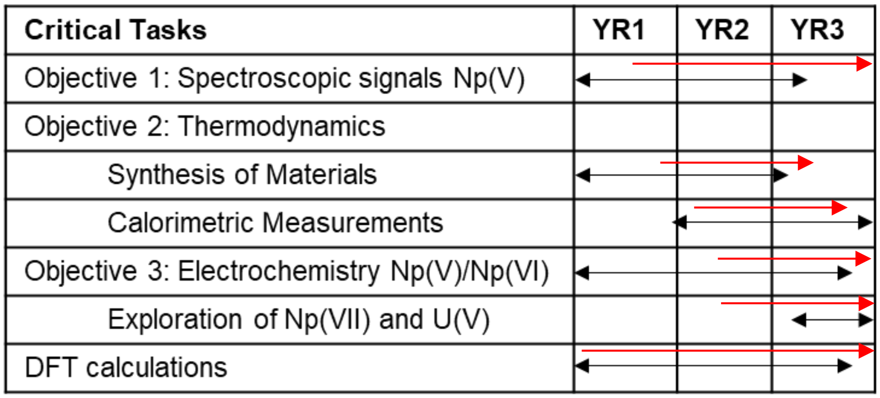
The work highlighted above includes the training of Ph.D. students to safely handle transuranium materials, while making progress on the overall goals of the proposed work. Graduate students trained on this project have transitioned into postdoctoral scholar positions working in transuranic laboratories (Dr. Mikaela Pyrch, Ph.D. 2022, Arnold Research Group UC Berkeley/LBL; Dr. Dmytro Kravchuk, Ph.D. 2023, Heavy Elements Group, ANL) based upon their experience on this project. I have three current students, Grant Benthin, Harindu Rajapaksha, and Emma Markum who also received training as part of this project and are projected to graduate with their Ph.D. in Spring 2026 (Grant and Harindu) and 2027 (Emma). In addition, my co-advised computational graduate student (Dr Logan Augustine, Ph.D. 2023) was able to apply DFT methodologies to uranium systems and then received a DOE Graduate Student Research (SCGSR) fellowship working with Drs. Ping Yang and Enrique Batista (LANL) to develop his skills in transuranic computational chemistry. His SCGSR project built off of ideas in Objective 3 of this project where they utilized DFT methods to evaluate the impact of electron donating and withdrawing ligands on the redox reaction between Np(V) and Np(VI) (Augustine I., 2023c). This opportunity led to his current position as a postdoctoral scholar at LANL.

Another important aspect of our work is to introduce undergraduate students to the field of radiochemistry and actinide chemistry through hands-on research experience. Their initial training with U-238 contributed to the goals of the grant by working with UO_2^{2+} analogues to provide a complete understanding of the chemistry before we move into systems with higher radioactivity and serve as way to benchmark our Np complexes. In addition, undergraduate students who work with me for several years can assist in the Np-237 studies, which provides additional understanding of transuranic chemistry. These students are also included as authors on manuscripts associated with this funded project (Pyrch *et al.*, 2022a, Pyrch *et al.*, 2022b, Rajapaksha *et al.*, 2023b, Williams *et al.*, 2021). Three undergraduate students participated in work associated with this funded project (James Williams, Haley Lightfoot, and Maguire Kasperski). James graduated with his B.S. in chemistry in 2021 and began a position at PAE National Security Solutions. Maguire graduated with his B.S. degree in Environmental Science and is currently a guide and photographer. Hailey attended the ACS/DOE Summer School in Nuclear and Radiochemistry in 2022 and then completed her B.S. in chemistry in 2023. She is currently in the Ph.D. program at Michigan State University working with Professor Alyssa Gaiser on radium chemistry.

3 Schedule delays and final progress on the proposed work

Scheduling challenges occurred due to delays related to the COVID-19 shutdown. We were able to stay relatively productive and meet many project goals by front loading the computational work and focusing on uranium studies that could be performed while social distancing. With relaxation of the distancing restrictions in 2021, we were able to begin the Np related studies. We have completed all experimental and computational studies on the proposed work. All work has been published, except for the collaborative work with Professor Blakemore, which is currently in preparation for submission in the next few weeks. Results from this funded effort provided insights into the importance of intermolecular interactions in controlling the chemical behavior of neptunium in aqueous solutions that have a direct impact on actinide separations and environmental transport modeling. We have provided a systems level understanding of the neptunyl cation to provide chemical controls within aqueous solutions and fill gaps in our knowledge of periodic trends along the actinide series. In addition, we further explored the relationships between subtle changes in electronic structures and related chemical and physical properties. This work also provided the opportunity for graduate students to receive training on the safe handling of transuranic compounds and supported the growing radiochemistry program at the University of Iowa.

Table 1. The projected (black arrows) and actual timelines (red arrows) for completion of the project.



Critical Tasks	YR1	YR2	YR3
Objective 1: Spectroscopic signals Np(V)			
Objective 2: Thermodynamics			
Synthesis of Materials			
Calorimetric Measurements			
Objective 3: Electrochemistry Np(V)/Np(VI)			
Exploration of Np(VII) and U(V)			
DFT calculations			

4 Published Manuscripts

Augustine, L. J., M.M.F. Pyrch, D. V. Kravchuk, J. M. Williams, S.E. Mason, and **T. Z. Forbes**. "Density Functional Theory Guided Investigation of Ligand-Induced Neptunyl-Neptunyl Interactions." (2023a) *European Journal of Inorganic Chemistry,* 26, e202200693

Augustine, L. J., H. Rajapaksha, **T. Z. Forbes,** and S. E. Mason "Periodic Density Functional Theory Calculations of Uranyl Tetrachloride Compounds Engaged in Uranyl-Cation and Uranyl-Hydrogen Interactions: Electronic Structure, Vibrational, and Formation Energy Analysis", (2023b) *Inorganic Chemistry,* 62, 372-380.

Augustine, L. J., J. M. Kasper, **T. Z. Forbes**, S. E. Mason, E. R. Batista, P. Yang. "Influencing properties of the neptunyl (V, VI) cations with electron-donating and withdrawing Groups." (2023c) *Inorganic Chemistry*, 62, 6055-6064.

Hutchison, D.C., D.V. Kravchuk, H. Rajapaksha, S. Stegman, **T. Z. Forbes** and R. E. Wilson

“Synthesis of Li_2NpO_4 and Li_4NpO_4, from aqueous lithium hydroxide solutions under mild hydrothermal conditions and determination of their single-crystal structures.” (2023) *Inorganic Chemistry,* 62, 16564-16573.

Kravchuk, D. V., L. J. Augustine, M.M.F. Pyrch, G. C. Benthin, H. Rajapaksha, S. E. Mason, E. R. Batista, P. Yang and **T. Z. Forbes** “Insights into the mechanism of neptunium oxidation to the heptavalent state”, (2024) *Chemistry – A European Journal*

Kumar, A., R. R. Golwankar, M. M. F. Pyrch, K. P. Carter, A. G. Oliver, V. W. Day, **T. Z. Forbes***, and J. D. Blakemore*. “Macrocyclic control of electron transfer to high valent uranium in heterobimetallic complexes”, In preparation, *Chemical Science*

Pyrch, M. M., L. J. Augustine, J. M. Williams, S.E. Mason and **T. Z. Forbes**. “Use of vibrational spectroscopy to identify the formation of neptunyl-neptunyl interactions: A paired Density Functional Theory and Raman spectroscopy study.” (2022a) *Dalton Transactions (invited article - listed as Hot Paper)*, 51, 4772-4785.

Pyrch, M. M. F., J. Bjorklund, J. M. Williams, M. Kasperski, S.E. Mason, and **T. Z. Forbes**. “Investigations of the Cobalt Hexamine Uranyl Carbonate System: Understanding the Influence of Charge and Hydrogen Bonding on the Modification of Vibrational Modes in Uranyl Compounds.” (2022b), *Inorganic Chemistry*, 61, 15023-15036.

Rajapaksha, H. G. Benthin, E.L. Markin, S.E. Mason, and **T. Z. Forbes***, “Synthesis, characterization, and Density Functional Theory investigation of $(CH_6N_3)_2[(NpO_2)Cl_3]$ and $Rb[(NpO_2)Cl_2(H_2O)]$ chain structures”, (2024) *Dalton Transactions* 53, 3397-3406.

Rajapaksha, H., L. J. Augustine, S. E. Mason, and **T. Z. Forbes** “An experimental and computational effort to determine the formation enthalpy and vibrational properties of solid-state uranyl halides and its influence by hydrogen bonds.” (2023a) *Angewandte Chemie International Edition (designated VIP by the editors)*, 33, e202305073

Rajapaksha, H. G. Benthin, D. V. Kravchuk, H. Lightfoot, S.E. Mason, and **T. Z. Forbes**, “Three-Dimensional Non-Covalent Interaction Network within $[(NpO_2)Cl_4]^{2-}$ Coordination Compounds: Influence on Thermochemical and Vibrational Properties”, (2023b) *Inorganic Chemistry,* 62, 17265-17275.

Rajapaksha, H. S.E. Mason, and **T. Z. Forbes**, “Synthesis, characterization, and Density Functional Theory Investigation of the $[(UO_2)Cl_4H_2O]^{2-}$ Complex”, (2023c) *Inorganic Chemistry* 62, 35, 14318-14325.

Williams, J. M., M. M. Pyrch, D.K. Unruh, H. Lightfoot, and **T. Z. Forbes** “Influence of heterocyclic N-donors on the structural topologies and vibrational spectroscopy of uranyl selenate phases.” (2021) *Journal of Solid-State Chemistry*, 304, 122619.

5 Invited and contributed seminars

Forbes, T.Z. “Chemistry with a dash of spice: Intersection of actinides and high radiation fields.” Plenary Lecture, Actinide Conference, Golden, CO, June 2023. (invited)

Forbes, T.Z. “Identifying structural features that result in activation and coupling of vibrational modes in actinyl materials.”, AVS International Symposium and Exhibition, Virtual, October 24-29,

2021. (invited)

Forbes, T.Z. “Identifying structural features that result in activation and coupling of vibrational modes in actinyl materials.” Spring/Fall Materials Research Society Meeting, Virtual meeting November 2020. (invited)

Forbes, T. Z. “The Transport of Nuclear Waste in Natural Aqueous Systems.” Grinnell College, March 3, 2022 (Graduate student recruiting talk)

Forbes, T. Z. “The Transport of Nuclear Waste in Natural Aqueous Systems.” University of Dubuque, September 24, 2021 (Graduate Ssudent recruiting talk)

Rajapaksha, H., L. Augustine, M. Pyrch, T. Z. Forbes, S.E. Mason. “Paired synthetic and computational efforts exploring hydrogen bonding effects on bonding, electronic structure and spectroscopic signals of uranyl halides.” Midwest Regional ACS meeting, Iowa City, IA, October 2022

Forbes, T. Z. “Impacts of Li^+ interactions on the vibrational features of Np(V), Np(VI), and NP(VII) solutions and solid materials.” Fall ACS meeting, Chicago, IL August 2022.

Kravchuk, D. V., Pyrch, M. M., Augustine, L., Mason, S. E., Forbes, T.Z. “Complete vibrational analysis of neptunium(VII) compounds: Point group symmetry challenge” Spring ACS Meeting, San Diego, CA, March 2022.

Pyrch, M. M. F., Kravchuk, D. V., Forbes, T.Z. “Formation of actinyl-cation interactions and the impacts of Li^+ on the actinyl cation vibrational bands in solution” Spring ACS Meeting, San Diego, CA, March 2022.

Forbes, T.Z. “Spectroscopic characterization of actinyl-actinyl interactions in aqueous solutions and solid-state materials” Fall Virtual ACS Meeting, Atlanta, GA, August 2021.