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Synthesis and Characterization of Tetravalent Actinide Amidinate Halide Complexes

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Introduction
Amidinate ligands have attracted considerable attention in the field of coordination chemistry over the last decades as versatile soft N-donor ligands to stabilize the transition metal complexes in various oxidation states. The additional advantages of employing amidinates over other ligand systems are their high modularity and easy access to a variety of analogs by altering the substitution patterns through straightforward synthetic procedures. There have been a number of studies on the transition metal amidinate complexes including lanthanides, and even some early actinides. Still, these studies are mainly limited to thorium(Th) and high valent(V, VI) uranium(U) complexes. Here we focused on the interaction of An(IV) complexes (An= Th, U, and Np) with benzamidinate ligands to provide a comprehensive understanding of the electronic properties of actinide compounds.

Results and Discussion
In this study, we successfully synthesized a series of tetravalent actinide tris-benzamidinate chloro complexes [AnCl(amid)₃] (An= Th, U, and Np; amid= iPr₂BA and (S)-PEBA). Furthermore, we also obtained additional halide complex series (F, Br) by halogen exchange reactions on chloro complexes as precursors to investigate the conformational stability of the complex. The crystal structures of the model actinide complexes were determined by SC-XRD, showing three benzamidinates and one halide ligand coordinated to the actinide metal center in a mono-capped distorted octahedral coordination geometry, resulting in a propeller-like shape with the halide lying on the rotation axis. The actinide complexes were also characterized in solution by using paramagnetic NMR spectroscopy to elucidate structural and chemical bonding situations with an increasing number of unpaired electrons along the 5f series.

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