
Selectivity in Ligand Binding to Uranyl Compounds: A Synthetic, Structural, Thermodynamic and Computational Study

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ABSTRACT

In aqueous solution, it is well known that uranium exists predominantly in the form of the uranyl ion UO_2^{2+} with the uranium atom in the +6 oxidation state. Aqueous uranium chemistry is of interest in two main areas. First, the large amount present in the world's oceans presents an opportunity to provide a long-term source of this important element. Second, there is an important need to control the movement of uranium in contaminated groundwater. This proposal outlines a plan of research aimed at developing our fundamental understanding of how donor ligands bind to the uranyl ion in aqueous solution. The results of these studies will impact the science and technology underlying our approaches to controlling the behavior of uranium in these systems.

Estimates of the total mass of uranium in seawater are on the order of 4.5 billion tons, one thousand times that present in terrestrial ores (the total ocean volume is approximately $1.4 \times 10^9 \text{ km}^3$; uranium is present at ca. 3 ppb). It is the 48th most abundant element in the earth's crust, i.e. higher than for antimony, tin, cadmium, mercury, or silver. Despite this abundance, there are several major impediments to using uranium from seawater as a viable economic source of this important element. First, the concentration of uranium is very low; second, there are relatively high concentrations of other ions that interfere with uranium separation; and third, the volumes involved are extremely high.

We propose to extend our recent work on non-aqueous *f*-element chemistry to study aqueous systems with a focus on the uranyl ion, UO_2^{2+} . We believe that the goal of developing a practical system for uranium separation from seawater will not be attained without new insights into our existing fundamental knowledge of actinide chemistry. We posit that detailed studies of the kinetic and thermodynamic factors that influence interactions between *f*-elements and ligands with a range of donor atoms is essential to any major advance in this important area. The goal of this research is thus to broaden the coordination chemistry of the uranyl ion by studying new ligand systems via synthetic, structural, thermodynamic and computational methods. We anticipate that this fundamental science will find use beyond actinide separation technologies in areas such as nuclear waste remediation and nuclear materials.