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Although the natural abundance of actinides is low, their chemistry is an active and broad field of research. Being one of the world's cheapest and most abundant sources of energy, nuclear power is, however, shadowed by its toxic waste products and the expensive and cumbersome reprocessing thereof. Understanding the physico-chemical behavior of actinides is crucial for different aspects of the nuclear fuel cycle, from the development of novel fuels to the characterization of waste products and remediation of radioactive contamination. Therefore, an essential element of actinide-based research is the prediction of the stability and properties of actinide compounds. Unfortunately, the acute toxicity, radioactivity, and instability of actinide compounds complicate experimental studies of their thermodynamics, kinetics, complexation, and reaction mechanisms. Theoretical approaches can be used to determine molecular properties and can provide a fundamental understanding of actinide reactivity and reaction mechanisms. This information can be used to discern "what's inside the soup" and "how to get it out". A reliable quantum-mechanical treatment of the chemical properties of actinide elements must satisfy two requirements: (i) it must provide a reliable treatment of relativistic effects and (ii) it must address the fact that electrons do not move independently, but in a correlated fashion.

Unfortunately, the computational resources required by standard quantum-mechanical methods grow exponentially with the size of the system, an effect known as the curse of dimension. Since the actinide-containing molecules of relevance to nuclear waste separation and reprocessing contain hundreds of electrons, innovative new approaches that break the curse of dimension must be developed. One such approach models many-electron molecules as collections of noninteracting electron pairs, called geminals. Standard geminal methods are inappropriate for nuclear waste chemistry, however, and must be extended to include (i) the effects of the fast movement of electrons (special relativity), (ii) correlations between electrons beyond electron-pairing effects, (iii) the modeling of electronically excited states, and (iv) the description of unpaired (open-shell) electrons. Achieving these goals within the context of an electron-pairing theory is immensely challenging, requiring new mathematical developments, software development and testing, and computational validation and applications to actinide chemistry. The extended geminal models thus developed will provide the first direct, atomistic, and quantitative computational model for understanding nuclear waste reprocessing and will provide the essential insights that are needed to guide the synthesis of new actinide compounds that can be used to separate Uranium and Plutonium from the other components in the soup of nuclear waste. The extended geminal models developed will also provide robust, inexpensive methods that can be used in many other areas of chemistry and materials physics like lanthanide and transition-metal chemistry, biochemistry, and semiconductor physics.