

Evaluation of the Accuracy of Computed Uranium Thermodynamic Properties with Dispersion-Corrected Density Functional Theory

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Density-functional theory (DFT) is a powerful tool to investigate properties of materials. However, there are deficiencies in DFT that result in poorly calculated formation enthalpies. Actinide elements in nuclear materials provide additional challenges due to the complex electronic structure of the f-orbitals that can result in the convergence to a meta-stable state electronic state, which can severely underestimate the calculated formation enthalpies and related thermodynamic properties. In this work, we present a study of several uranium containing compounds (e.g., U, UC, UCl₃, UN, UO₂, U₃Si₂) using DFT incorporating the exchange-hole dipole moment (XDM) dispersion model with different orbital electronic configurations. Our results show how incorporating a dispersion correction and orbital electron configuration affects the crystal structure and enthalpies. The results will be compared to experimental values where available.

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