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Thermodynamic Stability of Actinide-Dioxide Solid Solutions and Surface Interactions with Water[1]

MARK ASTA, Department of Materials Science and Engineering, University of California, Berkeley

Fluorite-structured actinide dioxides are the most common forms of fuel used in nuclear energy production worldwide. This talk will provide an overview of insights into the energetics of these compounds derived through the combination of density-functional-theory-based computational studies (including Hubbard-U corrections) and calorimetric measurements. The talk will focus on two main topics: the mixing energetics of cation solid solutions, and the energetics of water adsorption on the surfaces of these compounds. For the first topic, we present results for ThO_2 and UO_2 based solid solutions, highlighting the roles of elastic energy arising from cation size mismatch, electrostatic interactions, and charge-transfer reactions, in governing the sign and magnitude of the mixing energetics. For water adsorption, we contrast results for surface and adsorption energies on two fluorite-structured compounds, ThO_2 and CeO_2, that are relevant for understanding the behavior of water on actinide oxide surfaces more generally. Through a comparison between calorimetric measurements and computational results we assess the level of accuracy achieved in the computational modeling, and suggest areas where further experimental studies would be particularly useful.

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