Thermophysical properties of crystalline and amorphous uranium silicides from first-principles

Zhi-Gang Mei, Yinbin Miao, and Abdellatif M. Yacout

Argonne National Laboratory, Argonne, IL, USA

Uranium silicide compounds (e.g., U_3Si_2) have attracted great interest as fuels with enhanced accident tolerance for service in demanding reactor applications such as commercial light water reactors (LWRs) and more recently as low-enriched uranium (LEU) fuels for high-performance research reactors. This is mainly driven by its high uranium density and improved thermal conductivity compared to UO_2 fuel used in current LWRs. Uranium silicide compounds are known to become amorphous under irradiation at low temperatures. Recent studies by Miao et al found that Xe implanted U_3Si_2 decomposes into a Si-enriched nanocrystalline USi matrix phase and a U-enrich amorphous inclusion phase at 450 °C. Up to now there are very few studies of uranium silicide compounds at LWR temperatures, especially the fuel behavior under irradiation. On the other side, physics based computational modeling and simulation can provide unique perspectives on examining the performance of uranium silicide fuels at LWR conditions. To perform meaningful fuel performance simulations, fundamental material properties of nuclear fuels, such as thermophysical properties, are crucial. To this end, we systematically investigated the thermophysical properties of both crystalline and amorphous uranium silicide compounds (U_3Si_2, U_3Si, and USi) using first-principles calculations, including structural, electronic, mechanical, and thermodynamic properties. The atomic structures of the amorphous phases were generated using the melting and quenching method. The calculated electronic structures suggest that all the uranium silicide compounds are metallic phases. The predicted thermophysical properties of the crystalline phases compare well with the experimental measurement by White et al. We expect that the obtained material properties of uranium silicides will be useful to the fuel performance simulation in the future.