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Thermodynamics of Uranium Silicide, Nitride and Carbide

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Uranium silicide, nitride, and carbide have attracted considerable interest due to increasing demands for accident-tolerant nuclear fuels, especially after the Fukushima disaster in 2011. Uranium dioxide, UO_2, has been the major nuclear reactor fuel, but its thermal conductivity is undesirably low, resulting in large temperature gradients and thus excessive stored energies. Since UN, UC, and U-rich U-Si binary compounds (such as U_3Si_2) possess much higher thermal conductivities, and also contain higher amounts of U, they have great potential for nuclear fuel applications. However, to utilize these compounds as high-efficient accident-tolerant fuels, it is essential to have fundamental knowledge about their thermodynamic properties and phase stability at relevant conditions. In a series of studies, we have determined the thermodynamic properties of U_3Si_2, USi, U_3Si_5, UN, and UC using high-temperature drop calorimetry, low-temperature heat capacity measurements, and differential scanning calorimetry. Enthalpies of formations from constituent elements were measured by high-temperature transposed temperature drop and oxide melt calorimetry, standard entropies via low-temperature heat capacity measurements, and enthalpies of reactions during thermal oxidation processes by coupling differential scanning calorimetry with thermalgravimetric analysis and mass spectrometry. The obtained thermodynamic parameters are discussed in terms of crystal structure, chemical bonding, and electronic structure. The results have important implications for the utilization of these compounds as accident-tolerant fuels for the next-generation nuclear reactors.