

Energetics of Uranium Silicides

CHENG-KAI CHUNG^{1,2}, XIAOFENG GUO^{3,1}, JOSHUA WHITE⁴, ANDREW NELSON⁴, HAKIM BOUKHALFA¹, ROBERT ROBACK¹, HONGWU XU^{1*}, ALEXANDRA NAVROTSKY^{2*}

¹ Earth and Environmental Sciences Division, Los Alamos National Laboratory, Los Alamos, NM 87545, USA

(*correspondence: hxu@lanl.gov)

² Peter A. Rock Thermochemistry Laboratory and NEAT ORU, University of California Davis, Davis, CA 95616, USA

(*correspondence: anavrotsky@ucdavis.edu)

³ Department of Chemistry, Washington State University, Pullman, WA 99164, USA

⁴ Materials Science and Technology Division, Los Alamos National Laboratory, Los Alamos, NM 87545, USA

U-Si intermetallic compounds have drawn great attention for their applications as accident-tolerant nuclear fuels, since these materials generally possess superior thermal conductivities [1-4]. Further, U_3Si and U_3Si_2 have higher U densities than traditional UO_2 fuel, which would improve fuel performance without extensive U-enrichment [1]. However, as these materials are essentially line compounds and air sensitive, commercial production targeted to one specific phase with existing synthesis methods would yield minor secondary phases [1-3]. Experimental studies have also revealed discrepancies in the published U-Si phase diagram [2]. These suggest a better understanding of the thermodynamics of this system is needed to predict the relative-stabilities of the phases under conditions of interest. Transposed temperature drop calorimetry in a flowing oxygen atmosphere has been performed with a Setaram AlexSys Calvet-type calorimeter at LANL. The enthalpies of formation of these U-Si phases from elements are then derived. These data serve as benchmark parameters for predicting their phase stabilities as functions of composition, temperature, and pressure. Our results show that the standard formation enthalpy per mole atoms of U_3Si_5 is more exothermic than those of U_3Si_2 and USi . This trend is in agreement with computational results from first principles DFT and CALPHAD calculations [5].

[1] White et al. (2015) Journal of Nuclear Materials 464, 275-280. [2] White et al. (2015) Journal of Nuclear Materials 456, 442-448. [3] White et al. (2016) Journal of Nuclear Materials 471, 129-135. [4] White et al. (2014) Journal of Nuclear Materials 452, 304-310. [5] Noordhoek et al. (2016) Journal of Nuclear Materials 479, 216-223.