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Combining nanocalorimetry and ab-initio calculations for multilayer reaction studies

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The extraordinary sensitivity and extremely small thermal mass of nanocalorimetry sensors allow the study of solid-state reactions in thin films over a broad range of heating rates, from isothermal to 10^5 K/s. Ab-initio simulations provide insight in the phase transformation and diffusion behavior of a material at the atomistic scale. In the presentation, I am going to talk about the energetics, diffusion kinetics, and structural evolution of the solid-state reaction in Zr/B and Zr/B_4C reactive multilayered nano-laminates (RMNLs) using the combination approach of ab-initio computational methods and nanocalorimetry experiments.

On the experimental side, we examine the effects of heating rate (from 3,000 to 50,000 K/s) and bilayer period on the reactions in the RMNLs. The microstructural evolution of the multilayers during the reaction is revealed using transmission electron microscopy. We demonstrate low-temperature processing of ultra-high-temperature ceramics of highly textured ZrB_2 from the RMNLs. The simulations elucidate the effects of concentration (ZrB_x, $0<x\leq2$) and temperature on the diffusion kinetics and phase evolution of the Zr-B alloys in atomistic scale.

Both experimental and simulation results show that the synthesis of crystalline ZrB_2 using nanolaminates of Zr and B is facilitated by the formation of an intermediate amorphous phase that allows fast transport of boron. Also, the diffusion energy barriers determined using nanocalorimetry (0.52±0.17 eV) are in good agreement with the value determined from the ab-initio calculations (0.75±0.07 eV). We further investigated the kinetic effect of carbon in the reaction between Zr and B. A kinetic analysis shows that carbon lowers the energy barriers for both inter-diffusion and crystallization by more than 20%.

We believe that this study provides fundamental insight in the reaction kinetics of Zr and B and the kinetic role carbon plays in the synthesis of ZrB_2, and may bring about the possibility of low-temperature processing of ZrB_2. Furthermore, we believe that the combination of nanocalorimetry and first-principles modeling is a unique and powerful tool in materials research that can be used for a broad range of materials systems and phenomena.