

Exact theory of solute transport in zirconium

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While first-principles methods compute activated state energies, upscaling to mesoscale mobilities requires the solution of the master equation. A new automated Green function approach for transport both determines the minimum set of transition states to calculate from symmetry and computes the dilute-limit transport without additional approximations. This approach is both more accurate and efficient than stochastic approaches like kinetic Monte Carlo. It also permits straightforward evaluation of uncertainty quantification and derivatives of transport coefficients. We showcase the new functionality by computing transport coefficients for solutes in zirconium: Al, Be, Cr, Fe, Sn, Ni. Previous theoretical models to predict diffusivity from atomic jump frequencies had made uncontrolled approximations that affect their accuracy. Density-functional theory identifies nine different solute-vacancy configurations from which symmetry analysis determines 17 transition states corresponding to a 27-frequency model; we also consider interstitial diffusion mechanisms for Be, Cr, Fe, and Ni. Our Green function approach computes diffusivity for the six solutes using the density-functional theory data. We also study the effect of nonequilibrium vacancy concentration on solute diffusivity, where we find that Sn and Al diffusivities increase in response to high vacancy concentration, but counterintuitively the diffusivity of Be, Cr, Fe, and Ni decrease. This provides useful data for modeling alloy behavior under irradiation.