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Molecular Dynamics Analysis of Thermodynamic and Kinetic Properties of Bulk PdH_x

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ABSTRACT

This work uses molecular dynamics simulations to study PdH_x bulk properties relevant to hydrogen storage applications including lattice constants, Gibbs free energies, elastic constants, and diffusivities as a function of temperature and composition. During the course of the calculations, we demonstrated robust molecular dynamics methods to calculate highly converged finite temperature elastic constants, and highly converged overall diffusion properties accounting statistically for all possible atomic jump mechanisms. The robust calculations reveal ideally linear Arrhenius plots of hydrogen diffusion at low compositions, and abnormally nonlinear plots at high compositions. The fundamental cause for this behaviour has been identified. All of our calculated results are compared with available experimental data. Remarkably, the non-Arrhenius behaviour is validated by experiments. While elastic constants calculated at the 0 K temperature agree well with those measured at ~ 4 K, they differ at higher temperatures. Simulations using a vibrational loading condition suggests that this discrepancy may origin from the difference in the sonic methods typically used to measure elastic constants in experiments and the simulated mechanical testing conditions for PdH_x where significant hydrogen diffusion could occur in responding to deformation.

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