Hessisches Kompetenzzentrum
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Atomic Scale Modeling of Coherent Phase Transitions in Energy Storage Materials



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Project Term
2023 - 2024

Clusters
Lichtenberg II Cluster Darmstadt

Software
LAMMPS, Ovito

Additional Software
Atomsk

Institute
Fachgebiet Materialmodellierung

University
Technische Universität Darmstadt

Introduction

Hydrogen has emerged as a crucial element in the pursuit of decarbonization and the transition to a sustainable energy system. As the most abundant element in the universe, hydrogen's potential as a clean energy carrier is immense. Effective hydrogen storage materials are essential for their widespread adoption as an energy source. Among the various materials investigated for hydrogen storage, palladium has shown significant promise. When the pressure of surrounding hydrogen gas is elevated, hydrogen can be continuously absorbed into the palladium structure, forming an α dilute phase with a low concentration of hydrogen and a β phase with a higher concentration of hydrogen. This capability to store hydrogen efficiently makes palladium an attractive material for energy storage applications. The goal of this work is to study the type and mechanism of phase transition within the miscibility gap with atomistic simulations by applying different boundary conditions.

Methods

Molecular dynamics (MD) is a powerful computational technique used to study the behavior and properties of atoms and molecules at the molecular scale. It provides insights into the dynamics, thermodynamics, and structural characteristics of systems under study. In MD simulations, the motion of atoms is tracked over time based on classical mechanics principles. This method is invaluable for understanding how materials respond

to different conditions at an atomic level. Molecular Statics (MS) is another computational method used to explore the potential energy landscape and find stable configurations in materials simulations. Unlike MD, which considers the motion of atoms, MS involves treating atoms as if they have no kinetic energy (T = 0 K). This approach allows for the analysis of local minima and activation barriers, providing detailed insights into the stable configurations and possible transition states within the material.

Results

Through Molecular Statics (MS) simulations, we constructed the ”Energy-Volume” curve, revealing insights into equilibrium energy, bulk modulus, and lattice constants across various compositions of the palladium-hydrogen system. Our exploration extended to kinetics, focusing on hydrogen diffusion within the palladium structure. Analysis of Mean Squared Displacement (MSD) and Arrhenius plots provided critical insights into the diffusion mechanisms, although discrepancies compared to experimental data were observed. Investigations into phase separation within closed and open systems unveiled intriguing results. In closed systems, increased simulation cell dimensions did not lead to phase separation. In contrast, open surface boundary conditions revealed distinct decomposition mechanisms under NPT (constant pressure and temperature) and NVT (constant volume and temperature) regimes.

Discussion

The discrepancies observed in diffusion mechanisms compared to experimental data could be attributed to limitations in simulation conditions or approximations in capturing real system behaviors. Mechanical boundary conditions played a significant role in phase separation, aligning with thermodynamic predictions. These findings highlight the complexity and sensitivity of phase behavior to boundary conditions. Further simulations with thin film structures exhibited coherent phase separation due to their high surface-to-volume ratio. In these thin films, defects and dislocations migrated towards the surface, thereby reducing the energy costs associated with dislocation annihilation. This migration towards the surface indicates a mechanism by which the material can lower its overall energy, facilitating phase separation and potentially enhancing hydrogen storage capabilities. Overall, our study provides a comprehensive understanding of the phase transitions and diffusion mechanisms in palladium-hydrogen systems, offering valuable insights for the development of advanced hydrogen storage materials.

Last Update: 2024-06-17 14:57