Molecular Dynamics Study on the Effect of Hydrides on the Tensile Properties of Zirconium

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Abstract

Hydrides are common defects generated in zirconium alloy cladding tubes through the zirconium-water reaction with primary coolant during normal operation of nuclear power plants. This study employs molecular dynamics simulations with the COMB3 potential to construct zirconium-based models containing hydrides for uniaxial tensile testing, investigating the effect of hydride density on the mechanical properties of zirconium. The results demonstrate that when hydride density ranges from 0 to 1078 μg/g, the yield strength, strain, and Young' s modulus decrease with increasing hydride density. During the elastic stage, increased hydride density expands the stress concentration region, facilitating dislocation nucleation; during plastic deformation, higher hydride density causes initial dislocations to preferentially propagate around the hydrides. When hydride density ranges from 1078 to 2311 μg/g, the yield strength, strain, and Young' s modulus increase with hydride density, attributed to the generation of numerous dislocations and resultant dislocation pile-up at high hydride densities.

Full Text

Preamble

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Molecular Dynamics Study of the Influence of Hydrides on the Tensile Properties of Zirconium

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Abstract

[Background]: Hydride is a common defect formed through the zirconium-water reaction between zirconium alloy cladding tubes and primary coolant during normal operation of nuclear power plants. [Purpose]: This study investigates the effect of hydride density on the mechanical properties of zirconium. [Methods]: Using molecular dynamics simulations with the COMB3 potential function, we constructed zirconium matrix models containing hydrides and performed uniaxial tensile simulations. [Results]: The results show that when hydride density ranges from 0 to 1078 μg/g, yield strength, strain, and Young' s modulus decrease with increasing hydride density. When hydride density is between 1078 and 2311 μg/g, yield strength, strain, and Young' s modulus increase with increasing hydride density. Conclusions: In the elastic stage, increasing hydride density enlarges the stress concentration region, which facilitates dislocation nucleation. During plastic deformation, the initial dislocations tend to expand around the hydrides as hydride density increases. At higher hydride densities (1078–2311 μg/g), numerous dislocations are generated, leading to dislocation pile-up.

Keywords: Zirconium alloy; Hydride; Uniaxial tension; Molecular dynamics

Introduction

Zirconium alloys are widely used as structural materials for nuclear fuel cladding in light water reactors due to their excellent high-temperature mechanical properties, good corrosion resistance, and low thermal neutron absorption cross-section [1-6]. A critical issue during service is that zirconium cladding reacts with primary coolant, producing hydride precipitates through the zirconium-water reaction [7-9]. These hydride precipitates typically degrade the mechanical properties of zirconium alloys [10-12].

In recent years, many researchers have employed molecular dynamics (MD) methods to investigate the influence of hydrides on the mechanical properties of zirconium. A. L. Lloyd et al. [13] used the MEAM potential to study the effect of hydrogen concentration on Zr mechanical properties, finding that higher hydrogen concentrations reduce the ultimate tensile strength and fracture strain required for permanent deformation. Ghaffarian et al. [14] employed the MEAM potential to investigate the deformation mechanisms of hydrides embedded in polycrystalline Zr under tensile loading, elucidating the effect of hydride precipitation on the ductility of zirconium alloys. Poulami et al. [15] used the MEAM potential to study the mechanism of hydrogen concentration effects on zirconium, discovering that increased hydrogen content reduces the strain at fracture by promoting dislocation multiplication. More recently, Mark J. Noordhoek et al. [16] developed the COMB3 empirical potential for Zr, which reproduces stacking fault energies matching DFT calculations and accurately computes mechanical properties of Zr systems. Zhang et al. [17] used the COMB3 potential to predict the formation energy, lattice parameters, and bulk modulus

of δ-$ZrH_{1\cdot0\ 2\cdot0}$, showing good agreement with DFT values.

Building upon Zhang et al.' s [17] validation of the COMB3 potential for simulating hydrides formed through zirconium-water reactions, this study further employs the COMB3 potential to investigate the effect of hydride density on the mechanical properties of zirconium, examining the underlying mechanisms from the atomic scale.

1. Computational Methods

We performed molecular dynamics simulations using the LAMMPS code. The simulation model consisted of a 10.3 nm × 10.1 nm × 20.6 nm cubic box containing 92,160 Zr atoms arranged in an HCP single crystal, as shown in [Figure 1: see original paper]. The X, Y, and Z axes were oriented along the [11-20], [10-10], and [0001] crystallographic directions, respectively, with periodic boundary conditions applied in all three directions to minimize boundary effects. The interatomic interactions were described by the COMB3 potential developed by Noordhoek et al. [16].

To construct hydrides, we used the cubic δ-$ZrH_{1\cdot5}$ phase with a disk-shaped precipitate of radius 15 Å and height 4.785 Å. The lattice parameters were a, b, c = 4.785 Å (α, β, γ = 90.0°) with an FCC structure. The hydrides were embedded in the Zr matrix following the orientation relationship $(0001)\alpha//\{111\}\delta$. Multiple hydride layers (1–15 layers) were placed uniformly with a spacing of 15 Å, with each additional layer increasing the hydride density by 154 µg/g. lists the correspondence between hydride density and the number of hydride layers.

The initial unit cell used standard lattice parameters and structure [18]. A disk-shaped volume was cut from the center of the Zr system and replaced with an equal volume of δ-$ZrH_{1\cdot5}$ precipitate to match the hollow volume. [Figure 2: see original paper] shows schematic diagrams of Zr matrices containing one layer and five layers of hydrides.

The simulation proceeded in two steps. First, we performed energy minimization using the conjugate gradient (CG) method, followed by relaxation in the NPT ensemble at 300 K for 500,000 steps (timestep = 0.1 fs), corresponding to a total time of 50 ps. Second, uniaxial tensile loading was applied along the [0001] direction at a strain rate of 10^{10} s^{-1} at 300 K for 100,000 steps (timestep = 0.5 fs), corresponding to 50 ps and 50% strain. Atomic coordinates, forces in the X, Y, and Z directions, strain, stress, and atomic potential energies were output every 1000 steps. The resulting data were used to generate stress-strain curves in Origin. Young' s modulus was obtained by linear fitting of the slope in the elastic region of the stress-strain curve. For strain e and stress s, the relationship is s = Ee. Using least squares linear fitting, Young' s modulus was calculated as:

$E = \Sigma(e\ s\ ) - n\overline{es}\ /\ \Sigma(e^{\ 2}) - n\bar{e}^2$

where e and s are the mean values of strain and stress samples, and n is the number of samples.

Microstructural evolution during tensile deformation was analyzed using the adaptive common neighbor analysis to output structure type percentages and dislocation analysis (DXA) to quantify dislocation line lengths. The visualization software OVITO was used to display the microstructure and perform stress contour visualization through atomic color coding.

2.1. Stress-Strain Curves and Mechanical Parameters

[Figure 3: see original paper] shows the stress-strain curves for Zr matrices with different hydride densities. All models exhibit similar trends: stress increases with strain until reaching a peak stress that marks the transition from elastic to plastic deformation. Beyond the peak, stress drops abruptly to a plateau value and then fluctuates around this value with further strain increase.

[Figure 4: see original paper] illustrates the variation of yield stress and Young' s modulus with hydride density, revealing an inflection point at 1078 μg/g. When hydride density increases from 0 to 1078 μg/g, yield stress, yield strain, and Young' s modulus all decrease. At 1078 μg/g, the model reaches minimum values: yield stress of 7.69 GPa (42.22% lower than pure Zr), yield strain of 0.0895 (39.34% lower than pure Zr), and Young' s modulus of 112.18 GPa (8.94% lower than pure Zr). When hydride density increases from 1078 to 2311 μg/g, these mechanical parameters gradually increase, reaching 8.23 GPa yield stress, 0.086 yield strain, and 115.08 GPa Young' s modulus at 2311 μg/g.

The trend of hydride density effects on stress-strain curves calculated using the COMB3 potential is consistent with results from A. L. Lloyd [13] and Ghaffarian [14] using the MEAM potential for hydrogen concentration effects. The yield stress for pure Zr (13.31 GPa) shows less than 5% error compared to the value (14 GPa) reported by Amit [19] using the EAM potential, confirming that the COMB3 potential is suitable for zirconium alloy mechanical property calculations.

2.2. Microstructural Analysis

[Figure 5: see original paper] displays the microstructural evolution and stress contours during tensile simulations for hydride densities ranging from 154 to 770 μg/g. The microstructural evolution reveals that dislocation defects nucleate along hydride precipitate interfaces, and the contact area between hydrides and the Zr matrix increases with hydride density, generating more defects. As hydride density increases, dislocation density rises, with most being single dislocations, causing a continuous decrease in tensile strength. At the transition point of 1078 μg/g, the substantial increase in dislocations leads to entanglement and pile-up, causing tensile strength to increase. Since the pile-up is not severe, the mechanical parameters only increase slightly. The stress contours

show that stress concentration regions around hydrides expand with increasing hydride density, resulting from dislocation generation around hydrides. The continuous increase in dislocation density enlarges these stress concentration zones.

To investigate the role of different hydride densities in deformation mechanisms, we analyzed the stress values at hydride tips and dislocation line length variations during tensile loading, as shown in [Figure 6: see original paper] and [Figure 7: see original paper]. [Figure 6: see original paper] demonstrates that stress values at hydride tips increase with hydride density at the same strain level. The maximum stress at hydride tips reaches 5.899 GPa at 154 μg/g and 6.967 GPa at 770 μg/g, consistent with the microstructural evolution showing intensified stress concentration and expanded stress concentration zones around hydrides.

[Figure 7: see original paper] shows that the strain at which dislocations first appear decreases with increasing hydride density. For pure Zr (0 μg/g), dislocations initiate at 15.5% strain, which is 5.1% beyond the yield point (14.75% strain), with a peak dislocation line density of 1478.93 Å. At 154 μg/g, dislocations appear at 11% strain, 12.35% beyond the yield point (12.55% strain), with a peak density of 5532.16 Å. At 770 μg/g, dislocations emerge at 4.5% strain, 52.12% beyond the yield point (9.4% strain), with a peak density of 8377.27 Å. These results indicate that increasing hydride density advances the strain at which dislocations appear and increases dislocation line density, consistent with the microstructural evolution shown in [Figure 5: see original paper].

Conclusions

This work constructed hydride-containing Zr models and performed uniaxial tensile simulations using the COMB3 potential function to investigate the effect of hydride density on mechanical properties. The main conclusions are: (1) As hydride density increases, the yield strength, strain, and Young' s modulus of the Zr matrix initially decrease, reaching minimum values at 1078 μg/g, after which these mechanical parameters gradually increase with further hydride density increase. (2) Increasing hydride density enlarges the contact area between hydrides and the Zr matrix, intensifying stress concentration and facilitating dislocation nucleation, which reduces yield strength and yield strain. When excessive dislocations accumulate and become entangled, dislocation pile-up occurs, leading to increased matrix strength.

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Note: Figure translations are in progress. See original paper for figures.