

MM 4: Materials for the Storage and Conversion of Energy

Metal Hydrides, Hydrogen Embrittlement

Time: Monday 10:15–13:00

Location: H22

MM 4.1 Mon 10:15 H22

Atomic cluster expansion potential for the palladium hydride system — ●MINAAM QAMAR¹, APINYA NGOIPALA², MATOUS MROVEC¹, and MATTHIAS VANDICHE² — ¹ICAMS, Ruhr-University Bochum, Germany — ²Department of Chemical Sciences and Bernal Institute, University of Limerick, Ireland

Palladium (Pd) is extensively studied for metal-hydrogen interactions due to its remarkable ability to absorb large amounts of hydrogen under standard temperature and pressure conditions, forming stable palladium hydride structures. This property makes Pd a critical material for various hydrogen-related applications, including hydrogen storage and as a catalyst for hydrogen evolution reactions. We developed a highly accurate and computationally efficient Atomic Cluster Expansion (ACE) potential to unravel atomistic insights into the Pd-H interaction. This potential was utilized to perform molecular dynamics and Monte Carlo simulations, providing a detailed understanding of the formation mechanisms of palladium hydride nanoclusters and bulk structures.

MM 4.2 Mon 10:30 H22

Hydrogen kinetics in HPT-deformed bulk Mg and Mg-based alloys — ●GIORGIA GUARDI¹, SABINE SCHLABACH^{1,2,3}, JULIA IVANISENKO², STEFAN WAGNER¹, and ASTRID PUNDT¹ — ¹Karlsruhe Institute of Technology (KIT), Institute for Applied Materials (IAM-WK), Karlsruhe, Germany — ²Karlsruhe Institute of Technology (KIT), Institute of Nanotechnology (INT), Karlsruhe, Germany — ³Karlsruhe Institute of Technology (KIT), Karlsruhe Nano Micro Facility (KNMF), Karlsruhe, Germany

Magnesium (Mg) is a light and abundant element that can store hydrogen with a gravimetric density of 7.6 wt. % and a volumetric density of 110 kgH/m³. Therefore, it is a highly promising hydrogen storage material for a sustainable energy economy. However, its technical use is hindered by an inherent kinetic blockade in magnesium hydride. A high content of grain boundaries is expected to improve hydrogen kinetics in Magnesium samples, particularly in the magnesium hydride phase. This can be achieved in bulk samples through the use of severe plastic deformation techniques, such as High-Pressure Torsion (HPT).

High-pressure torsion is not fully effective in reducing the grain size in pure bulk magnesium due to dynamic recrystallization. To overcome this lower limit to bulk magnesium grain size, Mg- 2.3 at.% Zn- 0.17 at. % Zr alloys are used, reaching a final grain size of approximately 100 nm after HPT. This study examines the impact of the sample's microstructure on hydrogen kinetics. Hydrogen absorption is studied at room temperature by gas-phase and electrochemical hydrogen loading.

MM 4.3 Mon 10:45 H22

A first principles study on the free energy landscape of chemisorbed hydrogen atoms on a Pt (111) surface modified by transition metals — ●BINGXIN LI, SUDARSAN SURENDRALAL, MIRA TODOROVA, and JÖRG NEUGEBAUER — Max-Planck-Institut für Nachhaltige Materialien GmbH, Max-Planck-Straße 1, 40237 Düsseldorf

Platinum (Pt) is considered an excellent catalyst for the hydrogen evolution reaction (HER) due to its optimal hydrogen binding strength, as suggested by the Sabatier principle and the volcano plot of the HER. In surface-catalysed electrochemical reactions, the incorporation of adsorbates or dopants into a host surface can significantly enhance the catalytic selectivity, activity, and stability compared to the pristine surface while also introducing some novel properties not observed on the parent surfaces. Transition metals (TMs), such as Cu, Ag, Au, Pd, Ru, Rh, and Ir, form metal-hydrogen bonds with unique characteristics, resulting in varied H adsorption patterns and distinct hydrogen evolution behaviours. Consequently, doping these transition metals into a Pt (111) surface can significantly alter its catalytic properties for the HER, either by modifying H adsorption configurations or by impacting the reaction barrier of the fundamental steps for the hydrogen evolution, though the underlying mechanisms remain unclear. To this end, we employ ab initio molecular dynamics (AIMD) simulations to investigate H adsorption on TM-doped Pt (111) surfaces in the presence/absence of explicit water by constructing laterally resolved free

energy maps of chemisorbed H atoms.

MM 4.4 Mon 11:00 H22

Local hydrogen concentration and distribution in Pd nanoparticles: An in-situ STEM-EELS approach — ●SVETLANA KORNEYCHUK^{1,2}, STEFAN WAGNER¹, DARIUS ROHLEDER³, PHILIPP VANA^{3,4}, and ASTRID PUNDT¹ — ¹IAM-WK, Karlsruhe Institute of Technology — ²Karlsruhe Nano Micro Facility (KNMF), Karlsruhe Institute of Technology — ³Institute of Physical Chemistry, Georg-August-University Göttingen — ⁴Wöhler Research Institute for Sustainable Chemistry (WISCh), Georg-August-University Göttingen

Local detection of hydrogen concentration in metals is of central importance for many areas of hydrogen technology, such as hydrogen storage, detection, catalysis, and hydrogen embrittlement. We demonstrate a novel approach to measure the hydrogen concentration in a model system consisting of cubic palladium nanoparticles, with a lateral resolution down to 4 nm. By measuring the shift of the Pd bulk plasmon peak with scanning transmission electron microscopy combined with energy electron loss spectroscopy during in-situ hydrogen gas loading and unloading, local detection of the hydrogen concentration is achieved in TEM [1]. With this method, concentration changes inside the NPs at various stages of hydrogenation/dehydrogenation are observed with nanometer resolution. The versatility of in-situ TEM allows to link together microstructure, hydrogen concentration and local strain, opening up a new chapter in hydrogen research. [1] S. Korneychuk, et al., 'Local hydrogen concentration and distribution in Pd nanoparticles: An in-situ STEM-EELS approach', Small, accepted

MM 4.5 Mon 11:15 H22

Hydride formation in open thin film metal hydrogen systems: Cahn-Hilliard-type phase-field simulations coupled to elastoplastic deformations — ●STEFAN WAGNER¹, ALEXANDER DYCK², JOHANNES GISY², FREDERIK HILLE², ASTRID PUNDT¹, and THOMAS BÖHLKE² — ¹Institute for Applied Materials (IAM-WK), Karlsruhe Institute of Technology (KIT), Germany — ²Institute of Engineering Mechanics (ITM), Karlsruhe Institute of Technology, Germany

Metal-hydrogen systems are versatile model systems to study alloy thermodynamics and structural phase transitions. Hydrogen absorption and hydride formation in metals induce a volume expansion of the metal, leading to incompatibility stresses at internal and external interfaces. Resulting changes of the thermodynamic stability of phases in response to the mechanical constraint conditions can be understood by investigating the chemo-mechanical coupling [1,2]. Utilizing niobium-hydrogen thin films, combining a Cahn-Hilliard type phase-field theory and Finite-Element-Modeling (FEM) we study the feedback-loop of the local stress state, elasto-plastic deformations and hydrogen diffusion, determining the driving force of precipitation and growth of the hydride phase under open-system conditions. The simulation results are informed by and compared to measurements of the concentration-dependent stress state, chemical potential and phase separation in epitaxial niobium-hydrogen thin films adhered to sapphire substrates [3]. [1] A. Dyck, T. Böhlke, A. PunDT, S. Wagner, Scr. Mat. 247 (2024) 116117. [2] A. Dyck et al., Scr. Mat. 251 (2024) 116209. [3] A. Dyck et al., accepted to Mech. of Mat.

MM 4.6 Mon 11:30 H22

Hydrogen: a catalyst for abnormal grain growth in inert-gas condensed nanocrystalline Pd-Au? — ●FABIAN ANDORFER¹, JULES M. DAKE¹, MARKUS ZIEHMER¹, JOHANNES WILD², TORBEN BOLL², DOROTHÉE VINGA SZABÓ², STEFAN WAGNER², ASTRID PUNDT², and CARL E. KRILL III¹ — ¹Institute of Functional Nanosystems, Ulm University — ²Institute of Applied Materials - Materials Science and Engineering (IAM-WK), Karlsruhe Institute of Technology

Heat treatment causes some polycrystalline materials to manifest abnormal grain growth (AGG), during which a few grains acquire a significant growth advantage compared to the remaining matrix grains, resulting in a bimodal distribution of grain sizes. Inert-gas condensed nanocrystalline Pd-Au alloys undergo an extreme form of AGG. This

system is known to absorb hydrogen. When Pd-Au alloys are exposed to hydrogen below a certain gas pressure, hydrogen atoms primarily occupy grain boundaries and interstitial sites. This segregation is expected to lower the excess energy of the grain boundaries, which should have a noticeable effect on subsequent grain growth. To test this, Pd-Au samples were heat treated in argon and hydrogen atmospheres, respectively. The average size of the abnormally grown grains was determined, and the grain shape was characterized by calculating the circularity. Hydrogen appeared to increase the rate of abnormal grain “nucleation” without affecting the resulting grain shape.

MM 4.7 Mon 11:45 H22

The liquid intermediate phase for a hydrogen storage material — ●ANASTASIA THASE¹, OLIVER ALDERMAN², CHIARA MILANESE^{3,4}, ALESSANDRO GIRELLA⁴, CLAUDIO PISTIDDA⁵, MARTIN MÜLLER^{5,6}, and SEBASTIAN BUSCH¹ — ¹GEMS MLZ, Helmholtz-Zentrum Hereon WPN — ²ISIS Neutron and Muon Source — ³C.S.G.I. Department of Chemistry, Physical Chemistry Division, University of Pavia — ⁴Pavia Hydrogen Lab — ⁵Helmholtz-Zentrum Hereon WTN — ⁶CAU zu Kiel, Leibnitzstr. 19, 24098 Kiel, Germany

Hydrogen storage in light hydrides for mobile applications is a widely discussed but a highly controversial topic because of its ability to form explosive mixtures with oxygen. However, this danger is eliminated, if hydrogen is stored in complex hydrides, which provide hydrogen only under significant heat impact.

The main issue for complex hydride mixtures is the kinetics of the reversible reaction with hydrogen. One of prospective candidates is $6\text{Mg}(\text{NH}_2)_2:9\text{LiH}$, which promptly interacts with hydrogen when doped with LiBH_4 . Its catalytic impact is explained by the formation of low-melting intermediate phases with high Li-ion conductivity: a metastable $\text{Li}_2\text{BH}_4\text{NH}_2$ and a peritectically melting $\text{Li}_4\text{BH}_4(\text{NH}_2)_3$.

In the $\text{LiNH}_2 - \text{LiBH}_4$ phase diagram, the eutectic point is located at the 1:2 ratio and 90°C. This eutectic mixture was characterized by various techniques, including neutron total scattering, and its intrinsic structure under the reaction conditions was elucidated by EPSR, which gave a hint about its role as a liquid-phase catalyst in the corresponding hydrogen storage composition ($6\text{Mg}(\text{NH}_2)_2:9\text{LiH}:6\text{LiBH}_4$).

MM 4.8 Mon 12:00 H22

Machine Learning Interatomic Potentials for Studying Hydrogen Storage in TiCr2 Laves Phase Alloys — ●PRANAV KUMAR, BLAZEJ GRABOWSKI, and YUJI IKEDA — Institut für Materialwissenschaft, Universität Stuttgart, Pfaffenwaldring 55, 70569 Stuttgart, Germany

Efficient hydrogen storage is a crucial challenge for the widespread absorption of hydrogen as a clean energy carrier. TiCr2 Laves phase alloys, with their exceptional hydrogen absorption and diffusion properties, represent a promising class of materials for this purpose. This study employs a multiscale computational strategy, integrating density functional theory (DFT) and machine learning interatomic potentials (MLIPs), to investigate hydrogen behavior in TiCr2 Laves phases. Key areas of focus include hydrogen binding energies, diffusion pathways, and the complex effects of hydrogen clustering, which influence storage capacity and transport. Notably, our findings reveal possible equilibrium hydride structures consistent with experimental observations, offering valuable validation of the computational models. By leveraging large-scale DFT simulations within an active learning framework, we develop accurate MLIPs that enable efficient exploration of the phase space across diverse hydrogen concentrations. These MLIPs bridge atomic-scale simulations, facilitating precise calculations of hydrogen diffusion coefficients and shedding light on the mechanisms of hydrogen mobility. The insights derived from this work enhance our understanding of hydrogen-metal interactions and provide critical information for optimizing hydrogen storage materials.

MM 4.9 Mon 12:15 H22

The impact of Mn and Al on the trapping and diffusion of hydrogen in γ -Fe: An atomistic insight — ●BIKRAM KUMAR DAS¹, POULAMI CHAKRABORTY¹, MAURICIO RINCÓN BONILLA¹, and ELENA AKHMATSKAYA^{1,2} — ¹Basque Center for Applied Mathematics, Bilbao, Spain — ²Ikerbasque - Basque Foundation for Science, Spain

We propose a first principles-based framework to systematically unlock

the physical underpinnings of local distribution of the solute atoms on hydrogen trapping and diffusion in Mn/Al-alloyed γ -Fe. In our scheme, all thermodynamically stable substitutional solute sites were identified (≤ 5.4 wt% Mn; ≤ 4 wt% Al) up to the third nearest neighbour (NN) shell of a single H atom. The impact of Mn/Al on H-binding was quantitatively evaluated, indicating a surprisingly strong correlation with the local Al distribution regardless Mn content, and indirect stabilization by Al when present in the 2nd NN shell. Nonetheless, Al strongly repels H bonding. The contradictory role of Al was explained in terms of bonding/anti-bonding orbitals occupancy in H-M interactions ($M = \text{Al}, \text{Mn}, \text{Fe}$). The barriers to H hopping between adjacent local environments and the corresponding jump frequencies were subsequently calculated, providing insights into the limits imposed by the presence of Al and Mn on H mobility in Mn/Al-alloyed γ -Fe. Most notably, presence of Al in the 2nd NN shell of H severely reduces the H jump frequency, leading to irreversible trapping at high Al contents. Such behaviour may critically contribute to mitigate H-induced delayed fracture in Al-rich austenite steel.

MM 4.10 Mon 12:30 H22

Hydrogen - Microstructure Interaction in Ferritic Steels: From Ab-initio Simulations to Experiments — ●ONUR CAN ŞEN^{1,2,3}, SANTIAGO BENITO², SEBASTIAN WEBER², and REBECCA JANISCH³ — ¹IMPRS SusMet, Max Planck Institute for Sustainable Materials, Germany — ²Institute for Materials, Chair of Materials Technology, Ruhr-University Bochum, Germany — ³ICAMS, Ruhr-University Bochum, Germany

Hydrogen embrittlement (HE) is a phenomenon where hydrogen negatively impacts the mechanical properties of metallic materials. Mitigating HE requires minimizing hydrogen diffusion in microstructures and understanding how local heterogeneities influence this process across different length scales. While advanced experimental and computational methods exist, their application to varying systems complicates model validation. To address this, simplified ferritic Fe-based alloys were produced, and subjected to thermomechanical treatments, and their microstructures were characterized using EBSD. This approach aims to assess the hydrogen storage capacities of microstructural heterogeneities and their influence on the effective diffusion coefficient. Complementary ab initio density functional theory calculations were performed to evaluate hydrogen trapping energies and diffusion barriers under different chemical environments and CSL boundaries. These calculations inform predictions of hydrogen solubility and distribution within the experimental microstructures, to be validated experimentally. This work shows the importance of combining simulations and experiments to understand microstructural heterogeneities' role in HE.

MM 4.11 Mon 12:45 H22

Hydrogen Embrittlement in fatigue damage of ferritic steel: Theoretical investigation of failure mechanisms in varying hydrogen environments — ●ALEXANDRA STARK^{1,2}, PETRA SONNWEBER-RIBIC¹, and CHRISTIAN ELSÄSSER² — ¹Robert Bosch GmbH, CR, 71272 Renningen — ²Fraunhofer IWM, 79108 Freiburg

In this theoretical study, the influence of hydrogen on the fatigue behavior of a ferritic steel is examined by using a coupled hydrogen-diffusion and crystal-plasticity finite-element (CPFE) model. The damaging effect of hydrogen on structural materials, particularly ferritic and martensitic steels, poses a significant challenge for a wide range of technical applications. Despite being known for about 150 years, the underlying mechanisms of hydrogen embrittlement remain the subject of scientific debates. CPFE models are suitable for ensuring a microstructure-sensitive modelling and enabling the description of local plastic deformation, both essential factors in the complex process of metal fatigue. The presented study investigates the individual and combined effects of different damage models based on proposed hydrogen embrittlement failure mechanisms [1]. Furthermore, the work explores the impact of diverse environmental conditions on the cyclic material damage. The effects of varying local hydrogen concentration and distribution on the fatigue damage, as implemented in the CPFE model, are examined and discussed. [1] A. Stark, P. Sonnweber-Ribic, and C. Elsässer, Theoretical study of individual and combined effects of HELP- and HEDE-based damage models on the fatigue behavior of ferritic steel by hydrogen, submitted (2024).