MM 39: Hydrogen in Materials II

Time: Friday 10:15–12:45 Location: SCH/A215

MM 39.1 Fri 10:15 SCH/A215

Coulometric and (Magneto-)Optical Methods for Studying Hydrogen Absorption into Metals — •Felix Engelhardt, Karin Leistner, and Markus Gössler — Chemnitz University of Technology, 09107 Chemnitz, Germany

Hydrogen absorption into metals like palladium is a widely studied process with applications in hydrogen sensors, hydrogen storage systems, and recently magneto-ionics[1,2]. A major challenge in this field is to accurately quantify the hydrogen concentration within the metal. Traditionally, complex techniques such as nuclear reaction analysis or neutron scattering are used for this purpose. Alternatively, coulometric and (magneto-)optical methods are possible and offer the advantages of a simpler setup and operation under ambient conditions. We revisit the coulometric quantification of electrochemically absorbed hydrogen in metals and metallic multilayers, as well as the analysis of hydrogen absorption kinetics. Potential- and time-dependent coulometry was performed in a liquid-electrolyte flow cell designed to avoid side reactions. Complementary measurements of (magneto-)optical reflectivity of the metals upon hydrogen absorption were carried out to support findings from electrochemical methods. This presentation highlights common pitfalls in hydrogen absorption analysis and shows that, when these errors are minimized, the coulometry method yields accurate average hydrogen concentrations and reliable kinetic data. These results encourage the use of electrochemical methods in catalysis and hydrogen research. [1]M. Bischoff et al., Adv. Funct. Mater. 2024, 34, 2405323 [2]Huang, M. et al., Nat. Nanotechnol. 2021, 16, 981

MM 39.2 Fri 10:30 SCH/A215

Interfacial kinetics of hydrogen absorption in nanoporous palladium — •Seoyun Sohn[1,2], Nora Brunkhorst[2], Jürgen Markmann[2,1], and Jörg Weissmüller[1,2] — [1]Institute of Materials Physics and Technology, Hamburg University of Technology, 21073 Hamburg, Germany — [2]Institute of Hydrogen Technology, Helmholtz-Zentrum Hereon, 21502 Geesthacht, Germany

Sorption of hydrogen at metal surfaces is a key process controlling energy-conversion reactions, electrocatalysis, and battery operation. To focus on the interfacial sorption step and minimize bulk-transport contributions, we explore nanoporous (np) Pd as a model system, following the charging/discharging kinetics during electrochemical charging or exposure to H_2 gas at controlled partial pressure. Our observations point towards thermally activated transition across an internal barrier–between adsorbed and absorbed H–as the rate limiting step, and they exhibit anomalous kinetics, where absorption or desorption rates do not monotonously decrease with time. The command critiques the applicability of the Butler-Volmer equation–as a classic approach to the kinetics–and we advertise an alternative kinetic rate law that resolves the problematic issues of the classic approach. We show that the new rate law explains important features of the observations, specifically when admitting for a coherent initiation of the phase change.

MM 39.3 Fri 10:45 SCH/A215

Local hydrogenation of polycrystalline Pd nanodisks studied with in-situ TEM — •Svetlana Korneychuk[1,2], Carl Andersson[3], Stefan Wagner[1], Christoph Langhammer[3], and Astrid Pundt[1] — [1]IAM-WK, Karlsruhe Institute of Technology, Karlsruhe, Germany — [2]KNMFi, Karlsruhe Institute of Technology, Karlsruhe, Germany — [3]Chalmers University of Technology, Göteborg, Sweden

Hydrogen distribution and concentration in metals is of central importance for many areas of hydrogen technology, such as hydrogen storage, detection, and hydrogen embrittlement. In particular, arrays of Pd nanodisks can serve as optical hydrogen sensors [1] at room temperature owning to fast kinetics of hydrogen absorption and desorption of Pd. Here we demonstrate hydrogen distribution and concentration in polycrystalline Pd nanodisks with the lateral resolution down to 4 nm extending our previous study on the local measurement of hydrogen concentration in Pd nanoparticles with in-situ TEM [2]. 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. The method offers a way to observe hydrogen concentration at different sites: grain boundaries, surface and grains starting from early stages of hydrogenation to compete hydride formation identifying the pathways of hydrogenation of polycrystalline metals. [1] C. Wadell, et al., ACS Nano 2014 8 (12), 11925-11940 [2] S. Korneychuk, et al., Small 2025, 21, 2407092.

MM 39.4 Fri 11:00 SCH/A215

A comparative density functional theory study of hydrogen adsorption in metal organic frameworks — •Ikutaro Hamada — Department of Precision Engineering, Graduate School of Engineering, The University of Osaka , Suita, Osaka 565-0871, Japan

Metal-organic frameworks (MOFs) and covalent organic frameworks (COFs) have attracted considerable attention due to their design and synthesis flexibility, enabling applications in catalysis, gas adsorption, and separation. Among these, hydrogen storage is one of the most extensively studied properties of MOFs. However, their hydrogen storage capacity remains low, particularly at room temperature, which limits industrial and commercial applications. To design MOFs with enhanced hydrogen storage capability, it is essential to understand the microscopic details of hydrogen adsorption and absorption within these frameworks. In this work, we assess the accuracy of dispersion-inclusive exchange-correlation functionals against the coupled cluster method using hydrogen-benzene complexes, and we investigate hydrogen adsorption in UiO-66, UiO-67, and UiO-68 as prototypical MOFs. We will present details of our systematic exploration of the geometric and electronic structures of hydrogen molecules and their stability within UiO-66, UiO-67, and UiO-68.

MM 39.5 Fri 11:15 SCH/A215

Machine Learning Potential Approach to Complex Hydrogen-Surface Interactions: GRACE Applied to the H/Sn/Ru System — •Manuel Enns and Daniel F. Urban — Fraunhofer IWM, Wöhlerstraße 11, 79108 Freiburg, Germany

Understanding hydrogen interactions with contaminated metal surfaces remains a challenging problem in computational materials science, requiring methods that balance accuracy with computational efficiency to capture complex multi-component dynamics. We demonstrate the application of a GRACE machine learning interatomic potential to study hydrogen behavior on tin-contaminated ruthenium surfaces - a system that exemplifies the challenges of modeling reactive adsorbates on multi-element surfaces. Data from density functional theory calculations on hydrogen penetration mechanisms through ruthenium surfaces with varying tin coverages was used to fine-tune the GRACE potential, enabling computationally efficient molecular dynamics (MD) simulations. The resulting MD simulations successfully capture key dynamic processes including hydrogen diffusion and jump mechanisms, tin growth kinetics at a prior hydrogen coverage, and low-energy hydrogen deposition. This case study demonstrates GRACE's capability to handle complex multi-component systems involving reactive species, providing atomic-level insights into how surface contamination affects hydrogen penetration pathways and adsorbate interactions.

15 min. break

MM 39.6 Fri 11:45 SCH/A215

Diffusion of hydrogen in platinum during HER catalysis — Aparna Saksena[1], Bingxin Li[1], Yujun Zhao[1], J. Manoj Prabhakar[1], Mira Todorova[1], Jörg Neugebauer[1], Dierk Raabe[1], Baptiste Gault[1,2], and •Yug Joshi[1] — [1]Max Planck Institute for Sustainable Materials (MPI-SusMat), Max Max-Planck-Straße 1, 40237 Düsseldorf — [2]Univ Rouen Normandie, CNRS, INSA Rouen Normandie, Groupe de Physique des Matériaux, UMR 6634, F-76000 Rouen, France

Platinum remains the most common electrocatalyst used for hydrogen evolution reaction (HER) in acidic conditions. It is generally believed to act as a proton-blocking catalyst with adsorption of reaction intermediates limited to its surface. In this study, we thoroughly examine how Pt interacts with hydrogen and deuterium (H/D) within the bulk by monitoring the mass change of a Pt electrode in real-time during galvanostatic heavy/water splitting, using an electrochemical quartz crystal microbalance. Surprisingly, we observe an irreversible mass increase over time along with changes in the overpotential for both HER

and DER. Atom probe tomography of the Pt electrode after DER confirms that D diffuses into the bulk. The results are supported by density functional theory, showing the formation of an interstitial solid solution of H in Pt. These results challenge the conventional belief that Pt-proton interactions are only surface-limited. They suggest a need to revisit catalyst design strategies, which currently focus on surface-adsorbed species and the rate-limiting Volmer step, by also considering the bulk diffusion of H/D within the Pt matrix.

MM 39.7 Fri 12:00 SCH/A215

Hydrogen induced small polarons in iron oxides using occupation-matrix control DFT+U calculations — Ahmed Abdelkawy, •Mira Todorova, and Jörg Neugebauer — Max Planck Institute for Sustainable Materials, Max-Planck-Str.1, 40470 Düsseldorf

Over 6% of global CO_2 emissions come from using carbon-based reducing agents in iron production. Replacing these agents with hydrogen would result in water as the sole byproduct, thereby reducing harmful emissions. To understand H-based reduction of iron oxides, we focus on hydrogen interactions with bulk hematite (Fe_2O_3). Modeling transition metal oxides is challenging because conventional DFT-GGA has self-interaction errors and inadequately treats the Coulomb repulsion in partially filled d-orbitals. Corrections such as DFT+U or exchange-correlation functionals, which incorporate a fraction of the exact exchange, further complicate matters because they generate a multitude of metastable configurations that DFT cannot predict. In this study, we use DFT+U with occupation matrix control to map the energy landscape of the small electron polaron generated by H interstitials. We construct the three-dimensional potential energy surface of H interstitials using the resulting ground-state configurations. This enables us to gain detailed insight into hydrogen kinetics and rate-limiting steps.

MM 39.8 Fri 12:15 SCH/A215

An atomic-scale master-equation model study of the mobility of a dislocation core interacting with interstitial hydrogen in a metal — •Daniel Pfalzgraf[1], Daniel F. Urban[1,2], and Christian Elsässer[1,2] — [1]Fraunhofer IWM, Wöhlerstraße 11, 79108 Freiburg, Germany — [2]Freiburg Materials Research Center (FMF), University of Freiburg, Stefan-Meier-Str. 21, 79104 Freiburg, Germany

We examine the influence of interstitial hydrogen in a metal on the motion of a dislocation by means of a master equation model. This model takes as input atomic-scale information, such as material-specific defect-migration energy barriers and interatomic interaction energies, to produce the drift velocity of a mechanically driven dislocation core segment as a function of the local hydrogen concentration around it. Our approach can take detailed correlation effects into account, for example, that the dislocation core acts as a mobile hydrogen trap, resulting in an inhomogeneous distribution of hydrogen around it. The results can be used to estimate in which ranges of temperature or interaction strength the presence of hydrogen increases or decreases the mobility of the dislocation core, which is of interest in the context of hydrogen embrittlement mechanisms.

MM 39.9 Fri 12:30 SCH/A215

Hydrogen induced strain instability in Aluminum — •Ali Tehranchi[1,2], Bharathi Ganesh Ganesan Sekar[1], and Tilmann Hickel[1,2] — [1]Bundesanstalt für Materialforschung und -prüfung (BAM), D-12489, Berlin, Germany — [2]Max Planck Institute for Sustainable Materials, D-40237 Düsseldorf, Germany

Hydrogen embrittlement is a persistent form of metal degradation that arises from the complex interactions between hydrogen atoms and defects within metallic lattices. Environmental nanoindentation has become a standard experimental technique for probing these interactions, particularly the mechanisms underlying hydrogen-assisted deformation. In this work, we employ comprehensive large-scale molecular statics simulations in aluminum, complemented by ab initio calculations, to investigate the influence of hydrogen on homogeneous dislocation nucleation –the primary mechanism responsible for pop-in events observed under low-radius indenters. Our defect phase diagram analysis shows that, at hydrogen chemical potentials relevant to nanoindentation experiments, vacancy formation is significantly enhanced, leading to a substantial increase in the concentration of vacancy–hydrogen complexes. These complexes produce large local misfit strains, which in turn reduce the critical shear stress required for material instability and the onset of homogeneous dislocation nucleation.