O 56: Metal Substrates II

Time: Wednesday 15:00–16:00 Location: MA 043

O 56.1 Wed 15:00 MA 043

In situ observation of the on-surface thermal dehydrogenation of n-octane on Pt(111) — Daniel Arribas[1], Víctor Villalobos-Vilda[1], Ezequiel Tosi[1], Paolo Lacovig[2], Alessandro Baraldi[2], Luca Bignardi[2], Silvano Lizzit[2], José Ignacio Martínez[1], Pedro de Andres[1], Alejandro Gutiérrez[3], José Ángel Martín-Gago[1], and •Pablo Merino[1] — [1]Instituto de Ciencia de Materiales de Madrid — [2]Elettra-Sincrotrone Trieste S.C.p.A. — [3]Universidad Autónoma de Madrid

The catalytic dehydrogenation of alkanes constitutes a key step for the industrial conversion of these inert sp3-bonded carbon chains into other valuable unsaturated chemicals. To this end, platinum-based materials are among the most widely used catalysts. In this work, we characterize the thermal dehydrogenation of n-octane (n-C8H18) on Pt(111) under ultra-high vacuum using synchrotron-radiation X-ray photoelectron spectroscopy, temperature-programmed desorption and scanning tunneling microscopy, combined with ab initio calculations. At low activation temperatures, two different dehydrogenation stages are observed. At 330 K, n-C8H18 effectively undergoes a 100% regioselective single C-H bond cleavage at one methyl end. At 600 K, the chemisorbed molecules undergo a double dehydrogenation, yielding double bonds in their carbon skeletons. Diffusion of the dehydrogenated species leads to the formation of carbon molecular clusters, which represents the first step towards poisoning of the catalyst. Our results disclose the chemical mechanisms behind the first stages of alkane dehydrogenation on a Pt surface at the atomic scale.

O 56.2 Wed 15:15 MA 043

How to measure cones of reaction for single-molecule collisions — •Matthew James Timm[1], Stefan Hecht[2], and Leonhard Grill[1] — [1]Institute of Chemistry, University of Graz, Heinrichstraße 28/IV, 8010 Graz, Austria — [2]Department of Chemistry & IRIS Adlershof, Humboldt-Universität zu Berlin, Brook-Taylor-Str. 2, 12489 Berlin, Germany

Collisions between molecules are essential for chemistry, as they are required to form bonds. Their outcome depends on the collision energy, the relative alignment of reagents and on the impact parameter, which is the miss-distance between the centers of mass of the colliding species. Selection of impact parameter has been demonstrated for on-surface reactions by a "surface-molecular-beam" of CF2 projectiles, formed by dissociating CF3 molecules on a Cu(110) surface with a scanning tunneling microscope tip. These projectiles are directed along the underlying rows of Cu atoms toward a chemisorbed molecular target [1-2]. Control over the relative alignment of the target, however, has remained elusive so far. Here, a singly-debrominated molecular target has been chosen as it can adopt multiple possible adsorption alignments relative to the incoming CF2 projectile. This allows collision at a selected distance from, and alignment with, the center of mass of the target. Accordingly, it paves the way towards an unprecedented ability to map how collision geometry contributes to collision outcome. [1] Anggara, K.; Leung, L.; Timm, M. J.; Hu, Z.; Polanyi, J. C.; Sci Adv., 2018, 4, eaau2821. [2] Anggara, K.; Leung, L.; Timm, M. J.; Hu, Z.; Polanyi, J. C.; Faraday Discuss., 2019, 214, 89-103.

O 56.3 Wed 15:30 MA 043

Crystal growth of alkali and alkali earth metal on the transition metal surfaces — •Yuanyuan Zhou[1] and Chunye Zhu[2] — [1]Technical University of Denmark, Copenhagen, Denmark — [2]Guangdong University of Technology, Jieyang, China

Electrochemical nitrogen reduction is an attractive alternative to the Haber-Bosch process for making ammonia. The promising electrochemical system that produces large amounts of ammonia is the Li- and Ca-mediated process, which has achieved nearly 100% selectivity [Li *et al.* Joule 6, 1-19 (2022), Fu *et al.* Nat. Mater. (2023)]. However, the formation of the Li/Ca dendrites reduce the selectivity and deteriorate the stability of the Li- and Ca-mediated process. Therefore, a prerequisite for avoiding the dendrite formation is an in-depth understanding towards the growth mechanisms in an atomistic level.

We coupled our developed replica-exchange grand-canonical (REGC) [Zhou *et al.* Phys. Rev. B. 100, 174106(2019), Zhou *et al.* Phys. Rev. Lett. 128, 246101(2022)] method with machine-learned interatomic potentials (MLIPs) [Schütt *et al.* PMLR 139, (2021)] to simulate larger length scale and time scale. The MLIPs is constructed in an autonomous active learning during the REGC simulations. We investigated the Li/Ca growth on the transition metal surface (Cu and Fe) using REGC MLIPs-accelerated molecular dynamics. This framework vividly showcased growth process of Li/Ca on Cu/Fe, pinpointing the origin of different growth behavior between Li and Ca.

O 56.4 Wed 15:45 MA 043

Epitaxial CoCrFeNi films for surface investigations — Holger Schwarz[1], Jonathan Apell[1,2], Robert Wonneberger[1], Andreas Undisz[1], Julian Ledieu[3], Vincent Fournée[3], Peter Richter[1], and •Thomas Seyller[1] — [1]Chemnitz University of Technology, Chemnitz, Germany — [2]Friedrich Schiller University Jena, Jena, Germany — [3]Université de Lorraine, Nancy, France

High-entropy alloys (HEAs) are discussed for applications in the fields of corrosion, wear protection and electrocatalysis. Although the surface properties play a central role in these applications, they are still largely unexplored, which is caused by the unavailability of single-crystalline samples. In this presentation, recent progress is reported on the growth and subsequent characterization of epitaxial CoCrFeNi films [1], which were deposited by DC magnetron sputtering from spark-plasma sintered targets [2] on single-crystalline oxide substrates. A characterization of structural, chemical and electronic properties of the films was performed by different techniques including X-ray diffraction, scanning electron and transmission electron microscopy, energy-dispersive X-ray spectroscopy, photoelectron spectroscopy, low-energy electron diffraction and, more recently, by scanning tunneling microscopy. It is demonstrated that epitaxially grown HEA films have the potential to fill the sample gap, allowing for fundamental studies of properties of and processes on well-defined HEA surfaces over the full compositional space. [1] H. Schwarz, et al., Advanced Materials 35 (2023) 2301526. [2] H. Schwarz, et al., Coatings 11 (2021) 468.