ALBA II - Workshop on APXPS

Report of Contributions

https://indico.cells.es/e/workshop-APXPS

Contribution ID: **1** Type: **not specified**

Introduction

Thursday, 9 September 2021 10:15 (15 minutes)

Presenter: Dr ATTENKOFER, Klaus

Contribution ID: **2** Type: **not specified**

Aqueous Solution-Vapor Interfaces Investigated with Ambient Pressure X-ray Photoelectron Spectroscopy

Thursday, 9 September 2021 10:30 (30 minutes)

Aqueous solution-vapor interfaces govern important phenomena in the environment and atmosphere, including the uptake and release of trace gases by aerosols and CO2 sequestration by the oceans.[1] A detailed understanding of these processes requires the investigation of liquid-vapor interfaces with chemical sensitivity and interface specificity under ambient conditions, i.e., temperatures above 270 K and water vapour pressures in the millibar to tens of millibar pressure range. This talk will discuss opportunities and challenges for investigations of liquid-vapor interfaces using X-ray photoelectron spectroscopy and describe some recent experiments that have focused on the propensity of certain ions and the role of surfactants at the liquid/vapor interface.
[1] O. Björneholm et al., Chem. Rev. 116, 7698 (2016).

Presenter: Dr BLUHM, Hendrik (Fritz Haber Institute of the Max Planck Society, Department of Inorganic Chemistry)

Contribution ID: **3** Type: **not specified**

In Situ Characterization of Electrocatalysis at Electrified Interfaces

Thursday, 9 September 2021 11:00 (30 minutes)

Studying electrochemical reactions at interfaces between different states of matter has been a long-term interest for both experimentalists and theorists in wide-range research areas. Revealing the fundamental properties at such interfaces is critical for a complete description of relevant electrochemical processes and for future designs of advance materials. In this talk, we will present a brief review on our in situ investigations at electrified interfaces, including gas/solid interface of solid oxide electrochemical cells and liquid/solid interface of magnesium rechargeable batteries. 【1, 2】These examples highlight the importance of studying "living"interfaces in a dynamic environment and the value of correlative in situ methods. 【3】We will discuss our beamline at Shanghai Synchrotron Radiation Facility, which allows in situ studies at pressures up to 20 mbar with high spatial resolution. We will also share recent progress on our lab-based system dedicated for in situ investigations of liquid/solid interfaces. Other new experimental methods will be briefly discussed as well.

References:

[1] Zhang, C.; Yu, Y.; Grass, M. E.; Dejoie, C.; Ding, W.; Gaskell, K.; Jabeen, N.; Hong, Y. P.; Shavorskiy, A.; Bluhm, H.; Li, W. X.; Jackson, G. S.; Hussain, Z.; Liu, Z.; Eichhorn, B. W., J Am Chem Soc 2013, 135, 11572.
[2] Yu, Y.; Baskin, A.; Valero-Vidal, C.; Hahn, N. T.; Liu, Q.; Zavadil, K. R.; Eichhorn, B. W.; Prendergast, D.; Crumlin, E. J., Chemistry of Materials 2017, 29, 8504.
[3] Han, Y.; Zhang, H.; Yu, Y.; Liu, Z., ACS Catal. 2021, 11, 1464.

Presenter: Prof. LIU, Zhi (Center for Transformative Science, ShanghaiTech University and School of Physical Science and Technology, ShanghaiTech University)

Contribution ID: **4** Type: **not specified**

Dynamic Nanocatalysts: Environmental Effects

Thursday, 9 September 2021 12:00 (30 minutes)

In order to comprehend the catalytic performance of metal nanostructures, their dynamic nature and response to the environment must be taken into consideration. The working state of a nanocatalyst might not be the state in which the catalyst was prepared, but a structural and/or chemical isomer that adapted to the particular reaction conditions. Furthermore, deactivation phenomena taking place under reaction conditions can only be understood, and ultimately prevented, if sufficient information is available on the catalyst morphology, structure, chemical state, and surface composition while at work.
I will first describe novel approaches for the synthesis of size- and shape-controlled nanoparticle catalysts (Cu2O, Cu, Cu-M (M=Zn, Ni, Ga), NiGa and FeOx NPs). Subsequently, I will illustrate how to follow the evolution of their morphology and surface composition under different gaseous and liquid environments in the course of a catalytic reaction. Examples will be given regarding the dynamic transformations of the former nanocatalysts during the hydrogenation and the electrochemical reduction of CO2 via NAP-XPS and other complementary techniques such as XAS, or NAP-STM. Emphasis will be given to elucidating the role of the nanoparticle size, shape, composition, chemical state and support of the catalysts in their activity, selectivity and durability.

Presenter: Prof. ROLDÁN CUENYA, Beatriz (Department of Interface Science, Fritz-Haber Institute of the Max Planck Society)

Contribution ID: **5** Type: **not specified**

Time-resolved and event-averaged ambient pressure x-ray photoelectron spectroscopy: A new methodology for stroboscopic vision of dynamic catalyst surfaces at work

Thursday, 9 September 2021 12:30 (30 minutes)

Presenter: Dr KNUDSEN, Jan

Contribution ID: **6** Type: **not specified**

Evaluation of the impact of the in situ cell design on the interfacial speciation over C-based electrodes during room temperature CO2 electroreduction

Thursday, 9 September 2021 14:30 (30 minutes)

Electrochemically or photo-electrochemically activated, hydrogenation and dehydrogenation reactions of small molecules (e. g. H2O, CO2, O2, N2) are at the core of currently explored decarbonization technologies. These technologies are key to delivering net zero emissions. The major challenges to face for boosting innovation in this area are common to these reactions: reducing material costs; improving the energy efficiencies to practical levels and reducing material degradation during operation. The understanding of the underlying mechanisms, including restructuring and compositional change of the electrode surface upon polarization, activation, and deactivation is acknowledged to guide towards the synthesis of improved materials. This knowledge can be obtained with highest resolution of chemical states via surface-sensitive in situ spectroscopic techniques such as ambient pressure X-ray photoelectron and absorption spectroscopies that investigate the electronic structure at surfaces and interfaces. However, there are technical challenges to overcome for the realization of electron detection-based in situ studies of liquid/solid electrified interfaces: the inelastic mean free path of electron and the high absorption cross section in the soft X-ray regime limit the analysis to thin liquid films wetting the electrode surface.
In the last decade progress in this field has been gigantic and different approaches have been proposed, amongst which the suitability of electron-transparent graphene membrane to separate the liquid environment from the vacuum chamber has been the focus of recent investigations carried out in collaboration with the beamline research team at the ISISS beamline of the synchrotron facility BESSY II at Berlin.

In this contribution, I will focus on PEM-based systems to study the electrocatalytic conversion of CO2 to valuable products over carbon-based electrodes. I will describe the technical aspects of the various cell designs used, spanning different pressure ranges, from a humidified gas/solid interface [1] to a stagnant liquid film/solid interface [2]. I will compare the surface dynamics observed for chemical equivalent electrode systems in the different cell configurations, including the results obtained using a fluorescence yield based continuous flow cell. [3]

Supporting literature:
[1] V. Pfeifer, T. E. Jones, J. J. Velasco Vélez, R. Arrigo, S. Piccinin, M. Hävecker, A. Knop-Gericke, R. Schlögl, In situ observation of reactive oxygen species forming on oxygen-evolving iridium surfaces, Chem. Sci., 2017, DOI: 10.1039/C6SC04622C.
[2] J. Velasco-Vélez, E. Carbonio, C.-H. Chuang, C.-J. Hsu, J.-F. Lee, R. Arrigo, M. Hävecker, R. Wang, M. Plodinec, A. Centeno, A. Zurutuza, L. Falling, R. Mom, S. Hofmann, R. Schlögl, A. Knop-Gericke, T. Jones, Surface constrained electron-hole rich species active in the electrocatalytic water splitting, https://orcid.org/0000-0002-6595-0168.
[3] J.-J. Velasco-Vélez, C.-H. Chuang, D. Gao, Q. Zhu, D. Ivanov, H. Sang Jeon, R. Arrigo, R. V. Mom, E. Stotz, H.-L. Wu, T. E. Jones, B. Roldan Cuenya, A. Knop-Gericke, R. Schlögl, On the Activity/Selectivity and Phase Stability of Thermally Grown Copper Oxides during the Electrocatalytic Reduction of CO2, ACS Catalysis 2020 10 (19), 11510-11518.

Presenter: Dr ARRIGO, Rosa

Contribution ID: **7** Type: **not specified**

Nanomaterials under Operando Conditions

Thursday, 9 September 2021 15:00 (30 minutes)

Our group focuses on the development and application of state-of-the-art capabilities to synthesize and study functional surfaces and interfaces. In our research program, we emphasize the use of oxide and nanoporous ultrathin films such as 2D zeolites and metal organic frameworks (MOFs) to stabilize well-defined catalytic structures targeting the capture and conversion of small chemicals. I will present case studies showing how complementary in situ techniques including ambient pressure (AP) X-ray photoelectron spectroscopy (AP-XPS), infrared reflection absorption spectroscopy (AP-IRRAS) and AP-STM can be applied to study heterogeneous interfaces in model catalysts.

-"Enhanced Catalysis under 2D Silica: A CO Oxidation Study" Angew. Chem. Int. Ed., 60, 10888-10894 (2021)
-"Multi-modal surface analysis of porous films under operando conditions" AIP Adv. 10, 085109 (2020)
-"Tuning the Properties of Copper-Based Catalysts Based on Molecular in Situ Studies of Model Systems" Acc. Chem. Res. 48, 2151–2158 (2015)

Presenter: Prof. STACCHIOLA, Dario

Contribution ID: **8** Type: **not specified**

Round Table

Contribution ID: **9** Type: **not specified**

XPS above atmospheric pressure at POLARIS

Thursday, 9 September 2021 15:30 (30 minutes)

While many processes have been proposed to address climate change, from reducing the need for fossil fuels to changing the source of electricity, only one process can genuinely resolve the build-up of CO2 in the air: to synthesize fuel from the air. While a daunting task at the turn of the 20th century, the Haber-Bosch process accomplished a similar feat, converting nitrogen in the air to liquid ammonia for fertilizer. Today we require new tools to achieve more detailed knowledge of complex reactions on the relevant catalyst surfaces. To this end, POLARIS, the first high-pressure XPS, has been built. To measure the surfaces of catalysts at pressures over one atmosphere, we have made many technologic advances, discovering new and rediscovering old problems.

Since the installation of POLARIS at DESY in 2017, many experiments have taken place including, CO oxidation, methane oxidation, CO reduction, CO2 reduction, and N2 reduction. To reach the pressures required for these and other reactions, major shifts from conventional ambient pressure XPS are needed:
1. The aperture to the analyzer and the gap between the sample and aperture must be reduced to the micron scale to reduce scattering.
2. High energy x-rays must be used to reduce x-ray attenuation and provide enough kinetic energy for electrons to penetrate the gas.
3. Most importantly, the gas needs to flow from the analyzer to the chamber forming a virtual gas cell resulting in a much larger volume of gas required.
By incorporating these innovations, it is possible to measure XPS spectra at pressures over one bar. While the above process provides a framework, numerous other challenges arise, for instance, the lost surface sensitivity due to hard x-rays or the imprecise nature of maintaining micron-sized gaps between materials. Despite challenges, POLARIS has become a reliable tool for measuring the surface of catalysts in situ, providing the first look at the atomic species present during the most important catalytic reactions.

Ambient pressure XPS was first developed within years of the first XPS and had been limited to the millibar pressure range until POLARIS. At the core, POLARIS operates under the same process as all XPS; by using monochromatic x-rays of known energy to emit electrons, measure the kinetic energy and calculate the binding energy. Many adaptations needed to be made to build the high-pressure XPS, from determining the materials that can withstand the corrosive environment to designing a process to maintain a constant gap between the sample and aperture. Along with the development of the virtual gas cell, there are needed advances to the light source. Besides higher photon energies, highly focused light is required. The myriad problems and unique solutions have made POLARIS a complex yet highly exceptional instrument to measure XPS spectra approaching industrial pressures.

Presenter: Dr GOODWIN, Christopher