AKE 8: CO_2 Electro-Reduction

Time: Monday 17:15–18:00 Location: H3

AKE 8.1 Mon 17:15 H3

Monitoring the Chemical State of Catalysts for CO2 Electroreduction: An in operando Raman spectroscopic Study — •Akiyoshi Kuzume, Abhijit Dutta, Motiar Rahaman, and Peter Broekmann — Department of Chemistry and Biochemistry, University of Bern, Bern, Switzerland

A major concern of electrocatalysis research is to assess the structural and chemical changes that a catalyst may itself undergo in the course of the catalyzed process. These changes can influence not only the activity of the studied catalyst, but also its selectivity towards the formation of a certain product. An illustrative example is the electroreduction of carbon dioxide on tin oxide nanoparticles, where under the operating conditions of the electrolysis (that is, at cathodic potentials) the catalyst undergoes structural changes which in an extreme case involves its reduction to metallic tin. This results in a decreased Faradaic efficiency (FE) for the production of formate (HCOO*) that is otherwise the main product of CO2 reduction on SnOx surfaces. In this study we utilized potential and time dependent in operando Raman spectroscopy in order to monitor the oxidation state changes of SnO2 that accompany CO2 reduction. Investigations were carried out at different alkaline pH levels, and a strong correlation between the oxidation state of the surface and the FE of HCOO* formation was found. Understanding the transition of phases, such as oxide to metallic or vice versa, during CO2 electrolysis can offer distinct benefits of the catalyst material in terms of activity, selectivity, and stability.

AKE 8.2 Mon 17:30 H3

Activity of Cu-Au alloy NPs towards the electrochemical reduction of CO2: A compositional dependence study — •Motiar Rahaman, Abhijit Dutta, and Peter Broekmann — Department of Chemistry and Biochemistry, University of Bern, Freistrasse 3, Bern 3012, Switzerland

Our work focuses on the optimization of Cu-Au catalyst composition towards the electro-reduction of CO2 in aqueous media. We have synthesized various compositions of Cu-Au alloy nano-particles (NPs) directly on a technical vulcan carbon support where the chemical composition of the NPs were systematically varied from pure Au to pure Cu. The structure, composition and the electrochemical activity of these NPs with regard to the CO2 electro-reduction were studied by a combination of microscopic/spectroscopic methods (TEM, EDAX, XR and XPS) and electrochemical measurements (LSV and chronoamperometry). The formed gaseous and liquid products from the CO2 electroreduction have been analyzed by gas chromatography (GC) and ion chromatography (IC) techniques respectively. The catalytic activity of Cu-Au alloy composition towards CO2 reduction and the product distribution is found to be strongly dependent on the particular Au content. In case of Cu17Au83 alloy, CO is observed as main reduction product whereas, in case of Cu84Au16 alloy, hydrocarbons are dominantly produced with formate as side product. However the best hydrocarbon efficiency is observed for the Cu67Au33 alloy where ethylene and ethane are produced with substantial faradaic efficiencies.

AKE 8.3 Mon 17:45 H3

Enhancing Process Stability for the Electrolysis of Carbon Dioxide with Ionic Liquids — •Sebastian S. Neubauer[1], Ralf K. Krause[2], Joachim Wecker[2], Dirk M. Guldi[1], and Guenter Schmid[2] — [1]Department of Chemistry and Pharmacy, FAU, Egerlandstr. 3, 91058 Erlangen, Germany — [2]Siemens AG, Günter-Scharowsky-Str. 1, 91058 Erlangen, Germany

The production of valuable synthetic fuels and / or chemical feedstock out of waste CO_2 constitutes a key challenge in the contemporary field of energy storage. CO_2 is converted in a single step via a direct electrocatalytic process affording high efficiencies.
The conversion of CO_2 to carbon monoxide (CO) is a hot topic and the focus of this work. Studies, which were conducted in recent years, utilized mainly aqueous electrolytes. Rather short process stabilities and the competing hydrogen evolution reaction (HER) remain as major problems. In this context, ionic liquids have evolved as versatile solvent for the electrocatalytic conversion.[1] Considering their unique properties such as high ion conductivity, wide electrochemical window, and low Henry constant for CO_2, they potentially inhibit HER and shift the electron transfer towards CO_2 reduction. We identified ideal operation conditions for selected ionic liquids in the matter of working electrode potential and electrolysis stability. To this end, understanding and controlling electrophoresis is crucial with regard to the stability. The major conclusion of our work is the realization of high Faraday efficiencies for CO formation within a stable process, while HER is inhibited. [1] B. A. Rosen et al., Science 2011, 334, pp. 643-644