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Using APXPS to determine the fundamental reaction processes of the CO2 reduction reaction at the solid/gas and solid/liquid interface

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The valorization of carbon dioxide is one of the main research topics in the current time, since it is highly relevant for the development of sustainable energy cycles within the scope of net-zero CO_2-emissions for the future. The reaction of CO_2 on silver catalysts can take place either electrochemically (mainly to CO) or thermochemically (mainly to methanol). It is fundamentally interesting to determine the similarities and differences between the molecular activation processes and intermediates defining the product selectivity in each of these reaction environments.

We analyzed the thermochemical reaction (CO_2 + 3H_2 ⟶ CH_3 OH+H_2 O on Ag) using the solid/gas endstation at the In Situ Spectroscopy (ISS) beamline and the electrochemical reaction (CO_2 ⟶CO+0.5 O_2 in electrolytic cell with Ag cathode) using the solid/liquid dip-and-pull method at the PHOENIX beamline, both situated at the Swiss Light Source (SLS).

Understanding the response of a model reaction catalyst (here Ag foil) is crucial before increasing the catalyst complexity. The response of the Ag foil surface states to different gases and educt/product gas mixtures was evaluated as a function of temperature and time. Time resolved data are generated by using transient photoelectron emission measurements (TPM). By fast XPS scans during iterated gas switching, time-dependent measurements allow the detection of short-lived species and the determination of reaction kinetics.

The evolution of the electrochemical system over time on the other hand is characterized by pulling the working electrode out of the electrolyte under potential control, preserving the operational surface condition and present surface contaminations accumulated during CO_2-reduction. Performance improvements achieved e.g. by electrochemical reconditioning can then be directly related to changes of the surface species.

To obtain the best possible correlation between model experimental results and processes under industrially relevant conditions, the performed APXPS experiments are combined with ex situ reactor/electrochemical cell studies. Methanol synthesis from the state-of-the-art Cu-based catalyst is industrially performed at ≈50 bar and ≈250 ℃, which can be mimicked in a high-pressure lab-scale reactor.

For the economically viable electrochemical conversion of CO_2, current densities ≫100 mA/cmˆ2 geometric surface area are required. The results obtained by in situ APXPS CO_2-reduction (≤1 mA/cm_geomˆ2) are thus compared to ex situ liquid-electrolyte electrolyzer cells (≤50 mA/cm_geomˆ2) and gas diffusion electrode containing bipolar-membrane cells (>100 mA/cm_geomˆ2) in the presented project.

if ”Other”, please specify:

I apply for a travel grant

No

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