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Structure sensitivity in the electrocatalytic reduction of CO₂ with gold catalysts

Mezzavilla S. 1, Duarte R. 1, Maagaard T. 1 Stephens I.E.L. 2, Horch S. 1, Seger B. 1, Chorkendorff I. * 1

1: SurfCat, Department of Physics, Technical University of Denmark, DK-2800 Kgs. Lyngby, Denmark
2: Department of Materials Imperial College London, 2.03b, Royal School of Mines London SW7 2AZ, England
*Corresponding author email: ibchork@fysik.dtu.dk

The electrocatalytic reduction of CO₂, that is, the conversion of CO₂ to CO, hydrocarbons and alcohols in an electrochemical cell, represents a very promising strategy to store renewable electricity in chemical compounds and to facilitate the introduction of renewable energy sources in the chemical industry. Gold is one of the most active electrocatalysts capable to produce CO at moderate overpotentials and high selectivity. Many strategies, such as nanostructuring, the exploitation of catalyst-support effects and grafting with organic ligands, have been recently proposed to further enhance its performance. Even so, small improvements have been achieved so far in comparison to the performance of bulk Au electrodes and little is known about the nature of the catalytic active sites. Theoretical studies predict that stepped surfaces – and, more in general, under-coordinated sites – are the most active sites for the CO₂ electro-reduction. Even so, no experimental evidence has been brought forward in support of these hypotheses. In this work, a thorough experimental investigation of Au single crystals having preferential surface orientations and well-defined features is presented. The performance of terrace-rich crystals, such as (111) and (100), and a steps-rich (211) electrode is compared to assess their differences in selectivity and reaction rates. Furthermore, the presence of adsorbed reaction intermediates and spectators will be discussed. The findings of this study will guide the design and synthesis of efficient catalysts. Furthermore, the results obtained with these model catalysts may provide important elements to optimize the theoretical description of the phenomena occurring at the electrochemical interface and therefore improve the prediction accuracy of future screening investigations.

References