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Energypolis, Rue de l'Industrie 17, CH-1950 Sion, Switzerland

ENERGYPOLIS SEMINAR

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Calculations of the Electrochemical Reduction of CO_2 and the Competing Hydrogen Evolution Reaction

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Results of theoretical calculations of electrochemical CO_2 reduction to formate, hydrocarbons and alcohols will be presented. The mechanism for the formation of various products is established and the rate evaluated and compared with experimental measurements. The rate of the main side reaction, the hydrogen evolution reaction, is also estimated. The calculations are based on a detailed atomistic model of the electric double layer (metal slab and water layer) and density functional theory calculations to evaluate not only the free energy of intermediates as a function of applied voltage but also the activation energy for each elementary step, both Heyrovsky and Tafel reactions [1]. Comparison is also made with calculations using an implicit solvation model [2]. A range of close packed metal surfaces are compared, including Cu, Ag, Au, Ni, Fe, Rh, Ir and Pt. The results are in remarkably good agreement with the available measurements. A two parameter descriptor is established that can help identify improved catalysts for this important reaction. The elucidation of the reaction mechanism of CO_2 electroreduction to hydrocarbons and alcohols as well as the competing hydrogen evolution reaction is an important step towards the design of a selective and energy efficient catalyst for small scale, decentralized fuel production using renewable energy sources and CO_2 as reactant.

References:

[1] J. Hussain, H. Jónsson and E. Skúlason, "Calculations of Product Selectivity in Electrochemical CO_2 Reduction", ACS Catalysis 8:6 (2018), pp. 5240 - 5249

[2] M. Van den Bossche, E. Skúlason, C. Rose-Petruck and H. Jónsson, "Assessment of Constant-Potential Implicit Solvation Calculations of Electrochemical Energy Barriers for H2 Evolution on Pt", J. Phys. Chem. C 123:7 (2019), 4116 - 4204

CV: Prof. Hannes Jónsson

Born in 1957 in Reykjavík, Iceland, Hannes Jónsson graduated with a B.S. in Chemistry from the University of Iceland in 1980. He then went on to pursue graduate studies as a Fulbright Scholar at the University of California at San Diego, where he obtained his PhD degree in 1986. His doctoral thesis focused on calculations of quantum mechanical scattering of light atoms and molecules from surfaces of solids, in particular surfaces with defects. He then worked at Stanford University as post-doctoral fellow on computer simulations of liquids and glass transition. In 1988 he became Assistant Professor at the University of Washington in Seattle, and was later promoted to Associate Professor and Professor. In 2005 he moved back to Iceland and has been Professor of Theoretical Chemistry at the University of Iceland since then.