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“Carbon-dioxide Conversion via (Photo)electrolysis: from Novel Catalysts to Electrolyzer Development”

Dr. Csaba Janáky

ERC Starting Grant holder
Department of Physical Chemistry and Materials Science
University of Szeged

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Carbon-dioxide Conversion via (Photo)electrolysis: from Novel Catalysts to Electrolyzer Development

Csaba Janáky

University of Szeged Department of Physical Chemistry and Materials Science Rerrich Square 1., Szeged, 6720

janaky@chem.u-szeged.hu

Electrochemical reduction of CO_2 is a promising method for converting a greenhouse gas into value-added products, utilizing renewable energy. Novel catalysts, electrode assemblies, and cell configurations are all necessary to achieve economically appealing performance. In this talk, I am going to talk about two of these aspects, targeted by our laboratory. Finally, I will mention some aspects of the photoelectrochemical approach.

First, I am going to present a zero gap electrolyzer cell, which converts gas phase CO_2 to products without the need for any liquid catholyte. This is the first report of a CO_2 electrolyzer cell, where multiple stacks are connected, thus scaling up the electrolysis process.[1,2] The operation of the cell was validated using both silver nanoparticle and copper nanocube catalysts, and the first was employed for the optimization of the electrolysis conditions. Upon this, CO formation with partial current densities above 250 mA cm^{-2} were achieved routinely, which was further increased to 300 mA cm^{-2} (with ~95 % Faradaic efficiency) by pressurizing the CO_2 inlet. Evenly distributing the CO_2 gas among the stacks (parallel connection), the operation of the multi-stack cell was identical to the sum of multiple single-stack cells. When passing the CO_2 gas through the stacks one after the other (serial gas connection), the CO_2 conversion efficiency was increased remarkably. Importantly, the presented electrolyzer *simultaneously* provides high partial current density, low cell voltage (−3.0 V), high conversion efficiency (up to 40 %), and high selectivity for CO production; while operating at up to 10 bar differential pressure.

In the second part of my presentation I will shed light on the importance of catalyst morphology, using N-doped carbon (N–C) catalysts as a model system.[3] We found that CO2R activity, selectivity, and stability of N–C electrodes are highly dependent on their porosity. The presence of mesopores was demonstrated to be beneficial in achieving high CO selectivity and current density, with an optimal pore size around 27 nm. Even after convoluting factors other than morphology (e.g., surface chemistry, level of graphitization, surface area), the reasons behind the observed trends are complex. CO_2 adsorption properties, wetting characteristics, and geometric effects are jointly responsible for the massive difference in the CO2R performance. All these properties must be taken into consideration when we aim to understand the reduction mechanism on different catalysts and while improving the performance further to a technologically relevant level (as alternatives to precious metal catalysts).

In the last part of my talk, I will show two selected examples on how to boost photoelectrochemical CO_2 reduction and water oxidation on bioinspired hybrid photoelectrodes.[4,5] I will present examples for Cu_2O, and Fe_2O_3-based electrodes, where the nanocarbon-containing photoelectrodes outperformed the pure semiconductor counterparts, both in terms of the achieved current densities and stability.

References:

(1) Patent pending, PCT/HU2019/095001, Modular electrolyzer cell and process to convert carbon dioxide to gaseous products at elevated pressure and with high conversion rate.

(2) B. Endrődi, E. Kecsenovity, A. Samu, F. Darvas, R. V. Jones, V. Török, A. Danyi, C. Janáky: Multilayer Electrolyzer Stack Converts Carbon Dioxide to Gas Products at High Pressure with High Efficiency
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(3) Dorottya Hursán et. al: Morphological Attributes Govern Carbon Dioxide Reduction on N-Doped Carbon Electrodes, Joule 3 (2019) 1–15

(4) E. Kecsenovity, B. Endrődi, P. S. Tóth, Y. Zou, R. A. W. Dryfe, K. Rajeshwar, C. Janáky: Enhanced Photoelectrochemical Performance of Cuprous-oxide/Graphene Nanohybrids
Journal of the American Chemical Society, 139 (2017) 6682-6692

(5) C. Janáky et. al.: Bioinspired Design: NiFeOOH/α-Fe_2O_3/graphene Photoelectrodes with Advanced Water Oxidation Performance, submitted