



המעבדה לכימיה אורגנית ואי-אורגנית

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הפקולטה לכימיה ע"ש שוליק, הטכניון, חיפה

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בנושא:

**" Strain as an Activity Descriptor in the
Electrooxidation of Urea Over Nickel"**

מחקר זה בוצע בהנחייתו של ד"ר דוד איזנברג. ❖

Strain as an Activity Descriptor in the Electrooxidation of Urea Over Nickel

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Electrolyzers and fuel cells are examples of electrochemical technologies with great promise to aid in the energy transition, for example in the seasonal storage of renewable electricity. Urea is a nitrogen-based hydrogen carrier with low toxicity that can already be found in high concentrations in biowaste, and hence requires minimal new infrastructure for distribution. The urea oxidation reaction (UOR) is a multi-electron, multi-proton reaction which occurs preferentially over nickel-based catalysts. Nickel oxyhydroxide currently has the best known electrocatalytic performance for the urea oxidation reaction both in current density, and onset potential. Nevertheless, the instability of this phase at ambient conditions limits the implementation of any electrochemical technologies based on UOR. The catalytic reaction mechanism and its relationship to catalyst structure (structure-performance relationships), can potentially aid catalyst design in a bottom-up fashion. As such, recent research efforts have focused on elucidating the reaction mechanism of UOR over nickel, showing that UOR occurs spontaneously after nickel oxyhydroxide is formed by the electrooxidation of nickel hydroxide, implying this oxidation of the catalyst itself is the rate determining step. However, nickel, nickel hydroxide, and nickel oxyhydroxide all have several crystalline and amorphous phases that are thus likely intimately linked to catalyst activity. Here, we systematically study the electrochemical activity of three different nickel-based electrocatalysts which we tune electrochemically to be models allowing to disentangle the effect of different nickel phases, oxidation state, morphology, and structural disorder. By employing several characterization techniques, such as Raman spectroscopy, and in-situ spectroelectrochemical attenuated total reflection infrared spectroscopy (ec-ATR-IR), the structure-performance relationships obtained for the different nickel phases are related to different reaction mechanisms. We ultimately show that the strain of the nickel hydroxide to oxyhydroxide transition in the catalysts is negatively correlated to lower onset potential and catalyst stability, and negatively correlated to surface coverage of C-containing species which is believed to be the main deactivating mechanism in nickel-based UOR. We also show that the volumetric and longitudinal strain which are intrinsic parameters to the nickel catalyst materials influence the activity and stability. The insights obtained here into the effect of phase transition strain, and its relation to nickel particle size and structural disorder on the formation of nickel oxyhydroxide are likely to be relevant to nickel-based electrooxidation catalysts as a whole, including for ammonia oxidation, and the oxygen evolution reaction which are deemed important reactions in a sustainable society.