Breaking the scaling relations in electrocatalysis by creating controlled, dynamic artificial strain

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Catalysis is one of the pillars of industry especially heterogeneous catalysis plays a prominent role in synthesis of various commodity chemicals. A main parameter, which is important in controlling the catalytic activity of the material is the binding energy of the reaction intermediates on its surface. In brief, for an efficient catalysis the catalyst surface should have an optimal binding energy, neither too strong nor too weak. Current strategies for optimizing the catalytic activity involve static effects introduced by alloying or by changing the catalyst surface structure. The main idea of those traditional approaches lies on generation of a “static” strained catalyst surface. Moreover, a strain is used to govern the catalytic activity through the binding energy of the reaction intermediates. However, Nature does the catalysis with nearly 100% efficiency by continuously changing and adapting the geometry of the catalyst centers, so that the catalyst and the reactants have an optimal interaction during the process. By mimicking the nature, we aim to design a system where the catalyst surface can be tuned and altered during the reaction to increase the catalytic activity of various electrocatalysts. The current project is a collaboration within theoreticians and experimentalists.

Within the current proposal the theoretical work contribution is twofold:

1. Screen the different catalysts in terms of surface, strain and ligand effects to find promising electrocatalysts for Oxygen Reduction Reaction (ORR)/Oxygen Evolution Reaction (OER).
2. Identify the most promising Metal-Oxide-Framework-derived (MOF) materials.

Both targets will be achieved applying, in conjunction, atomistic calculations (based on density functional theory) and data driven methods implementing machine learning (ML) models.

Target 1: To individuate the most promising catalysts we will extend the work on paper [1] to go beyond simple metallic systems. To do so, we plan to implement a new Deep Neural Network. The success of a supervised ML method is strongly related to three points: (i) the size and quality of the training dataset, (ii) the correct choice of the data representation for the machine and (iii) the most effective network architecture. (i) We plan to collect dataset from direct measurements (by experimentalist partners) as well as from in house atomistic calculations using Quantum Espresso. (ii) To implement efficient ML architectures based on Graph Neural Networks (GNN). The trained ML model will help to predict, starting from unrelaxed structures, the binding energies for *O, *OH and *OOH that will be used, applying the scaling theory, to predict the electrochemical activity for OER/ORR.

Target 2: Once the most active catalysts are found a second target will be to help to identify the best MOF to synthesize them. The problem of finding the correct MOFs relies on selecting the right metallic nodes, linkers and topology in order to generate the correct cage for synthesis of the nanostructured catalyst.

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Required skills: programming, machine learning, material science.
Contract Type/Length: 75% E13 position (3 years).