Carbon foams as catalyst for electrochemical CO_2 reduction reaction (eCO_2RR)

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Transformation of waste materials into valuable products is the key pillar for building sustainable economy. Therefore, utilization of environmentally harmful CO_2 emissions as a resource is of interest (1) For recycling of CO_2 as an energy carrier, thus reducing its concentration in the atmosphere (2) As a convenient means of storing electrical energy in chemical form via electroreduction of CO_2 (eCO_2RR), and (3) For the production of renewable hydrocarbon fuels from abundant renewable electricity, CO_2 and water.

So far, large scale implementation of the eCO_2RR process has been hindered by the lack of stable and inexpensive catalysts, which also should be sufficiently active and selective at low overpotentials. Noble metal catalysts such as Ag, Pd and Au based electrodes have traditionally been considered as the best performing materials for CO_2 to CO conversion reactions. Unfortunately, large-scale application of noble metal catalysts is hampered by their scarcity and high price. On the other hand, Cu and Sn which are abundant and inexpensive have been studied extensively as catalysts for eCO_2RR. Cu is active toward the formation of various C1-C3 products, while Sn exhibits superior selectivity towards formate with Faradaic efficiency up to 98%. However, a rapid performance degradation is usually observed in both Sn- and Cu-based electrodes in less than an hour from the start of the CO_2 electrolysis.

Metal-free catalysts for eCO_2RR are attractive due to their lower cost, potentially longer lifetime and higher durability. Recently, nitrogen-doped carbon-foams were prepared and confirmed as high activity catalysts for eCO_2RR. However, the detailed atomistic mechanism for this catalytic activity remains elusive. Fundamental understanding of the catalytic activity of the nitrogen-doped carbon foams is important because it might suggest routes for further improvements, or clearly show the limits of these materials.

Objective of my study is to gain atomistic insights into CO_2 binding into variety of graphene defects and Nitrogen-doped defects. Main assumption in the simulation is that the turnover frequency, which is key characteristic of every catalyst, should depend on the binding energy of CO_2 to the active site of the carbon foam. The DFT simulations aim to answer three fundamental questions:

-What is the binding energy of CO_2 to nitrogen-free defects?

-Is nitrogen-doping always beneficial to CO_2-binding, or binding depends on the structure of the defect?

-Could we change work function of the carbon foam by Nitrogen-free defects?

The answer to the above three questions will be presented in the talk and directions for future work will be highlighted.