17th International Conference on Nanosciences & Nanotechnologies (NN20)
7-10 July 2020, Thessaloniki, Greece

CO_2 electroreduction toward C_2H_4 at Cu decorated graphene nanofilm: Prediction from first principles

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Abstract: The electrochemical reduction of CO_2 is a promising green approach to convert CO_2 into usable form of carbons. The monodisperse Cu nanoparticles assembled on a single layered graphene can support mass- and size-dependent catalysis for the selective electrochemical reduction of CO_2 to ethylene (C_2H_4). The properly treated (e.g. N-doped) graphene itself can catalyse the CO_2 reduction to formate, but the composite graphene-Cu structure functions as a CO_2 and proton absorber, facilitating hydrogenation and carbon–carbon coupling reactions on Cu-nanocluster for the formation of C_2H_4. In this study, we employ density functional theory calculations to examine different active sites in Cu-decorated graphene, to give an outline of the trends in its catalytic activity. We found that adsorption energies of CO_2 and CH_2 do not follow the linear scaling relationships observed for the pure transition metals, and this unique scaling is rationalized through differences in electronic structure between transition metals and graphene. This finding rationalizes most of the experimental observations on the carbon-based materials which present promising catalysts for the two-electron reduction of CO_2 to CO. With thermodynamic analysis, we identify active sites that are expected to exhibit a comparable or even better activity to the state-of-the-art copper catalysts, and the best configurations are suggested to be selective for CO_2 reduction reaction.

Funding from European Union`s Horizon 2020 Research and Innovation Program project under grant agreement No 768789 is greatly acknowledged.