

Computational electrochemistry of doped graphene as electrocatalytic material in fuel cells

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In this work, we present an overview on how density functional theory calculations can be used to design novel electrocatalytic materials for fuel cells. In particular, we focus the attention on non-metal doped graphene systems, which were reported to present excellent performances as electrocatalysts for the oxygen reduction reaction (ORR) at the cathodic electrode of fuel cells and are, thus, considered promising substitutes of platinum or platinum alloys electrodes[1]. The methodology, originally proposed by Nørskov et al. [2] for electrochemical processes at metal electrodes, is revisited and applied specifically to doped graphene. Finite molecular models of graphene are found to perform as well as periodic models for localized properties or reactions [3,4]. Therefore, the sophisticated molecular quantum mechanics methodologies can be safely used to compute reliable Gibbs free energies of reaction in an aqueous environment for the various steps of reduction (at the cathode) or of oxidation (at the anode). Details of the reaction mechanisms and accurate cell onset- or over-potentials can be derived from the Gibbs free energy diagrams. The latter are computational quantities which can be directly compared to experimentally obtained cell overpotentials. Modeling electrocatalysis at fuel cells is, thus, an extremely powerful and convenient tool to improve our understanding of how fuel cells work and to design novel potentially active

electrocatalytic materials. In this work, we present two specific applications of B-doped graphene [5], as electrocatalysts for the ORR at a half-cell cathode [6] and for the methanol oxidation reaction (MOR) at a half-cell anode.

References

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Figures

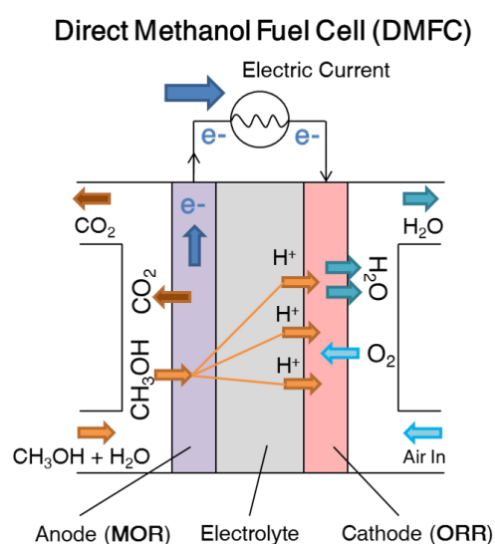


Figure 1: Scheme of a DMFC (Direct Methanol Fuel Cell) fuel cell: at the anode methanol is oxidized to CO₂, while at the cathode oxygen is reduced to H₂O.