

ABSTRACT

Within the overarching goal of increasing energy sustainability and decreasing emissions, the hydrogen fuel cell holds a key place. The proton exchange membrane fuel cell (PEMFC) has been commercialized in the Toyota Mirai. The PEMFC, however, uses a Pt catalyst and is economically unaffordable by many consumers.

Hydroxide exchange membrane fuel cells (HEMFC) enable the use of a broad variety of non-precious metal catalysts. However, a drawback to commercialization of HEMFC technology is the decreased rate of hydrogen oxidation reaction (HOR) in base. Herein, computational studies are performed to predict active catalysts for basic environments, as well as to elucidate the cause of the decreased rate of HOR.

Molecular simulations are performed for a variety of nanoparticles supported on graphene and graphene doped with nitrogen or boron. The presence of either nitrogen or boron leads to a greater nanoparticle-support interaction, and influences nanoparticle properties. The development of a predictive adsorption model, size effects, and the impact on HOR activity are discussed.

To explain the dependence of the HOR kinetics on pH, a new thermodynamic descriptor for HOR, the apparent Gibbs free energy of hydrogen adsorption, is introduced. This descriptor, which accounts for pH-dependent water adsorption, is computed and shown to be consistent with measured HOR kinetics.

Future directions include direct computation of HOR free energy barriers, as well as the dynamic simulation of the electrode interface. Experimentally, controlling nanoparticle size and tuning water adsorption should be points of emphasis.