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3D INTERFACE-ENGINEERED TRANSITION METAL OXIDE/CARBON HYBRID STRUCTURES FOR EFFICIENT TRI-FUNCTIONAL OXYGEN ELECTROCATALYSIS IN ALKALINE AND ACIDIC ENVIRONMENTS FOR NON-MANNED SPACE EXPLORATION DEVICES

Abstract

Many of the non-manned technologies at NASA use regenerative fuel cells (RFC) which require trifunctionality to decrease cost (reducing weight) for space exploration. Tri-functionality uses three reactions to supply energy: oxygen evolution reaction (OER), oxygen reduction reaction (ORR), and hydrogen evolution reaction (HER) to allow both water splitting and formation. Many RFC have used Pt and its alloys (Pt/Ir or Pt/Ru) due to their efficient catalytic surface area, selectivity of hydrogen and oxygen in aforementioned reactions, and stability in harsh environments. However, their fast degradation in durability tests, agglomeration, and methanol poisoning has driven researchers to find alternatives. Non-precious transition metal oxides (TMO) including MnOx, Fe3O4, and Co3O4 have attracted attention as TMOs can maximize catalytically active sites per mass and volume.

We utilized a composite structure where CeO2, ZrO2 or TiO2 is deposited onto conductive 3D carbon structures, such as graphene oxide (GO) or metal-organic frameworks (MOF), whose merits include high surface area and excellent electronic conductivity. However, GO only contains active anchorage sites for nanoparticle on wrinkles and edges, not the basal plane. MOF suffer from degradation after pyrolysis (common step) for which, a thin metal oxide could resolve. To combat these, we leverage acid treatments to functionalize the basal plane for GO (hydrobromic/oxalic acid) and MOF (phosphoric acid).

The hydroxylated CeO2/GO hybrids showed the best ORR and OER performance in both 0.1 M HClO2 and 0.1 M KOH, in terms of onset/half-wave potential, electron transfer number, and current density when comparing to the performance of IrO2 or Pt/C. From a series of material/experimental analyses, a strong tethering of metal oxides upon the basal plane of GO prohibits restacking, and that the particle-GO interfaces (as oppose to the particle or GO itself) dictates the performance and reaction route, as indicated in density functional theory calculations. In addition, a hybrid catalyst where TiO2 nanodots are uniformly anchored on phosphorylated MOF by atomic layer deposition (ALD) showed an even better ORR and OER performance in 0.1 M KOH when compared the aforementioned CeO2/GO hybrid. Materials characterization emphasizes a strong adhesion of metal oxides upon MOF structures, thus providing ample surface interactions for favorable reaction route is important.

However, further analysis was needed to see if an additive such as PdO2 via hydrothermal reaction could increase HER capabilities. Results show an increase in HER capabilities of the CeO2/GO/PdO2 hybrid, but not for the titania MOF based sample.