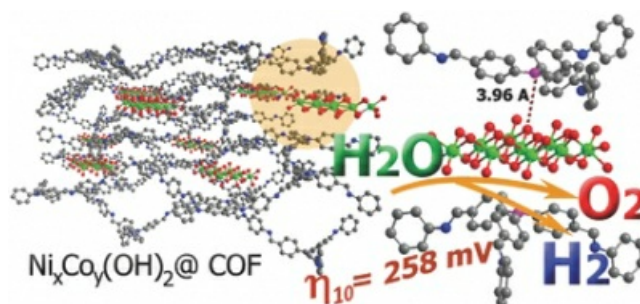


Low overpotential electrocatalytic water splitting with $\text{Co}_x\text{Ni}_y(\text{OH})_2$ NPs supported COFsDinesh Mullangi¹, Vaidhyanathan Ramanathan¹¹Iiser Pune, Pune, India

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Covalent Organic Frameworks (COFs) are highly ordered crystalline porous materials, have attracted a great deal of research interest due to their chemical diversity, high surface area and tenable functionalities¹. Presence of pre-designed nitrogen rich functional groups in COFs make them capable of strongly interacting with active metal nanoparticles to form excellent heterogeneous catalysts and therefore avoid metal NPs leaching.²⁻⁴ Here, we formed highly flexible tetrahedral sp^3 nitrogen rich COF which can serve as ideal supports for nanoparticles for electrocatalytic water splitting. This flexibility gives rise to structural changes which generate mesopores capable of confining very small (<2 nm) non-noble-metal-based nanoparticles (NPs). This dispersed small-size NPs in the mesopores of the COF enables high access to the catalytic sites resulting exceptional activity toward the Oxygen Evolution Reaction (OER) from alkaline water with overpotential of 258 mV at a current density of 10 mA cm^{-2} . The overpotential observed in the COF-nanoparticle system is the best in class, and is close to the current record of ≈ 200 mV. Also, it possesses outstanding kinetics (Tafel slope of 38.9 mV dec^{-1}) for the reaction. These values surpass most of the reported Ni/Co-based electrocatalyst in their supported as well as unsupported forms. DFT modeling shows the interaction between the hexagonal $\text{Ni}(\text{OH})_2$ NPs having sandwiched between the sp^3 nitrogens of the adjacent COF layers. This interaction could be crucial to maximizing their synergistic interactions for active catalyst.

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