Dear Editor,

Enclosed please find our manuscript, "**Chestnut-like Copper Cobalt Phosphide Catalyst for All-pH Hydrogen Evolution Reaction and Alkaline Water Electrolysis**", which we submit for publication in **Journal of Materials Chemistry A.** We declare that there are no related manuscripts submitted elsewhere, and the authors have no competing financial interests.

Hydrogen generation through water splitting has been widely regarded as a green and efficient way to meet the increasing demands for renewable and clean energy sources. So far, Pt is well known as the most effective catalyst for HER, while Ru/Ir oxides are acknowledged as the most efficient OER electrocatalysts. Unfortunately, the widespread usage of these catalysts is seriously impeded due to their scarcity and high cost. Many efforts have been devoted to transition-metal phosphides (TMPs) as an alternative to noble metals owing to their noble-metal-like surface reactivity, mechanical strength/hardness, and chemical stability in a corrosive environment. However, there is still a lack of efficient electrocatalysts that can simultaneously catalyze the HER and OER in the same electrolyte at low over-potential. Hence, it is urgent to develop bifunctional, non-noble metal catalyst that have both high catalytic and stability, which will simplify the cell structure and reduce the cost of the electrolyzed water technology. At present, the electronic structure of catalysts is an efficient way to accelerate the HER process by metal doping. In addition, the morphology of the catalysts is another critical factor to affect their HER activity.

In this work, we present an electronic structure and morphology control engineering of CoP for accelerating HER process in solutions with a full pH range. A novel type of chestnut-like $Cu_xCo_{1-x}P$ nanoarrays on carbon paper ($Cu_xCo_{1-x}P$/CP) for the splitting of water are synthesized through a simple hydrothermal followed by in situ phosphorization treatment. Its various analysis of structures and composition, detailed electrochemical analysis of kinetics and DFT calculations reveals that Cu doping can reduce the hydrogen adsorption free energy (ΔG_{H*}) and enhanced electrical conduction thus greatly enhancing the HER activity. In addition, Cu-doping reduces the Gibbs free energy (ΔG_2) of rate-determining step and the calculated adsorption energy of O^* drops dramatically for OER, thus leading to the enhanced reaction kinetics and excellent electrocatalytic activity. Moreover, the SEM images of the $Cu_{0.075}Co_{0.0925}P$/CP after HER and OER test proved the as prepared catalysts are of good structural stability.

This synthesize method presented in this work may be extended to other transition-metal phosphides, for the rational design and synthesis of non-noble metal bifunctional catalysts with unprecedented electrocatalytic activities and stabilities for next generation energy conversion and renewable energy technologies such as water splitting and zinc-air batteries.

Thank you very much for your attention to our paper.

Sincerely yours,
Yunyong Li on behalf of all authors.

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