Nature-Inspired Design of Nano-Architecture-Aligned Ni_5P_4-Ni_2P/NiS Arrays for Enhanced Electrocatalytic Activity of Hydrogen Evolution Reaction (HER)

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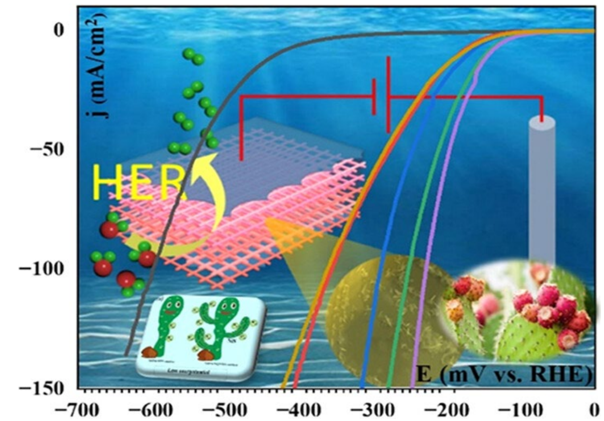
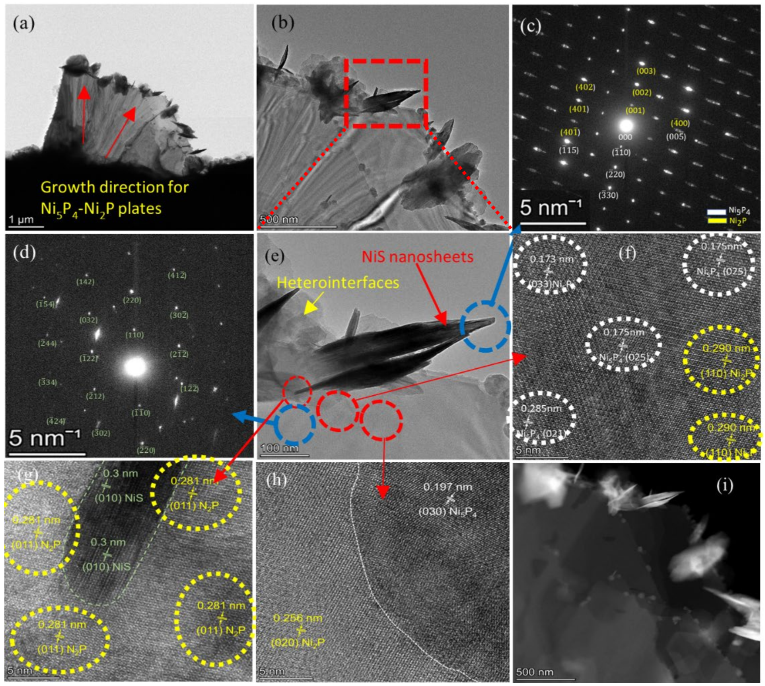
Herein, we demonstrate a controllable phospho-sulfidation process to synthesize the Ni_5P_4-Ni_2P/NiS (plates/nanosheets) electrocatalyst with abundant epitaxial heterointerfaces for hydrogen production. A regional prickly pear cactus with plentiful surface area motivates this study to design a synthesis strategy that led constructing a 3D heterostructure electrocatalyst with an abundant active catalytic area and enhanced intrinsic catalytic activity. We rationally employ an ultrathin nickel mesh to facilitate a uniform phase transformation and support spatial freedom for departing produced hydrogen. After the phosphidation process, the Ni_5P_4 and Ni_2P structures, known for their outstanding HER performance, form simultaneously on the mesh surface with two surface features, protrusions and vertically aligned plates. We realize that Ni_5P_4 and Ni_2P phases coexist in the vertical plates, making nanoscale heterointerfaces and reinforcing electrical connectivity. After the sulfidation process, the epitaxial NiS nanosheets nucleate and grow primarily at the edges, kinks, or steps observed on the facets of the Ni_5P_4-Ni_2P plates. The *in-situ* epitaxial interfaces nucleate at the root of the NiS nanosheets interfacing with the Ni_2P phase and induce the heterointerfaces formation. We deduce that the electrochemical surface area (ECSA) expansion and intrinsic catalytic activity enhancement are the two influential factors optimizing the Ni_5P_4-Ni_2P/NiS electrocatalytic performance. The electrochemical studies, including EIS and CV analyses, verify the importance of emerging heterostructures in reinforcing electron mobility and stability at higher overpotentials. Our findings emphasize the importance of the successful growth of epitaxial nanosheets in improving electrocatalytic activities of the Ni_5P_4-Ni_2P/NiS heterostructures.





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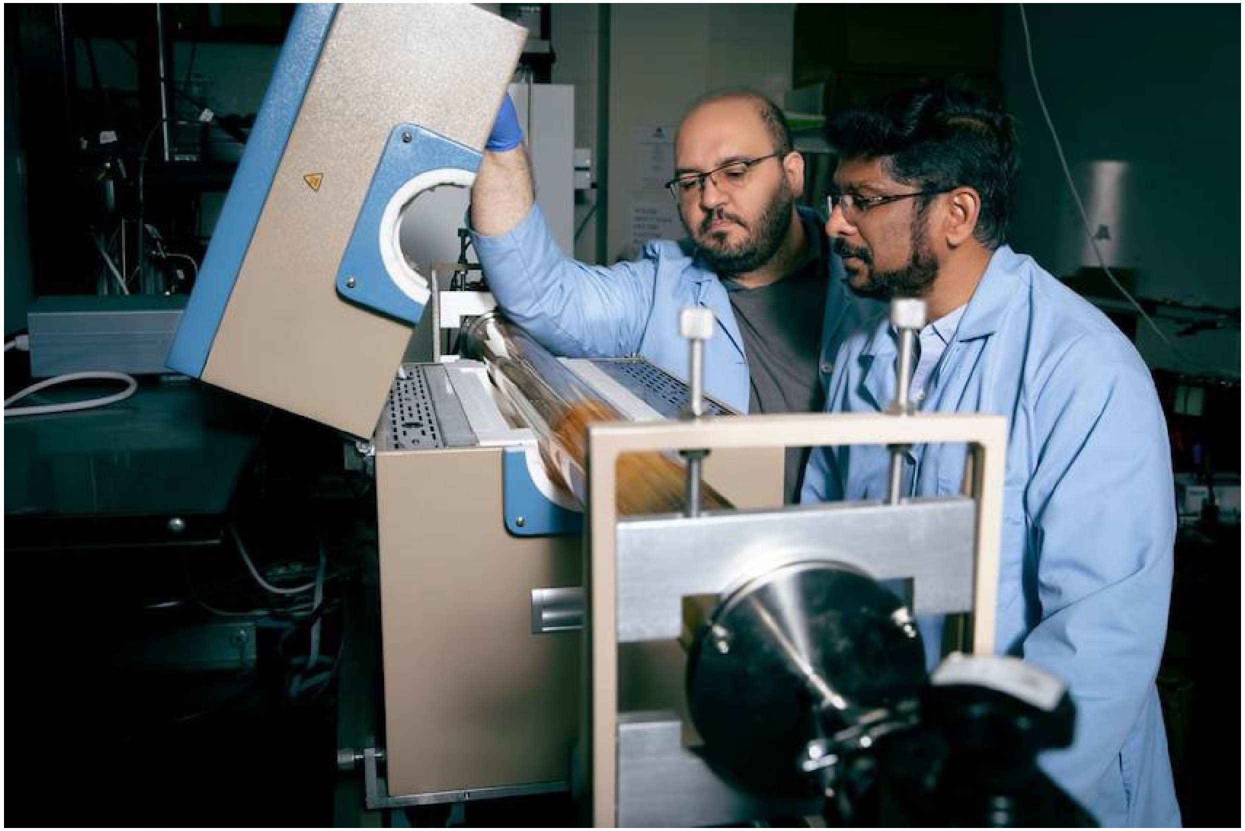

Preparing the next generation of scientists, particularly from underrepresented groups, is an ongoing priority for the PREM Program. Navid Attarzadeh, a doctoral student this year, has been awarded funding from the PREM program. Many younger scientists will look up to Navid as a mentor and inspiration for their own research careers. Mr. Navid is graduating with his Ph.D. in July/Aug. 2023.