

Rational Design of (Photo)electrocatalytic Materials for Artificial Photosynthesis

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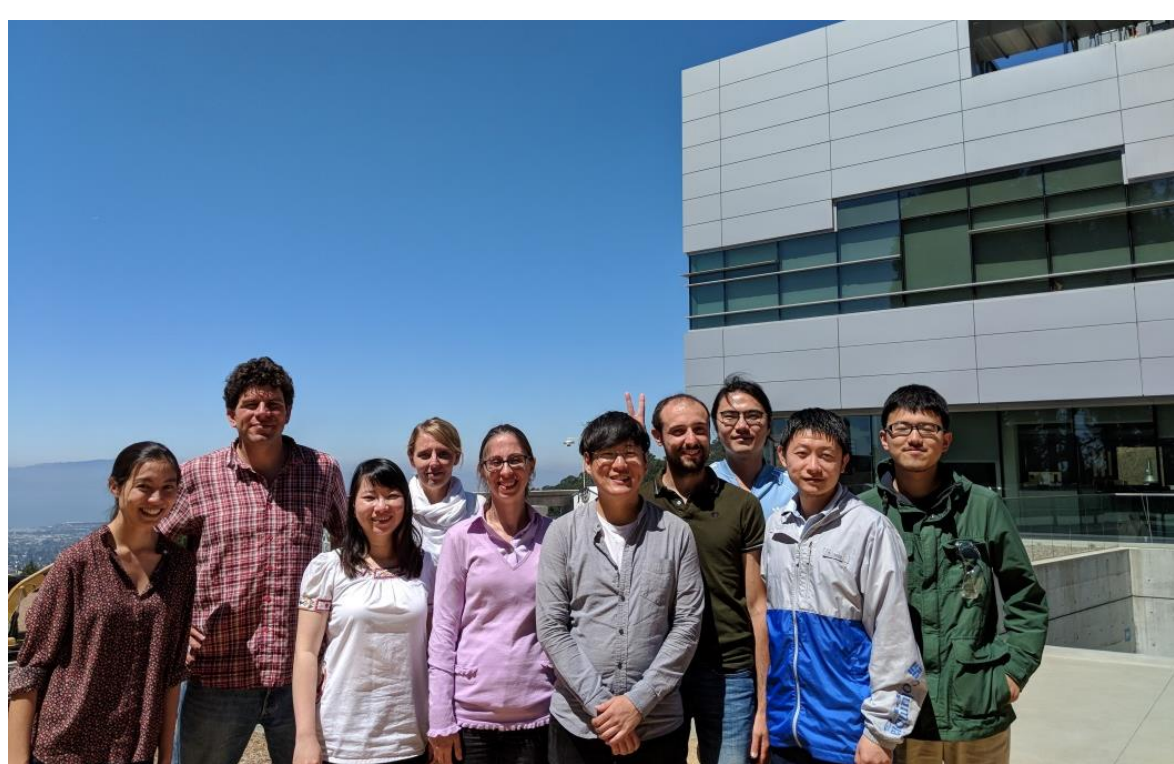
Abstract:

Due to the rapid depletion of fossil fuels and related environmental issues, developing technologies for producing renewable, clean fuels for our future is of great importance. Artificial photosynthesis via water splitting or CO₂ reduction offers an attractive and cost-effective route to achieve this goal. However, existing (photo)electrocatalytic materials generally suffer from low activity or instability, thus greatly impeding the feasibility of artificial photosynthesis technologies. Herein, we present design strategies for the synthesis and integration of novel (photo)electrocatalytic materials, which sheds light on rational design of photoelectrocatalytic materials for artificial photosynthesis.

Introduction

Industrial development and population growth have led to a surge in the global demand for energy in recent years. Sustainable energy technologies such as solar-fuels devices enable us to generate sustainable fuel from renewable energy, which constitute a promising approach to power our planet. Market commercialization of sustainable energy technologies requires the rational design of efficient and stable (photo)electrocatalytic materials

Team



Outlook

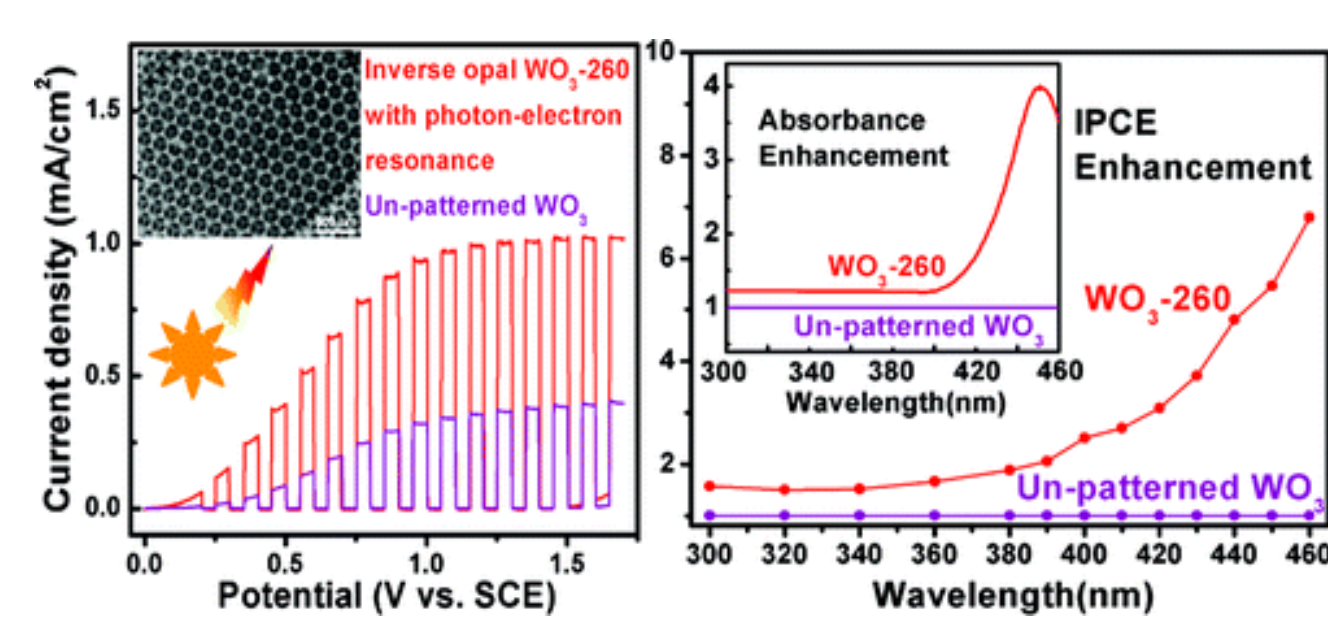
These work shed light on paths for rational design and implementation of advanced materials in viable technological devices.

Acknowledgments

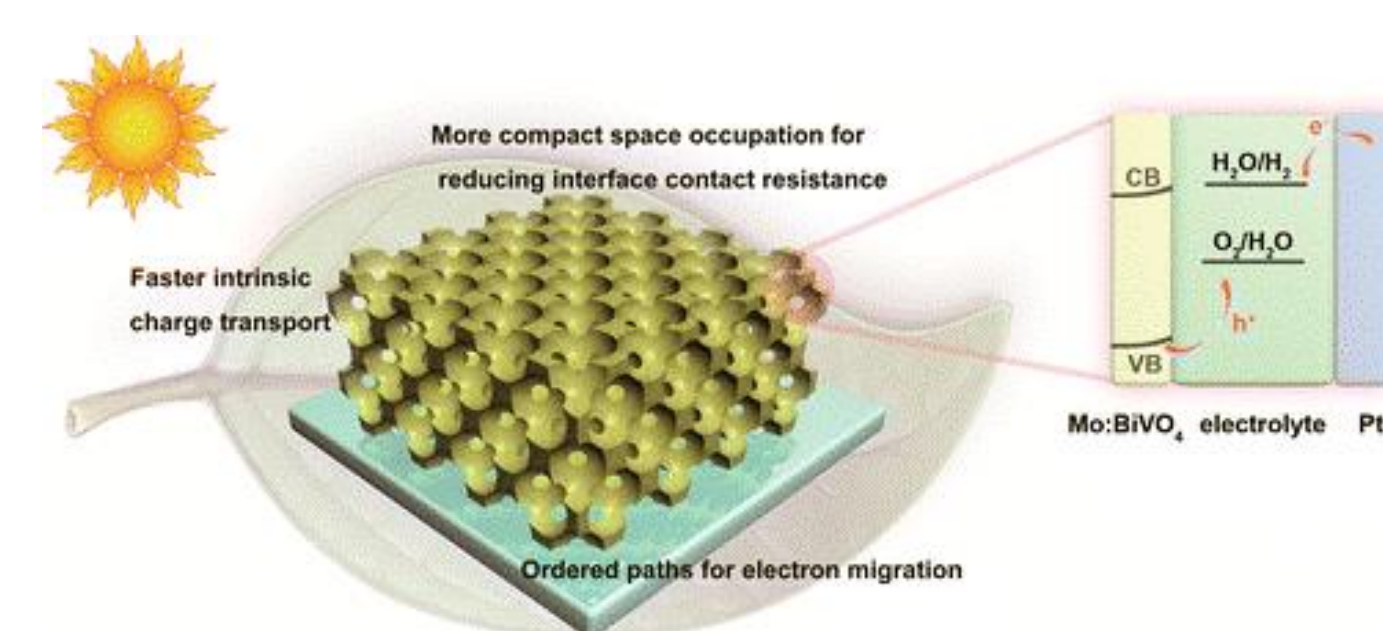
This material is based upon work performed by the Joint Center for Artificial Photosynthesis, a DOE Energy Innovation Hub, supported through the Office of Science of the U.S. Department of Energy under Award Number DE-SC0004993. The authors thank Dr. Joel Haber, Dr. John Gregoire and Dr. Ian Sharp for their insightful discussions and contributions to the work.

Results, Highlights, and Accomplishments

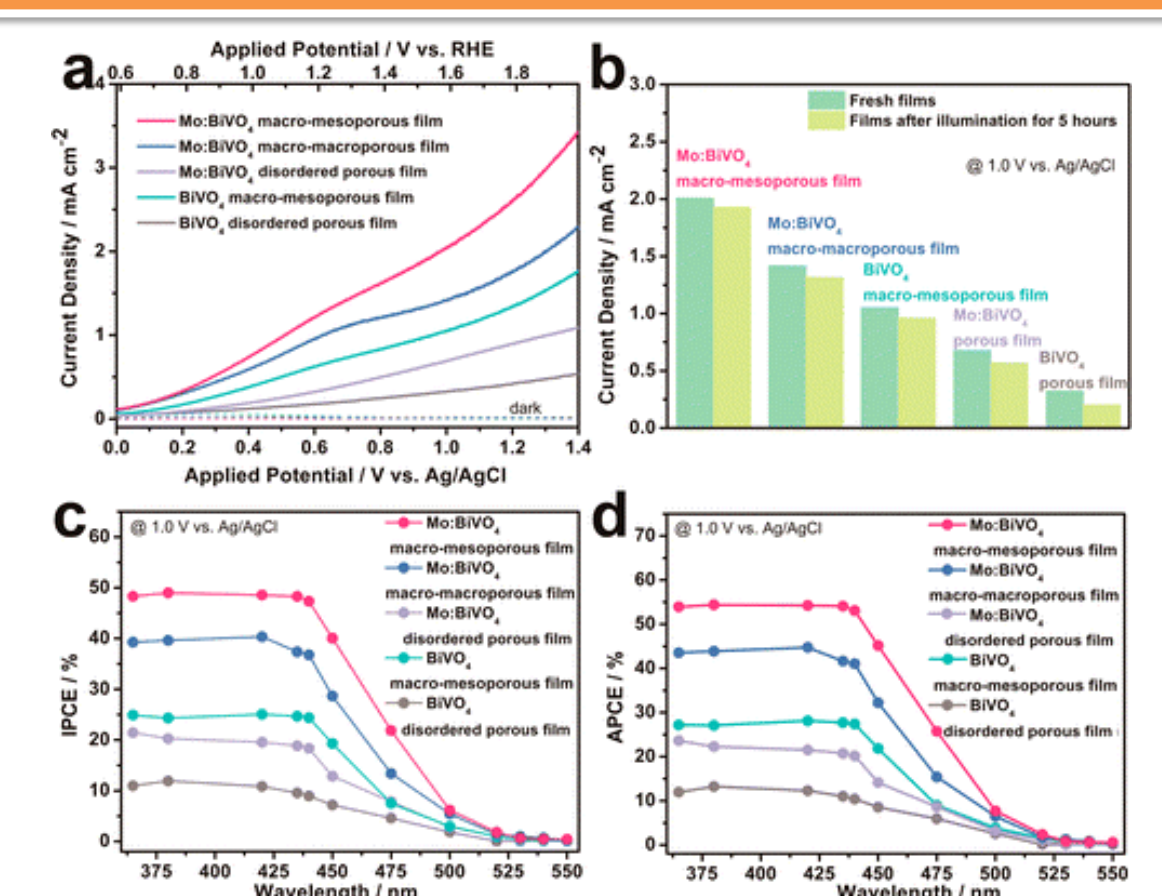
Fabrication and optical characterization of polystyrene opal templates for the synthesis of scalable, nanoporous (photo)electrocatalytic materials



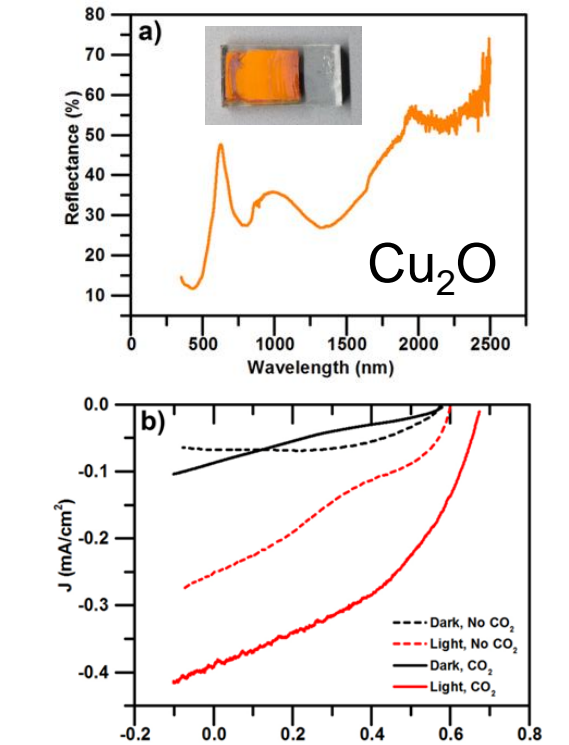
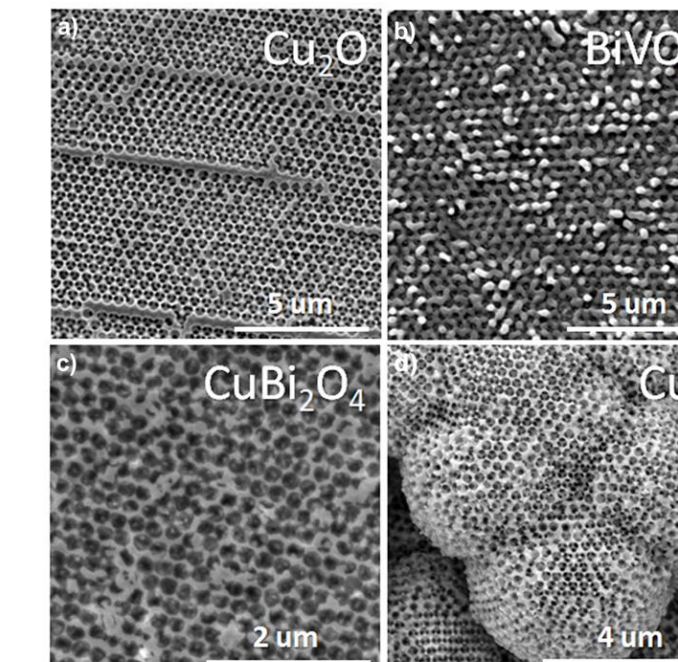
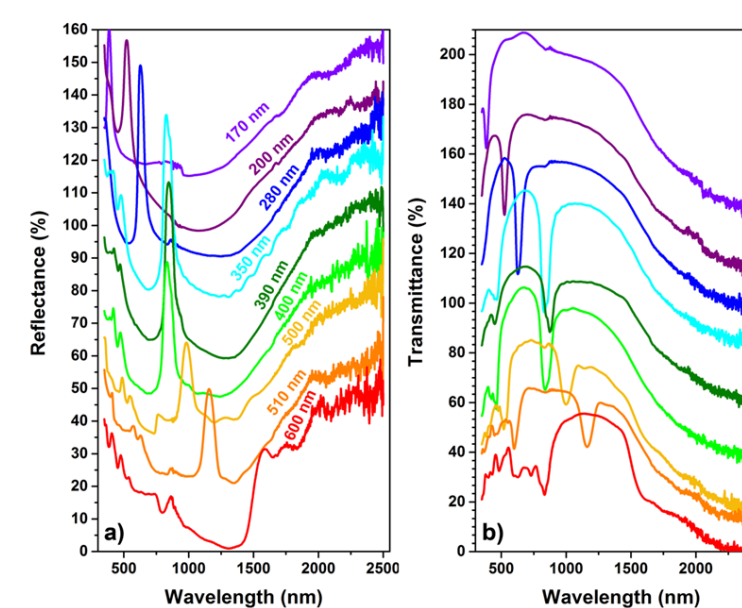
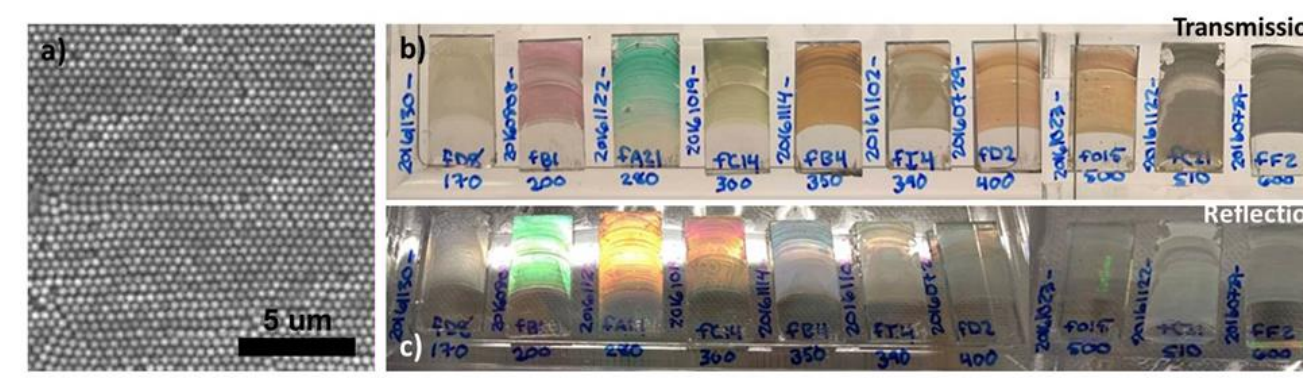
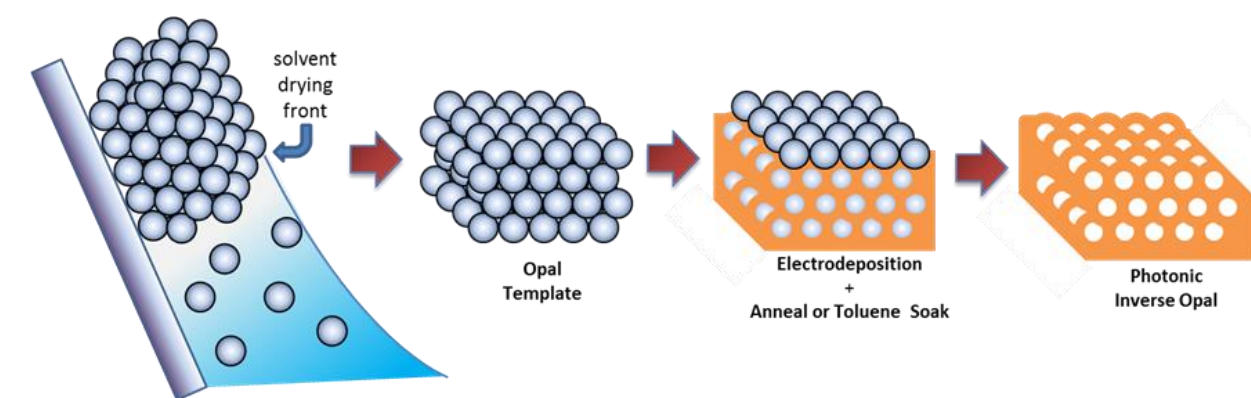
ACS Nano 2011, 5, 6, 4310.



ACS Nano 2014, 8, 7, 7088



Nanostructuring approaches for photoelectrocatalytic materials have the potential to reduce bulk recombination and improve electron-hole pair separation in semiconductor light absorbers, as well as to increase the active surface area and to influence the activity in catalytic systems. 3D inverse opals (IOs) provide highly ordered and uniform porous structures for generating photonic crystals for controlled manipulation of light propagation and charge transport in photoelectrocatalytic materials. However, it is challenging to fabricate large scale opal template films and effectively convert them into equally homogenous ordered replicas. Herein, we report the synthesis of scalable (cm²), highly reproducible polystyrene (PS) opal template for the fabrication of electrodeposited IOs of (photo)electrocatalytic materials.

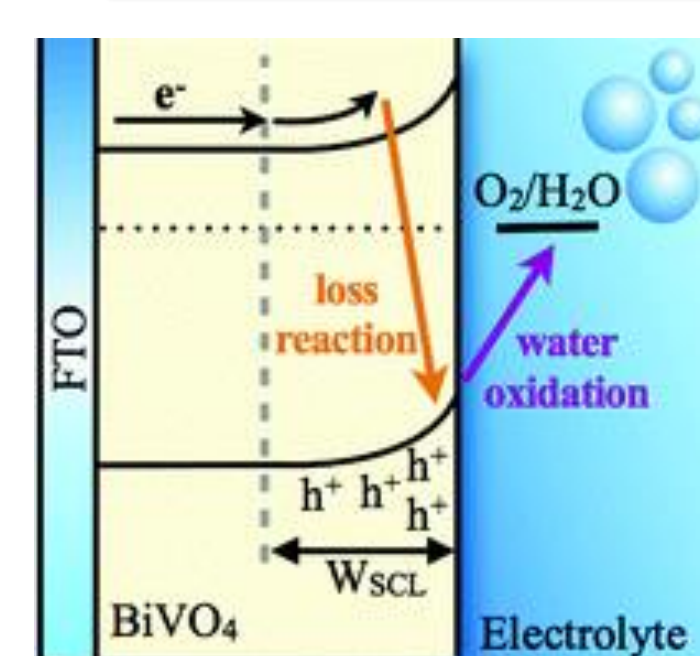


High quality polystyrene (PS) sphere templates

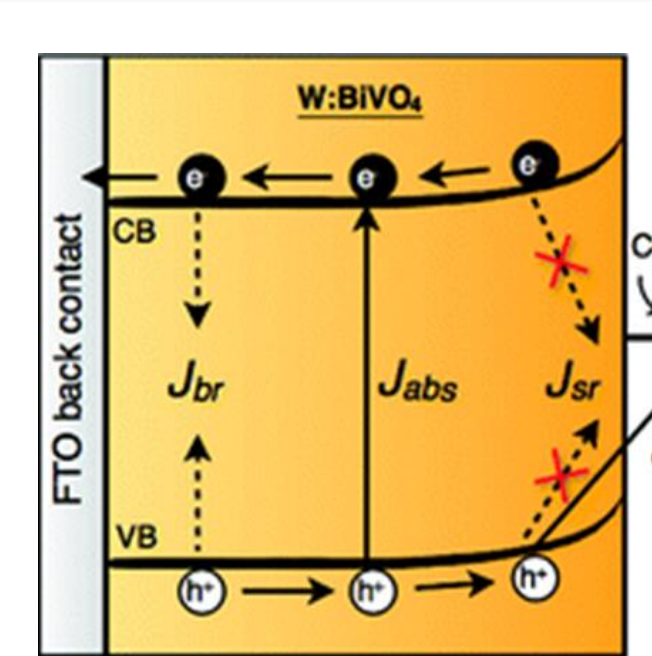
High quality PS opal films were fabricated to template bottom-up electrodeposition of IO porous structures comprising a variety of electrocatalytic and photoelectrocatalytic materials including Cu₂O, BiVO₄, CuBi₂O₄, and Cu.

J. Mater. Chem. A, 2017, 5, 11601.

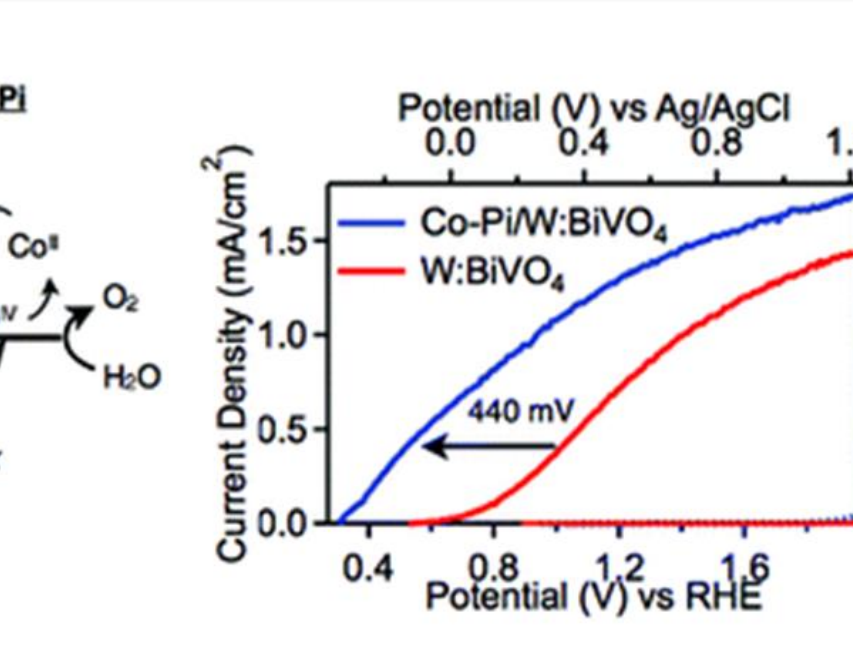
Interface engineering for light-driven water oxidation: unravelling the passivating and catalytic mechanism in BiVO₄ overlayers



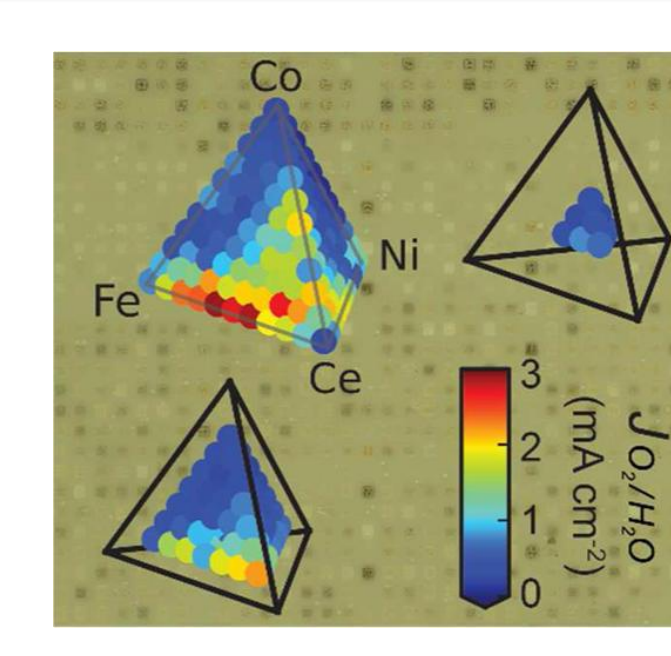
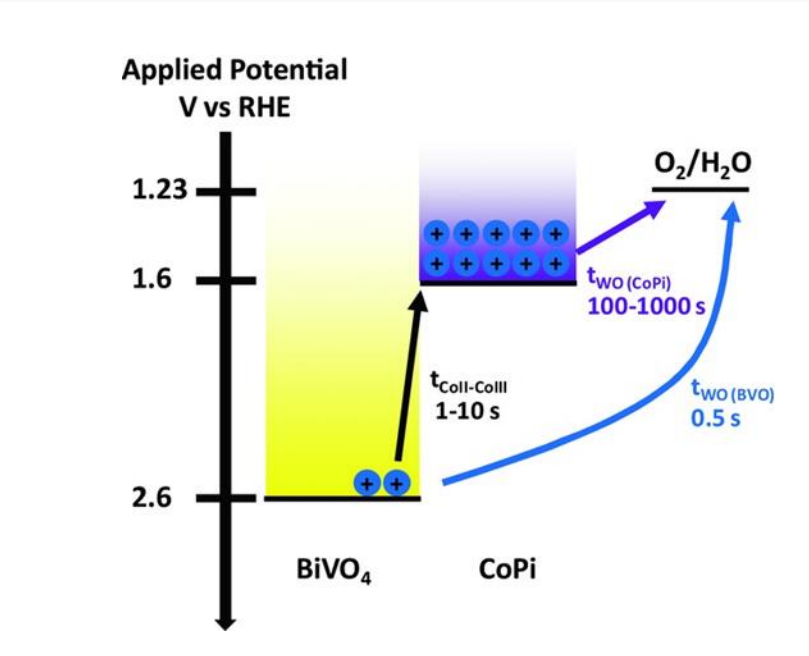
Chem. Sci., 2014, 5, 2964.



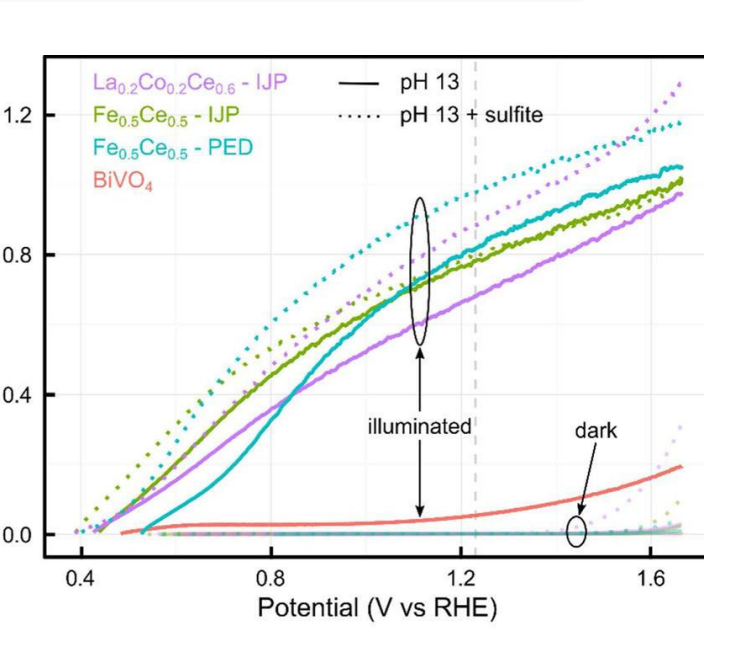
J. Am. Chem. Soc., 2011, 133, 45, 18370.



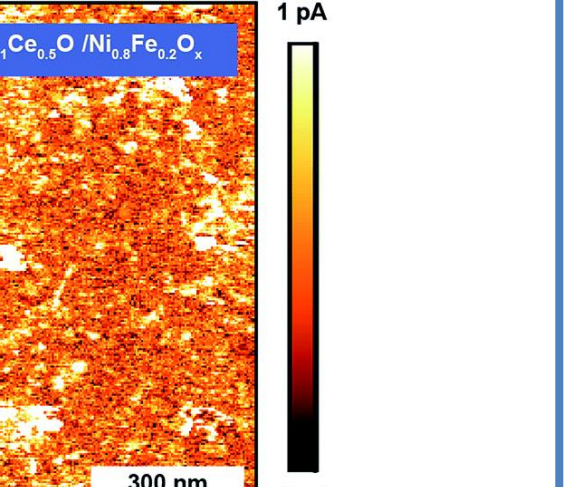
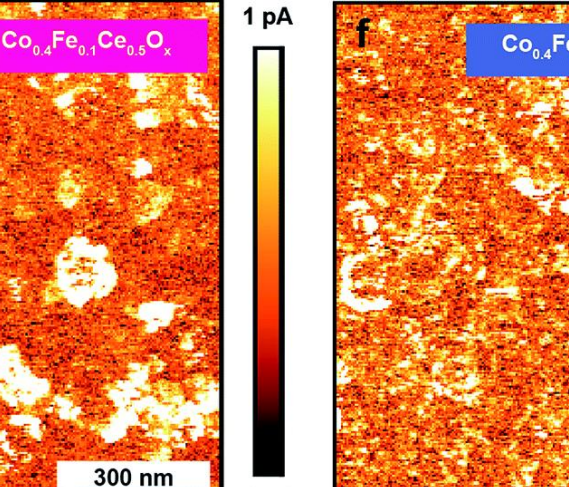
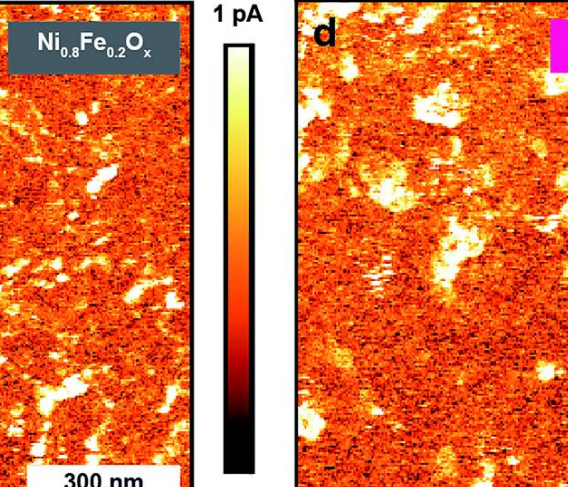
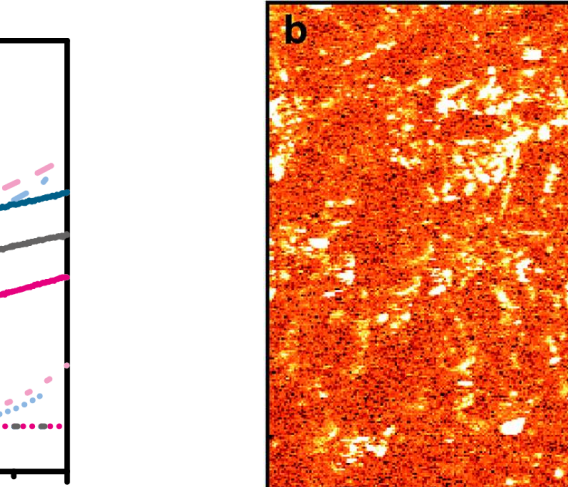
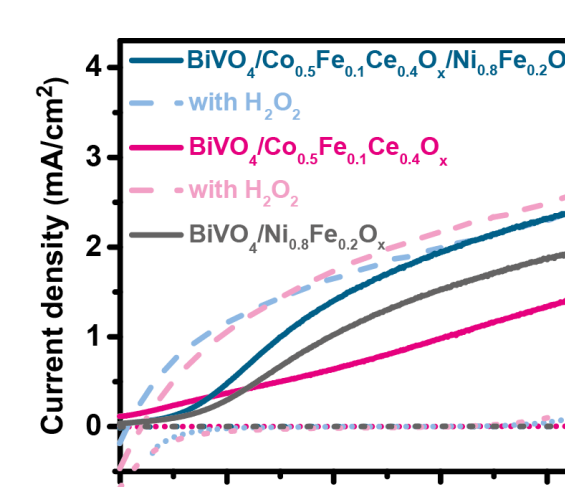
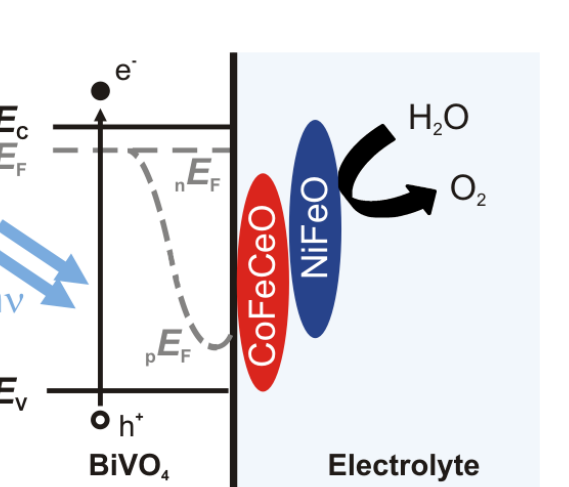
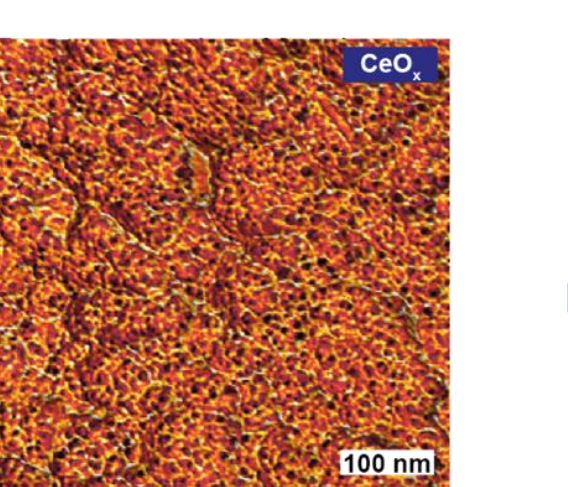
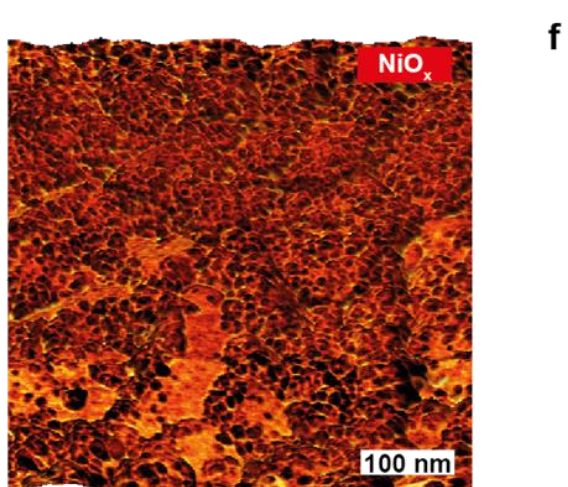
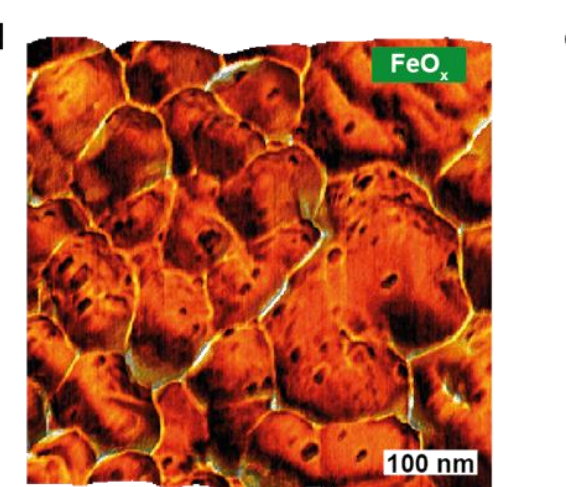
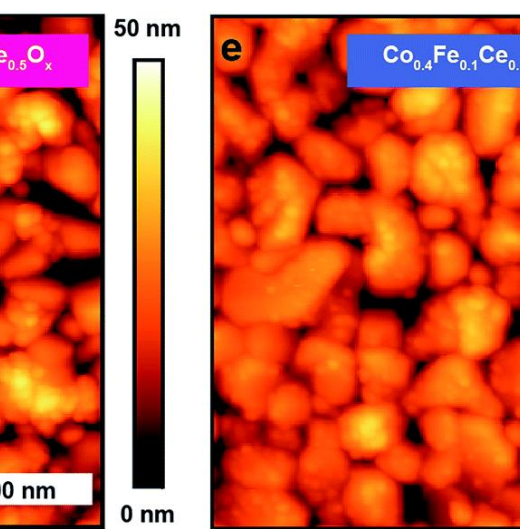
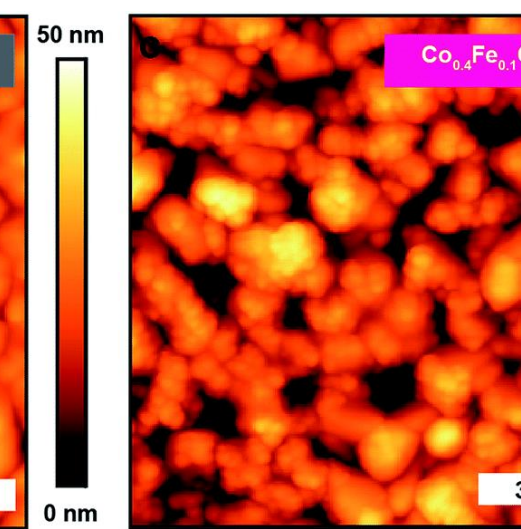
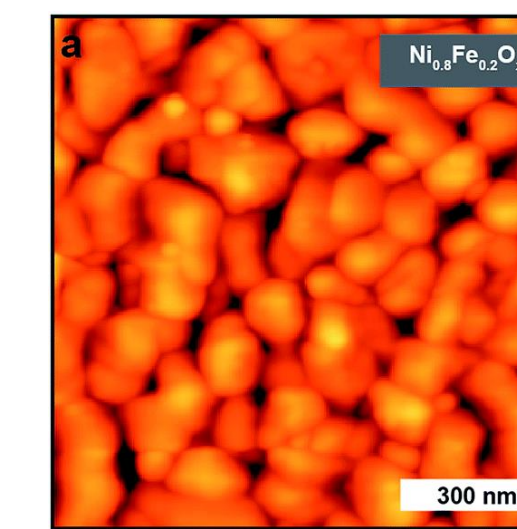
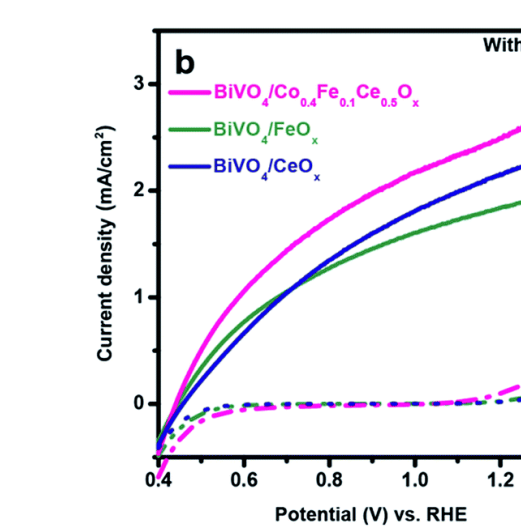
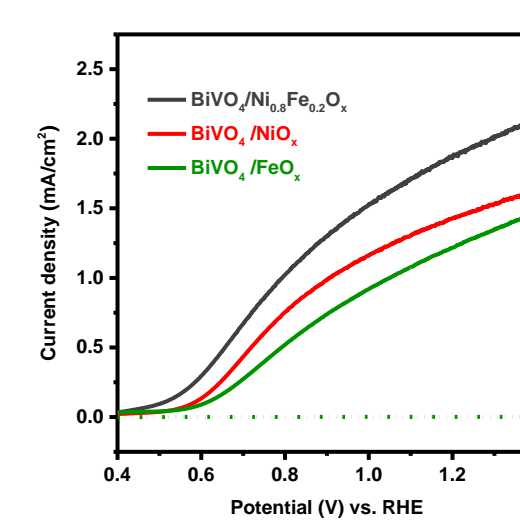
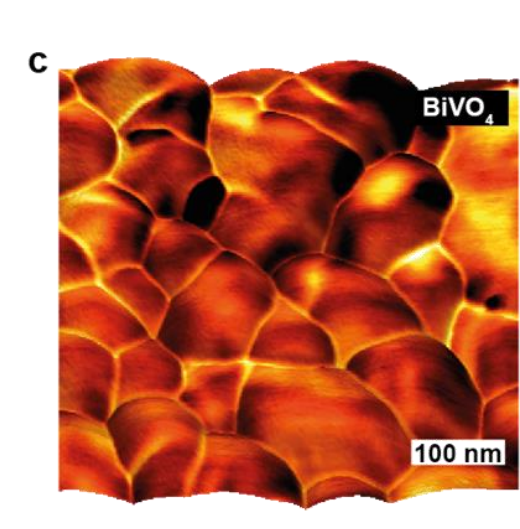
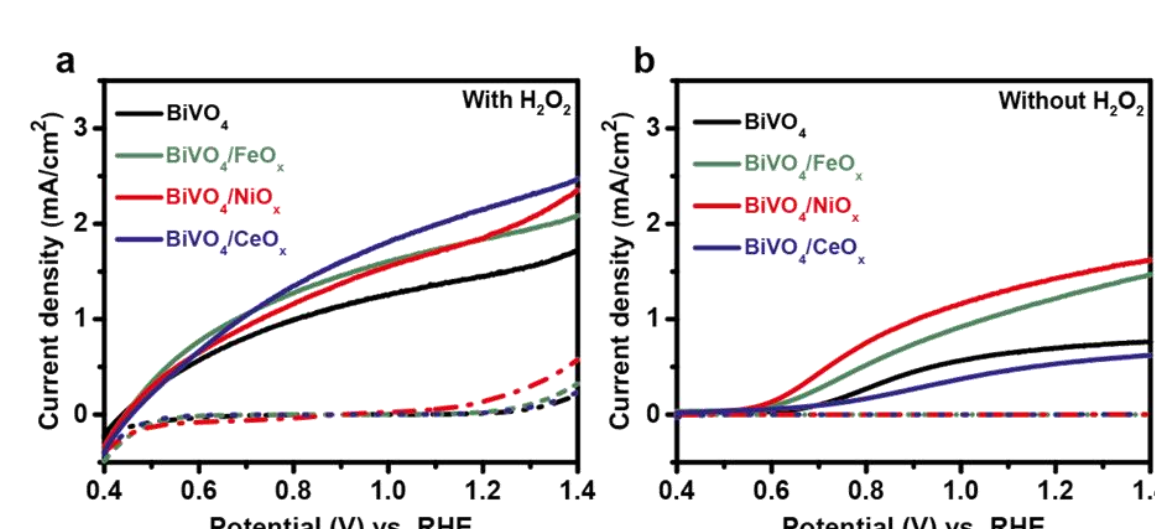
Adv. Funct. Mater. 2016, 26, 4951.



ACS Appl. Mater. Interfaces., 2016, 8, 23696.



The surface of BiVO₄ processes poor catalytic nature for oxygen evolution reaction (OER). Despite numerous efforts that have coupled various catalysts to light absorbing BiVO₄, the optimization of semiconductor/catalyst as well as catalyst/electrolyte interfaces and the identification of the role of the catalyst still remain a key challenge. In addition, different working mechanisms have been reported for these catalysts, depending on the physicochemical nature of the BiVO₄/catalyst interface. Herein, we assemble (NiFeCoCe)_x multi-component overlayers, interfaced with bismuth vanadate photoanodes, and determine the roles of different elements on promoting interfacial charge transfer and catalytic reaction over competitive photocarrier recombination loss processes.



- Individual (Ni, Fe, Co, Ce) oxides and their combinations in multi-component (Ni-Fe-Co-Ce)_x overlayer were studied by photoelectrochemical characterizations and photoconductive AFM.
- The BiVO₄/catalyst interface was optimized by sequentially integrating (Co-Fe-Ce)_x and (Ni-Fe)_x with BiVO₄, resulting in near-complete suppression of interface losses.

Sustainable & Energy Fuels, 2019, 3, 127; In collaboration with Dr. John Gregoire and Dr. Joel A Haber.