Designing Chemically Robust Metal Oxides for Visible-Light Photocatalysis: toward Solar Z-Scheme Water Splitting

Solar driven water splitting for large-scale hydrogen fuel production from semiconductor photo-electrodes has the potential to provide energy on large scale from renewable, sustainable sources. Our research focuses on the kinetically more demanding oxygen-evolution reaction, and we prepare thin film metal oxide photoanodes by low-temperature, solution-based processes. One promising light absorber is TiO_2:(Nb,N) where Nb and N substitute for Ti and O on their respective lattice sites in anatase. These materials are prepared by sol-gel processing followed by annealing in flowing ammonia. We observe a band-gap energy as low as 2.0 eV at 25% Nb and 2% N. In conjunction with a RuO_2 catalyst, powdered TiO_2:(Nb,N) evolves O_2. A second class of materials we study is the transition-metal tungstates, and we have prepared our most promising candidate, $CuWO_4$, by several routes: electrochemical deposition, sol-gel processing, and spray pyrolysis. These methods afford highly reproducible and robust $CuWO_4$ thin-film electrodes on transparent conducting substrates. $CuWO_4$ is an *n*–type semiconductor with a band-gap energy of ~2.4 eV. $CuWO_4$ thin films photooxidize water with simulated solar radiation with a nearly quantitative Faradaic efficiency for O_2 evolution at no applied bias in the presence of the sacrificial electron acceptor, $[Fe(CN)_6]^{3-}$. Most important, these thin-film electrodes are stable against photocorrosion when illuminated with visible light at neutral pH, a significant improvement to the more commonly studied photoanode, WO_3. Current efforts are aimed at preparing complex tungstates that absorb lower energy light to improve the quantum yield.

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MATERIALS SEMINAR

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3:30 p.m. in 1315 Chemistry