Polymer Chromophore-Catalyst Assembly for Solar Fuel Generation

Scientific Achievement

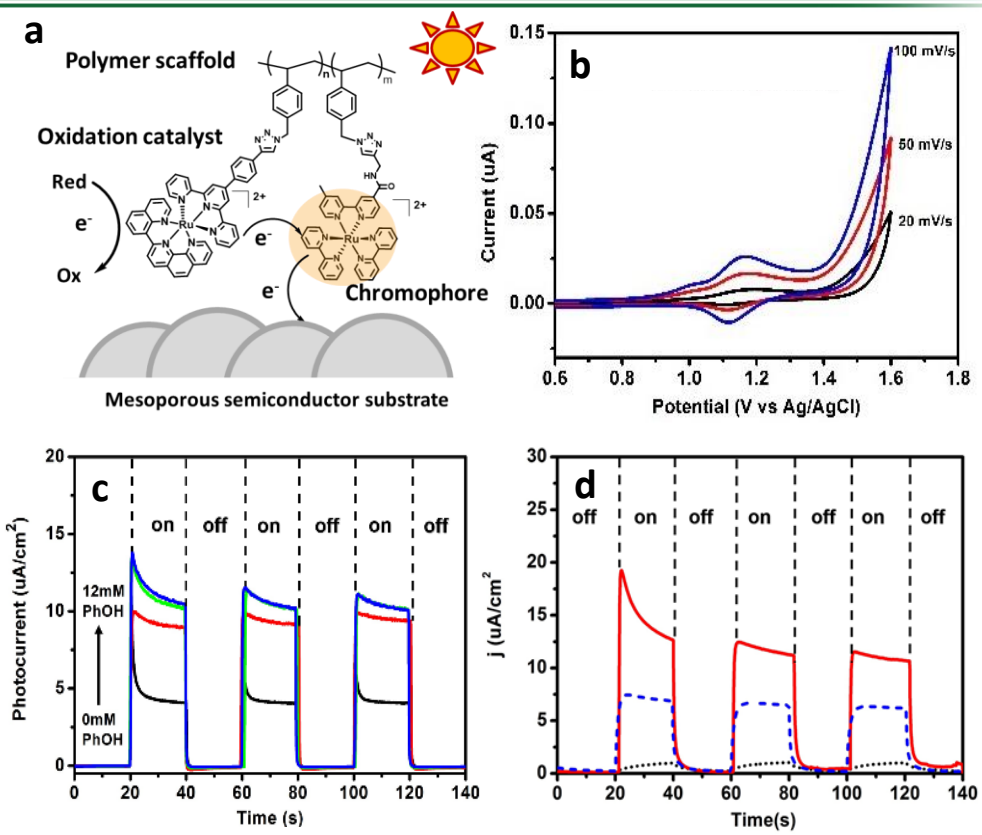
A novel approach toward design and synthesis of a polymeric chromophore–catalyst assembly, followed by its deposition onto a semiconductor via the Layer-by-Layer (LbL) self-assembly method to construct a DSPEC photoanode for photodriven water oxidation applications.

Significance and Impact

Developed the first polymeric chromophore–catalyst assembly specifically containing chromophore units (RuC) and an oxidation catalyst (RuCat) and demonstrated its use in light-driven water oxidation at a photoanode-solution interface.

Research Details

- Nitroxide-mediated radical polymerization (NMP)-click methodology is applied to construct well defined polystyrene scaffolds that can be used to assemble RuC chromophores and RuCat water oxidation catalysts onto a single polymer chain.
- The electrochemical properties of the chromophore–catalyst assembly on FTO provide evidence for catalytic water oxidation and hole transfer from RuC to RuCat in the films.
- Photocurrent response shows that the FTO//TiO_2//(PAA/PS-RuC-RuCat)$_{10}$ assembly gives an increased photocurrent with increasing phenol (PhOH) compared to the reference FTO//TiO_2//(PAA/PS-RuC)$_{10}$ assembly lacking the water oxidation catalyst.



(a) Light-driven oxidation by a polymeric chromophore–catalyst assembly on a mesoporous semiconductor. (b) Cyclic voltammetry of ITO//(PAA/PS-RuC-RuCat)$_5$ in 0.1 M $HClO_4$ aqueous solution. (c) Photocurrent–time traces for FTO//TiO_2//(PAA/PS-RuC-RuCat)$_{10}$ in PhOH solution and (d) FTO//(SnO_2/TiO_2)//(PAA/PS-RuC-RuCat)$_5$ (solid red line), FTO//(SnO_2/TiO_2)//(PAA/PS-RuC)$_5$ (dashed blue line), and bare FTO//(SnO_2/TiO_2) (dotted black line) in 0.5 M KNO_3 aqueous solution.

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Work was performed at the University of Texas at San Antonio and at UNC Chapel Hill

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