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Rational Design of Improved Metal-Organic Framework (MOF)-based Photoanode for Water Splitting

Ru-catalyst MOFs with Pt co-catalyst drive visible-light water splitting at high photocurrents and H_2 evolution rates under acidic conditions.

Researchers at Purdue have developed a novel, visible light (VL) driven catalytic system to produce hydrogen gas for the purpose of renewable energy generation and storage. Due to the threats of global warming, energy-dense, easy-to-produce, non-greenhouse-gas-producing fuels are needed. Much work has been done to solve this problem, including the development of artificial photosynthetic systems to produce hydrogen gas, from just water and solar energy. TiO2-based photon acceptors, one of the most studied photocatalytic water splitting systems, require UV light to drive the water splitting reaction forwards. Because UV light only makes up ~5% of the solar spectrum, a photon acceptor that can utilize VL (~50% of the solar spectrum) to split water will be substantially more efficient in producing hydrogen gas.

The Purdue researchers developed a water splitting system under acidic conditions (pH = 1) with two principal components, the Ru-based water oxidation catalyst demonstrated as [Ru(bpy)2(H2O)2]2+ and [Ru(tpy)(Qc)(H2O)]+, and an iron-oxide-based metal organic framework (MOF) as VL photon acceptors. The optimized system was able to achieve a photocurrent of 1.6*10^-3 A/cm^2, close to the highest reported in the field (~15*10^-3 A/cm^2), with a lower Ru catalyst density and without the need for complex dual-junction solar panels. Additionally, the researchers discovered that by integrating the platinum nanoparticles as a co-catalyst (Pt@MOF) with their optimized MOFs and irradiating with VL, they were able to observe an H2 evolution of 4.8 uM/g*min, without the need for a sacrificial electron donor, a first in the field.

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The photocurrent of the proposed system was evaluated by measuring the current density with the Ru-doped MOFs as the photoanode, platinum wire as the cathode, and an Ag/AgCl reference electrode immersed in 0.1M HNO3. The system was irradiated with VL and left in the dark for 1 min each, for a total of 10 cycles in acidic conditions (pH = 1). The measured photocurrent was 1.6*10^-3 A/cm^2. The stability of the MOFs was evaluated by measuring the infrared spectrum using Fourier transform infrared spectroscopy before and after Ru-doping, the spectrums collected were similar, indicating that the MOF structure is unaltered after doping. The stability of the Ru atoms in the photosystem were evaluated by measuring the electrolyte with an inductively coupled plasma mass spectrometer (ICPMS) before and after electrocatalysis. No Ru atoms were observed in the electrolyte in either sample. Pt@MOF hydrogen gas evolution was measured using a semiconductor-based hydrogen detection system upon exposure of the Pt@MOF to VL. H2 was found to evolve at 4.8 uM/g*min and a maximum evolution of 101 uM/g.

Advantages:

- High photocurrent

- Stable: no observed loss of costly Ru atoms in bulk solution or decomposition of MOFs

Applications:

- Sustainable energy generation/storage

- Hydrogen gas production for bulk synthesis of common chemical reagents

Related Publication: "Photoexcitation of Fe3O Nodes in MOF Drives Water Oxidation at pH=1 When Ru Catalyst Is Present"

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