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研究担当者	王 謙
研究機関名	名古屋大学
所属部署名	工学研究科
役職名	准教授
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研究成果の概要

We designed and developed a novel light-driven system for overall water splitting by integrating photocatalytic and photoelectrochemical technologies and employing two semiconductors with different bandgaps in a Z-scheme configuration. This system addresses two key challenges commonly associated with conventional photocatalytic water splitting: (1) severe side reactions caused by redox mediators used to shuttle electrons, which significantly reduce the solar-to-hydrogen (STH) conversion efficiency, often to below 0.6%; and (2) the simultaneous generation of H₂ and O₂ in a single reaction chamber, which requires subsequent gas separation for practical applications.

The system incorporating a photocatalytic H₂ evolution reaction (HER) cell separated from an O₂ evolution reaction (OER) cell mediated by the I₃⁻/I⁻ redox shuttle. Specifically, a mixed halide perovskite FAPbBr_{3-x}I_x (FPBI, FA = CH(NH₂)₂⁺; absorption edge 620 nm) loaded with molybdenum selenide (MoSe₂) was developed as the photocatalyst for H₂ production while splitting HI and oxidizing I⁻ to I₃⁻. A NiFe-layered double hydroxide modified BiVO₄ (NiFe-LDH/BiVO₄, absorption edge 520 nm) film grown on an FTO glass was employed for OER via water oxidation, with the linked carbon cloth electrode for I₃⁻ reduction. This design allows for the separate production of H₂ and O₂, completely avoiding back reaction (recombination of H₂ and O₂). Additionally, side reactions caused by the redox couple (the oxidation of I⁻ by the holes in BiVO₄) can be completely suppressed because the OER cell did not contain the redox mediates. Therefore, the system split water with an STH of 2.50%, which is the new record for Z-scheme photocatalytic overall water splitting.

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