Scalable and Efficient Pure-Water Splitting on Particulate Photocatalyst Sheets

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Solar-driven water splitting is a promising approach for producing hydrogen as a clean and sustainable energy carrier. Z-scheme water splitting systems, which employ two different semiconductors for H_2 and O_2 evolution, are capable of utilizing visible light more efficiently than a single-component photocatalyst because the energy required for driving each photocatalyst can be reduced. A major challenge in developing high-performing Z-scheme water splitting systems lies in achieving efficient transfer of electrons between the hydrogen evolution photocatalyst (HEP) and oxygen evolution photocatalyst (OEP). Herein, we report an all-solid-state device for redox-mediator-free Z-scheme water splitting, that is, a photocatalyst sheet consisting of HEP and OEP particles and a conductive layer for efficient electron relay.[1,2]

The $SrTiO_3$:La,Rh/Au/$BiVO_4$:Mo sheets were prepared by a particle transfer method. The SEM-EDX analysis of the photocatalyst sheet prepared revealed that both HEP and OEP particles contacted the gold layer. Thus, electrons could be transferred through the Au between HEP and OEP particles. Simultaneous evolution of H_2 and O_2 occurred at the H_2/O_2 ratio of two under visible light irradiation ($\lambda >$ 420 nm). The enhancement in the electric contact between Au and the semiconductor photocatalysts by annealing boosted the gas evolution rates, confirming the importance of the electron transfer through the Au layer. The $SrTiO_3$:La,Rh/Au/$BiVO_4$:Mo photocatalyst sheet exhibited an AQY of 33% at 419 nm in overall pure water (pH 6.8) splitting at 331 K and 10 kPa. The STH was calculated to be 1.1% on the basis of the average rate of gas evolution over the ten-hour reaction. Nevertheless, the photocatalytic water-splitting activity of the $SrTiO_3$:La,Rh/Au/$BiVO_4$:Mo sheets dramatically decreased with increasing background pressure because of reverse reactions involving molecular oxygen. Hence, surface modification using Cr_2O_3 and amorphous titanium oxide (a-TiO_2) layers are necessary in order to suppress the reverse reactions. Furthermore, a high water-splitting activity at ambient pressure were achieved by utilizing carbon as an oxygen-tolerant conductor layer in the photocatalyst sheet systems.[3] The $SrTiO_3$:La,Rh/C/$BiVO_4$:Mo sheets exhibited a STH of 1.0% during unassisted pure-water (pH 6.8) splitting at 331 K and 91 kPa, as shown in Figure 1, because the sputtered carbon conductor was less active with regard to reverse reactions.

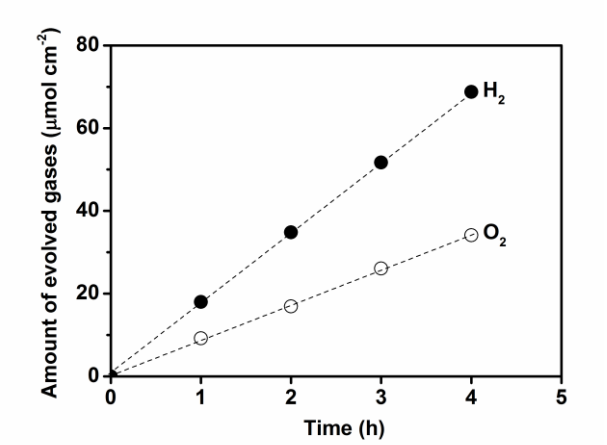

Fig. 1. A time course of water splitting on a $SrTiO_3$:La,Rh/C/$BiVO_4$:Mo photocatalyst sheet modified with Cr_2O_3/Ru under simulated sunlight (AM 1.5G) at 331 K and 91 kPa.

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