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## 2D/3D heterojunction of MAPbI<sub>3</sub>/OA<sub>2</sub>PbI<sub>4</sub> thin films for Photovoltaic Hydrogen Evolution

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Three-dimensional (3D) halide perovskites have emerged as a promising class of inexpensive and easy-to-make semiconductors for photocatalytic application in hydrogen evolution due to their exceptional visible light response. Their high sensitivity to humidity, however, resulted in significant instability issues. This led to the development of 2D/3D halide perovskites (HaP), which showed efficient charge-carrier transport, humidity resistance, and higher stability in ambient conditions. In this study, a pure 3D MAPbI<sub>3</sub>, 2D OA<sub>2</sub>PbI<sub>4</sub>, and 2D/3D OA<sub>x</sub>MA<sub>1-x</sub>PbI<sub>3</sub> HaP were synthesized using the spin-coating method. From the X-ray diffraction (XRD) results, a 3D HaP was confirmed at 14.1° and 28.4°, whilst a 2D HaP depicted its characteristic peaks in the low 2θ area at 5.5°. Also, the 2D/3D HaP powder shows identifiable reflections assigned to the OA<sub>2</sub>PbI<sub>4</sub>. The morphological studies of the 2D, 3D, and 2D/3D powders were studied using a scanning electron microscope (SEM). The HaP materials exhibit particle sizes of 100 nm for 3D, 400 nm for 2D, and around 200 nm for 2D/3D HaP with mixed phases. The pristine 3D nanoparticles with clear grain boundaries have relatively different appearances, while the 2D/3D particles with different cation ratios all show good crystallinity as confirmed by XRD. In contrast to the MAPbI<sub>3</sub>, which exhibits a significant absorption over the entire visible range, the OA<sub>2</sub>PbI<sub>4</sub> exhibits a major absorption peak in the UV spectrum at 375 nm, which is above the visible range. Furthermore, the 2D/3D displays a longer onset than the pristine 3D HaP and similar strong visible light absorption across the entire visible range. Pure 2D perovskite exhibits the lowest photocatalytic hydrogen evolution (PHE) rate of 192.1 mol g<sup>-1</sup> h<sup>-1</sup> while the pristine perovskite exhibits a PHE rate of 323 mol g<sup>-1</sup> h<sup>-1</sup> within 5 hours. The PHE rates are, however, 219.3, 430.2, 960.2, and 274.5 mol g<sup>-1</sup> h<sup>-1</sup> for the 2D/3D HaP with octadecylamine (OA) concentrations of 5, 10, 15, and 25%, respectively. The outcomes show that the 2D/3D perovskite with the ideal OA content of 15% had the best performance. The 2D/3D HaP show the same characteristic peaks before and after the PHE, indicating a high stability of the materials. The transient photocurrent responses (i-t curves) and electrochemical impedance spectroscopy (EIS) of the perovskites coated on fluorine-doped tin oxide (FTO) were performed under illumination with a single wavelength of light at 420 nm. The results showed that the OA<sub>0.15</sub>MA<sub>0.85</sub>PbI<sub>3</sub> exhibits the strongest photocurrent, indicating the most effective electron-hole separation and transportation.

**Apply to be considered for a student ; award (Yes / No)?**

Yes

**Level for award;(Hons, MSc, PhD, N/A)?**

PhD

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