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Hybrid Organic-Inorganic H2 evolving Photocathodes: Understanding the Route towards High Performances Organic Photoelectrochemical Water Splitting

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Direct conversion of solar energy into H_2 fuel at a low cost semiconductor/water junction is still a challenge. Despite its theoretical simplicity, limitations in suitable semiconductors have hindered its development. We present different architectures of hybrid organic-inorganic photocathodes based on semiconducting polymeric absorbers. PVD methods were used to synthetize different inorganic interfacial layers (MoO_3,WO_3,CuI and TiO_2/Pt) and their influence on device performances has been assessed. The relevance of this study can be summarized in few key points: (i) high performances with photocurrents up to 8mA/cm^2 at 0V_{RHE}; (ii) optimal process stability with 100% faradaic efficiency along electrode's lifetime; (iii) excellent energetics with onset potential as high as +0.7V_{RHE}; (iv) promising operational activity of more than 10 hours and (vi) by-design compatibility with a tandem architecture. Collectively, these features establish organic photoelectrochemical systems as promising candidates for efficient solar fuel production. We present a study of different architectures investigating the role of each interface, enlightening their working principles and limiting factors. We show the photocatalityic activity and long-term stability of a catalysed bulk heterojunction and the effect of selective contactsis investigated separately. Introduction of electron selective layers increases the photocurrent response while hole blocking layers shift the onset potential towards positive voltages allowing operation with tandem photoanodes. This work opens the way to a new generation ofdevicesexploitating organic semiconductors for low cost conversion of solar energy into H_2.

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