2-Photon tandem device for water splitting: comparing photocathode first versus photoanode first designs

Within the field of photocatalytic water splitting there are several strategies to achieve the goal of efficient and cheap photocatalytic water splitting. This work examines one particular strategy by focusing on monolithically stacked, two-photon photoelectrochemical cells. The overall aim of the analysis is to compare the relative merits of two fundamentally different designs: one, where the photoanode is the large bandgap material (light-facing side), and the other, where the photocathode is the large bandgap material. Even though the former design is often shown in the literature, the present analysis shows that the latter design has several advantages. This is particularly true when considering designs that incorporate protection layers to protect the photoabsorbers. A high throughput computational screening was used to filter materials databases in search of candidates with the correct properties. These results show that without protective layers there are scarcely any materials which seem viable as photoabsorbers whereas with protection layers there are significantly more candidates. Since the protection layer (and redox catalysts) on the light facing side should not interfere with light absorption, this is the more difficult side to optimize. Nevertheless, by using TiO2 as a transparent cathode protection layer in conjunction with known H2 evolution catalysts, protection is clearly feasible for a large bandgap photocathode. This suggests that there may be promising strategies for photocatalytic water splitting by using a large bandgap photocathode and a low bandgap photoanode with attached protection layers.

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