Protonated imine-linked covalent organic frameworks for photocatalytic hydrogen evolution

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Covalent organic frameworks (COFs) have emerged as an important class of organic semiconductors for the photocatalytic hydrogen evolution reaction (HER). When tuning the photocatalytic activity of COFs, efforts are typically invested on choosing the proper combination of linkers. However, the effect of the linkage on the photocatalytic performance has rarely been studied. Herein, we demonstrate that upon protonation of Schiff-base moieties, donor-acceptor (D-A) type imine-linked COFs can produce hydrogen with a rate as high as 20.7 mmol g^{-1} h^{-1} under visible light irradiation. A significant red shift in light absorbance, largely improved charge separation efficiency, as well as an increase in hydrophilicity triggered by protonation of the Schiff-base moieties in the imine-linked COFs, are responsible for the improved photocatalytic performance. The protonation phenomenon, which has not been investigated within the context of imine COFs photocatalytic HER before, arose as a key aspect to improve the photocatalytic performance. On the other hand, it is also instructive to broaden the application of COFs as semiconductors in light harvest and transformation, such as organic solar cells.