学 位 論 文 の 要 旨

Hollow Multi-shelled Structured TiO_2/$MAPbI_3$ Composites: Preparation and Application in Photocatalytic Hydrogen Evolution

（中空マルチシェル構造 TiO_2/$MAPbI_3$ 複合体：調製と光触媒水素発生への応用）

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Hydrogen with the advantages of clean energy and environmentally friendly has been widely studied. Photocatalytic hydrogen generation is realized by photocatalysts under light illumination. In this thesis, loading narrow bandgap semiconductors as a light sensitizer inside hollow multi-shelled structures (HoMSs) provides a way to improve photocatalytic activity. Design and synthesis of heterogeneous interfaces between TiO_2 hollow multi-shelled structures and $MAPbI_3$ perovskite were carried out in the first. The method of synthesis of hollow multi-shelled structures is the sequential templating approach (STA). The effect of pretreatment of carbonaceous microspheres (CMSs) by ethanol on morphology was studied. With the prolongation of pretreatment time, the pore size of CMSs increases. Turning the adsorption duration time, pretreatment duration time, and heating rate can control the shells number of TiO_2.

Then the $MAPbI_3$/Pt/TiO_2 composites were synthesized and applied in the photocatalytic hydrogen evolution reaction. The impacts of morphology and heterogeneous interfaces of photocatalysts on photocatalytic performance have been studied. Interfacial photogenerated charge separation and transport have been demonstrated as a great impactor on photocatalytic performance. Thanks to the thin shell structure, it can also reduce the transmission distance of the carriers to reduce the charge recombination and improve the charge utilization. As a result, samples of $MAPbI_3$/Pt/triple-shelled TiO_2 hollow structure displayed an H_2 evolution rate of 6856.2 $\mu mol\ h^{-1}\ g^{-1}$ under visible-light illumination, which is much faster than that of Pt/$MAPbI_3$ (268.6 $\mu mol\ h^{-1}\ g^{-1}$).

In the end, to optimize electron transfer from $MAPbI_3$ to TiO_2 surface, the different phase ratios of anatase and rutile were synthesized and used for photocatalytic reaction. The contrast of electrons transport between anatase and rutile was characterized by EIS, transient photocurrent response, and linear sweep voltammetry method. The experimental results prove that electrons transfer in $MAPbI_3$/anatase is faster than $MAPbI_3$/rutile. $MAPbI_3$/anatase possesses smaller transfer resistance and higher transient photocurrent value.