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Electronically Integrated Mesoporous Ag–TiO_2 Nanocomposite Thin films for Efficient Solar Hydrogen Production in Direct Sunlight

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The synthesis of mesoporous TiO_2 by a solution-based assembly process and Ag/TiO_2 nanocomposites is provided. The efficacy of Ag/TiO_2 nanocomposite as photocatalyst in thin-film form is demonstrated for solar hydrogen generation in sunlight. Integration of Ag with TiO_2 dramatically enhanced the H_2 production: with 1 wt% Ag on TiO_2 (TiAg-1), the H_2 yield was observed to be 4.59 mmol h^{-1} g^{-1}, which is 2.3 (30) times larger than 0.5 wt% Ag on TiO_2. TiAg-1 shows 4.3 times higher activity in film form compared with its powder counterpart. High photocatalytic efficiency is attributed to the surface plasmon resonance effect of Ag, electronic integration of Ag with TiO_2, and subsequent valence band broadening, large distribution of Ag nanoparticles and abundant Ag-TiO_2 Schottky junctions, and the later minimizes electron–hole recombination. Interparticle mesoporous network increases necking and the high surface area offers easy accessibility of the reactants to a large number of active sites.

1. Introduction

The concern pertaining to the large use of nonrenewable and diminishing fossil fuels, the associated aggravating pollution issues and global warming have made the researchers worldwide to focus on sustainable energy resource-based process technologies. Among various sustainable energy processes, renewable H_2 production that effected through photocatalytic water splitting with suitable metal or metal oxide deposited semiconductor composite is considered as one of the potential technology.[1] Although TiO_2 is state-of-the-art material for photocatalysis and photovoltaic applications, its large bandgap (3.2 eV) and the high rate of photogenerated electron–hole recombination makes it less effective to use as a visible-light-driven photocatalytic material for practical applications.[2]

Significant efforts have been made over the past few decades to modify TiO_2 by reducing its band gap and suppressing the electron–hole recombination and also improving the charge transfer so as to enhance the visible-light utilization and photocatalytic performance.[3] One of the effective strategies to achieve a high-efficiency electron–hole separation in TiO_2 and extend its light absorption capacity to visible region is to integrate TiO_2 with noble metal nanoparticles (NPs).[4] Especially the metal–semiconductor Schottky heterojunction formed with noble metals, such as Pt, Pd, Au, and Ag, facilitate the generation and easy separation of charge carriers; also surface plasmon resonance (SPR) effect observed with Au and Ag improved the charge transfer efficiency of photogenerated carriers during the photocatalytic reaction.[4] In particular, Ag nanoparticles is considered to be an attractive candidate in designing high-performance visible-light-driven photocatalysts due to its SPR properties in visible-light range, interesting physicochemical properties and lower cost.[5–8]

The SPR effect of Ag nanoparticles is largely explored with Ag/TiO_2 photocatalyst for the decomposition of organic molecules and pollutants removal photocatalytically.[9,10] There are also ambiguous arguments about the nature of active components and factors that are responsible for the enhanced photocatalytic activity of Ag/TiO_2 nanocomposites. The role of Ag and oxygen vacancies in Ag/TiO_2 nanocomposites in the surface modification of TiO_2 and promoting the electron–hole separation as well as conductivity have been investigated both theoretically and experimentally.[10,11] The combined effect of ionic-Ag^+ and metallic Ag species in enhancing the photocatalytic activity of TiO_2 has been reported in the literature.[8] Patra et al. studied the effect of Ag by preparing Ag–Au alloy and integrated with titania to study the photocatalytic properties for hydrogen production under visible light and observed that highest H_2 production with specific alloy composition by optimizing the SPR absorption from both metals.[7] In a separate study, Nanaji et al. found that both doped- and metallic-Ag on TiO_2 contribute for an enhanced photocatalytic activity.[5] It is clear from the above the potential associated with the Ag/TiO_2 photocatalyst;

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