Mesoporous Materials for Catalysis Applications: Water Splitting and Cyclohexene Epoxidation

Abstract

The nanocrystalline mesoporous TiO_2 photocatalyst was successfully prepared by a new surfactant-assisted templating sol-gel process of laurylamine hydrochloride/tetraisopropyl orthotitanate modified with acetylacetone system and applied for photocatalytic H_2 evolution from water splitting reaction. The synthesized mesoporous TiO_2 thermally treated under optimum conditions of 600 °C for 4 h exhibited higher photocatalytic H_2 evolution activity than non-mesoporous commercial TiO_2 powders, approximately four times higher than Ishihara ST-01 and ten times higher than Degussa P-25. In order to improve the photocatalytic activity of the mesoporous TiO_2, loadings of Ni and Pt cocatalysts were performed. The single-step sol-gel (SSSG) process with surfactant template, in which the desired amount of cocatalyst precursors was incorporated into the TiO_2 sol during sol-gel preparation, was proposed for the first time to load the desired cocatalysts onto the mesoporous TiO_2. In case of Ni cocatalyst, the SSSG method was performed in comparison with the conventional incipient wetness impregnation (IWI) method. In case of Pt cocatalyst, in addition to the IWI method, the widely used conventional photochemical deposition (PCD) method was also comparatively investigated. The photocatalytic H_2 evolution results showed that the loaded photocatalysts prepared by the SSSG method provided superior photocatalytic activity than those prepared by the mentioned conventional methods. As the SSSG method was proved to be an efficient synthetic route for loaded photocatalysts, it was expanded to prepare Cu-, Pd-, and Au-loaded mesoporous TiO_2 photocatalysts. The experimental results revealed that these cocatalysts enhanced the photocatalytic activity as well. The surfactant-assisted templating sol-gel process was also used to prepare nanocrystalline mesoporous Ta_2O_5 by controlled hydrolysis and condensation of tantalum pentaethoxide modified with acetylacetone in the presence of laurylamine hydrochloride surfactant aqueous solution. The unloaded mesoporous Ta_2O_5 showed the maximum photocatalytic H_2 evolution activity after being thermally treated at 700 °C. The photocatalytic activity of the mesoporous Ta_2O_5 calcined at the optimum temperature was improved when Ni cocatalyst was loaded by the proposed SSSG method. Moreover, the synthesized nanocrystalline mesoporous TiO_2 was also evaluated its catalytic performance via cyclohexene epoxidation with H_2O_2 in *tert*-butanol. The mesoporous TiO_2 showed both higher cyclohexene conversion and cyclohexene oxide selectivity than non-mesoporous commercial TiO_2 powders, ST-01 and P-25, as well. Aiming to enhance the cyclohexene oxide selectivity, various metal oxide additives were loaded onto the mesoporous TiO_2. RuO_2 was proved to be the best additive for the mesoporous TiO_2, exhibiting cyclohexene selectivity up to 80%. The plausible mechanisms for such the catalytic reaction were also proposed.