

## Coupling of semiconductors for photocatalytic oxidation and CO<sub>2</sub> reduction reactions under visible-light

Hark Jin Kim and Wan In Lee\*

Department of Chemistry, Inha University, Incheon, 402-752, Republic of Korea  
(wanin@inha.ac.kr)

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Removal of environmental pollutants through photocatalytic reaction has drawn increasing attention over the last few decades. Photocatalysts have also been designed and investigated for the purpose of water splitting and CO<sub>2</sub> reduction to generate clean energies. We fabricated several heterojunction structures between TiO<sub>2</sub> and other visible-light absorbing semiconductors, and found that relative energy band locations between TiO<sub>2</sub> and sensitizer are a crucial factor in determining the efficiency of the photocatalytic reactions. First, we investigated several coupled structures of TiO<sub>2</sub> and sensitizers, whose VB are lower than that of TiO<sub>2</sub> (denoted to "type-B heterojunction"). With visible light irradiation, the electrons in the VB of the semiconductor are excited to its CB. Thereby, its VB is rendered partially vacant, and the electrons in the VB of TiO<sub>2</sub> can be transferred to that of the semiconductor, since its VB is located at lower level. As a result, the holes generated in the VB of TiO<sub>2</sub> have sufficient lifetime to initiate the photocatalytic oxidation reactions. Some of the coupled systems exhibited significantly higher photocatalytic efficiency than the typical N-doped TiO<sub>2</sub> in decomposing gaseous 2-propanol and several organic pollutants in aqueous solution. For further enhancement of visible-light catalytic efficiency, we doubly combined the two different sensitizers with TiO<sub>2</sub>. That is, sensitizer-B with lower VB position than that of TiO<sub>2</sub> was designed to be located in the core of the TiO<sub>2</sub> structure, whereas sensitizer-A with higher CB was loaded onto the TiO<sub>2</sub> surface. Under visible-light irradiation, both the electrons and holes are generated in CB and VB of TiO<sub>2</sub>, and these active species induces remarkably high photocatalytic efficiency in evolving CO<sub>2</sub>. Second, we also found that some of the coupled photocatalytic systems can be used for the reduction of CO<sub>2</sub> under visible-light irradiation. We monitored the evolution of methanol and carbon monoxide, and it was also found that relative energy band positions between two semiconductors were critical for the photocatalytic CO<sub>2</sub> reduction reactions.