Computational Materials Science, vol. 138: 462-473

Enhancing photocatalytic activity for hydrogen production and pollutant degradation by modifying tetragonal ZrO_2 with monolayers slab surface of $BiVO_4$, Ag_3PO_4, $SrTiO_3$ and WO_3: A first-principles study

Opoku F
Govender KK
van Sittert CGCE
Govender PP

ABSTRACT:

Semiconductor-based photocatalysis has received increasing attention in energy storage and environmental remediation process due to the abundant solar energy. For this purpose, heterostructures of ZrO_2 coupled with $BiVO_4$, Ag_3PO_4, $SrTiO_3$ and WO_3 monolayers are designed to examine their potential applications in hydrogen production and degradation of pollutants using density functional theory (DFT) + U method for the first time. The results revealed that the calculated band gaps of the heterostructures are reduced compared to the pure ZrO_2, which favour redshift absorption. A type-I band alignment is attained for the $BiVO_4/ZrO_2$, Ag_3PO_4/ZrO_2 and WO_3/ZrO_2 heterostructures. More importantly, a type-II staggered band alignment formed in the $SrTiO_3/ZrO_2$ heterostructure restrained the charge recombination rate of photoinduced charge carriers, as well as enhancing the photocatalytic activity. In particular, suitable band alignment of $SrTiO_3/ZrO_2$ with enough driving forces for charge carrier transfer show overall water splitting and degradation of pollutant in which $SrTiO_3$ acted as charge separation centre. Furthermore, h+, and radicals played a major role in the photocatalysis process of the $SrTiO_3/ZrO_2$ heterostructure. These results reveal that the ZrO_2 acts as an oxidation site so that better access of electron acceptor to the interface is a significant factor that improves the photocatalytic activity of $SrTiO_3/ZrO_2$ heterostructure towards H_2 evolution.