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Various Shaped ZnO and ZnO/Graphene Oxide Nanocomposites and Their Use in Water Splitting Reaction

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Abstract : Exploring strategies for oxygen vacancy engineering under mild conditions and understanding the relationship between dislocations and photoelectrochemical (PEC) cell performance are challenging issues for designing high performance PEC devices. Therefore, it is very important to understand that how the oxygen vacancies (VO) or other defect states affect the performance of the photocatalyst in photoelectric transfer. So far, it has been found that defects in nano or micro crystals can have two possible significances on the PEC performance. Firstly, an electron-hole pair produced at the interface of photoelectrode and electrolyte can recombine at the defect centers under illumination of light, thereby reducing the PEC performances. On the other hand, the defects could lead to a higher light absorption in the longer wavelength region and may act as energy centers for the water splitting reaction that can improve the PEC performances. Even if the dislocation growth of ZnO has been verified by the full density functional theory (DFT) calculations and local density approximation calculations (LDA), it requires further studies to correlate the structures of ZnO and PEC performances. Exploring the hybrid structures composed of graphene oxide (GO) and ZnO nanostructures offer not only the vision of how the complex structure form from a simple starting materials but also the tools to improve PEC performances by understanding the underlying mechanisms of mutual interactions. As there are few studies for the ZnO growth with other materials and the growth mechanism in those cases has not been clearly explored yet, it is very important to understand the fundamental growth process of nanomaterials with the specific materials, so that rational and controllable syntheses of efficient ZnO-based hybrid materials can be designed to prepare nanostructures that can exhibit significant PEC performances. Herein, we fabricated various ZnO nanostructures such as hollow sphere, bucky bowl, nanorod and triangle, investigated their pH dependent growth mechanism, and correlated the PEC performances with them. Especially, the origin of well-controlled dislocation-driven growth and its transformation mechanism of ZnO nanorods to triangles on the GO surface were discussed in detail. Surprisingly, the addition of GO during the synthesis process not only tunes the morphology of ZnO nanocrystals and also creates more oxygen vacancies (oxygen defects) in the lattice of ZnO, which obviously suggest that the oxygen vacancies be created by the redox reaction between GO and ZnO in which the surface oxygen is extracted from the surface of ZnO by the functional groups of GO. On the basis of our experimental and theoretical analysis, the detailed mechanism for the formation of specific structural shapes and oxygen vacancies via dislocation, and its impact in PEC performances are explored. In water splitting performance, the maximum photocurrent density of GO-ZnO triangles was 1.517mA/cm-2 (under UV light ~ 360 nm) vs. RHE with high incident photon to current conversion Efficiency (IPCE) of 10.41%, which is the highest among all samples fabricated in this study and also one of the highest IPCE reported so far obtained from GO-ZnO triangular shaped photocatalyst.

Keywords : dislocation driven growth, zinc oxide, graphene oxide, water splitting
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