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Creation of three-dimensional textured graphene/Si Schottky junction photocathode for enhanced photoelectrochemical efficiency and stability

Abstract
The environmentally friendly technology for clean energy has become the hottest research topic over last decades. Photoelectrochemical (PEC) water-splitting is a promising technique, which directly converts solar to hydrogen energy. Due to its relatively narrow bandgap and mature industry, silicon has successfully been applied in photoelectrode. However, the corrosion in acidic or alkaline electrolyte severely limits Si catalytic efficiency. Compared with the bare p-Si photocathode, graphene/p-Si photocathode is effective to protect the PEC device. To overcome the large reflection of planar Si, textured Si photocathodes, such as pyramids-like Si, can significantly enhance the light harvesting efficiency to produce hydrogen. Herein, we present the three-dimensional graphene/pyramid-like Si photocathode for highly efficient hydrogen generation. Through the flexible and stretchable polymer, ethylene-vinyl acetate (EVA), we successfully transfer graphene onto textured surface etched by KOH. The formation of 3D pyramid-like graphene/p-Si Schottky junctions exhibit enhanced electrochemical activity and promote charge separation efficiency compared with the bare pyramid Si surface without graphene. The inherent chemical inertness of graphene significantly improves the operational stability of 3D graphene/p-Si Schottky junction photoelectrochemical cells. The 3D pyramid-like graphene/p-Si Schottky junction photocathode delivered an onset potential of 0.04 V and a saturated photocurrent density of -37.5 mA/cm² with excellent stability comparable to values reported for textured or nanostructured p-Si photocathodes coated with ultrathin oxide layers by the conventional atomic layer deposition (ALD) technique. Our results suggest that the formation of graphene/Si Schottky junctions with a 3D architecture is a promising approach to improve the performance and durability of Si-based photoelectrochemical systems for water splitting or solar-to-fuel conversion.

References
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Figure 1: The schematic illustration of a novel 3D pyramid-like graphene/p-Si Schottky junctions device in PEC conversion for H$_2$ production.

Figure 2: (a) Cross-sectional scanning TEM images of graphene/p-Si photocathode. (b)-(d) The zoom-in images of the areas labeled as 1,2,3, respectively, in (a). The green lines in (b) indicates the orientation of Si (111) lattice plane.