Title:	Silicon nanowire-Ta2O5-NGQD heterostructure: an efficient photocathode for photoelectrochemical hydrogen evolution.
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Abstract:	Photoelectrochemical (PEC) water splitting has propelled broader research interest for the large-scale and facile entrapment of solar energy in hydrogen fuel. It offers the most favorable and environment-friendly approach to harvest renewable energy under solar radiation for solar-to-hydrogen fuel conversion. However, for superior hydrogen evolution reaction (HER) and enhanced overall efficiency, it is inevitable to design a suitable low-cost, active, scalable and durable photocathode. Although silicon (Si) is the backbone of the photovoltaic industry, an alternate drive is under development to establish its applicability towards PEC-HER. Nevertheless, bare Si with a metal catalyst suffers due to its instability, high reflectance loss, surface oxidation, and sluggish kinetics. These issues can be addressed by passivating the Si surface with the appropriate protection layer anchored with a low-cost metal-free catalyst to design the pertinent photocathode. Herein, vertically aligned p-silicon nanowires (p-SiNWs) with a conformal coating of tantalum pentoxide (Ta2O5) passivation layer and N-doped graphene quantum dots (NGQD) as a metal-free catalyst (p-SiNWs–Ta2O5–NGQD) have been designed, which impart efficient, stable, and scalable photocathodes for PEC-HER. The photocathode exhibits an applied bias photon-to-current conversion efficiency (ABPE) of ~21.1%, which is 6-fold higher than that of the p-SiNWs–NGQD matrix with a low overpotential (−449 mV @ 5 mA cm−2) and Tafel slope (78 mV dec−1) indicating the superiority of the catalyst. The role of Ta2O5 is not only to act as the passivation layer via reducing the lattice mismatch but it also facilitates the charge transfer process from Si to the electrolyte, minimizing the conduction band offset. Moreover, the non-corrosive nature of NGQD, having various N-functionalities, significantly enhances the active site densities and thereby imparts prolonged durability towards the HER performance for at least 10 hours @ 10 mA cm−2 with negligible potential loss. The simplistic and compatible design strategy can promote a new-fangled pathway for the commercialization of large-scale Si-based metal-free photocathodes towards next-generation green fuel technology. Graphical abstract: Silicon nanowire–Ta2O5–NGQD heterostructure: an efficient photocathode for photoelectrochemical hydrogen evolution
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