Three-dimensional graphene networks and reduced

graphene oxide nanosheets co-modified dye-sensitized solar

cells

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1. Calculation of graphene size and defect density

Raman spectroscopy is one of non-destructive tools to obtain the thickness, average size and defect density of the graphene samples. The average size of graphene sheets can be calculated by following equation [1, 2].

$$L_a = 43.5 \times \frac{I_G}{I_D} \tag{1}$$

Defect density can be roughly defined as $(1/L_a)^2$ in cm⁻² [3-5]. Based on the ratio of I_D/I_G of the RGO nanosheets (0.53), the average size is ~80 nm. Similarly, based on the value of I_D/I_G (0.16), the defect densities of the 3DGN is 1.36×10^9 cm⁻².

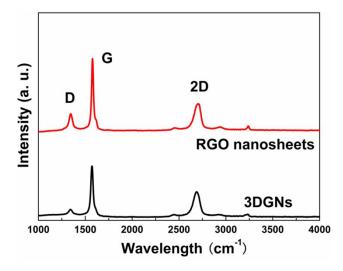


Fig. S1 Raman curves of the adopted RGO nanosheets and 3DGN.

2. Electrical structure of dye molecules, TiO₂, graphene and conductive substrate

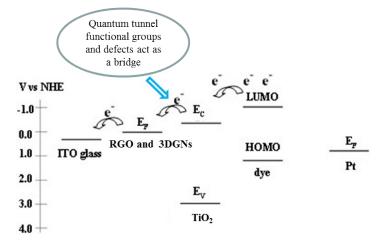


Fig. S2 Schematic diagram of electron transport in the whole photoanode system.

3. Preparation of transport layer

The beforehand RGO nanosheets and TiO₂ nanosheets were handled by LBL selfassembly technology to prepare the transport layer [6, 7]. Briefly, a conductive glass (coating ITO) was immersed into a protonic Polyethyleneimine (PEI) aqueous solution for 15 min to introduce positive charges, and followed by washing with deionized water for 2 times (step 1). Afterwards, the conductive glass was immersed into a colloidal suspension of the TiO₂ nanosheets solution for 15 min and then washed for 2 times (step 2). After that, the sample was immersed into the PEI solution for 15 min and rinsed for 2 times (step 3). The resulted sample was immersed into a graphene nanosheets solution for 15 min and washed for 2 times (step 4). The steps 1-4 were repeated until the desired number of the bi-layer was obtained.

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