Supporting information

Modulation of Interfacial Charge Transfer by Self-assembly of Single-layer Graphene Enwrapped One-dimensional Semiconductors Toward Photoredox Catalysis

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Fig. S1. Zeta potentials of (a) GO and (b) WO₃ NRs aqueous solutions.
Fig. S2. XRD pattern of pristine WO$_3$ NRs.

Fig. S3. XRD patterns of GO and GR.
Fig. S4. High-resolution XPS spectra of (a) W 4f and (b) Ti 2p for pristine WO$_3$ NRs and TNBs, respectively.
Fig. S5. High-resolution C 1s spectra of (a) GO and (b) TNBs/WO$_3$ NRs/GR ternary heterostructure.
**Fig. S6.** FTIR spectra of (a) TNBs, (b) TNBs/WO3 NRs/GR and (c) GO.

**Note:** GO displays several characteristic bands at 1730, 1633, 1401, 1226, and 1058 cm\(^{-1}\). Specifically, the carboxyl group appears at 1730 cm\(^{-1}\) (i.e., C=O stretching vibration)\(^1\) along with the presence of C-O vibration of epoxy groups at 1058 and 857 cm\(^{-1}\).\(^2\) The hydroxyl group (-OH) is located at ca. 3400 and 1401 cm\(^{-1}\), the epoxy group (C-O-C) at 1226 cm\(^{-1}\), and the alkoxy group (C-OH & C-O) at 1050 cm\(^{-1}\).\(^3\) The peak located at 1633 cm\(^{-1}\) is associated with the aromatic C=C bond in GO.\(^4-5\)

**Fig. S7.** (a) High and (b) low-magnified FESEM images of WO3 NRs.
Fig. S8. High-magnified FESEM images of (a) TNBs matrix, (b) TNBs/WO$_3$ NRs and (c) TNBs/GR binary heterostructures.
Fig. S9. (a) AFM image of GO with corresponding height profile in the inset and (b) UV-absorption spectrum of GO aqueous solution with photograph in the inset, (c) FESEM and (d) TEM images of GO.
**Fig. S10.** HRTEM image of TNBs/WO₃ NRs/GR ternary heterostructure with corresponding SAED pattern in the inset.

**Fig. S11.** Variations of TOC percentage as function of irradiation time over different catalysts.
Fig. S12. Survey XPS spectra of TNBs/WO₃ NRs/GR ternary heterostructure before and after cyclic reactions.

![Survey XPS spectra](image)

Fig. S13. High-resolution N 1s spectrum of TNBs/WO₃ NRs/GR ternary heterostructure after cyclic reactions.

![High-resolution N 1s spectrum](image)

**Note:** It is apparent that peak intensity of high-resolution N 1s spectrum of TNBs/WO₃ NRs/GR ternary heterostructure is still rather weak and comparable to that of before cyclic reaction, indicating PDDA layer intercalated within the interfacial region of TNBs and WO₃ NRs was not removed during the photocatalytic reaction and this is beneficial for retaining the structural integrity of the ternary heterostructure.
**Fig. S14.** Control experiments by bubbling N\textsubscript{2} and O\textsubscript{2} gas into the reaction system toward the photocatalytic degradation of RhB over TNBs/WO\textsubscript{3} NRs/GR ternary heterostructure with the same irradiation time of 60 min.

**Experimental section**

**Synthesis of Graphene Oxide (GO)**

GO nanosheets were synthesized from crystalline graphite nanosheets powder (99.5 \%) using a modified Hummers method\textsuperscript{6,7}. In a typical synthesis, 10 g of graphite powder was added to 230 mL of concentrated H\textsubscript{2}SO\textsubscript{4} (98 \%) which was cooled in an ice bath. Following, 30 g of KMnO\textsubscript{4} was added slowly to the above solution with gentle stirring for 10 min. Temperature of the mixed solution was then raised to 35 °C and kept for 2 h to completely oxidize graphite. Afterwards, the mixture was diluted with 460 mL of DI H\textsubscript{2}O and further diluted by 1.4 L of DI H\textsubscript{2}O 5 min later under vigorous stirring. 50 mL of H\textsubscript{2}O\textsubscript{2} was then added to the mixture and a brilliant yellow product was formed along with bubbling. The mixture was filtered and washed by 1 : 10 HCl aqueous solution to remove metal ions and by DI H\textsubscript{2}O to remove residual acid. The filter cake thus-obtained was dispersed in DI H\textsubscript{2}O with a mechanical agitation and centrifuged at 4500 rpm for 2 min and 5 cycles to remove small GO pieces and water-soluble byproducts. The final sediment was dialyzed for two-
weeks to remove impurity atoms and then re-dispersed in DI H₂O with mechanical agitation or mild sonication to result in exfoliated GO aqueous solution.

References