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Supporting Information

Self-assembly of Graphene-Encapsulated Antimony Sulfide Nanocomposites for Photoredox Catalysis: Boosting Charge Transfer via Interface Configuration Modulation

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Experimental section

Synthesis of graphene oxide (GO) by a modified Hummers' method.1-7

GO was synthesized from natural graphite powder by a modified Hummers' method. In detail, 2 g of graphite powder (supplied from Sinopharm Chemical Reagent Co., Ltd., China) was put into a mixture of 12 mL of concentrated H₂SO₄, 2.5 g of K₂S₂O₈, and 2.5 g of P₂O₅. The solution was heated to 80 °C in an oilbath kept stirring for 24 h. The mixture was then carefully diluted with 500 mL of deionized (DI) water, filtered, and washed until the pH of rinse water became neutral. The product was dried under ambient condition overnight. This pre-oxidized graphite was then subjected to oxidation described as follows. In a typical procedure, pre-oxidized graphite powder was added to a mixture of 120 mL of concentrated H₂SO₄ and 30 mL HNO₃ under vigorous stirring, and the solution was cold to 0 °C. Then, 15 g of KMnO₄ was added gradually under stirring and the temperature of the mixture was kept to be below 20 °C by cooling. Successively, the mixture was stirred at room temperature for 96 h, and then diluted with 1 L of DI water in an ice bath to keep the temperature below 50 °C for 2 h. Shortly after the further diluted with 1 L of DI water, 20 mL of 30% H₂O₂ was then added to the mixture and a brilliant yellow product was formed along with bubbling. The mixture was filtered and washed with 1:10 HCl aqueous solution to remove metal ions followed by DI water to remove the acid. The filter cake was then dispersed in water by a mechanical agitation. Lowspeed centrifugation was done at 1000 rpm for 2 min. The supernatant then underwent two more high-speed centrifugation steps at 8000 rpm for 15 min to remove small GO pieces and water-soluble byproduct. The final sediment was redispersed in water with mechanical agitation or mild sonication using a table-top ultrasonic cleaner, giving a solution of exfoliated GO.



Scheme S1. Schematic flowchart for fabrication of Sb₂S₃ NRs-GR⁻ nanocomposites.



Fig. S1. Zeta potentials of (a) GO^{-} and (b) surface-modified GO^{+} aqueous solutions as a function of pH.



Fig. S2. UV-vis absorption spectra of (a) GO^- and (b) surface-modified GO^+ aqueous solutions.



Fig. S3. X-ray diffraction patterns (XRD) of Graphite and GO.



Fig. S4. High-resolution C 1s spectrum of GO.



Fig. S5. High-resolution N 1s spectra of (a) Sb₂S₃ NRs-3%GR⁺ and (b) blank Sb₂S₃ NRs.



Fig. S6. FTIR spectra of GO^- and surface-modified GO^+ aqueous solutions.



Fig. S7. FESEM, TEM and AFM images of (a, c, e) GO⁻ and (b, d, f) surface-modified GO⁺.



Fig. S8. Elemental mapping results of blank Sb_2S_3 NRs for (b) Sb and (c) S signals from (a); elemental mapping results of Sb_2S_3 NRs-3%GR⁻ for (e) Sb, (f) S and (g) C signals from (d); EDX results of (h) Sb_2S_3 NRs-3%GR⁻ and (i) Sb_2S_3 NRs.



Fig. S9. EDX result of Sb₂S₃ NRs-3%GR⁺ nanocomposite.



Fig. S10. TEM image of Sb₂S₃ NRs-3%GR⁺ nanocomposite.



Fig. S11. PL spectra of (a) blank Sb₂S₃ NRs, (b) Sb₂S₃ NRs-3%GR⁻ and (c) Sb₂S₃ NRs-3%GR⁺ nanocomposites as a function of irradiation time with an excitation wavelength of 312 nm using TA as a probe molecule under visible light irradiation (λ >420 nm).

Element	Sb ₂ S ₃ NRs	Sb2S3 NRs-3% GR ⁺	Chemical Bond Species
C 1s A	284.5	284.6	C-C, C=C & C-H ⁸
C 1s B	285.9	286.3	С-ОН
C 1s C	286.9	287.7	C-O-C & C=O
C 1s D	287.3	288.8	OH-C=O
Sb 3d5/2	529.6	529.3	${ m Sb}^{3+9,10}$
Sb 3d _{3/2}	539.0	538.6	Sb ³⁺
S 2p _{3/2}	161.5	161.2	S ^{2-9,10}
S 2p _{1/2}	162.6	162.5	S ²⁻
N 1s	N.D.	400.1	C-N ^{11,12}

 Table S1. Chemical bond species vs. B.E. for different samples.

N. D.: Not Detected.

 Table S2.
 Information obtained from the BET measurements.

Samples	Specific Surface Area (m²/g)	Pore Volume (cm³/g)	Pore Size (nm)
Sb ₂ S ₃ NRs	6.6711	0.008297	4.97501
Sb ₂ S ₃ NRs-3% GR ⁻	10.0460	0.008297	5.74458
Sb ₂ S ₃ NRs-3% GR ⁺	18.9763	0.033158	6.98926

 $\label{eq:stables} \begin{array}{l} \mbox{Table S3. Peak positions along with the corresponding functional groups for blank Sb_2S_3 NRs, Sb_2S_3 NRs-3%GR^+$ nanocomposites. \\ \end{array}$

Peak position (cm ⁻¹)	Vibration mode
733	Sb-S
1380	СООН
1470	СООН
1650	СООН
2850	CH ₂
2920	CH ₂

Table S4. Peak positions along with the corresponding functional groups for GO⁻ and surface-modified GO⁺.

Peak position (cm ⁻¹)	Vibration mode
857	C-0
1057	С-ОН & С-О
1226	C-O-C
1626	C=C
1732	C=O
2851	CH ₂
2919	CH_2
3437	О-Н

Table S5. Kinetic rate constants of different samples toward photoreduction of Cr (VI) under visible lightirradiation (λ >420 nm).

Kinetic	No	In dark	Sb ₂ S ₃						
rate	catalyst		NRs	NRs-1%	NRs-3%	NRs-5%	NRs-1%	NRs-3%	NRs-5%
(min ⁻¹)				GR ⁻	GR ⁻	GR ⁻	GR ⁺	GR ⁺	GR ⁺
Cr (VI)	0	0	0.6908	0.1088	0.1427	0.1542	0.3772	0.9210	0.3650

Table S6. Kinetic rate constants of different samples toward mineralization of MO under visible lightirradiation (λ >420 nm).

Kinetic rate (min ⁻¹)	No catalyst	In dark	Sb ₂ S ₃ NRs	Sb ₂ S ₃ NRs-3% GR ⁻	Sb ₂ S ₃ NRs-3% GR ⁺
МО	0	0	0.0206	0.0351	0.0408

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