Supporting Information

Simultaneous precipitation and discharge plasma processing for one-step synthesis of α-Fe_2O_3-Fe_3O_4/graphene visible light magnetically separable photocatalysts

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Catalyst recycling process

After the photocatalyst experiment, the material was maintained in the vial for 30 min. Then, an external magnet was applied to attract the material aside as shown in figure S1(a). The mixing Solution in the vial was transferred to further treatment and absolute ethanol was dropped into the vial. The as-used material was sonicated for 5 minutes to become homogeneous and transferred to a flask. As-used material was washed at least 3 times with absolute ethanol and DI water with the collection by a magnet at the bottom of the flask (Figure S1(b)). The catalyst material was dried at 50 ^{o}C for 5 hours. The recycled material was used for the next photocatalyst experiment.

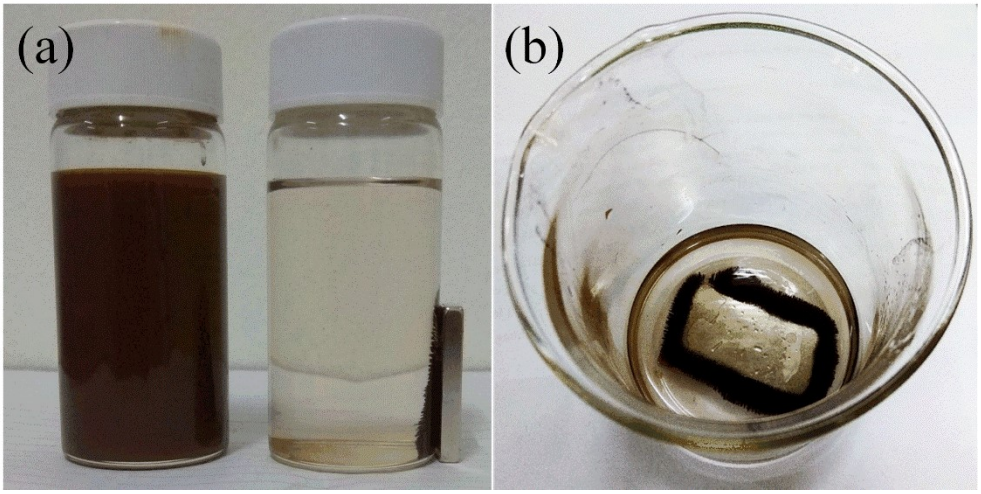

Figure S1. (a) The magnetic recovery by applying an external magnet in the (a) solution and (b) after annealing process

The amount of Fe was determined by mixing 2 mg of GF with a dilute solution of 1 mL HCl and 3 mL HNO_3. The mixture was shaken until the Fe_xO_y is completely dissolved. The concentration of Fe in the solution was measured 3 times by using an Atomic Absorption Spectrophotometer (AAS- PinAAcle 900T PerkinElmer). The amount of iron in each photocatalysis experiment time is determined by 64.8 wt% through the following table:

Table S1. Fe amount determined by AAS

Times	C_0 (mg/L)	Fe (mg)	Fe (wt%)
1st	844.75	12.67	63.4
2nd	848.25	12.72	63.6
3rd	897	13.46	67.3
Mean	863.3	12.95	64.8

We determined the leaching of Fe in the material after each recycling cycle. At each recycling cycle, 4 mL of the photocatalyst solution and 1 mL of a mixture of dilute acidic solution ($HCl:HNO_3$ = 3:1) were filtered through a PVDF membrane with a pore size of 0.2 μm. After each photocatalytic cycle, the mass of leaching iron was calculated the following formula:

$$m_{Fe} = \frac{5C}{4}.\frac{150}{1000}(mg)$$

Table S2. The iron leaching after each photocatalytic cycle

Cycles	Fe concentration (C) (mg/L)	Mass of leaching Fe (mg)	Ratio (wt %)	Photocatalytic efficiency
1	0.102	0.019	0.15	91%
2	0.127	0.024	0.18	89.7%
3	0.106	0.020	0.15	87.6%
4	0.109	0.021	0.16	85%
5	0.14	0.026	0.20	84%

Table S2 shows the ratio of iron leaching after each photocatalytic cycle. Obviously, the amount of iron leaching after each photocatalytic cycle is less than 0.2% related to the stability of

graphene/Fe_xO_y composite through a photocatalyst process. A minor amount of iron leached could be attributed to the material adhesion or to the material washing during recovery step.

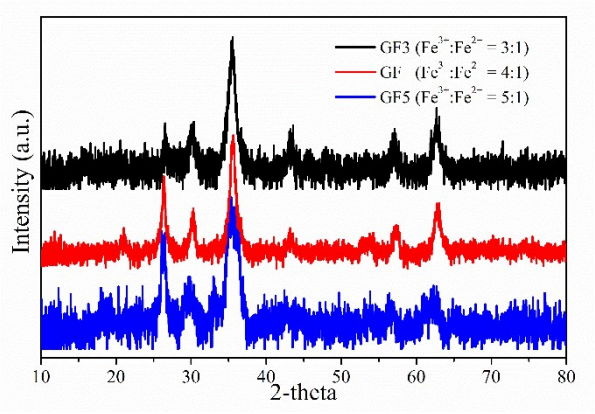

Figure S2. XRD pattern of GF with different initial precursor ratios

To investigate the influence of the Fe^{3+}/Fe^{2+} ratio on the photocatalytic properties of the materials, different initial precursor ratios of 3:1, 4:1, and 5:1 were performed by denoted as GF3, GF, and GF5, respectively. Figure S2 displays the diffraction pattern of GF3, GF, and GF5. The typical peaks of Fe_3O_4 were clearly observed in these samples. However, the typical peak of graphene locates at 26^0 in the GF and GF5 samples are stronger than that of GF3. This result can attribute to the stronger cathodic plasma energy of higher Fe ion concentration. Besides, there are no any others special peak appearances in both 3 sample which confirmed the purity of these samples.

Table S3. Photodegradation comparison with different Fe^{3+}/Fe^{2+} ratio

Samples	C/C_0	Efficiency
GF3 (Fe^{3+}/Fe^{2+} = 3:1)	0.17	83%
GF (Fe^{3+}/Fe^{2+} = 4:1)	0.09	91%
GF5 (Fe^{3+}/Fe^{2+} = 5:1)	0.12	88%

The photocatalytic experiment of these samples was performed as shown in table S3. GF sample presents a photodegradation performance of 91% slighter higher than those of GF3 (83%) and GF5 (88%).