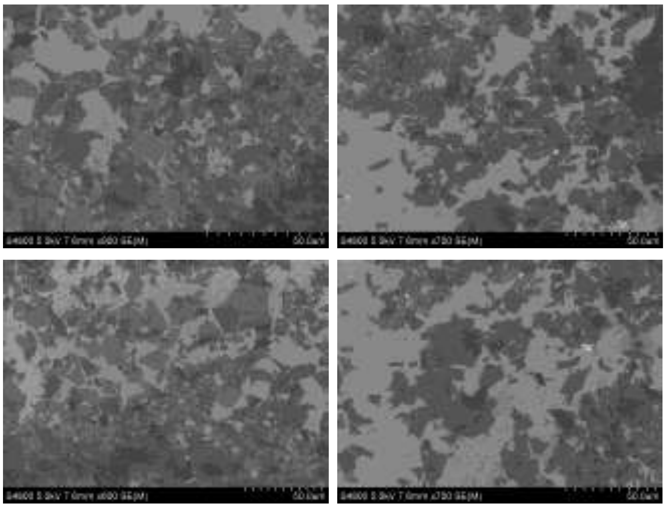
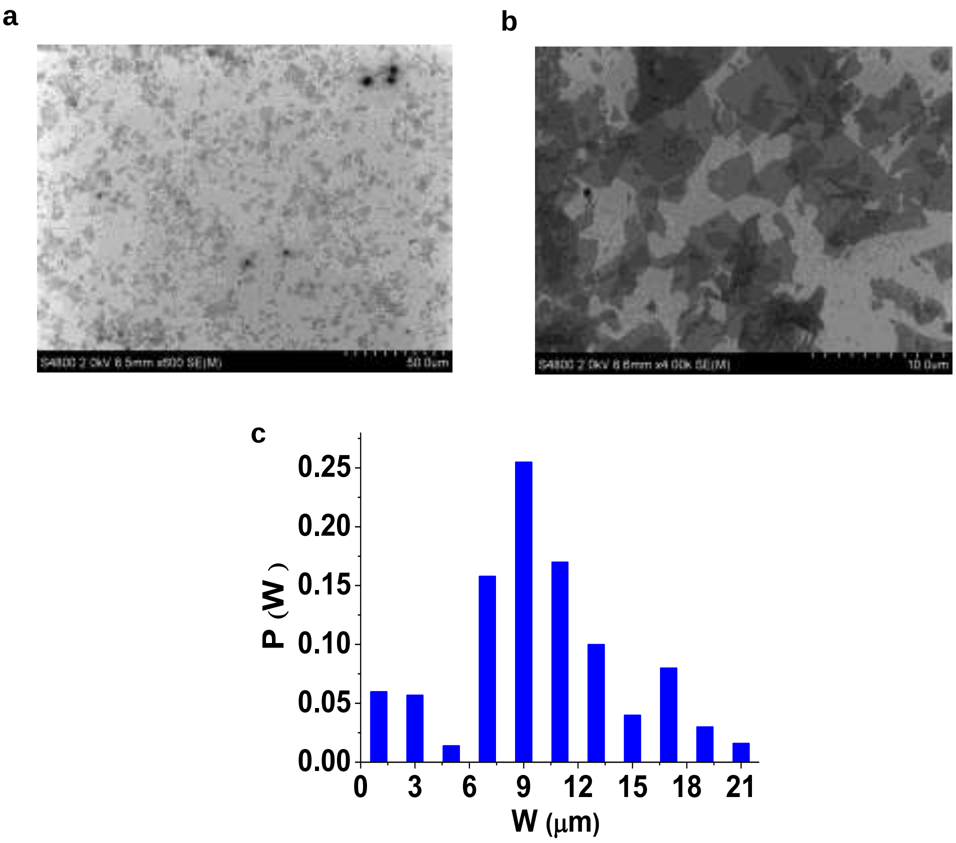
Supplementary Figures

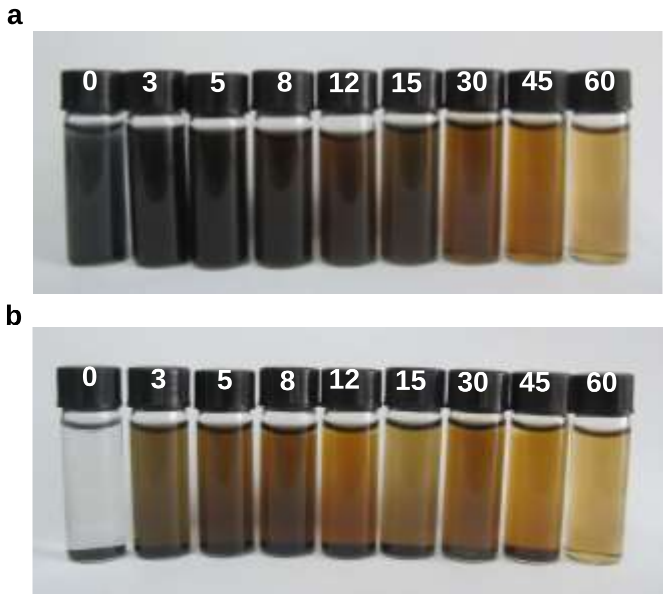
Supplementary Figure 1 | Picture of reactor (20 L) for the scalable preparation of GO^{Fe}.



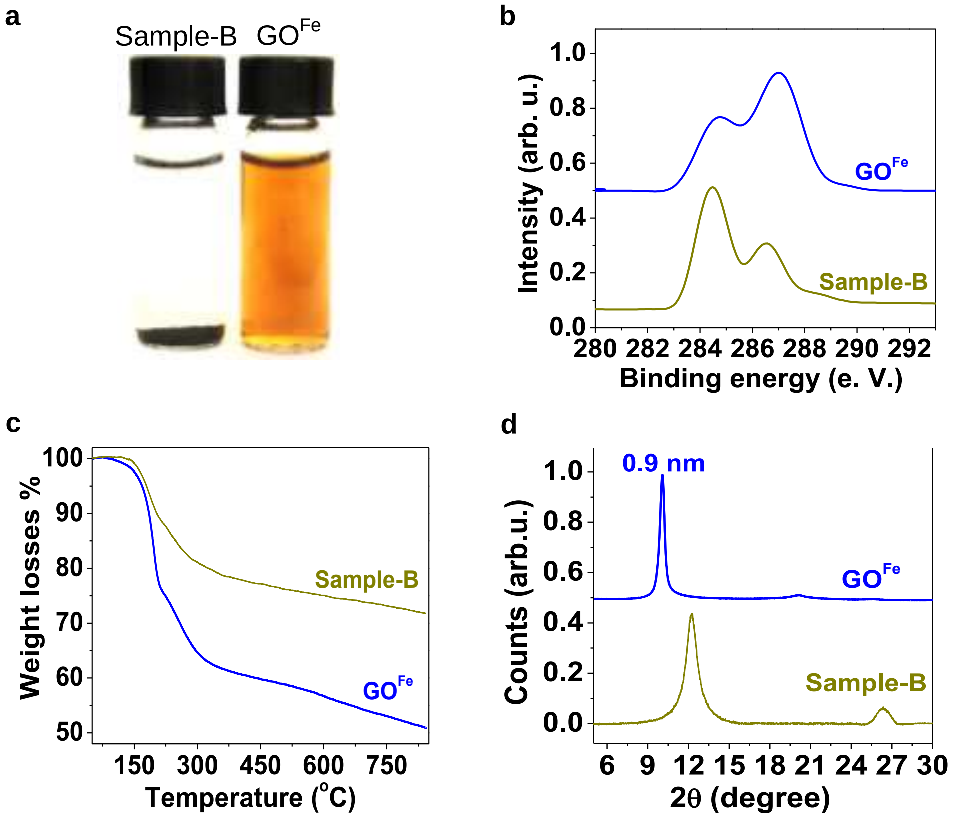
Supplementary Figure 2 | SEM images of GO^{Fe} sheets.



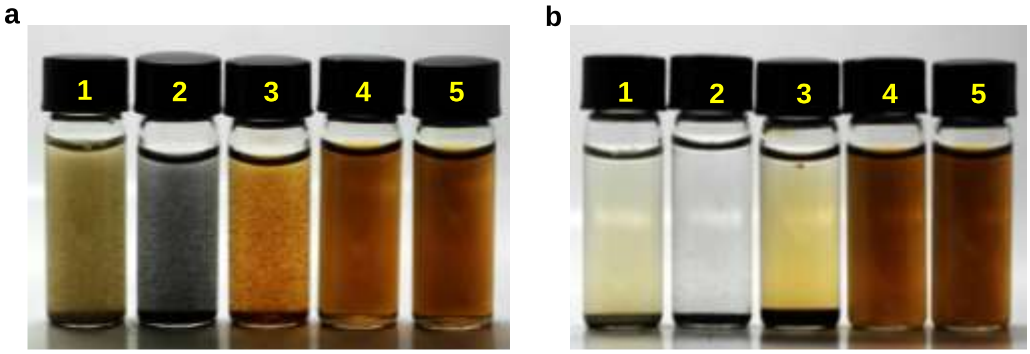
Supplementary Figure 3 | (**a, b**) SEM images of GO^{Mn} on Si/SiO_2 substrate. (**c**) Size distribution of GO^{Mn}, counted and calculated from (**a, b**).



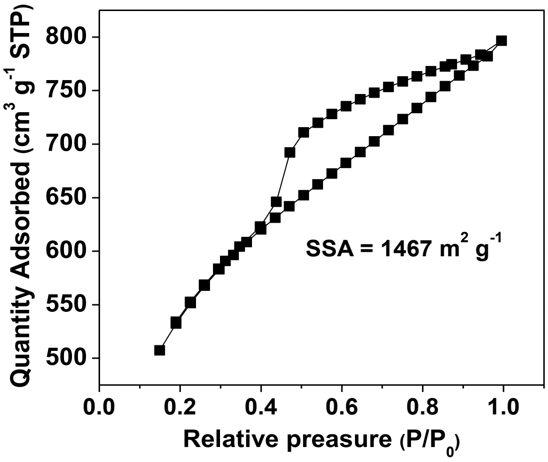
Supplementary Figure 4 | (**a**) Graphite oxide dispersions after 0 min, 3 min, 5 min, 8 min, 12 min, 15 min, 30 min, 45 min and 60 min reaction with a concentration of 0.5 mg mL^{-1}. (**b**) Counterparts of (**a**) after staying for 24 h.



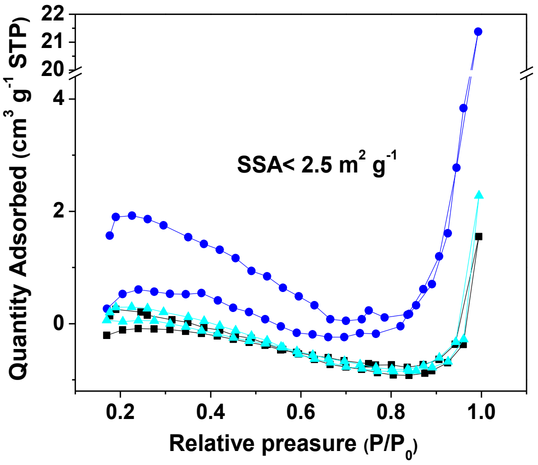
Supplementary Figure 5 | Comparison of GO^{Fe} and Sample-B ($KClO_3$ oxidant for 1 h reaction). (**a**) Photograph of GO^{Fe} and Sample-B placed in water after staying for 2 h. (**b**) C1s XPS spectra, (**c**) TGA plots, and (**d**) XRD spectra of GO^{Fe} and Sample-B.



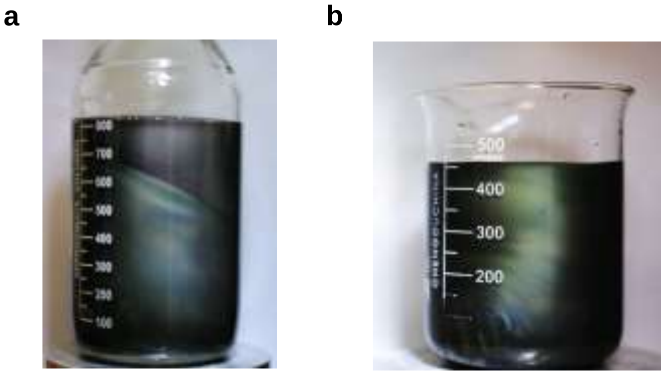
Supplementary Figure 6 | (**a**) Re-dispersed GO in water from commercial dried GO powders (**1**, **2**, **3**) after ultrasound for 12 h, our spray-dried GO^{Fe} powders (**4**) and the fresh GO^{Fe} solution (**5**). (**b**) Counterparts of (**a**) after staying for 20 minutes.



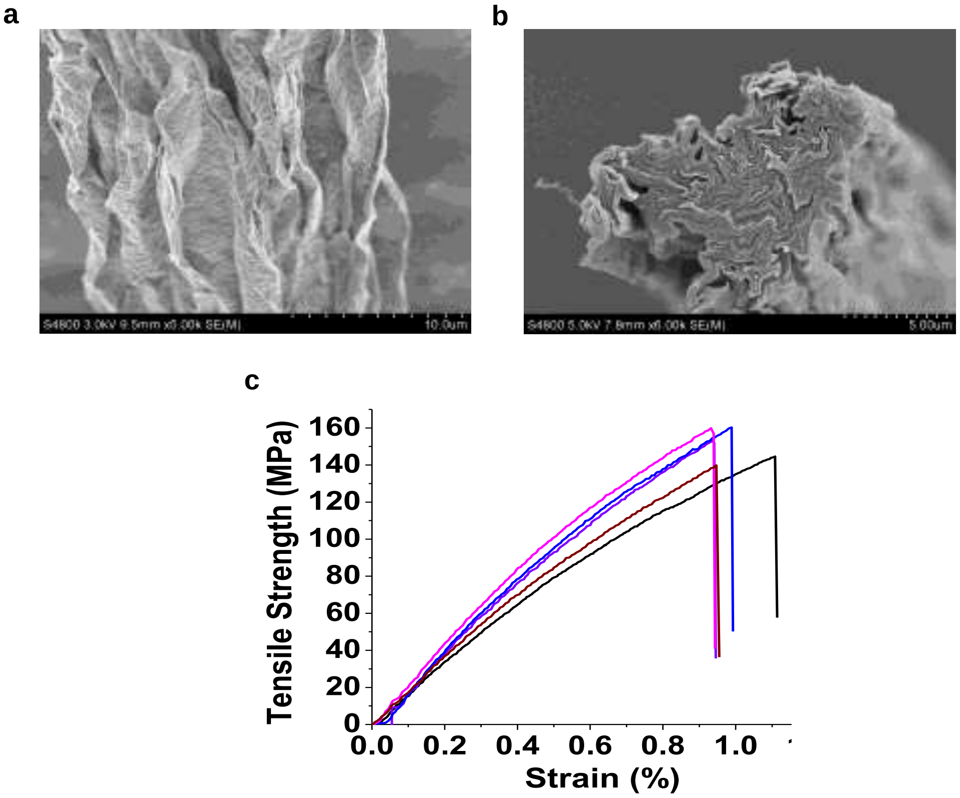
Supplementary Figure 7 | Nitrogen adsorption/desorption isotherm for the GOFe powders.



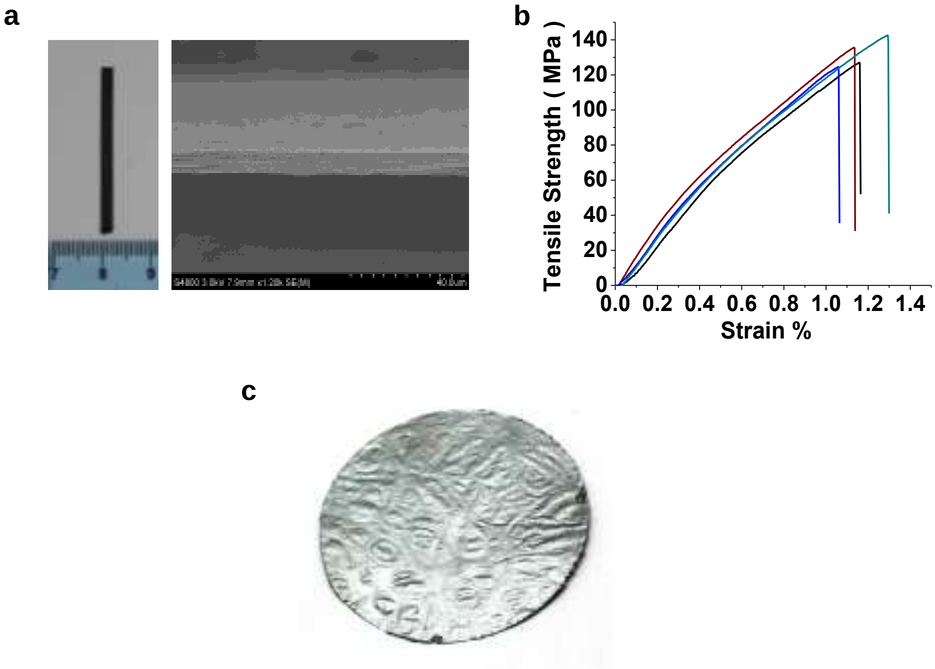
Supplementary Figure 8 | Nitrogen adsorption/desorption isotherm for three commercial dried GO powders.



Supplementary Figure 9 | (**a**) GO^{Fe} solution and (**b**) re-dissolved GO^{Fe} solution with a concentration of 6 mg mL^{-1}, indicating macroscopic liquid crystalline fluid for the whole solution.



Supplementary Figure 10 | (**a, b**) SEM images of GO^{Fe} fibre and (**c**) its strength-strain curve.



Supplementary Figure 11 | (**a**) The sample of GO^{Fe} film for the tensile tests. (**b**) Strength-strain curve of GO^{Fe} film. (**c**) Silver-like GO^{Fe} film reduced by HI.

Supplementary Figure 12 | (**a**) Aerogel without reduction prepared by freeze-drying aqueous solution of CNTs and GO^{Fe} sheets. (**b**) GO^{Fe} aerogel reduced by N_2H_4 and the demonstration of its excellent elasticity after compression.

Supplementary Tables

Ref.	Method	Size of graphite	Preparation steps (Sonication)	Reaction temperature	Ingredients for 1g graphite	Reaction time
Our work	K_2FeO_4-based method	40 µm (1g)	1	r. t.	40 mL H_2SO_4 6 g K_2FeO_4	1h
1[1]	Staudenmaier method	45 µm (1 g)	1	r. t.	17.5 mL H_2SO_4 9 mL $fHNO_3$ 11 g $KClO_3$	>96 h
2[2]	Staudenmaier method	45 µm (1 g)	1	r. t.	17.5 mL H_2SO_4 9 mL HNO_3 11 g $KClO_3$	>96 h
3[3]	modified Hummers method	4 µm (1 g)	1 (Y)	98 °C	82 mL H_2SO_4 0.75 g $NaNO_3$ 4.5 g $KMnO_4$ 3 mL H_2O_2	>120 h
4[4]	modified Hummer's method	1 g	1	105 ºC	50 g NaCl 23 mL H_2SO_4 6 g $KMnO_4$ 10 mL H_2O_2	>14.5 h
5[5]	modified Hummers method	45 µm (12 g)	2	80 ºC	44 mL H_2SO_4 0.83 g $K_2S_2O_8$ 0.83 g P_2O_5 5 g $KMnO_4$ 4.2 mL H_2O_2	>6.5 h
6[6]	modified Hummers method	150 µm (3 g)	1	50 °C	120 mL H_2SO_4 13.3 mL H_3PO_4 6 g $KMnO_4$ 1 mL H_2O_2	>12 h

7[7]	modified Hummers method	250 µm (1 g)	1	r. t.	17.5 mL H_2SO_4 9 mL HNO_3 11 g $KClO_3$	>120 h
8[8]	modified Hummers and Offeman’s method	45 µm (3 g)	1 (Y)	80 °C	40 mL H_2SO_4 8.3 g $K_2S_2O_8$ 8.3 g P_2O_5	>6 h
9[9]	—	5 g	1 (Y)	40 °C	18 mL H_2SO_4 6 mL HNO_3	>96 h
10[10]	modified Hummers method	49 µm (5 g)	1	98 °C	0.5 g $NaNO_3$ 24 mL H_2SO_4 3 g $KMnO_4$ 10 mL H_2O_2	>32 h
11[11]	modified Hummers method	45 µm (20 g)	1	35 °C	24.5 mL H_2SO_4 0.5 g $K_2S_2O_8$ 0.5 g P_2O_5 3 g $KMnO_4$ 2.5 mL H_2O_2	>16 h
12[12]	modified Brodie method	74 µm (1 g)	1 (Y)	—	8.5 g $NaClO_3$ 20 mL fHNO_3	>24 h
13[13]	modified Hummers method	5 g	1	—	0.5 g $NaNO_3$ 13 mL H_2SO_4 3 g $KMnO_4$ 10 mL H_2O_2	>8 h
14[14]	modified Hummers method	1 g	1 (Y)	40 °C	20 g NaCl 0.1 g $NaNO_3$ 23 mL H_2SO_4 0.5 g $KMnO_4$ 10 mL H_2O_2	>26 h
15[15]	Hummers method	4 g	1 (Y)	r. t.	29 mL H_2SO_4 2 g $K_2S_2O_8$ 2 g P_2O_5 3 g $KMnO_4$ 0.5 g $NaNO_3$ 2.5 mL H_2O_2	>8 h

16[16]	modified Hummers method	>150 µm (15 g)	1 (Y)	r. t.	0.5 g $NaNO_3$ 23.3 mL H_2SO_4 3 g $KMnO_4$ 10 mL H_2O_2	21 h
17[17]	Staudenmaier method	5 g	1	r. t.	17.5 mL H_2SO_4 95 mL $fHNO_3$ 11 g $KClO_3$	>96 h
18[18]	Hummers method	45 µm (4 g)	1	80 °C	82.5 mL H_2SO_4 1.5 g $K_2S_2O_8$ 1.5 g P_2O_5 8.75 g $KMnO_4$ 25 mL H_2O_2	>10 h
19[19]	modified Hummers method	1 g	1	40 °C	50 g NaCl 23 mL H_2SO_4 0.75 g $KMnO_4$ 10 mL H_2O_2 0.1 g $NaNO_3$	>25 h
20[20]	modified Hummers method	4 µm (1 g)	1	98 °C	0.75 g $NaNO_3$ 82 mL H_2SO_4 4.5 g $KMnO_4$ 6 mL H_2O_2	>120 h
21[21]	modified Hummers method	12 g	1	80 °C	42.5 mL H_2SO_4 0.83 g $K_2S_2O_8$ 0.83 g P_2O_5 5 g $KMnO_4$ 4.2 mL H_2O_2	>10 h
22[22]	modified Hummers method	1 g	1 (Y)	80 °C	24.5 mL H_2SO_4 0.5 g $K_2S_2O_8$ 0.5 g P_2O_5 3 g $KMnO_4$ 2 mL H_2O_2	>8 h

23[23]	Staudenmaier method	5 g	1	r. t.	17.5 ml H_2SO_4 9 mL HNO_3 11 g $KClO_3$	>96 h
24[24]	Hummers method	45 µm (20 g)	1	80 °C	24.5 mL H_2SO_4 0.5 g $K_2S_2O_8$ 0.5 g P_2O_5 3 g $KMnO_4$ 2.5 mL H_2O_2	>8 h
25[25]	Hummers method	45 µm (2 g)	1	80 °C	6 mL H_2SO_4 1.5 g $K_2S_2O_8$ 1.5 g P_2O_5 12.5 g $KMnO_4$ 19.5 mL H_2O_2	>9 h
26[26]	modified Hummers method	1 g	1	98 °C	0.75 g $NaNO_3$ 85 mL H_2SO_4 6 g $KMnO_4$ 6 mL H_2O_2	>168 h
27[27]	modified Hummers method	4 µm (5 g)	1	98 °C	0.75 g $NaNO_3$ 82 mL H_2SO_4 4.5 g $KMnO_4$ 6 mL H_2O_2	>120 h
28[28]	modified Hummers method	4 µm (1 g)	1	98 °C	0.75 g $NaNO_3$ 82 mL H_2SO_4 4.5 g $KMnO_4$ 6 mL H_2O_2	>120 h
29[29]	modified Hummers method	0.3 g	1	80 °C	48 mL H_2SO_4 1.67 g $K_2S_2O_8$ 1.67 g P_2O_5 5 g $KMnO_4$ 6.7 mL H_2O_2	>8.5 h

30[30]	Hummers method	45 μm (20 g)	1	35 °C	24.5 mL H_2SO_4 0.5 g $K_2S_2O_8$ 0.5 g P_2O_5 3 g $KMnO_4$ 2.5 mL H_2O_2	>8 h
31[31]	modified Hummers method	1 g	1	70 °C	40 g NaCl 23 mL H_2SO_4 0.5 g $KMnO_4$ 0.1 g $NaNO_3$ 10 mL H_2O_2	>25 h
32[32]	modified Hummers method	45 μm (6 g)	1	90 °C	42.5 mL H_2SO_4 0.83 g $K_2S_2O_8$ 0.83 g P_2O_5 5 g $KMnO_4$ 4.2 mL H_2O_2	8.5 h
33[33]	modified Hummers method	2 g	1	35 °C	48 mL H_2SO_4 0.5 g $NaNO_3$ 3 $KMnO_4$ 2.5 mL H_2O_2	>18 h
34[34]	modified Hummers method	1 g	1	70 °C	23 mL H_2SO_4 0.1 g $NaNO_3$ 3 g $KMnO_4$ 10 mL H_2O_2 50 g NaCl	>24 h
35[35]	modified Hummers method	0.3 g	1	80 °C	48 mL H_2SO_4 1.67 g $K_2S_2O_8$ 1.67 g P_2O_5 5 g $KMnO_4$ 6.7 mL H_2O_2	8.5 h
36[36]	modified Hummers method	24 μm (10 g)	1	r. t.	62.1 g H_2SO_4 0.75 g $NaNO_3$ 4.5 g $KMnO_4$ 3 g H_2O_2	>120 h

37[37]	modified Hummers method	0.3 g	1	80 °C	48 mL H_2SO_4 1.67 g $K_2S_2O_8$ 1.67 g P_2O_5 5 g $KMnO_4$ 6.7 mL H_2O_2	8.5 h
38[38]	modified Hummers method	45 µm (5 g)	1	r. t.	36 mL H_2SO_4 12 mL fHNO_3 5 g $KMnO_4$ 6 mL H_2O_2	>120 h
39[39]	modified Hummers method	0.2 g	1	r. t.	80 mL H_2SO_4 0.875 g $NaNO_3$ 4.5 g $KMnO_4$ 3 mL H_2O_2	>120 h
40[40]	Hummers method	0.15 g	1	80 °C	867 mL H_2SO_4 33.3 g $K_2S_2O_8$ 33.3 g P_2O_5 400 g $KMnO_4$ 333 mL H_2O_2	>9.5 h
41[41]	modified Hummers method	3 g	1	80 °C	44 mL H_2SO_4 0.83 g $K_2S_2O_8$ 0.83 g P_2O_5 5 g $KMnO_4$ 6.7 mL H_2O_2	6.5 h
42[42]	modified Hummers method	24 µm (10 g)	1	r. t.	62.1 g H_2SO_4 0.75 g $NaNO_3$ 4.5 g $KMnO_4$ 3 g H_2O_2	>120 h
43[43]	modified Hummers method	45 µm (3 g)	1	80 °C	44 mL H_2SO_4 0.83 g $K_2S_2O_8$ 0.83 g P_2O_5 5 g $KMnO_4$ 6.7 mL H_2O_2	8.5 h

44[44]	modified Hummers method	3000-5000 μm (2 g)	1 (Y)	80 °C	66 mL H_2SO_4 7.5 g $KMnO_4$ 10 mL H_2O_2	8.5 h
45[45]	modified Hummers method	45 μm (3 g)	2	80 °C	44 mL H_2SO_4 0.83 g $K_2S_2O_8$ 0.83 g P_2O_5 5 g $KMnO_4$ 6.7 mL H_2O_2	>8.5 h
46[46]	modified Hummers method	1 g	1	98 °C	46 mL H_2SO_4 1 g NaNO3 6 g $KMnO_4$ 20 mL H_2O_2	6.25 h
47[47]	modified Hummers method	1.5 g	1	90 °C	30 mL H_2SO_4 0.67 g $NaNO_3$ 4 g $KMnO_4$ 6.7 mL H_2O_2	>23 h
48[48]	modified Hummers method	5 g	1	r. t.	38.8 mL H_2SO_4 0.9 g KNO_3 4.5 g $KMnO_4$ 3 g H_2O_2	>120 h
49[49]	modified Hummers method	5 g	1	r. t	38.8 mL H_2SO_4 0.76 g $NaNO_3$ 4.5 g $KMnO_4$ 3 g H_2O_2	>120 h
50[50]	modified Hummers method	< 20 μm (1 g)	1	80°C	98 mL H_2SO_4 2 g $K_2S_2O_8$ 2 g P_2O_5 15 g $KMnO_4$ 10 mL H_2O_2	8.25 h
51[51]	modified Hummers method	1.5 g	1	80°C	46.7 mL H_2SO_4 0.83 g $K_2S_2O_8$ 0.83 g P_2O_5 5 g $KMnO_4$ 6.7 mL H_2O_2	6.5 h

52[52]	modified Hummers method	4 g	1	80°C	29 mL H_2SO_4 2 g $K_2S_2O_8$ 2 g P_2O_5 3 g $KMnO_4$ 2.5 mL H_2O_2 0.5g NaNO3	>13 h
53[53]	modified Hummers method	0.3 g	1	80°C	48 mL H_2SO_4 1.67 g $K_2S_2O_8$ 1.67 g P_2O_5 5 g $KMnO_4$ 6.7 mL H_2O_2	8.5 h
54[54]	Hummers method	3 g	2	98°C	12.3 mL H_2SO_4 0.83 g $K_2S_2O_8$ 0.83 g P_2O_5 1 g $KMnO_4$ 0.17 g $NaNO_3$ 1 mL H_2O_2	>60 h
55[55]	Hummers method	1 g	2	80°C	24.5 mL H_2SO_4 0.5 g $K_2S_2O_8$ 0.5 g P_2O_5 3 g $KMnO_4$ 2 mL H_2O_2	>8 h
56[56]	modified Hummers method	0.3 g	1	80°C	48 mL H_2SO_4 1.67 g $K_2S_2O_8$ 1.67 g P_2O_5 5 g $KMnO_4$ 6.7 mL H_2O_2	8.5 h
57[57]	modified Hummers method	3 g	1	80°C	44 mL H_2SO_4 0.83 g $K_2S_2O_8$ 0.83 g P_2O_5 5 g $KMnO_4$ 6.7 mL H_2O_2	8.5 h

The methods come from the most cited papers describing slGO obtained by the oxidation-exfoliation method.

Supplementary Table 1｜Oxidation exfoliation methods to obtain slGO.

Supplementary Methods

Experimental materials.

Natural graphite flakes (40 μ m) were purchased from Qingdao Henglide Graphite Co., Ltd. K_2FeO_4 was obtained from Hubei CSW-China Chemistry Co., Ltd. All the other reagents were purchased from Sinopharm Chemical Reagent Co., Ltd. and used as received.

Scalable preparation of GO^{Fe}.

K_2FeO_4 (25 kg) was added into concentrated H_2SO_4 (93%, 16 L) slowly. Then graphite (500 g, 40 μm) was added slowly and the mixture was stirred for 1 h. The mixture was centrifuged (10000 rpm for 3min) to recycle the concentrated sulfuric acid. The paste-like product was collected by repeated centrifugation and water washing until the pH of the decantate approached 7, yielding 750 g of spray-dried powder.

Synthesis of GO^{Mn}.

The preparation process of GO^{Mn} was according to reference 24.

Synthesis of sample-T, sample-H, and sample-B.

The preparation processes of sample-T, sample-H, and sample-B were according to references of 14, 13 and 8.

Preparation of GO fibre, film and carbon aerogel.

The preparation process of GO fibres, films and carbon aerogels followed the references of 22-24 in the main text.

Measurement of O_2.

The mass of O_2 released from the reaction system of K_2FeO_4, H_2SO_4 and graphite was measured by the mass loss of the reaction system.

Typically, K_2FeO_4 (6 g) was added into concentrated H_2SO_4 (40 mL) to carry on the control experiment between K_2FeO_4 and H_2SO_4 in the absence of graphite until no gas releasing from the system. The mass loss before and after reaction is about 0.67 g, corresponding to the oxygen generated by the reaction between K_2FeO_4 and H_2SO_4.

In another experiment, graphite (1 g, 40 μm) and K_2FeO_4 (6 g) were added to concentrated H_2SO_4 (40 mL). The mixture was stirred for 1 h. In this process, 0.116 g mass loss was measured, corresponding to the oxygen decomposed from K_2FeO_4. The suspension was poured slowly into 60 mL water and stirred for 2 h, and 0.084 g of mass loss was found, corresponding to the oxygen generated by the remained K_2FeO_4 after production of GO.

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