Facile Two-step thermal annealing of graphite oxide in air for graphene with a higher C/O ratio

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Table S1. The C/O ratio of thermal reduced graphene from the literatures

C/O ratio	Temperature (℃)	Conductions (time, atmosphere, pressure, catalyst)	Ref.
7.7	100	6h, Ar	6
6.95-10.2	100-200	3h, vacuum, metal Al as catalyst	7
9.3	100-200	6h, Air, benzyl alcohol	6
4	200	1h, Ar	8
7.3-9.9	500-700	60s, vacuum	4
6.3/12.4	500/1000	5min, Ar (80%) / H_2 (20%)	9
10	650	Ar/4%H_2	10
25	750	Ar/4%H_2	10
3.9-14.1	200-1000	under Ar, Ar/H_2 or UHV	11
110	1000	Ar/4%H_2	10
5.6-7.9	1050	30s, Ar;	12
10	1050	30s Ar	13
24-340	1100	with different time or second thermal treatments	4

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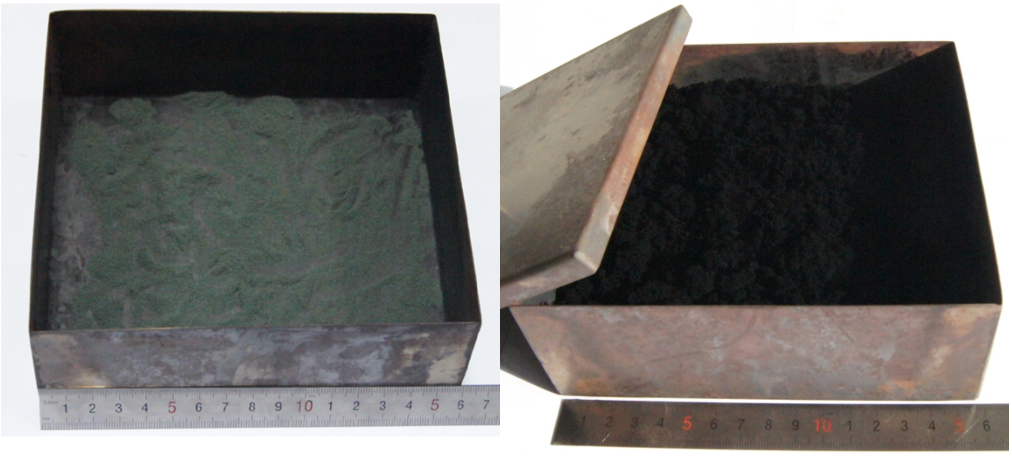

Fig. S1- the self-designed container for the thermal exfoliation and reduction: (a) 1g graphite oxide in the container, (b) the obtained graphene after thermal annealing under 550 ℃ 30s in air.

Table S2. The yield of graphene thermal annealed in air under 250-1050 ℃ for 10 to 120s. 1g graphite oxide was used as precursor.

Temp. \ Time	10s	30s	60s	120s
250 ℃	/	0.53	/	0.58
350 ℃	/	0.56	/	/
450 ℃	0.46	0.57	0.56	0.48
550 ℃	0.53	0.44	0.50	0.46
650 ℃	0.56	0.55	0.46	0.25
750 ℃	0.48	0.36	0.17	0.10
850 ℃	0.43	0.35	0.23	0.09

950 ℃	/	0.32	/	/
1050 ℃	/	0.29	/	/

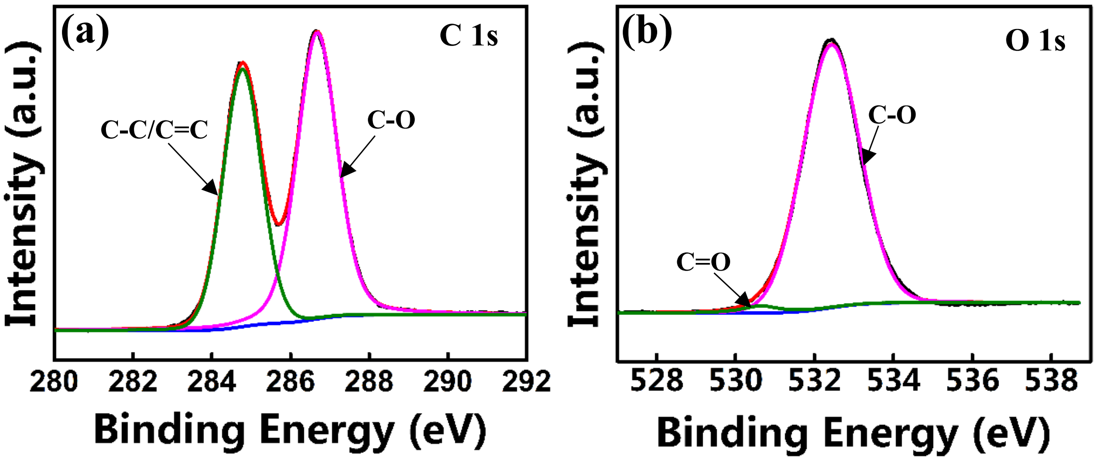

Fig. S2 - XPS C1s and O1s spectrum of graphite oxide. (a) C1s spectrum of GO, the proportion of C-C/C=C and C-O groups is 47.9% and 52.1% respectively. (b) O1s spectrum of GO, the proportion of C-O and C=O groups is about 97.7% and 2.3%.

BET	Temperature (℃)	Conductions (time, atmosphere, pressure, catalyst)	Ref.
18	100	6h, Ar	S1
758	135	24h, vacuum	S2
500-750	130-200	8min, ambient atmosphere, volatile HCl	S3
368-382	200-400	5h, High vacuum	S4
62.2-403	300-600	2h, N_2	S5
960	500	1h, inert gas, metal agglomeration, many steps	S6
600-1500	1050	30s, Ar	S7
1200	1050	30s, Ar (pretreat for 10min in Ar)	S8
569-703	450	10-120s, air	This work

Table S3. The BET data from literatures about thermal annealing

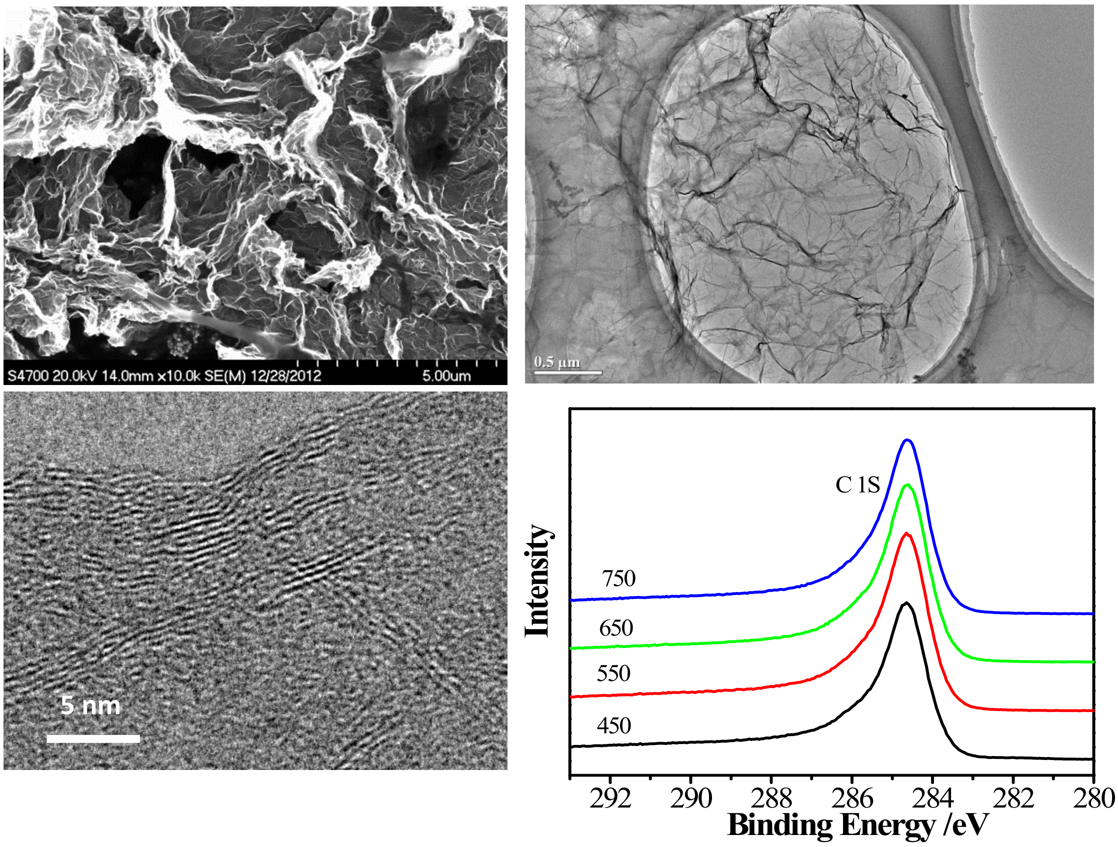

Fig. S3 - the SEM image (a), the TEM image (b), The HRTEM image (c) of the samples annealed under 550 ℃ for 30s, and XPS C1s of samples under 450-750 ℃.

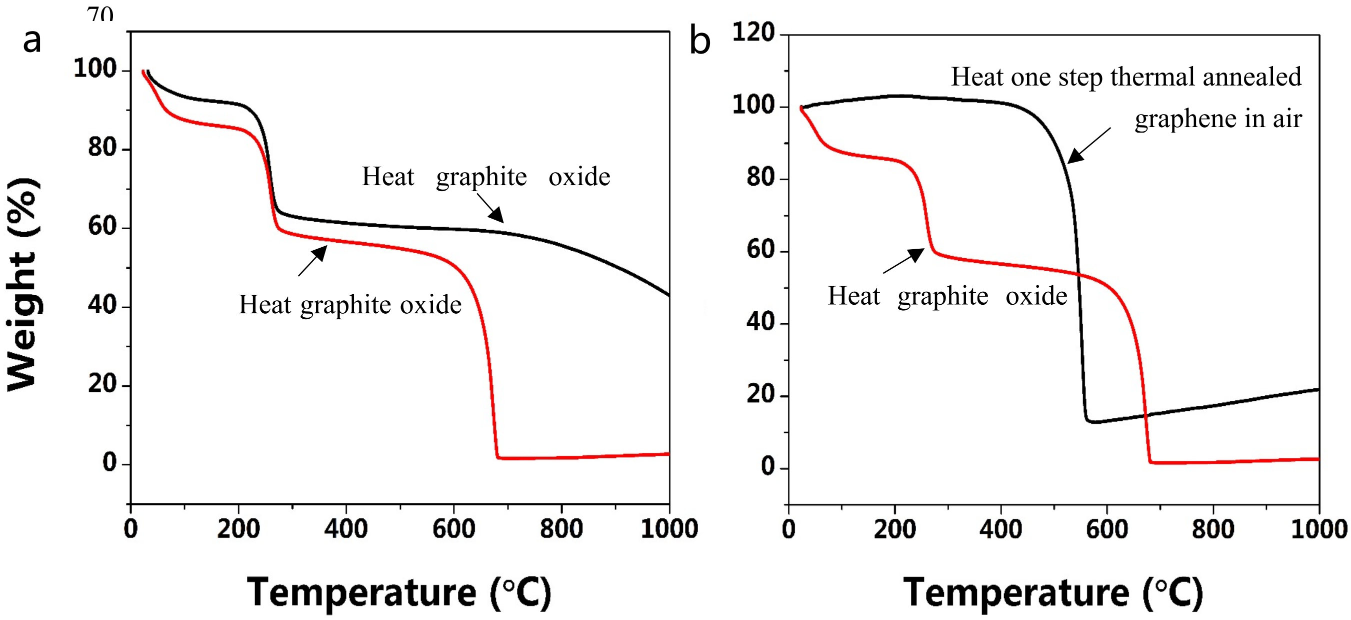

Fig. S4 – TGA data: (a) TGA spectra of the graphite oxide in air and in N_2. (b) TGA spectra of the graphite oxide and one step thermal annealed graphene in air.

The TGA spectra of graphite oxide and one-step annealed graphene samples are shown In Fig. S4a. Heating graphite oxide in air, the spectra exhibits three stage of weight losing: the first stage happens below 100 ^{o}C due to the H_2O loss; The second stage happens between 250 and 300 ^{o}C due to the transformation from oxygen-containing groups into CO and CO_2; The third stage is around 550 ^{o}C due to the further transformation and removal of residual functional groups. However, for heating graphite oxide in N_2, because of the protection of N_2 atmosphere, it shows a slow and gradual weight loss from 400-1000 ^{o}C. These results indicate the important effect of oxygen for the thermal annealing, especially when the temperature over 400 ^{o}C. We also did the test to compare the TGA of reduced graphene oxide and graphite oxide in air, as shown In Fig. S4b. For the one-step thermal annealing graphene, the spectra have only a weight loss around 550 ^{o}C. It doesn't have the obvious weight loss before 400 ^{o}C, and the weight loss around 550 ^{o}C is due to the carbon burning in air. The fast weight loss around 550 ^{o}C on the TGA curve of one-step annealed graphene indicates that one cannot treat graphene in air when the temperature is higher than 550 ^{o}C. We can get reduced graphene from 450 to 850 ^{o}C without air protection due to the released gas protection inside the box.

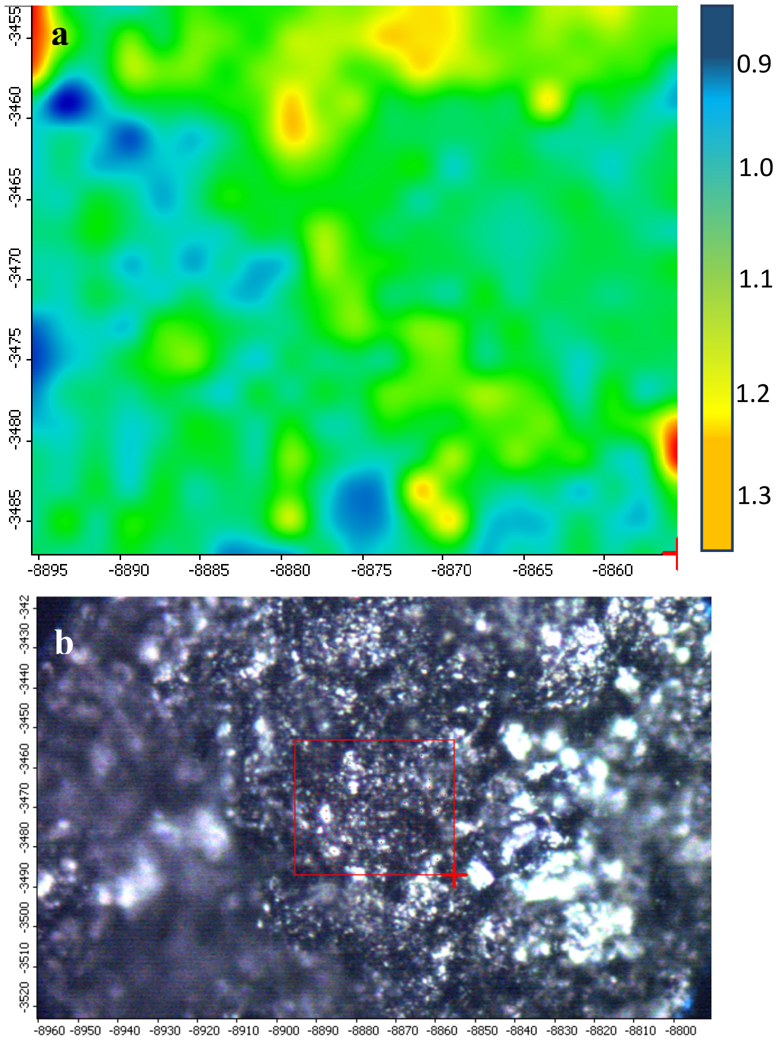

Fig. S5 – a) I_D/I_G Raman mapping of two-step annealed graphene on 30 μm*30 μm surface of a graphene bulk prepared by pressing fluffy graphene powder.

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