Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Informations

Towards the continuous production of high crystallinity graphene via electrochemical exfoliation with molecular in-situ encapsulation

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Table S1. The electrolyte recipes with various concentrations of melamine additives.

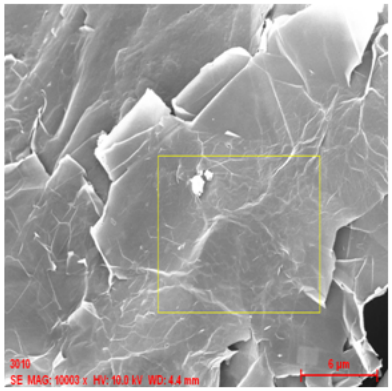
Number	H_2SO_4 (g/100ml)	Melamine (mg/10 ml)	Siring (RPM)	Current(A)	Exfoliated Graphene (g)
1	2.4	0	150	3	------- *
2	4.8	0	150	3	0.437
3	6.5	0	150	3	0.414
4	2.4	20	150	3	------- *
5	4.8	20	150	3	0.597
6	6.5	20	150	3	0.424
7	6.5	22	150	3	0.548
8	6.5	22	0	3	0.340
9	6.5	10	150	3	0.624
10	6.5	20	150	3	0.469
11	6.5	60	150	3	0.479
12	6.5	100	150	3	0.424
13	6.5	200	150	3	0.319

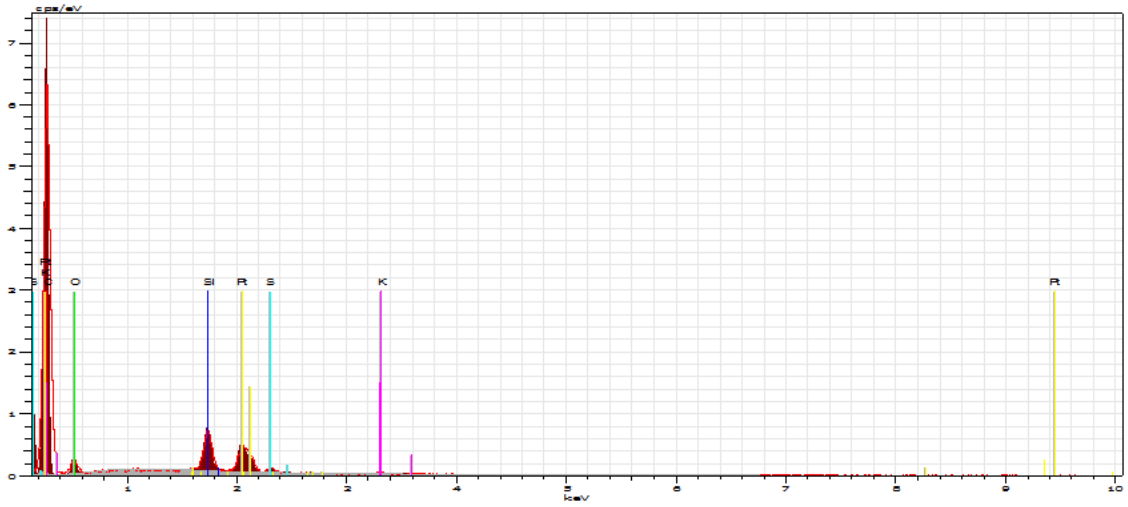
***: The amount of exfoliated graphene is much lower and unavailable for measurement.**

Figure S2. The characterizations of energy-dispersive X-ray spectroscopy(EDS) on as-prepared graphene

Spectrum:

Element	Series	unn. C [wt.%]	norm. C [wt.%]	Atom. C [at.%]	Error (3 Sigma) [wt.%]
Carbon	K-series	73.13	80.55	92.95	27.22
Oxygen	K-series	3.75	4.13	3.57	2.48
Silicon	K-series	4.46	4.91	2.42	0.67
Sulfur	K-series	0.45	0.50	0.21	0.16
Potassium	K-series	0.43	0.47	0.17	0.17
Platinum	M-series	8.58	9.45	0.67	1.15
	Total:	90.79	100.00	100.00	





S3 Comparison with other graphene production methods

Here, we analyzed 56 reported papers that involve the production of graphene via a liquid processing method, including intensive ultrasonication, GO and rGO, ball milling, etc. All of the studied papers use graphite powder as the starting material. However, most of these papers reported a production rate below 0.01 g/hr, which is lower compared with the production rate of our method (>0.02g/hr). Moreover, the level of quality of graphene produced, such as oxidation degree and D/G ratio, are included for this discussion. It is clear seen that the flake size prepared by our method is 10 to 100 times larger than other works.

Table S3. The comparison with graphene production methods

Ref	Method	Flake size	Raman D/G ratio	Oxidation degree(O%)	Production rate (g/hr)
This work	Electrochemical Exfoliation in the presence of melamine	12~35 μm	0.53 (532 nm)	3.68 (by XPS)	1.54
1	Wet milling in SDS	0.1-1.4 μm	0.6-0.7 (532 nm)	N/A	1.5
2	Shear exfoliation in organic solvent	300-800 nm	0.17~0.37	None	5.3
3	Liquid phase exfoliation(in pyrene)	2-2.5 μm	0.33 (633 nm)	N/A	0.02
4	GO by Hummers' method	1~5	>1	16	0.45

Reference

[1] Catharina Knieke et al., Scalable production of graphene sheets by mechanical delamination. Carbon , 3196-3204, (2010)

[2] Keith R et al., Scalable production of large quantities of defect-free few-layer graphene by shear exfoliation in liquids, Nature Materials, 624-630, (2014)

[3] Parviz, D. *et al.* Dispersions of Non-Covalently Functionalized Graphene with Minimal Stabilizer. *ACS Nano* 6, 8857-8867, (2012).

[4] Ken-Hsuan Liao et al., Aqueous Only Route toward Graphene from Graphite Oxide. ACS Nano , 1253-1258, (2011)

S4 . XRD patterns of (a) EC-graphene and (b) graphite