Electronic Supplementary Information

Facile synthesis of battery waste-derived graphene for transparent and conductive film application by electrochemical exfoliation method

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S1. Experimental set up for the synthesis of graphene

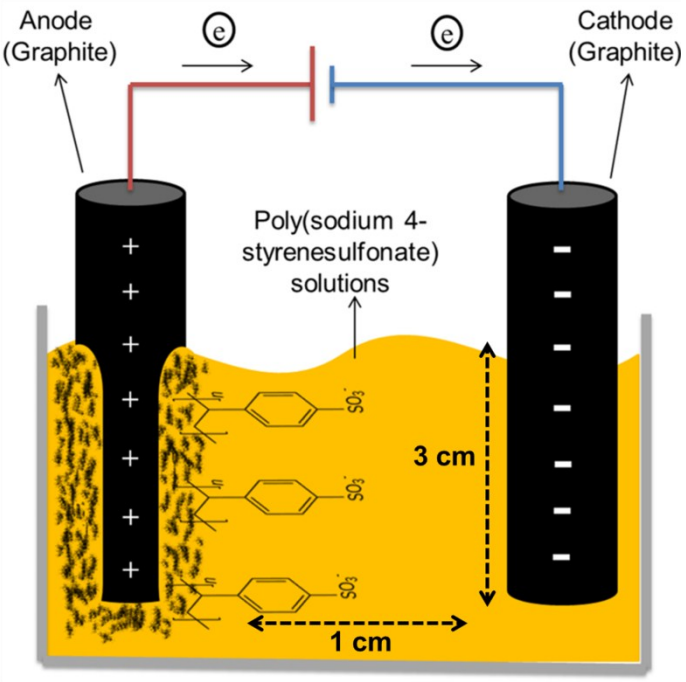


Fig. S1 Schematic of the electrochemical set up during the electrochemical exfoliation of graphite.

S2. Digital images of all graphene-PSS dispersions

Large PSS concentration and high applied voltage during the synthesis process lead to the formation of a dark brownish color of graphene-PSS dispersion. This fact indicates the escalation of exfoliation process which results in the higher concentration of exfoliated graphene.

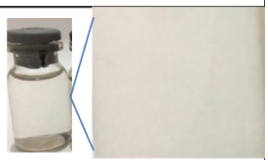
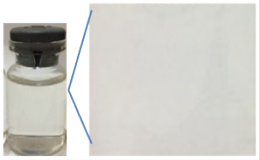
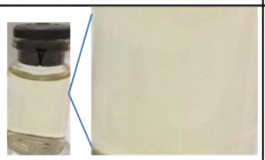
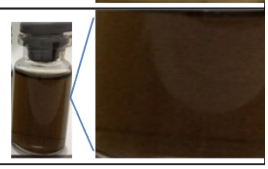
		Voltage (V)		
		3	5	8
Concentration (M)	0.001	-	-	
	0.02			
	0.1			
	0.5			

Fig. S2 Digital images of all graphene-PSS dispersions

S3. The UV Vis spectra of all graphene-PSS dispersions

The intensity of the absorption peaks of graphene-PSS dispersion grows with the increase of the applied voltage and PSS concentration. This is because that larger applied voltage and higher PSS concentration will escalate the intercalation process of polystyrenesulfonate anion into the graphite rod, resulting in a high concentration of exfoliated graphene in the PSS solution. Furthermore, the absorption peak of graphene-PSS dispersions is observed to be red-shifted with the increase of applied voltage and PSS concentration due to the strong interaction of graphene with PSS.

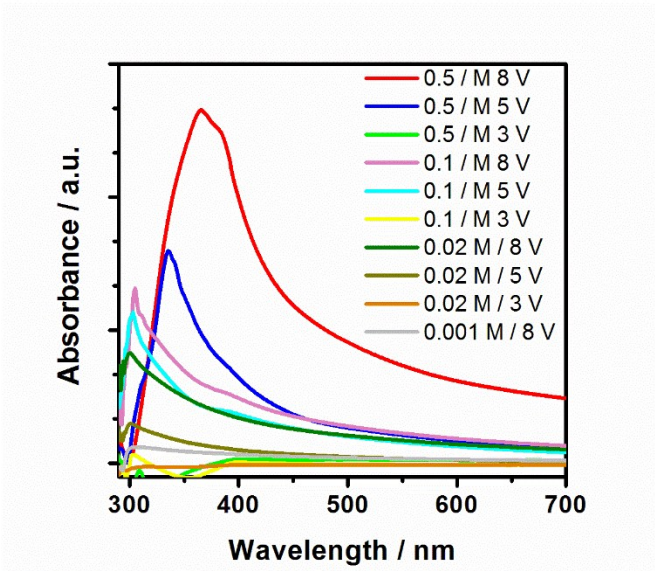


Fig. S3 The UV Vis spectra of all graphene-PSS dispersions

S4. Raman spectrum of Graphene 0.02/5

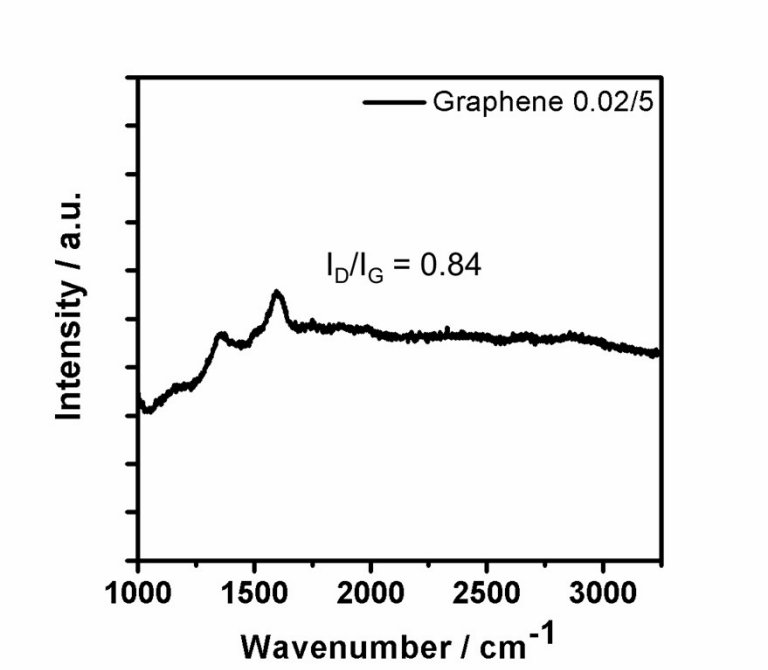


Fig. S4. Raman spectrum of Graphene 0.02/5

S5. XPS spectrum of Zn2p, Mn2p, Fe2p, and Cl2p in graphene 0.5 M / 5 V

XPS spectrum of graphene sample confirm no peak of impurities from Zn-C battery elements (e.g. Zn2p, Mn2p, Fe2p and Cl2p) observed.

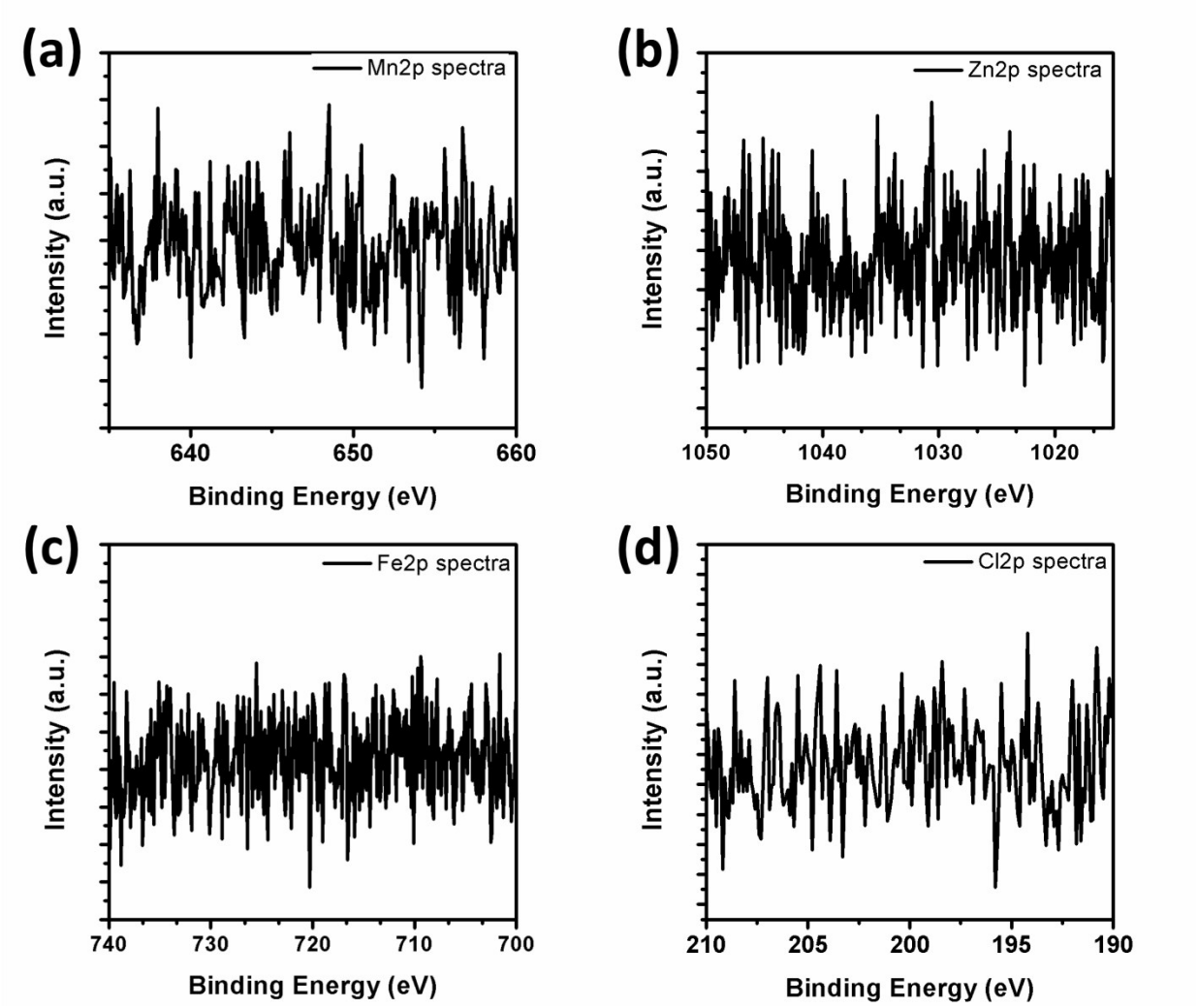


Fig. S5 XPS spectrum of a) Mn2p, b) Zn2p, c) Fe2p, and d) Cl2p elements in graphene 0.5 M / 5V

S6. SEM image of graphene

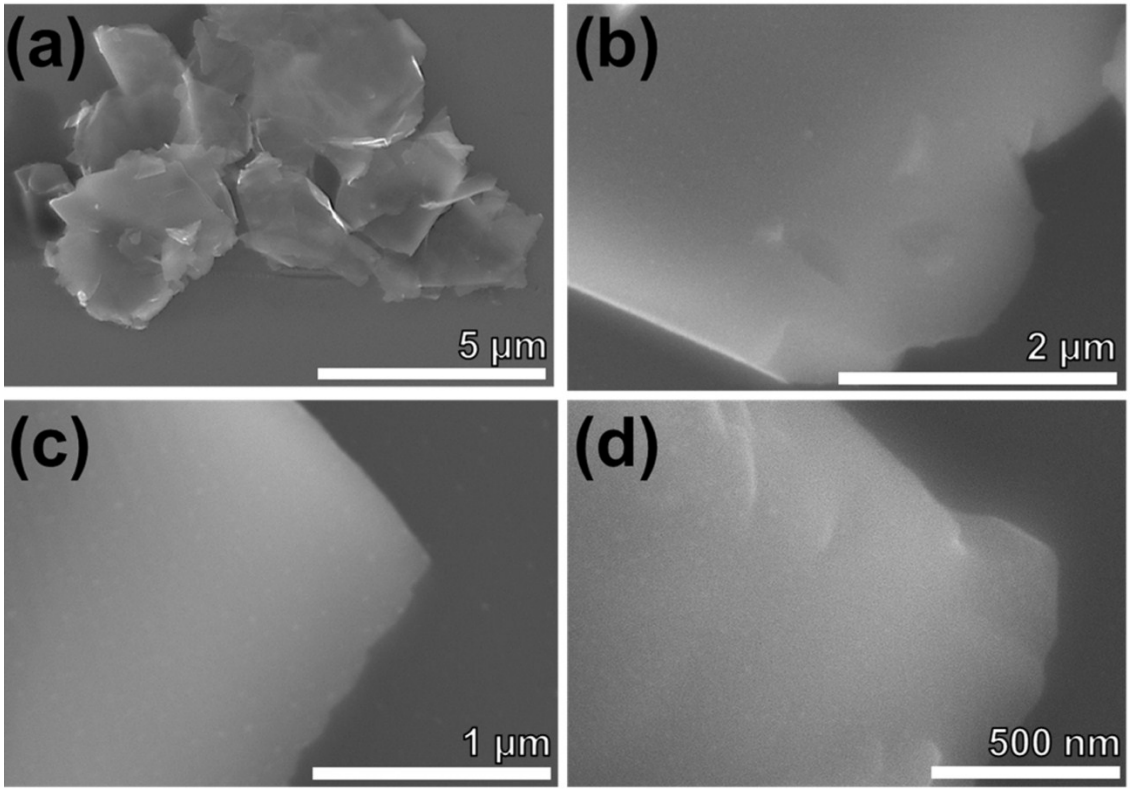


Fig. S6 SEM images of graphene sample under several magnifications.

S7. HRTEM image of graphene

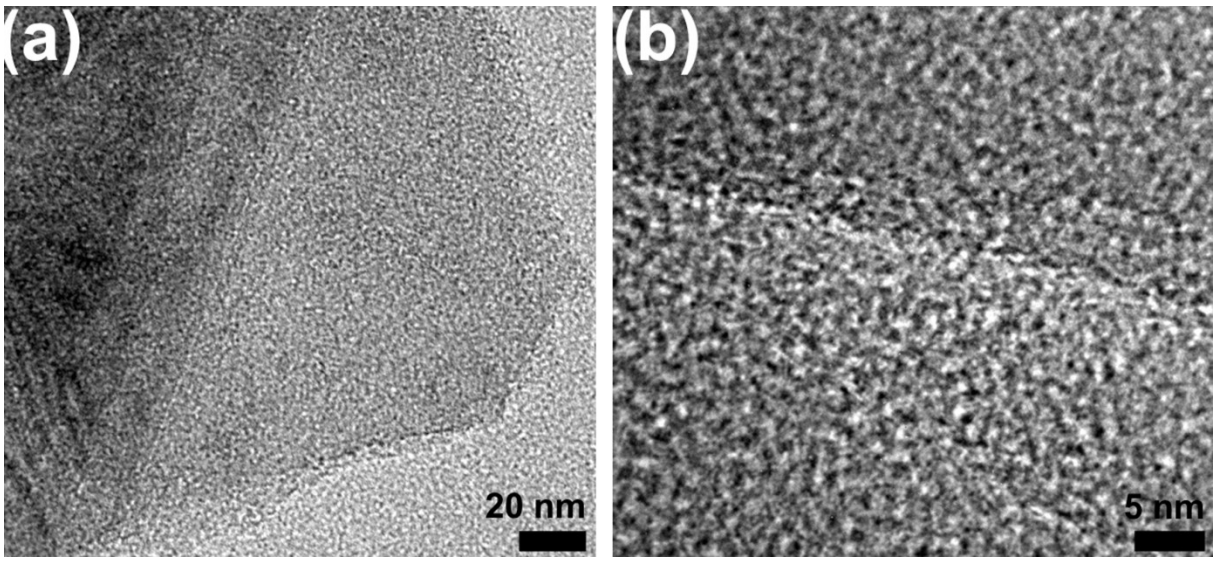


Fig. S7 a) TEM and b) HRTEM images of the graphene sample.

S8. Defect ratio (I_D/I_G) measurement of various graphene samples

Table S1. Defect ratio (I_D/I_G) of the graphene prepared by electrochemical exfoliation in the literature

Electrolyte	Source material	Voltage (V)	Defect ratio $(\frac{I_D}{I_G})$	Reference
PSS	Graphite of spent battery	5	0.86	This work
H_2SO_4	Graphite of spent battery	10	0.95	[1]
$(NH_4)_2SO_4$	Graphite foil	10	1.9	[2]
SDBS	Graphite of spent battery	4.5	0.2	[3]
H_2SO_4	Graphite foil	3	0.97*	[4]
$(NH_4)_2SO_4$	High purity graphite & HOPG	12	1-1.03	[5]
H_2SO_4 melamine	Graphite	20	0.2-0.54	[6]
TCNQ/DMSO	High purity graphite	25	1.02	[7]
SDBS	Graphite	25	0.98-1.05	[8]
$BMTF_2N$	HOPG	-20	0.05-1.7	[9]
Na_2SO_4 & SDS	HOPG	5-6	1.02*	[10]
Ionic liquid	High purity graphite	20	1.41	[11]
SDS	High purity graphite	1.4 - 2	0.124 – 1.370	[12]

Note: * Graphene obtained without further modification

S9. Sheet resistance and transmittance value of various graphene thin films

Table S2. Sheet resistance and transmittance value of various graphene thin films in the literature

Source material	Synthesis method	Sheet resistance ($k\Omega\ sq^{-1}$)	Transmittance (%)	Reference
Graphite of spent battery	Electrochemical exfoliation	1.1	89	This work
Graphite foil	Electrochemical exfoliation	2.7	-	[2]
Graphite foil	Electrochemical exfoliation	100	94.4	[13]
Graphite flake	Electrochemical exfoliation	22.31	73	[14]
Natural graphite rocks	Hummers	22	88	[15]
Expandable graphite	LPE	7.3	-	[16]
High purity graphite & HOPG	Electrochemical exfoliation	0.32	< 20	[5]
Graphite foil	Electrochemical exfoliation	3.91	85	[17]
HOPG	Electrochemical exfoliation	0.44 *	75.8	[10]
HOPG	Electrochemical exfoliation	0.66 *	96	[18]
CH_4	CVD	0.125	97.4	[19]

Note: * Graphene film modified by thermal annealing or chemical doping

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