

## Polymer Casting of Ultralight Graphene Aerogels for the Production of Conductive Nanocomposites with Low Filling Content

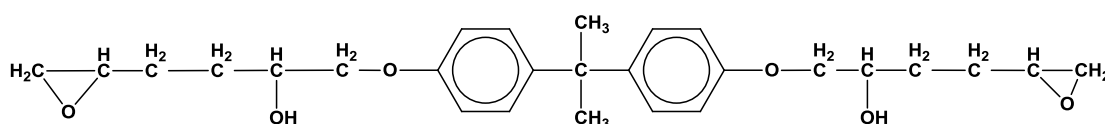
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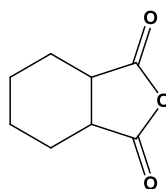
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**Table S1.** The amount of GO and agents for synthesis of UGLA with desired properties

GO dispersion		EDA/ $\mu\text{L}$	Inner diameter of Reactor/ mm
Amount/ mL	Concentration/ $\text{mg}\cdot\text{mL}^{-1}$		
5	1.5	10	18
5	2	13	18
5	3	20	18
5	4	24	18
2	3	8	12
75	3	300	28



E44



hexahydrophthalic anhydride

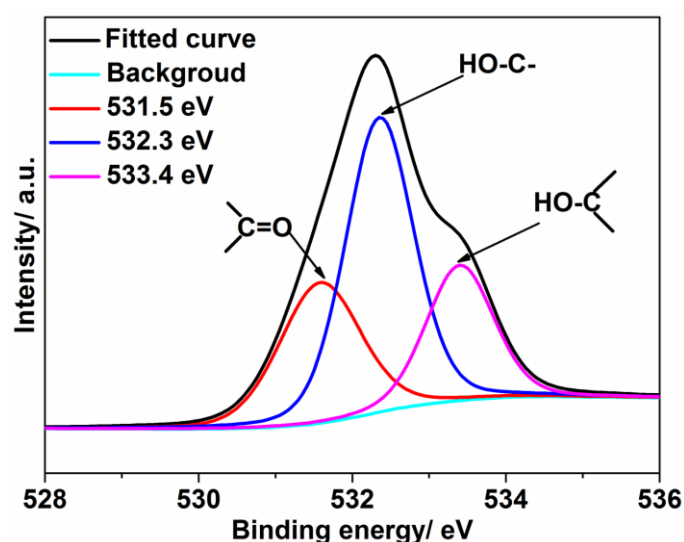
**Fig.S1.** Chemical structure of monomer of E44 epoxy resin and curing agent of hexahydrophthalic anhydride. The structure shown above containing aromatic rings, hydroxyl and CH that can provide several kinds of interactions with the abundant  $\pi$ -electrons of graphene.



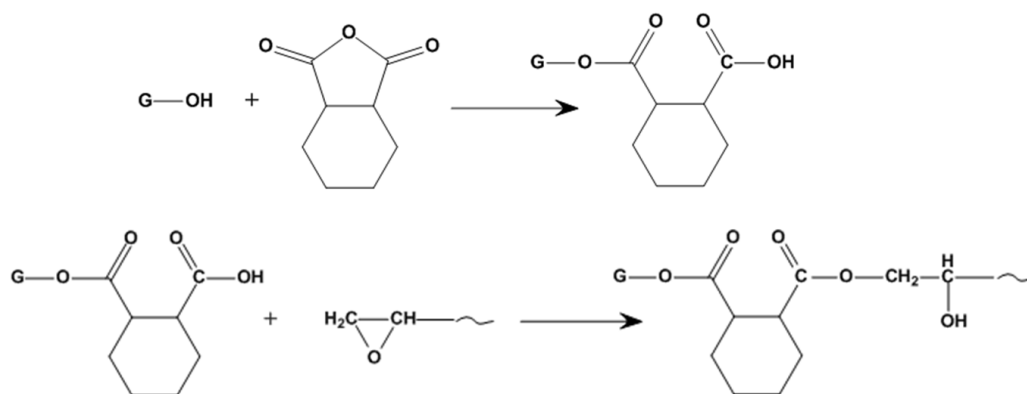
**Fig.S2.** Digital images of ER/ULGA composites with different sizes.

**Table S2.** Summary of peaks appearing in Raman spectra<sup>1</sup> in Fig. 2d

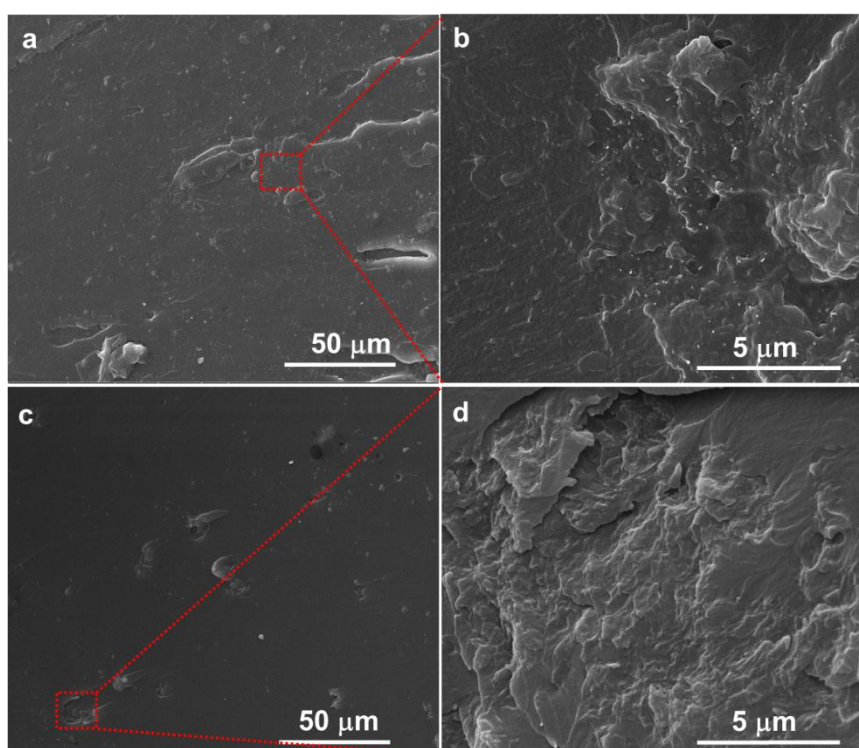
Raman shift/ $\text{cm}^{-1}$	Mode
824	Aromatic C-H bend
1114	Aromatic C-H bend
1187	Aromatic C-H bend
1226	Epoxide C-O-C stretch
1350	$\text{sp}^3$ carbon
1463	Epoxide C-H bend
1576, 1585	$\text{sp}^2$ carbon
1610	aromatic C=C ring stretch
2871	$\text{CH}_3$ stretch
2928	$\text{CH}_2$ stretch
3069	aromatic C-H stretch



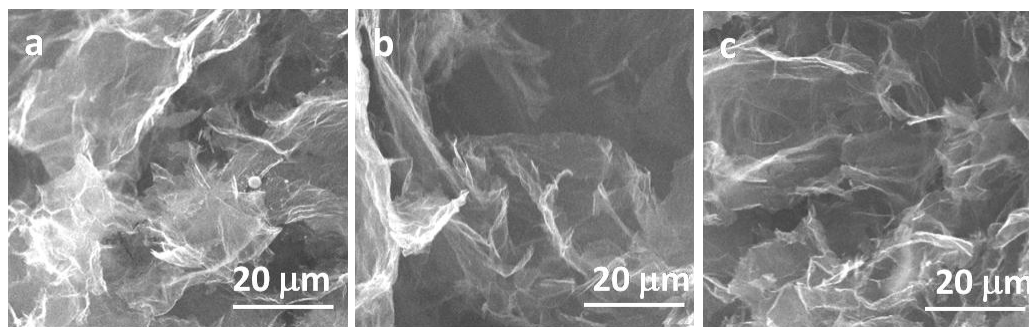
**Fig.S3** High resolution of O 1s spectrum of ULGA. The fitted results show three kind of oxygen-containing functional groups which are carbonyl group at 531.5 eV, hydroxyl group attached to aliphatic carbon at 532.3 eV and hydroxyl groups attached to aromatic carbon at 533.4 eV.<sup>2</sup>



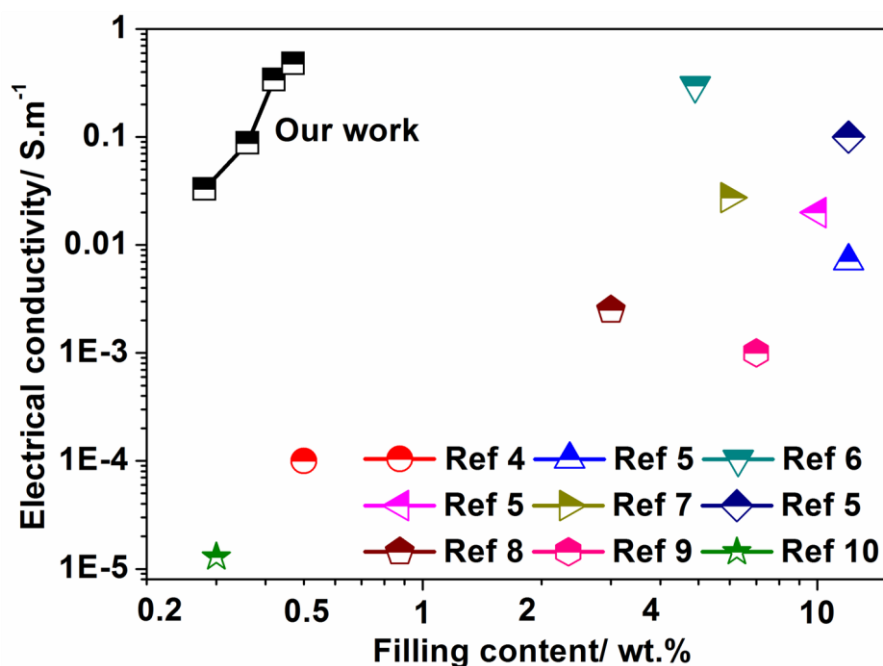
**Fig.S4** The possible reactions involving graphene in the curing process. In these reactions, graphene has been abbreviated to G. In a possible route, the anhydride ring may be opened by hydroxyl groups on graphene, giving rise to half-ester grafted on graphene. Then, the acid group of the half-ester further reacts with epoxide ring producing a hydroxy diester.<sup>3</sup> Via a serial of reactions shown above, strong chemical bonding may be formed between graphene and epoxy resin



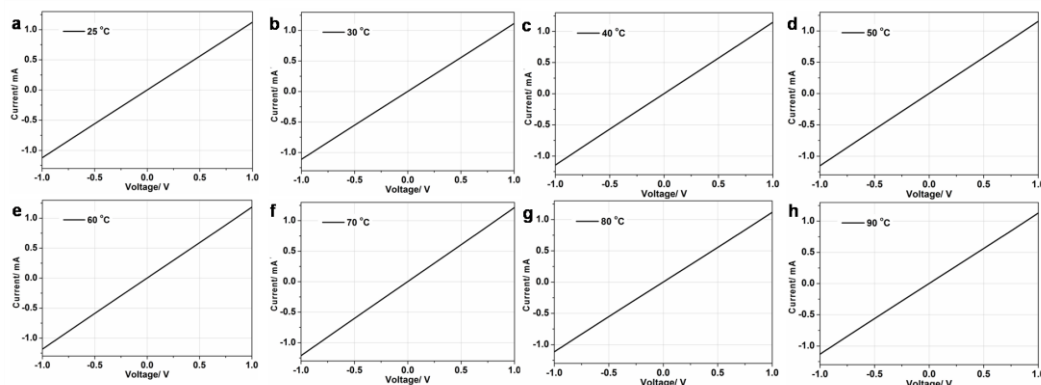
**Fig.S5.** (a) and (b) SEM images of ER/CNT(p), (c) and (d) SEM images ER/G(p)



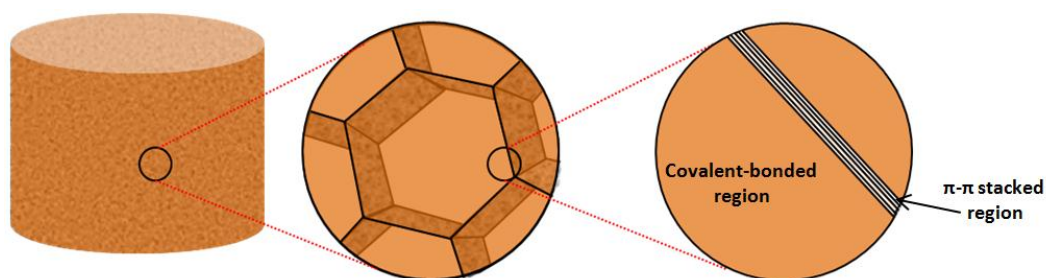
**Fig.S6.** SEM images of graphene aerogels synthesized via L-ascorbic acid-mediated reduction (a), sodium bisulfite-mediated reduction (b) and hydrothermal-mediated reduction (c) of GO



**Fig.S7** Summary of conductivity of different polymer-graphene composites



**Fig.S8** *I-V* curves of the composite tested at different temperature



**Fig.S9** Different bonded region within the composite

The matrix and interface of graphene and polymer are bonded with strong covalent bonds while the graphene layers of the cellular walls are hold together via van der Waals force. The continuous network of these nanosheets are highly important for the conductive performance. However, the interaction among different layers is unsuitable to the enhancement of the mechanical performance as the van der Waals force is too weak to transfer the external load effectively.

## References

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