## Solvent-Controlled Formation of Reduced Graphite Oxide gel via Hydrogen Bonding

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Figure S1 The photos of GO suspension in different solvents



Figure S2 The SEM images of rGO gels synthesized in different solvents.





Figure S3 The C1s XPS spectra of GO (a) and products obtained from reduction of GO with VC in different solvents: rGO-H<sub>2</sub>O (b), rGO-EG (c), rGO-DMF (d), rGO-NMP (e).

In Figure S3, the C1s XPS spectra of GO and rGO gels prepared in different solvents were shown clearly. For GO, four different peaks centered at 284.6, 286.5, 287.9 and 289.2 eV are observed, corresponding to C–C/C=C in aromatic rings, C–O (epoxy and alkoxy), C=O and O–C=O groups respectively.<sup>1</sup> After the deoxidization of GO, the peak intensities centered at 286.5, 287.9 and 289.2 eV significantly decrease respectively. Nonetheless, some peaks of oxygen-containing groups do not disappear completely, indicating that there are a lot of residual oxygenic groups on rGO sheets. Due to the existence of these oxygenic groups, the compact 3D networks between these oxygen-containing groups and solvents is intensive to be built in the rGO gels by the hydrogen bonding.



Figure S4 The FT-IR spectra of GO before and after being reduced in various solvents, NMP, DMF, EG and water.

The reduction of GO can be seen clearly in Figure S4. The intensities of C=O stretching vibrations peak at 1724 cm-1 and O-H in-plane bending vibrations from hydroxyl groups at 1410 cm-1 decreased remarkably after reduction.<sup>2</sup> Furthermore, the C-O (epoxy) stretching vibration peak at 1226 cm-1 and C-O stretching vibration associated to alkoxy peak at 1025 cm-1 almost disappeared. These observations confirmed that a large proportion of oxygen functionalities in GO were removed.



Figure S5 Raman spectra of GO, and rGOs synthesized in various solvents, NMP, DMF, EG and water.

Table S1 The D/G parameters of GOs and rGOs from Raman spectra.					
Sample	D (cm <sup>-1</sup> )	I <sub>D</sub>	G (cm <sup>-1</sup> )	I <sub>G</sub>	$I_D/I_G$
GO	1349	219	1596	208	1.05
rGO-H2O	1341	293	1586	268	1.09
rGO-EG	1342	239	1588	175	1.37
rGO-DMF	1344	172	1589	152	1.13
rGO-NMP	1344	230	1592	206	1,12

Raman spectra of rGO could also show the reduction of GO. As depicted in Figure S5, the peaks at around 1590 and 1345 cm<sup>-1</sup> attributed to G (the  $E_{2g}$  mode of sp<sup>2</sup> carbon atoms) and D (the symmetry  $A_{1g}$  mode) bands respectively.<sup>3, 4</sup> The Raman spectrum of GO, as expected, displayed a prominent G peak at 1596 cm<sup>-1</sup> and the D band at 1349 cm<sup>-1</sup>; after deoxidization, the broad G band and D band of GO all shifted toward lower wavenumber indicating the restoration of sp<sup>2</sup> domains. The increased D/G intensity ratio compared to that in GO demonstrated the removal of oxygen moieties and restoration of the sp<sup>2</sup> network during reduction (Table S1).

## Reference

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