

Supporting Information

High-Strength Graphene Composite Films by Molecular Level Couplings for Flexible Supercapacitors with High Volumetric Capacitance

Jun Cao,^a Chen Chen,^a Kena Chen,^a Qiongqiong Lu,^a Qingrong Wang,^a Pengfei Zhou,^a Daobin Liu,^c Li Song,^c Zhiqiang Niu*^a and Jun Chen^{ab}

^a *Key Laboratory of Advanced Energy Materials Chemistry (Ministry of Education), College of Chemistry, Nankai University, Tianjin, 300071, P.R. China.*

^b *Collaborative Innovation Center of Chemical Science and Engineering (Tianjin), Nankai University, Tianjin, 300071, China.*

^c *National Synchrotron Radiation Laboratory, CAS Center for Excellence in Nanoscience, University of Science and Technology of China, Hefei, Anhui 230029, P. R. China.*

*E-mail: zqniu@nankai.edu.cn

Supplementary Figures

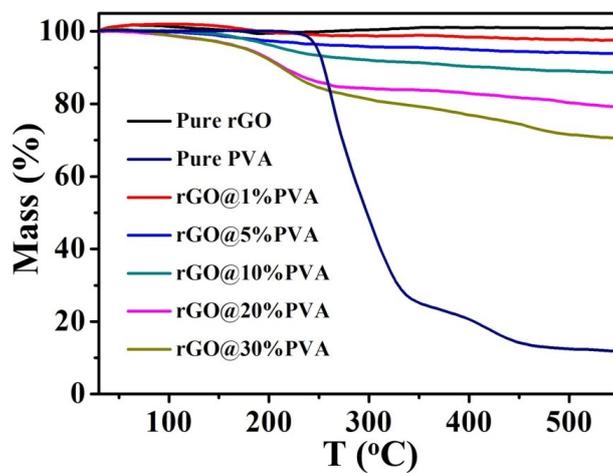


Fig. S1 Thermogravimetric curves of pure PVA, pure rGO and rGO@PVA composite films with different PVA contents in N₂ with a heating rate of 5 °C min⁻¹.

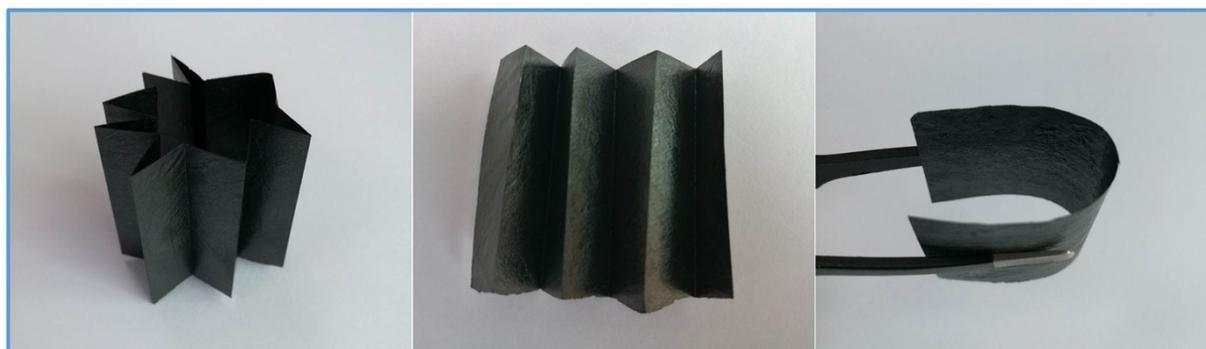


Fig. S2 Optical images of rGO@PVA composite films bent or twisted to different shapes.

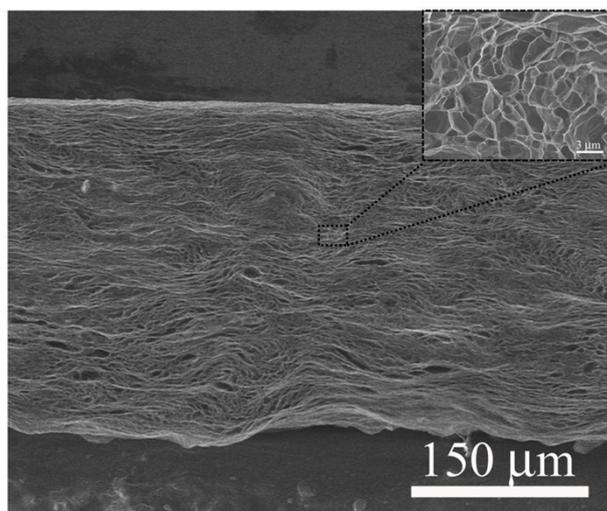


Fig. S3 SEM image of rGO@20%PVA composite film with porous structure. The insert shows the high magnification SEM image of the composite film.

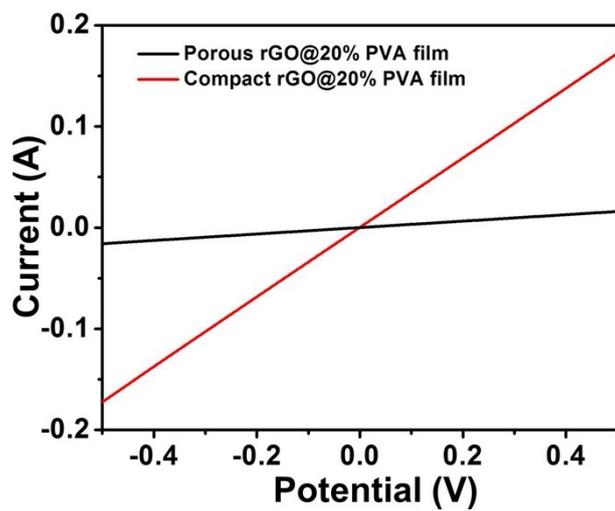


Fig. S4 I-V curves of porous and compact rGO@20%PVA composite films.

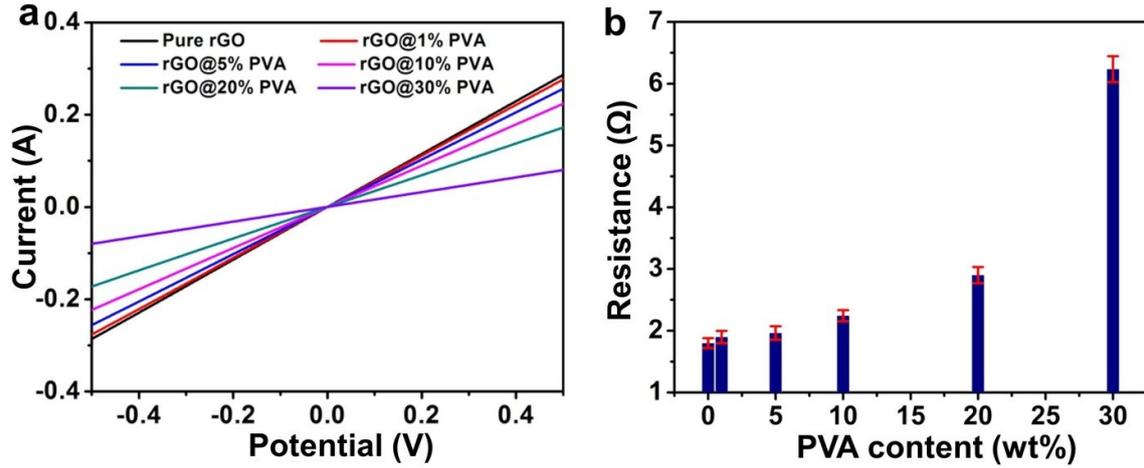


Fig. S5 (a) I-V curves and (b) sheet resistances of pure rGO and rGO@PVA composite films with different PVA contents.

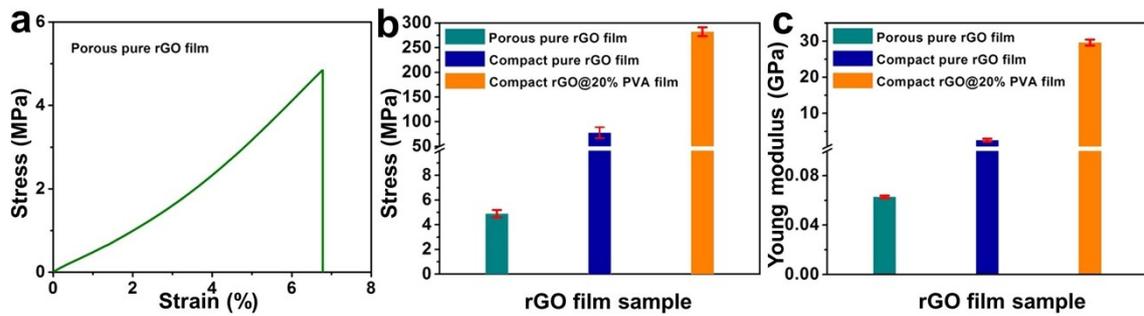


Fig. S6 (a) Stress-strain curves, (b) yield stresses and (c) Young's moduli of porous pure rGO films, compact pure rGO films and rGO@20%PVA composite films.

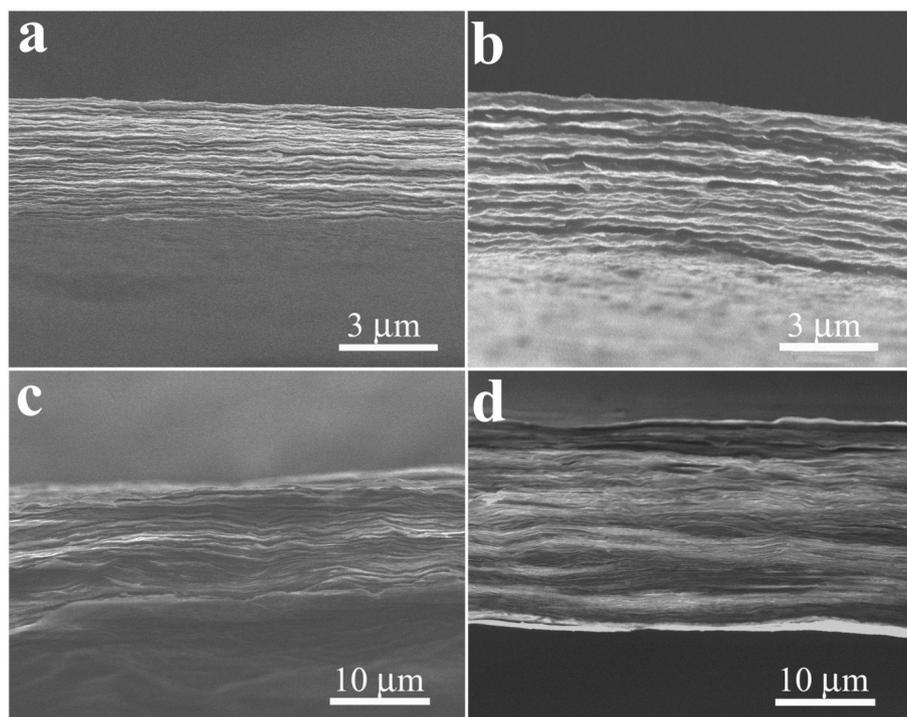


Fig. S7 Cross-sectional SEM images of rGO@20%PVA composite films with different H_2SO_4 concentrations: (a) 0.1 M (b) 0.2 M (c) 1.0 M (d) 2.0 M.

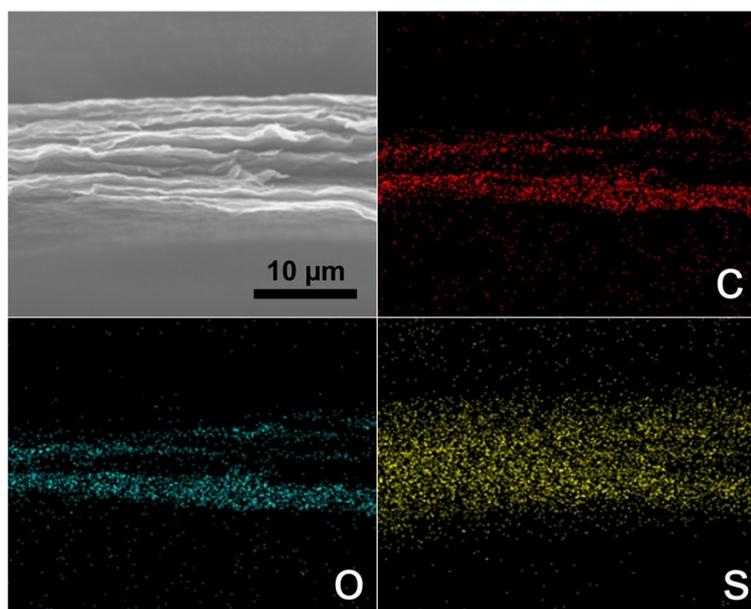


Fig. S8 SEM elemental mappings of elemental C, O, and S in the rGO@PVA- H_2SO_4 composite film.

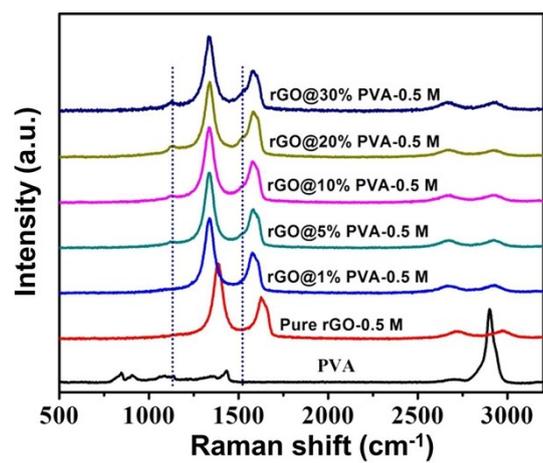


Fig. S9 Raman spectra of pure PVA and rGO@PVA-0.5 M composite films with different PVA contents.

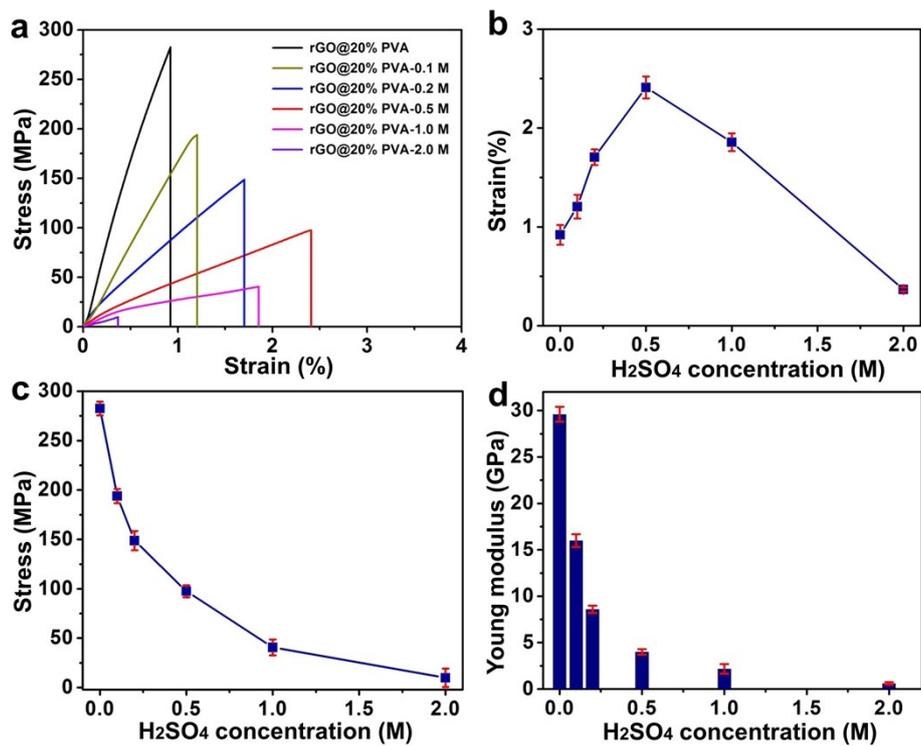


Fig. S10 (a) Stress-strain curves, (b) yield strains, (c) yield stresses and (d) Young's modulus of rGO@20%PVA composite films with different H₂SO₄ concentrations.

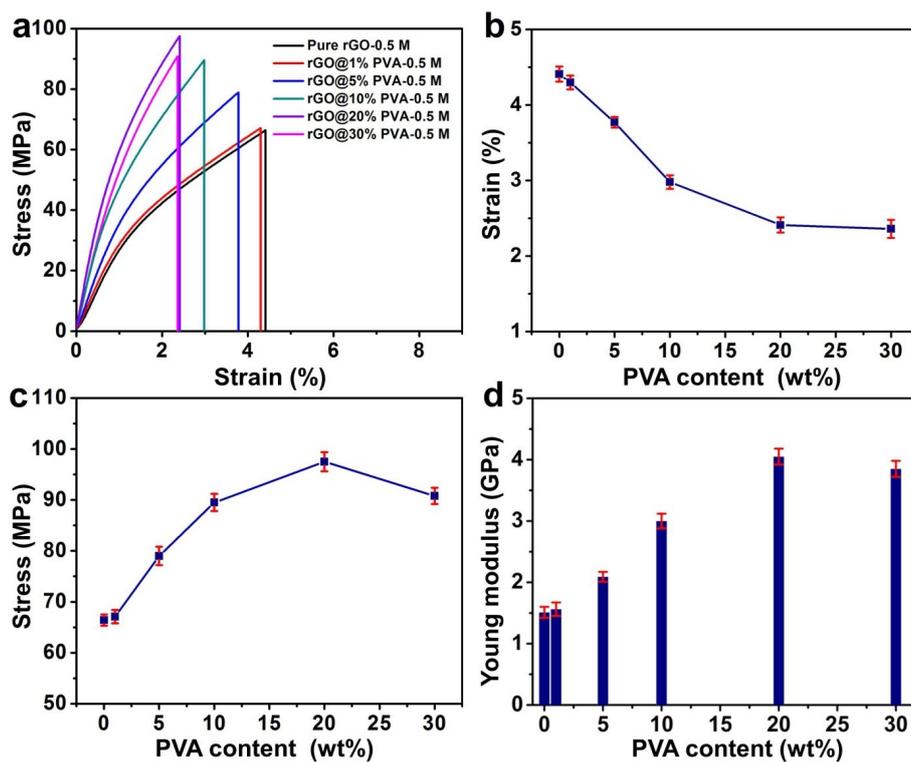


Fig. S11 (a) Stress-strain curves, (b) yield strains, (c) yield stresses and (d) Young's modulus of rGO@PVA-0.5 M composite films with different PVA contents.

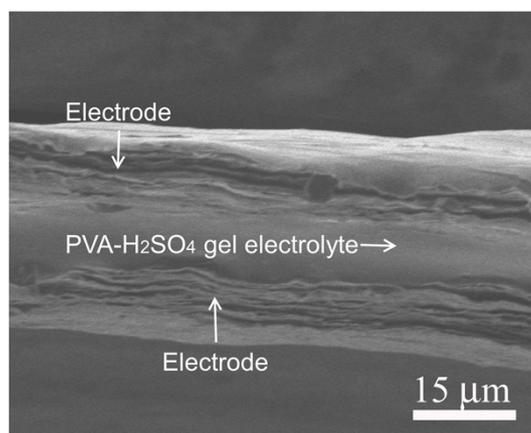


Fig. S12 Cross-sectional SEM image of the all-solid-state supercapacitor based on rGO@20%PVA-0.5 M composite films.

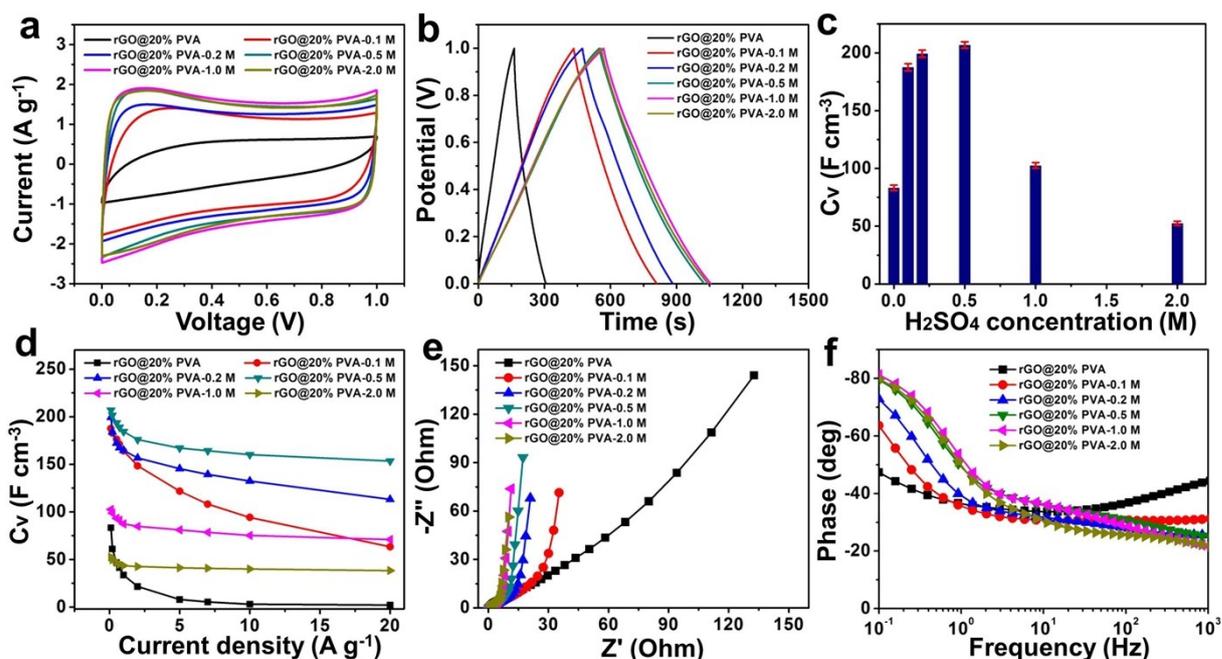


Fig. S13 Electrochemical performance of rGO@20%PVA composite films with different H_2SO_4 concentrations: (a) CV curves at 50 mV s^{-1} , (b) CD curves at 0.1 A g^{-1} , (c) volumetric capacitances at 0.1 A g^{-1} , (d) specific capacitances at different current densities, (e) Nyquist plots, and (f) Bode plots of phase angle versus frequency.

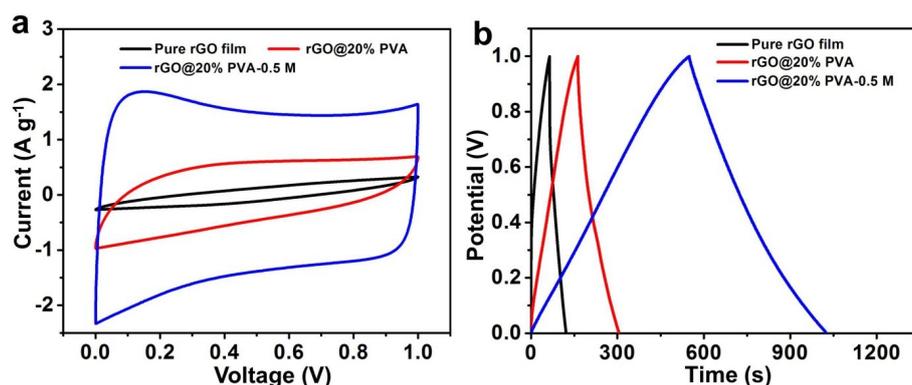


Fig. S14 Electrochemical performance of pure rGO film, rGO@20%PVA and rGO@20%PVA-0.5 M composite films: (a) CV curves at 50 mV s^{-1} , (b) CD curves at 0.1 A g^{-1} .

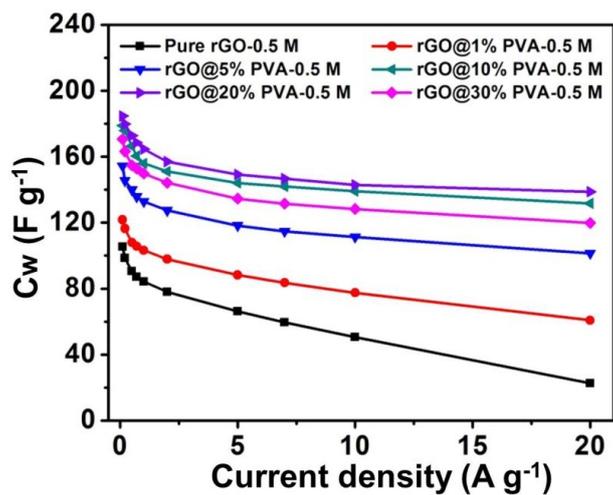


Fig. S15 Specific capacitance of rGO@PVA-0.5 M composite films with different PVA contents at different current densities.

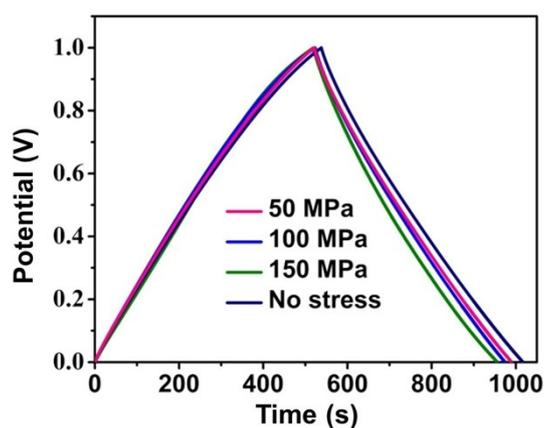


Fig. S16 CD curves of the supercapacitor under different stresses at a current of 0.1 A g⁻¹.

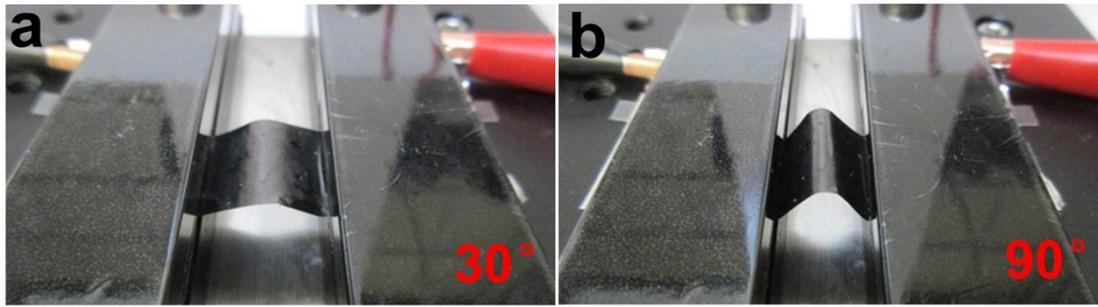


Fig. S17 Optical images of a supercapacitor devices at different bending states.

Table S1. The d-spacing parameters of rGO@PVA samples with different PVA contents.

PVA contents (%)	0	1	5	10	20	30
d-spacing parameters (Å)	3.68	3.69	3.72	3.77	3.85	3.94

Table S2. The packing densities of the composite rGO@PVA-H₂SO₄ films with different H₂SO₄ concentrations.

H ₂ SO ₄ Concentration (M)	0	0.1	0.2	0.5	1	2
Packing Density (g cm ⁻³)	1.46	1.37	1.27	1.12	0.55	0.29

The packing densities were calculated through dividing the volume of the rGO@PVA-H₂SO₄ film by the mass of the rGO in the composite film. The volume of the rGO@PVA-H₂SO₄ films was calculated through multiplying the thickness by area of the composite film. The mass of the rGO in the composite film was measured and calculated by the TGA data.