Supplementary Information

Facile Preparation of Flexible Binder-Free Graphene Electrodes for High-Performance Supercapacitors

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**Fig. S1.** Schematic illustration of hydrogen bonds in graphene oxide (GO).

**Fig. S2.** Optical and SEM (inset) images of (a) GO film and (b) Reduced GO film prepared by thermal treatment at 500 °C.
Table S1 Raman Peaks of GO film, GO/TRGO film and reduced-GO/TRGO film.

| Sample                     | Peak Index | Raman Shift (cm\(^{-1}\)) | \(\frac{I_D}{I_G}\) Central Peak Ratio |
|----------------------------|------------|-----------------------------|----------------------------------------|
| GO film                    | D          | 1364                        | 0.93                                   |
|                            | G          | 1584                        |                                        |
| GO/TRGO film               | D          | 1370                        | 0.96                                   |
|                            | G          | 1588                        |                                        |
| reduced-GO/TRGO film       | D          | 1359                        | 0.98                                   |
|                            | G          | 1586                        |                                        |

Fig. S3. Raman spectrum of TRGO.
Fig. S4. (a) XPS survey spectra and (b) XPS high-resolution C 1s spectra of TRGO.

Fig. S5. FTIR spectra of GO, GO/TRGO, and reduced-GO/TRGO films.

Table S2 Specific surface area of GO/TRGO films before and after reduction.
**GO/TRGO Film**

| GO/TRGO (Mass ratio) | Specific Surface Area (m$^2$/g) |
|----------------------|----------------------------------|
| 1:0                  | 8.9                              |
| 3:1                  | 97.5                             |
| 1:1                  | 218.0                            |
| 1:3                  | 253.1                            |
| 0:1                  | 357                              |

**Fig. S6.** Pore size distribution of GO film and GO/TRGO films with different composition (1:0, 3:1, 1:1, 1:3).
Fig. S7. CV curves of supercapacitors with (a) free-standing reduced-GO/TRGO (1:1) film and (b) filtered TRGO film electrodes at various scanning rate from 10 to 200 mV s\(^{-1}\).

Fig. S8. Galvanostatic charge–discharge curves of supercapacitors with (a) free-standing reduced-GO/TRGO (1:1) film and (b) filtered TRGO film electrodes at different current densities from 0.2 to 10 A g\(^{-1}\).
**Fig. S9.** Cross-sectional SEM image of filtered TRGO film.

**Fig. S10.** (a) Nitrogen adsorption/desorption isotherms and (b) Pore size distribution of filtered TRGO film.
Fig. S11. Photograph of fabricated solid state supercapacitor.

Fig. S12. Capacitance retention of solid state supercapacitor with free-standing reduced-GO/TRGO (1:1) film at bend angle of 180° under different bending cycle.
Table S3 Comparison of electrochemical performance with relevant graphene-based materials reported in literature.

| Material                        | Electrolyte          | Test Condition | Specific Capacitance (F g⁻¹) | Energy Density (Wh kg⁻¹) | Reference |
|---------------------------------|----------------------|----------------|------------------------------|--------------------------|-----------|
| RGO/AC/CNT                      | Organic (1M LiClO₄ EC/DEC) | 0.2 A/g       | 101                          | 30                       | Ref. 2    |
| N-doped holey graphene aerogel  | EMIMTFSI             | 1 A/g         | 142.3                        | 60.5                     | Ref. 3    |
| PCSG-60                         | TEABF₄               | 1 A/g         | 221                          | 38.2                     | Ref. 4    |
| IL-induced RGO                  | BMIMPF₆             | 0.5 A/g       | 150                          | 25.3                     | Ref. 5    |
| EDA/RGO                         | BMIMBF₄             | 2 mV/s        | 119                          | 51                       | Ref. 6    |
| N-doped graphene                | BMIMBF₄             | 1 A/g         | 137                          | 55                       | Ref. 7    |
| N/O co-doped hierarchical porous carbon | EMIBF₄              | 1 A/g        | 51.9                         | 22.1                     | Ref. 8    |
| UHFG                            | TEABF₄               | 1 A/g         | 182                          | 46                       | Ref. 9    |
| Flexible graphene               | EMIBF₄               | 0.2 A/g       | 180                          | 85.6                     | This work |

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