Supporting Information

Biotemplated hierarchical polyaniline composite electrode with high performance for flexible supercapacitors

Cheng-Ming Chang, Zh-Hao Hu, Ting-Yin Lee, Yi-An Huang, Wei-Fu Ji, Wei-Ren Liu,

Jui-Ming Yeh and Yen Wei*

Dr. C. M. Chang, Z. H. Hu, T. Y. Lee, Y. A. Huang, Prof. J. M. Yeh
Department of Chemistry and Center for Nanotechnology, Chung-Yuan Christian University
Chung Li 32023, Taiwan, R.O.C.
*E-mail: juiming@cycu.edu.tw
Prof. W. R. Liu
Department of Chemical Engineering, R&D Center for Membrane Center, Chung-Yuan Christian University
Chung Li 32023, Taiwan, R.O.C.
Prof. Y. Wei
Department of Chemistry and Key Lab of Organic Optoelectronic & Molecular Engineering of Ministry of Education, Tsinghua University
Beijing, 100084, China

Materials needed:

Aniline (Sigma-Aldrich) was distilled prior to use. 4,4'-Diaminodiphenylamine sulfate (APS, Aldrich) , N,N-dimethylacetamide (DMAc) and N-methyl-2-pyrrolidinone (NMP, Aldrich)and polydimethylsiloxane (PDMS, Dow Corning, Sylgard 184) were used as received without further purification. All the reagents were reagent grade unless otherwise stated.

The RGO purify process:

GO derived from SFG44 synthetic graphite powders (TIMCAL) was synthesized by a modified Hummers' method. 8.0 g synthetic graphite powder and 4.0 g $NaNO_3$ were put into 560 ml concentrated H_2SO_4 solution with stirring for 2 h. Then 32 g $KMnO_4$ was slowly added into the flask with ice bath for 2 h. The mixture was diluted by 800 ml deionized water. After that, 5% H_2O_2 was added into the solution until the color of the mixture changed to brown to ensure that $KMnO_4$ was fully reduced. The as-prepared GO slurry was re-dispersed in de-ionized water. Then, the mixture was washed with 0.1 M HCl solution to remove SO_4^{2-} ions. Finally, the GO solution was washed with distilled water to remove the residual acid until the solution pH reached ~5. Reduction processes were adopted for GO to form RGO. Thermally reduced graphene oxides (RGO) were prepared by calcination at different temperatures under 15% H_2/N_2 atmosphere for 2 h with a heating rate of 0.5^oC min^{-1}.

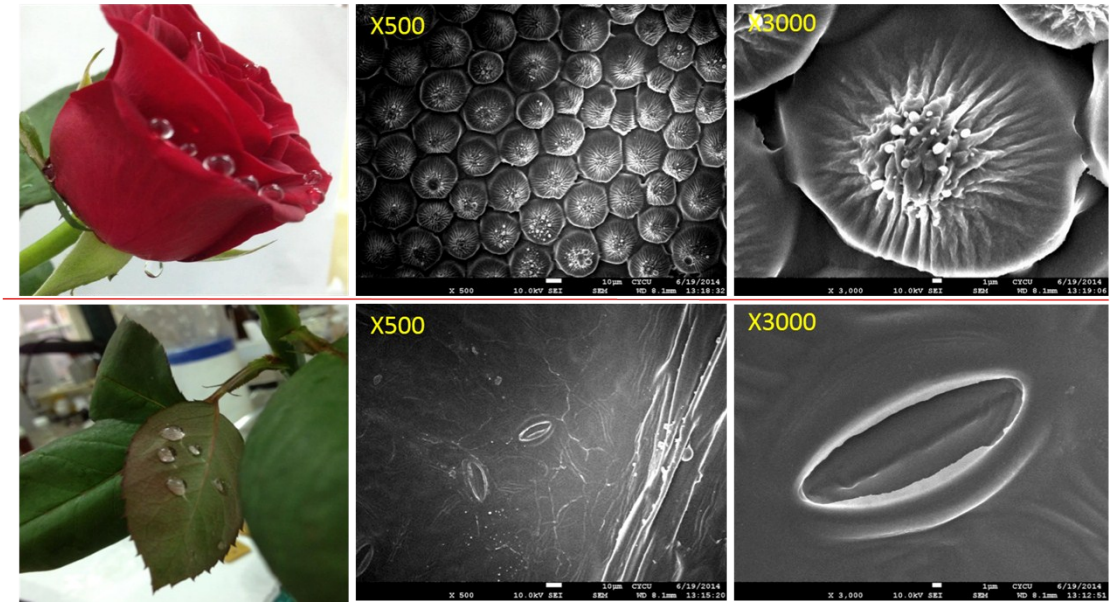
PDMS template method:

The PDMS pre-polymer was prepared from a 10:1 mixture (by weight) of the base and curing agent. After thoroughly mixing the base and curing agent, the pre-polymer was poured into the template of fresh Rose flower petal and leaves and then cured at 50 ^oC for 4 h and 90 ^oC overnight to ensure the completion of the PDMS cross-linking process. After curing, the resulting PDMS was separated from the template of the

petal and leaf and used as a negative template for preparing tri-dimentional construction PANI coatings.

Support information Fig. S1

The petal and leaves structure from *Rose flower* was characterized by photograph and SEM image.

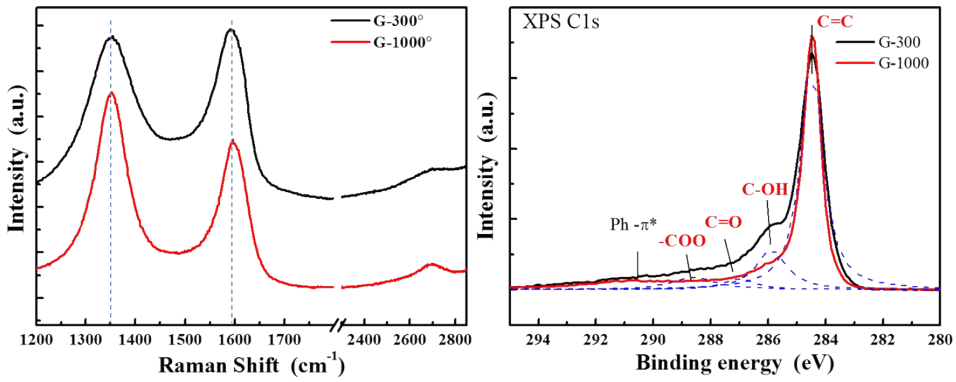


Support information Fig. S2

RGO structure change undergo in thermally calcination process was characterized by Raman and XPS

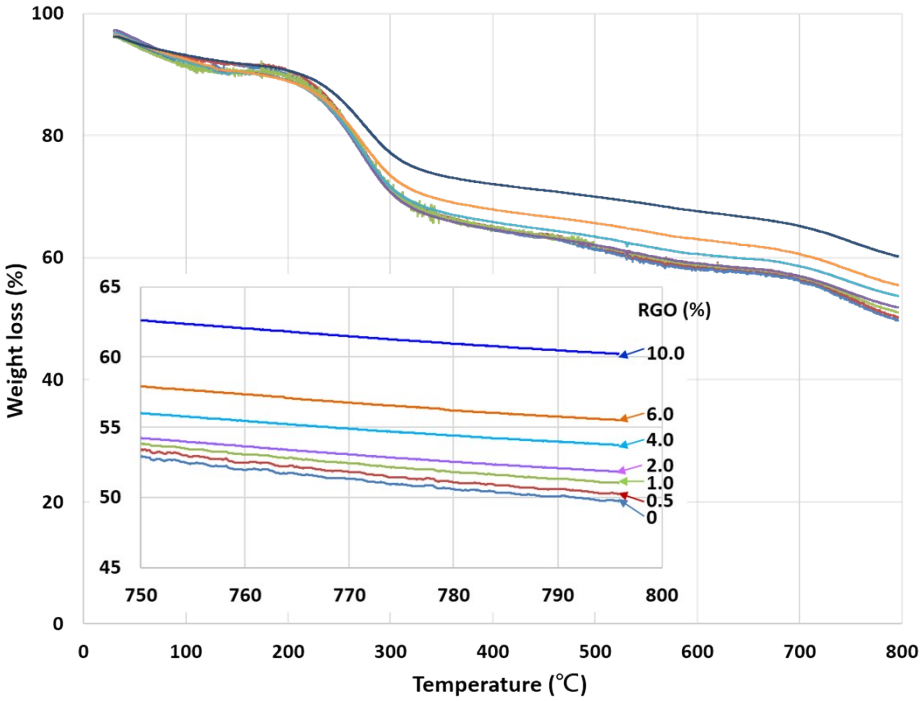
G-300°: thermally calcination temperature at 300°

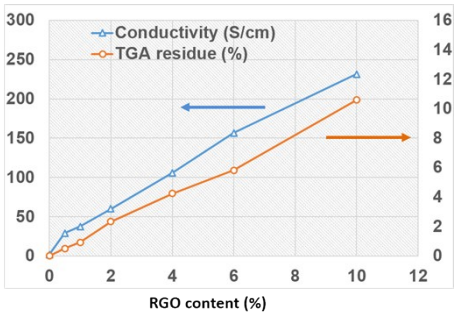
G-1000°: thermally calcination temperature at 1000°



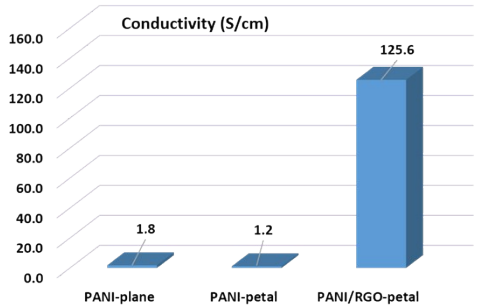
Support information Fig. S3

The RGO real content and conductivity of PANI composite film as shown in follow





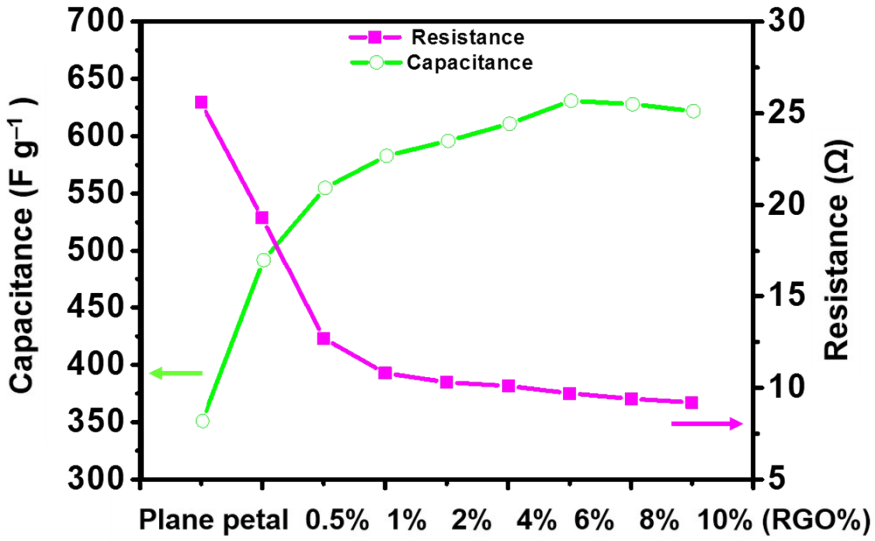
RGO (%)	0	0.5	1	2	4	6	10
Resistance (Ohm)	18.17	1.14	0.888	0.557	0.315	0.213	0.144
Conductivity (S/cm)	2	29	38	60	106	156	231
TGA residue (%)	0	0.51	0.92	2.33	4.24	5.82	10.59



PANI composite	PANI-plane	PANI-petal	PANI/RGO-petal
Resistance (Ohm)	18.17	28.3	0.2654
Conductivity (S/cm)	1.83	1.18	125.60
TGA residue (%)	0.00	0.03	6.07

Support information Fig. S4

The electrochemical capacities of reduced graphene oxide (RGO) content in the PANI/RGO-petal composite.



The conductive resistance of the PANI/RGO-petal hybrid decreased as the RGO loading content increased from 0.5 to 10 wt% (The petal-like hybrid film was easily cracked by peeling from template substrate at RGO content over 10 wt%);

Support information Fig. S5

The coulombic efficiency of PANI/RGO-petal composite and inset graphic is charge discharge curves between start and the end in the period of cycle testing