

Electronic Supplementary Information

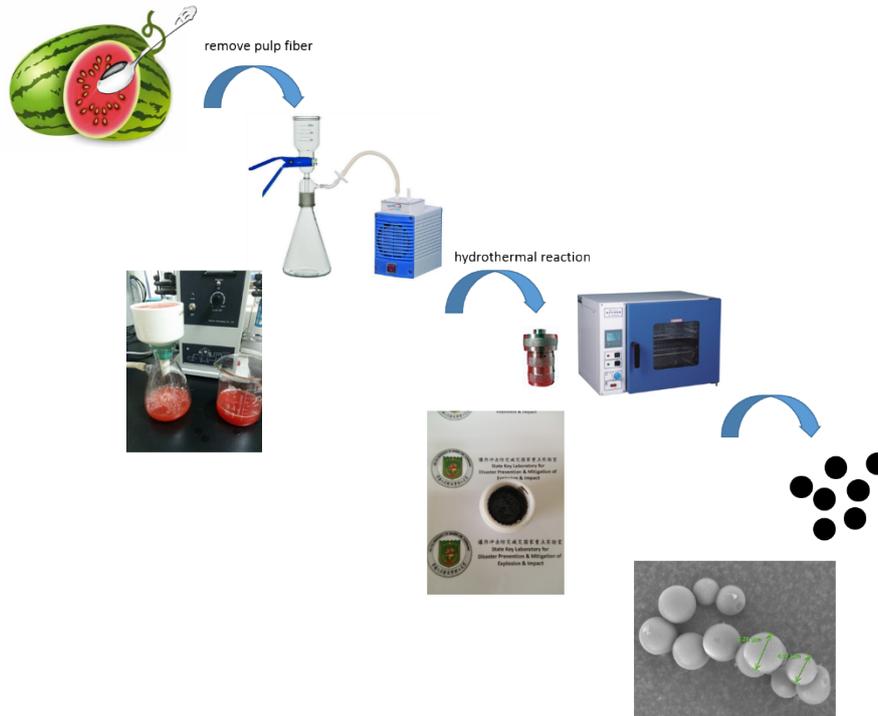
Materials

Watermelon was purchased from the supermarket in Nanjing. Ethanol (95%) was purchased from GENERAL-REAGENT, Titan Scientific Co., Ltd, Shanghai, China.

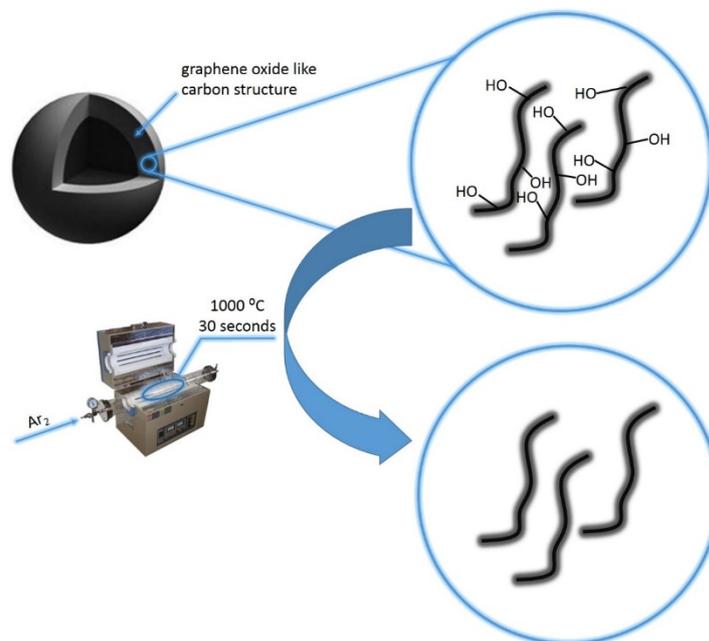
Distilled water was obtained from Direct-Q3 UV, Millipore.

Synthesis process

First, the pulp of watermelon was scooped and squeezed into juice. After that, the juice was filtered to remove the pulp fiber. Then 15 ml of juice and 45 ml of distilled water was mixed, and the above mentioned solution was sealed in a 100-mL Teflon-lined autoclave and maintained at 180 °C for 20 hours. Then the autoclave was naturally cooled to room temperature and the as-prepared dark purple puce slurry was obtained. A rinsing process involving several cycles of washing and re-dispersion was performed with hot water (60 °C) and ethanol (95%), respectively. The final sample of carbonaceous spheres (CS) were obtained after oven-drying at 50 °C. This synthesis process has been summarized in scheme 1. To remove the oxygen groups on the CS surface, a method, similar to that of McAllister et al.¹, was used to prepare graphene sheets containing less oxygen. These CS were putted into a tubular furnace at 1000 °C for 30 seconds under argon flow. This synthesis process has been summarized in scheme 2.



Scheme 1. Synthesis strategy for biomass-derived CS.



Scheme 2. Annealing process to remove the oxygen groups from CS surface.

Characterization and measurement

The size and morphology of the carbonaceous spheres and energy dispersive X-ray (EDX) analysis were measured using a scanning electron microscope (SEM, Phenom

Prox, Philips) and field emission high resolution transmission electron microscope (FE- HRTEM, Tecnai G2 F30 S, FEI). X-ray photoelectron spectra (XPS) was carried out in a Thermo Scientific ESCALAB 250Xi X-ray photoelectron spectrometer equipped with a monochromatic Al K α X-ray source (1486.6 eV). The crystal structure of the as-synthesized samples was identified by X-ray diffractometer (XRD, D8 Advance, Bruker AXS) from 5° to 50°, using Cu K α ($\lambda = 1.54 \text{ \AA}$) radiation. Raman spectroscopy was carried out on a Renishaw in Via Raman Microscope, equipped with 532 nm laser.

S 1.4 Dielectric and electromagnetic absorption property characterization

The relative complex permittivity (ϵ_r) and permeability (μ_r) wer measured by a vector network analyzer (VNA, N5242A PNA-X, Agilent) in the frequency range of 2-18 GHz. The measured samples were prepared by uniformly mixing 5.0 wt%, 15.0 wt% and 25.0 wt% of carbonaceous spheres with a paraffin matrix at 100 °C. The mixture was then pressed into toroidal shaped samples with an outer diameter of 7.00 mm and inner diameter of 3.04 mm. In a coaxial wire analysis, ϵ_r of the dielectric material has been calculated from the experimental scattering parameters S_{11} (or S_{22}) and S_{21} (or S_{12}) using the standard Nicolson-Ross-Weir (NRW) algorithm.^{2,3}

Due to the frequency range is from 2-18 GHz, the source-to-shield distance be greater than the free-space wavelength, so the measurements are considered under far field.⁴

According to the transmission line theory,⁵ the input impedance (Z_{in}) on the interface can be expressed as

$$Z_{in} = Z_0 \sqrt{\frac{\mu_r}{\epsilon_r}} \tanh\left(j \frac{2\pi f d}{c} \sqrt{\epsilon_r \mu_r}\right) \quad (1)$$

Where Z_0 is the impedance of free space, μ_r is the complex permeability, $\mu_r = \mu' - j\mu''$,

ε_r is the complex permittivity, $\varepsilon_r = \varepsilon' - j\varepsilon''$, f is the frequency, d is the thickness of material, c is the speed of light.

On the basis of the model of metal backplane, the reflection loss (RL) of a sample is determined from Z_0 and Z_{in} according to the following equation

$$RL(dB) = 20 \lg \left| \frac{Z_{in} - Z_0}{Z_{in} + Z_0} \right| \quad (2)$$

When the RL is lower than -10 dB, 90% of the electromagnetic energy is absorbed.

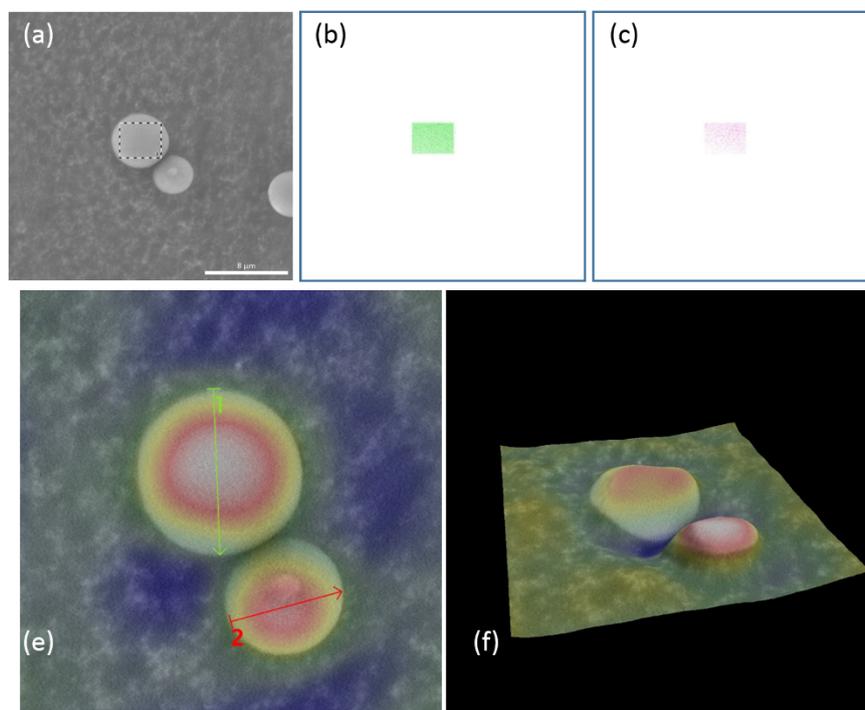


Fig. S1 SEM image of CS (a); EDX mapping of elements of C (b) and O (c); 3D simulation of CS (e, f)

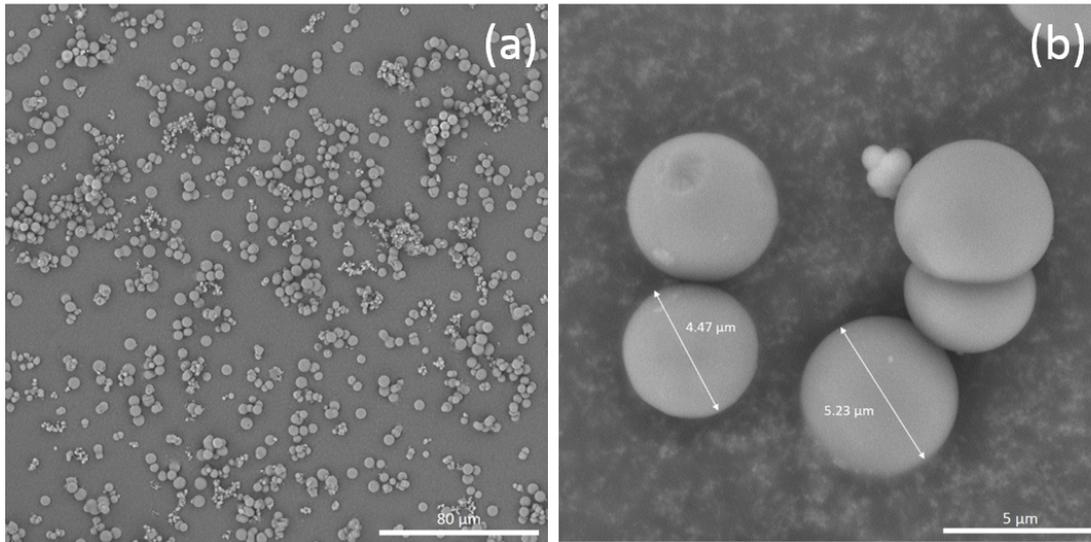


Fig. S2 SEM images of CS.

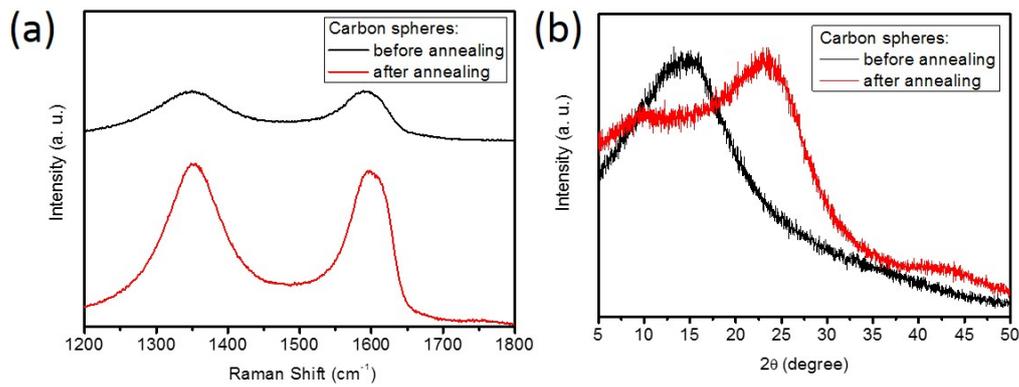


Fig. S3 Raman (a) and XRD spectrum of CS before and after annealing.

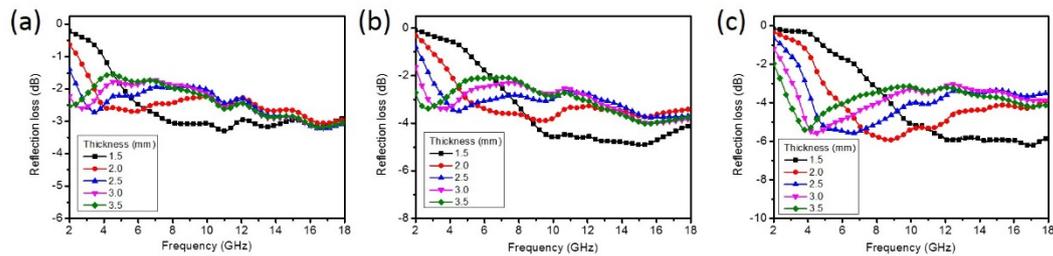


Fig. S4 The calculated RL for paraffin composites with (a) 5.0 wt%, (b) 15.0 wt% and (c) 25.0 wt% of CS (before annealing).

References

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Alonso, D. L. Milius, R. Car, R. K. Prud'homme and I. A. Aksay, *Chem. Mater.*, 2007, **19**, 4396.

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