Electronic Supplementary Information (ESI)

Efficient electromagnetic interference shielding of lightweight graphene/polystyrene composite

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Experimental

The functionalized graphene sheet (FGS) was prepared by directly exfoliating graphene oxide (GO) at 900 °C for 30s under the protection of nitrogen. The oxygen content in FGS was 8.5% using XPS measurement. PS was a commercial polystyrene (5250) kindly

- ¹⁵ supplied from Taiwan. FGS was suspended in absolute alcohol (C_2H_5OH) and the mixture was subjected to ultrasound and strong stirring for 60 min to create a homogeneous dispersion. At the same time, PS was completely dissolved in dichloromethane (CH_2Cl_2). By dropping the FGS/ C_2H_5OH dispersion into the PS/ CH_2Cl_2 hybrid, coagulated material precipitated continuously. Subsequently, C_2H_5OH was poured into the mixture until no more coagulated material precipitated. The dried FGS/PS particle was measured to own the particle diameter in the range of 20–40 µm. The following procedures were mentioned in the text. The cellular morphology of the samples was
- 20 observed under a Field-emission scanning electron microscopy (Inspect F, FEI, Finland) with an accelerating voltage of 20 kV. The specimens were frozen in liquid nitrogen for 1 hour, and then quickly impact fractured, sputter coated with gold. Transmission electronic microscopy observations were carried out using a FEI Tecnai F20 at an acceleration voltage of 200 kV. The composite sample was prepared with a thickness of 50–60 nm via a Leica EMUC6/FC6 microtome. EMI shielding characteristics of the porous composites were evaluated in the frequency range of 8.2-12.4 GHz (X band) by an Agilent vector network analyzer (VNA) in 201 data points. All the
- ²⁵ porous samples with 10 mm diameter and 2.5 mm thickness were placed between two X-band waveguide parts, which were connected to separate ports of the VNA. The VNA sent a signal down the waveguide upon the sample and the scattering parameter (S_{11} and S_{21}) were gained to calculate the EMI shielding effectiveness (SE_{total}, SE_R and SE_A). Electrical properties of the conductive porous composites were carried out using a four-point probe instrument (SDY-6, Guangzhou, China). Thermal conductivity was measured by the transient plane source (TPS) technique using a Hot Disk 2500-OT equipment at room temperature according to ASTM C518-91.

X-ray photoelectron spectroscopy (XPS) analysis of FGS.



Fig. SI1 XPS spectra of FGS after reaction at 900° C for 30 s in nitrogen atmosphere.

³⁵ **EMI** shielding effectiveness (SE_{total}), microwave reflection (SE_R) and microwave absorption (SE_A) from scattering parameters From the measured scattering parameters (S11 and S12), the power coefficients of reflectivity (R), transmissivity (T), and absorptivity (A) can be obtained, with their relationship described as R+T+A = 1. Then The EMI SE SE_{total}, SE_R, SE_A can be calculated as follows:

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$SE_{total} = -10\log T$	(1)
$SE_R = -10\log(1-R)$	(2)
$SE_A = SE_{total} - SE_R - SE_M$	(3)

where SE_M is the microwave multiple internal reflections, which can be can be negligible when SE_{total} \geq 15 dB.

Effect of pressure and temperature on electrical and EMI properties

Table SI1 The effect of pressure and temperature on electrical and EMI properties of the composites was shown in the supplementary information

Molding condition	235 °C	235 °C	235 °C	180 °C	180 °C
Physical properties	1000 MPa	100 MPa	10 MPa	1000 MPa	10 MPa
Electrical Conductivity (S m ⁻¹)	1.25	0.89	1.05	0.94	0.68
Average EMI SE (dB)	29	23	25	24	21

The effect of pressure and temperature on electrical conductivity and average EMI SE of the composites with a density 0.45 g cm⁻³. Was ¹⁰ were shown in Table S1, which indicated that the molding pressure and temperature had positive influences on both the electrical conductivity and average EMI SE of the porous composites. High pressure and temperature were benefit to ensure the sufficient molecular diffusion and good bonding of FGS/PS particles, building more perfect interconnected graphene network in the polymer matrix, which resulted in the improvement of the electrical conductivity and average EMI SE. Thus we chose 235 °C and 1000 MPa to prepare our porous composites.

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Electrical and thermal conductivity of porous composites

Table SI2 Electrical and thermal conductivity of pure PS and porous FGS/PS composite.

Mark	Sample	Electrical	Thermal conductivity
		conductivity (S m ⁻¹)	$(W m^{-1} K^{-1})$
PS	PS	10-16	0.08
GPS045	FGS/PS	1.25	0.11
GPS027	FGS/PS	0.22	0.07

As shown in Table SI2, the introduction of FGSs to the porous composites brings out a considerable increase of electrical conductivity $_{20}$ (σ) up to 1.25 and 0.22 S m⁻¹, 15 to 17 magnitudes higher than that of pure PS (10^{-16} S m⁻¹), however, the maximum thermal conductivity (κ) is only 1.5 times larger than that of pure PS. The low κ is ascribed to high air volume and the open-cell structure. The high σ and low κ endow the porous FGS/PS composite with the thermoelectric performance and potential application as thermoelectric materials.

25 Comparative data of storage modulus and electrical conductivity for porous pure PS, porous FGS/PS and graphite/PS composite



Fig. SI2 Temperature dependence of storage modulus (a) electrical conductivity (b) for porous pure PS, 30 wt% FGS/PS and 30 wt% graphite/PS composites at the same density 0.45 g cm⁻³.

³⁰ The results indicated that FGSs work more effectively than graphite in mechanical and electrical properties enhancement of the porous PS composites, which should be attributed to the higher aspect ratio and specific surface area of FGSs than graphite. Thus the FGSs were more easily to build perfect intensive and interconnected conductive network in the porous PS system, resulting in higher mechanical and electrical properties. In addition, the result indicated that storage modulus of 30 wt% FGSs/PS porous composites was only 10% lower than that of porous pure PS at the same density 0.45 g cm⁻³. The inconspicuous deterioration of the mechanical property even at the very ³⁵ high loading of 30 wt % should be attributed to the high pressure and temperature in the molding process, which ensured the sufficient

³⁵ high loading of 30 wt % should be attributed to the high pressure and temperature in the molding process, which ensured the su molecular diffusion and good bonding of FGS/PS particles to improve the mechanical properties of the composites.