Electronic Supplementary Information (ESI)

Graphene/Carbonyl Iron Cross-Linked Composites with Excellent

Electromagnetic Absorption Properties

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Fig.S1 Thermogravimetric curves of RGO-SCI (2) composite and SCI (inset) at a rate of 10 $^{\circ}$ C min⁻¹ in air.

The thermostability is an important property of electromagnetic wave absorption materials, which is related to practical applications. Fig.S1 shows thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) curves of RGO-SCI (2) composite and SCI at a heating rate of 10 °C min⁻¹ in air. The weight increase step of SCI is mainly attributed to the oxidation of Fe, corresponding to several exothermic processes can be seen in DSC curves.

The TGA/DSC curves of RGO-SCI exhibit a small weight loss below 150°C, corresponding to the desorption of water molecules located between the RGO nanosheets.¹ The steady weight loss from 150 to 400 °C is assigned to the decomposition of the oxygen functional groups remaining in RGO and the formation

of CO, CO₂, H₂O and C, corresponding to a sharp exothermic peak at 195 $^{\circ}$ C in the DSC curve of RGO-SCI.²⁻⁴ The weight increase process after 470 $^{\circ}$ C may be caused by the oxidation of Fe accompanied by an exothermic DSC peak.



Fig.S2 FTIR spectra of SCI and APS modified SCI.

To character the APS was successfully grafted onto the surface of SCI during our process, FTIR spectra of SCI and APS modified SCI is measured and shown in Fig.S2. The characteristic IR peaks at 1056 and 794 (552) cm⁻¹ assigned as Si-O-Si and SiO-H bending vibration, indicating that APS was grafted onto SCI successfully.³² The other characteristic peaks of APS such as N-H and C-N are not obvious due to its tiny addition.



Fig.S3 EDS image of RGO-SCI (4) composite.

Energy-dispersive X-ray spectroscopy (EDS) was tested to confirm the elemental composition of RGO-SCI (4) in Fig.S3. The measurement data shows the presence of Fe, C and Si. It indicates a weight ratio of Fe/C \approx 7.7/1, smaller than the initial mass ratio of 20/1, suggesting the partial loss of Fe in the preparation procedure. Besides, A Silicon content of 0.02 wt% can be neglected which was incorporated from APS.



Fig.S4 Frequency dependence of (a) complex permittivity, (b) complex permeability of RGO-SCI (2) and None-APS; (c) The calculated reflection losses of None-APS paraffin wax composites versus frequency and (d) comparison of electromagnetic wave reflection losses of the two samples with the thickness of 2 mm. (None-APS is the contrast experiment of RGO-SCI (2) without APS).

In order to explain the APS effect on the electromagnetic wave absorption property of RGO-SCI, the property of a contrast experiment of RGO-SCI (2) without APS denoted as None-APS was provided to compare with RGO-SCI (2), as shown in Fig.S4. Obviously, both ε' and ε'' of None-APS are higher than that of RGO-SCI (2) and the complex permeability is barely changed. The calculated reflection losses of None-APS with different thickness are shown in Fig.S4 (c) and the comparision of the two samples with the thickness of 2 mm is shown in Fig.S4 (d). The RL of None-APS is poor to compare with RGO-SCI (2). We believe that the decrease absorption property of None-APS is caused by the unbalanced electromagnetic parameters due to the increase permittivity, deriving to the lacking of APS. Therefore, the addition of APS has significantly improved the electromagnetic wave absorption properties of the composites.



Fig.S5 Raman spectra of SCI and H-SCI (SCI treated by APS and HCl).

Raman spectra of SCI and H-SCI are shown in Fig.S5. For α -Fe₂O₃, seven phonon lines are expected in the Raman spectrum, namely two A_{1g} modes (225 and 498 cm⁻¹) and five E_g modes (247, 293, 299, 412 and 613cm⁻¹), shown in the Raman spectra of H-SCI. The peaks at 300, 530 and 660 cm⁻¹ are attributed to the present of α -Fe₃O₄, and the other band corresponds to α -Fe₂O₃, indicating that the surface of pristine SCI is oxidized to the mixture of α -Fe₃O₄ and α -Fe₂O₃. And α -Fe₃O₄ was oxidized to α -Fe₂O₃ on the surface of H-SCI.³⁷ Here, it could be seen when adding SCI in the composite, the peaks corresponding to SCI disappear due to the covering of RGO networks.

References

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