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Supporting Information

Sandwich-type phase change composites with dual-function of efficient heat management and temperature-regulated electromagnetic interference shielding performance

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S1 Experimental section

S1.1 Materials

Expanded graphite (EG), average particle size 50 mesh, purchased from Qingdao Tengshengda carbon graphite. Multiwalled carbon nanotubes (MWCNTs) are purchased from Nanjing Jicang Nanotechnology Co., Ltd. Octane (ES, ρ=0.8±0.1g/cm3, melting point 35-37°C) was purchased from Shanghai Aladdin Biochemical Technology Co., Ltd. Polyvinylidene fluoride (PVDF, molecular weight Nw = 1 million) was purchased from Zhanyang Polymer Company, Dongguan. Iron oxide (II, III) (Fe_3O_4, 50 – 150 nm) was provided from Aladdin Chemistry Co. Ltd., China.

S1.2 Characterizations

Cu-Kα radiation (40 KV, 30 mA) was used at room temperature, and the scanning speed was 10°/min. The crystal structures of the materials from 10°to 90°were recorded by X-ray diffraction (XRD, X'Pert, PANalytical, Netherlands). Scanning electron microscopy (SEM, SU8020, Hitachi, Japan) was used to observe the microstructure at 5KV accelerating voltage. Before SEM testing, all surfaces were sputtered with gold to provide enhanced conductivity. Differential scanning calorimetry (DSC, DSC-Q2000, TA Instrument Company, USA) was used to test the phase transition temperature and latent heat in the range of 0°C to 80°C at 10°C/min. The uncertainty of DSC measurement for temperature and enthalpy was 0.05°C and 3.0%, respectively. Thermogravimetric analyzer (TGA, SDT650, TA Discovery, USA) was used to heat from 25°C to 600°C at 10°C/min in nitrogen atmosphere. The thermal diffusivity of each sample was measured by a laser thermal conductivity meter (LFA 457, NETZSCH, Germany). The thermal conductivity was calculated by the formula, where is the thermal diffusivity, is the density, and is the constant pressure specific heat capacity. The volume conductivity was measured by a four-probe

instrument (ST2258C, Suzhou Lattice Electronics Co., Ltd.). Each sample was tested at four different points, and the average value of the results was taken. At room temperature, the magnetic measurement system (MPMS XL5, Quantum Design, USA) was used to test the magnetization of Blend and PVDF/Fe_3O_4 spinning membrane with Fe_3O_4. The electromagnetic interference of samples in X-band (8.2-12.4GHz) was measured by vector network analyzer using waveguide method. Thermal management performance of sandwich structure materials is recorded by infrared thermal imager (Fluke TIs75, USA). Thermal cycling test was carried out in a dry incubator (JX-20, Shanghai Jingxin Industrial Development Co., Ltd.) between 20°C and 70°C.

S2 Results and discussions

S2.1 Attribution of diffraction peaks

According to Fig. S1A, the diffraction peak at 2θ=27° in PCCs belongs to the result of the interaction between eicosane and expanded graphite. Compared with the crystal plane at 2θ=27.06° (111) of eicosane, it has a slight left shift. The diffraction peak 2θ=26.56° of the expanded graphite belongs to the (002) crystal plane, and the similar diffraction peak 2θ=26° corresponds to the characteristic peak of the (002) crystal plane of the multi-walled carbon nanotubes belonging to the graphite. The peaks 2θ=20.1°, 24.15°and 27.06° of n-octadecane belong to (010), (100) and (111) crystal planes, respectively.

S2.2 XRD of PCCs

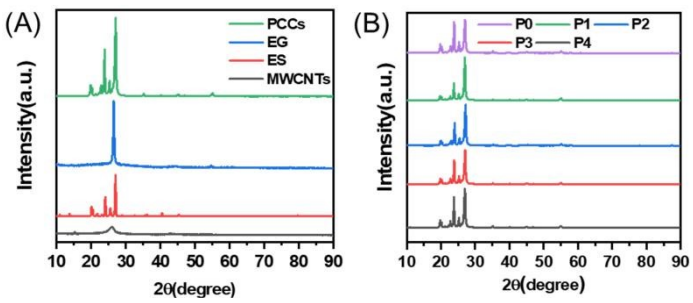

Fig.S1 (A) XRD of PCCs, EG, ES and MWCNTs. (B) XRD of P0, P1, P2, P3, P4.

S2.3 SEM and EMI SE of Magnetic spinning film (MSF)

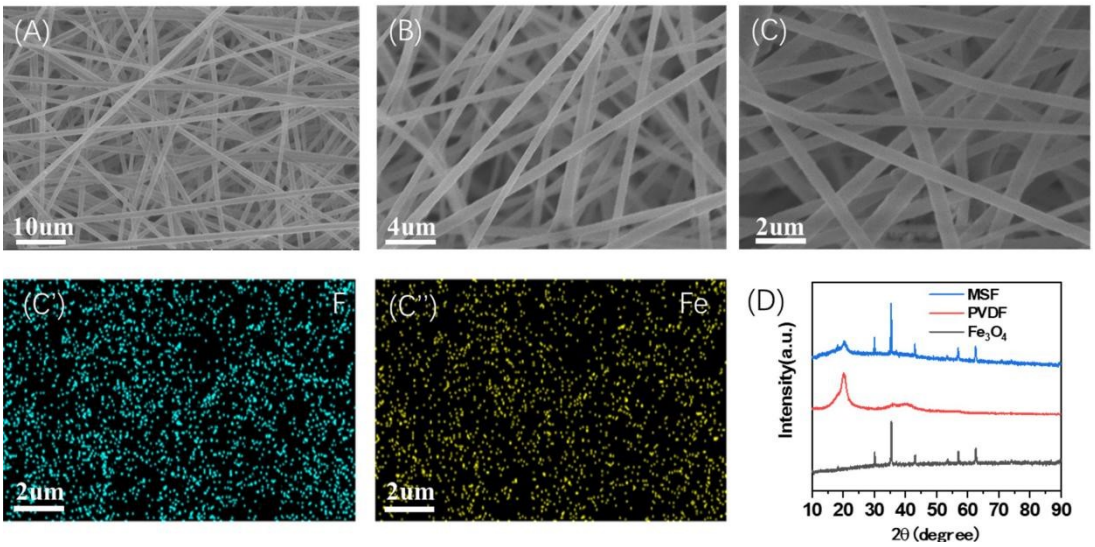

Fig.S2 (A-C) SEM images of Magnetic spinning film (MSF), (C', C'') EDS mapping of elements F and Fe, (D) XRD of MSF, Fe_3O_4 and PVDF.

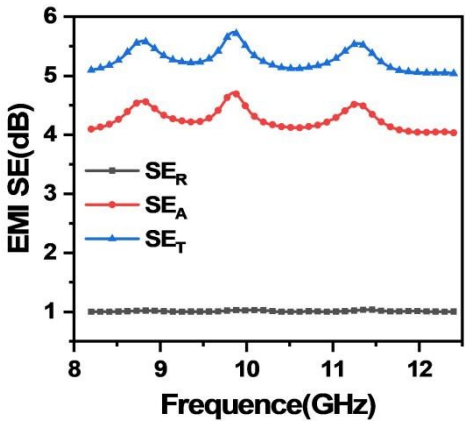

Fig.S3 EMI SE of MSF.

S2.4 Magnetism of MSF, BC and Fe_3O_4

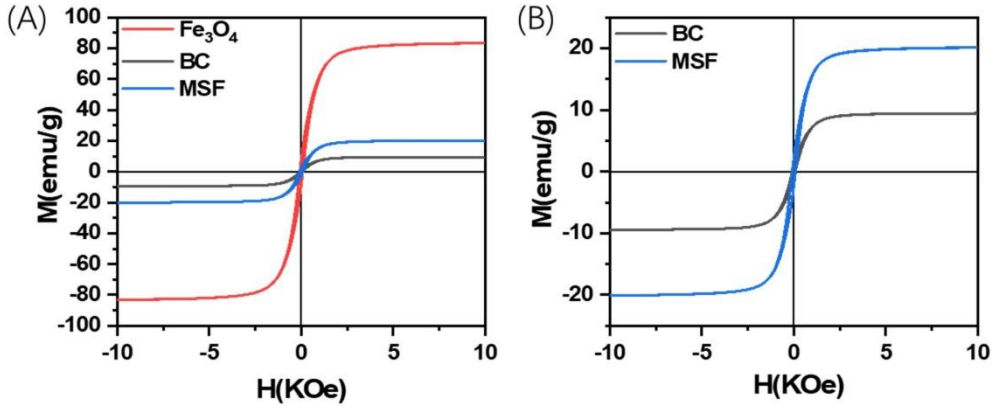

Fig. S4 (A) Field dependent magnetization curves of Fe_3O_4, BC and MSF. (B)Field dependent magnetization curves of BC and MSF.

S2.5 SEM and EDS of BC

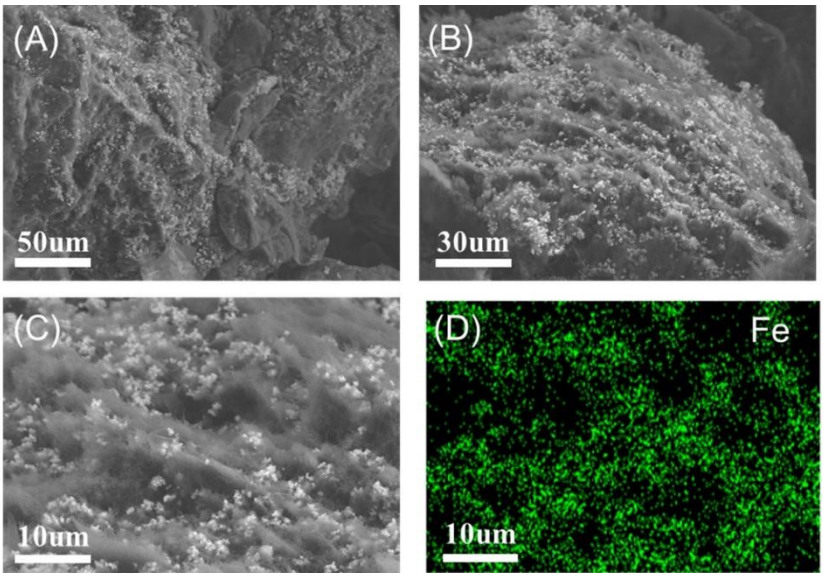

Fig. S5 (A-C) SEM images of BC, (D) EDS mapping of elements Fe.

S2.6 Heat-resistance index (THRI) values of ES and STPCCs

Table.S1 Heat-resistance index (THRI) values of ES and STPCCs

Samples	Weight loss temperature/°C		THRI */ °C
	T_5	T_{30}	
ES	167.9	211.5	95.1
SC0	179.2	222.2	100.5
SC1	185.4	225.3	102.6
SC2	184.6	224.6	102.2
SC3	185.4	225.3	102.6
SC4	180.8	228.2	102.5

*THeat-resistance index=0.49×[T_5+0.6×(T_{30}-T_5)]. T_5 and T_{30} represent the corresponding decomposition temperatures of 5 wt% and 30 wt% weight loss, respectively.

S2.7 Shape stability of ES, PCCs, STPCCs, and BC

$$\omega = \frac{m_1 - m_n}{m_1} \times 100\%$$

m_1 is the initial weight of the sample and m_n is the weight of the sample after each time it is kept for a

certain heating time.

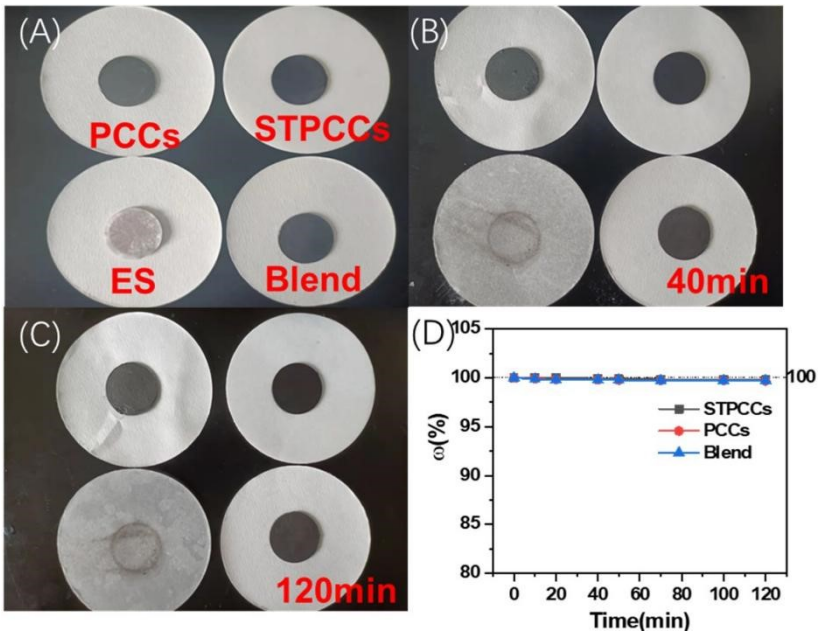

Fig. S6 (A-C) Digital photos of ES, PCCs, STPCCs, and BC maintained at 70°C in an oven for 2 hours, (D) Weight statistics of ES, PCCs, STPCCs, and BC during leakage experiments

S3 The EMI shielding and Thermal conductivity properties of the reported works.

Table.S2 The EMI shielding properties of the reported works

Materials	Thick/mm	SE_T/dB	Thermal conductivity（W/mK）	Ref.
PP/CNTs/Fe_3O_4/PW	2	39.2	0.59	1
WPC/PEG/Fe_3O_4	4.91	55.09	-	2
MXene/PCM	3	64.7	0.74	3
PW@CNS	3	29.4	1.85	4
Scaffold/PW	1.5	37.7	0.38	5
MF@MA/PEG	2	30.5	0.196	6
MPAχ-PEG PCM	1.5	82.02	0.42	7
NiM/PCM	2	34.6	0.39	8
PP/CNT&EPDM/PW SSPCC	1	57.2	0.469	9
MSF/PCCs/MSF	**1**	**98.39**	**3.61**	**This word**

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