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## **Supplementary Information**

## Fe<sub>3</sub>O<sub>4</sub>-functionalized graphene nanosheet embedded phase change material composites: efficient magnetic- and sunlight-driven energy conversion and storage

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Fig. S1. The FT-IR spectra of (a)  $Fe_3O_4$ -GNS synthesized with 10 mmol FeCl<sub>3</sub>, (b) PCM and (c)  $Fe_3O_4$ -GNS/PCM-4.

Fourier transform infrared spectrophotometer was recorded on Thermo Scientific Nicolet 6700 using KBr pellet and Nujol.

Fe<sub>3</sub>O<sub>4</sub>-GNS (Fig. S8-a): IR (KBr pellet): 1577 (v C=C), 1219 (v C-C), 578 (vs Fe-O).

PCM: ITR (Nujol) (Fig. S8-b): 3294 (v NH), 2885 (v CH2), 1727(v C=O), 1599 and 1538(v C=C aromatic), 1467 (δas CH2), 1359 (vas C-N aromatic), 1343 (δs CH2), 1280 (vas C-O-C), 1241 (vs C-N aromatic), 1112 (vs C-O-C).

Fe<sub>3</sub>O<sub>4</sub>-GNS/PCM-4: ITR (Nujol) (Fig. S8-c): 3287 (v NH), 2887 (v CH2), 1727(v C=O), 1599 and 1536(v C=C aromatic), 1467 (δas CH2), 1359 (vas C-N aromatic), 1343 (δs CH2), 1280 (vas C-O-C), 1241 (vs C-N aromatic), 1111 (vs C-O-C).



Fig. S2. The O1s spectrum of Fe<sub>3</sub>O<sub>4</sub>-GNS nanocomposites synthesized with 10 mmol FeCl<sub>3</sub>.



Fig. S3. HRTEM image of Fe<sub>3</sub>O<sub>4</sub>-GNS nanocomposite synthesized with 10 mmol FeCl<sub>3</sub>



Fig. S4. The AFM image of Fe<sub>3</sub>O<sub>4</sub>-GNS nanocomposite synthesized with 10 mmol FeCl<sub>3</sub>.



Fig. S5. (a) Room temperature magnetization loops in an applied magnetic field of up to 20 kOe for  $Fe_3O_4$ -GNS nanocomposites synthesized with 10 mmol  $FeCl_3$ , (b) The respective expanded plots for field between 2 k and 2 kOe, (c) The ZFC-FC curves at 500 Oe for  $Fe_3O_4$ -GNS nanocomposites synthesized with 10 mmol  $FeCl_3$ .



**Fig. S6.** (a) Magnetothermal measurement under the alternating magnetic field (1.36 MHz, 550  $A \cdot m^{-1}$ ) for Fe<sub>3</sub>O<sub>4</sub>-GNS nanocomposites synthesized with 10 mmol FeCl<sub>3</sub>, (b) UV-vis-NIR absorption spectrum in ethanol (0.06 mg·mL<sup>-1</sup>) for Fe<sub>3</sub>O<sub>4</sub>-GNS nanocomposites synthesized with 10 mmol FeCl<sub>3</sub>.



Fig. S7. Cross-sectional SEM images of PCM (a) and Fe<sub>3</sub>O<sub>4</sub>-GNS/PCM-4 composites (b).



Fig. S8. A schematic of the magnetic-to-thermal energy conversion and storage measuring system.



**Fig. S9.** (a) Magnetic-to-thermal energy conversion curves of the samples (0.5 g) under the alternating magnetic field (1.36 MHz, 550 A·m<sup>-1</sup>). (b) Calculated magnetic-to-thermal energy storage efficiencies for the Fe<sub>3</sub>O<sub>4</sub>-GNS/PCM composites with different content of Fe<sub>3</sub>O<sub>4</sub> in Fe<sub>3</sub>O<sub>4</sub>-GNS.



**Fig. S10.** (a) Solar-to-thermal energy conversion curves of the samples (3.0 g) under sunlight radiation (12:10-13:08, October 5, 2016, P=0.26 W, at an ambient temperature of around 19.5 °C, Dalian, China). (b) Calculated solar-to-thermal energy storage efficiencies for the Fe<sub>3</sub>O<sub>4</sub>-GNS/PCM composites with different content of Fe<sub>3</sub>O<sub>4</sub> in Fe<sub>3</sub>O<sub>4</sub>-GNS.



**Fig. S11.** DSC curves of Fe<sub>3</sub>O<sub>4</sub>/PCM, GNS/PCM and Fe<sub>3</sub>O<sub>4</sub>-GNS/PCM with different content of Fe<sub>3</sub>O<sub>4</sub> in Fe<sub>3</sub>O<sub>4</sub>-GNS.



**Fig. S12.** (a) Magnetic-to-thermal energy conversion curves of Fe<sub>3</sub>O<sub>4</sub>-GNS/PCM-2 (0.5g) before (black line) and after (red line) 100 cycles of magnetic heating (1.36 MHz, 550 A·m<sup>-1</sup>), (b) Light-to-thermal energy conversion curves of Fe<sub>3</sub>O<sub>4</sub>-GNS/PCM-2 (3.0 g) before (black line) and after (red line) 100 cycles under simulated sunlight irradiation (P=0.68 W, at an ambient temperature of around 20.5 °C).



**Fig. S13.** DSC curves of Fe<sub>3</sub>O<sub>4</sub>-GNS/PCM-2 before and after 100 magnetothermal and photothermal conversion and storage cycles.

**Table S1.** Phase change behavior of Fe<sub>3</sub>O<sub>4</sub>-GNS/PCM-2 before and after 100 magnetothermal and photothermal conversion and storage cycles.

Cycle	ΔH (J/g)			T <sub>r</sub> (°C)	
	Heating cycle		Cooling cycl	e Heating cycle	Cooling cycle
0	98.8		100.9	55.9	35.5
100 photothermal	101.6		102.1	56.5	34.5
100 magnetothermal	102.6		103.7	55.0	35.6
	30 °C	PEG 10000	PCM	Fe <sub>3</sub> O <sub>4</sub> -GNS/PCM nanocomposite	
	65 ℃				
	100 °C				

**Fig. S14.** Macroscopic morphology of the pure PEG, PCM, and Fe<sub>3</sub>O<sub>4</sub>-GNS/PCM nanocomposites at different temperatures.