Supplementary Information

Fe$_3$O$_4$-functionalized graphene nanosheet embedded phase change material composites: efficient magnetic- and sunlight-driven energy conversion and storage

Wentao Wang, Bingtao Tang,* Benzhi Ju, Zhanming Gao, Jinghai Xiu and Shufen Zhang

State Key Laboratory of Fine Chemicals, Dalian University of Technology, Dalian 116024, China

*E-mail: tangbt@dlut.edu.cn
Fig. S1. The FT-IR spectra of (a) Fe₃O₄-GNS synthesized with 10 mmol FeCl₃, (b) PCM and (c) Fe₃O₄-GNS/PCM-4.

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

Fe₃O₄-GNS (Fig. S8-a): IR (KBr pellet): 1577 (ν C=C), 1219 (ν C-C), 578 (vs Fe-O).

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

Fe₃O₄-GNS/PCM-4: ITR (Nujol) (Fig. S8-c): 3287 (ν NH), 2887 (ν CH₂), 1727(ν C=O), 1599 and 1536(ν C=C aromatic), 1467 (δ as CH₂), 1359 (vas C-N aromatic), 1343 (δ s CH₂), 1280 (vas C-O-C), 1241 (vs C-N aromatic), 1111 (vs C-O-C).

Fig. S2. The O1s spectrum of Fe₃O₄-GNS nanocomposites synthesized with 10 mmol FeCl₃.
**Fig. S3.** HRTEM image of Fe₃O₄-GNS nanocomposite synthesized with 10 mmol FeCl₃.

**Fig. S4.** The AFM image of Fe₃O₄-GNS nanocomposite synthesized with 10 mmol FeCl₃.

**Fig. S5.** (a) Room temperature magnetization loops in an applied magnetic field of up to 20 kOe for Fe₃O₄-GNS nanocomposites synthesized with 10 mmol FeCl₃, (b) The respective expanded plots for field between 2 k and 2 kOe, (c) The ZFC-FC curves at 500 Oe for Fe₃O₄-GNS nanocomposites synthesized with 10 mmol FeCl₃.
Fig. S6. (a) Magnetothermal measurement under the alternating magnetic field (1.36 MHz, 550 A·m$^{-1}$) for Fe$_3$O$_4$-GNS nanocomposites synthesized with 10 mmol FeCl$_3$, (b) UV-vis-NIR absorption spectrum in ethanol (0.06 mg·mL$^{-1}$) for Fe$_3$O$_4$-GNS nanocomposites synthesized with 10 mmol FeCl$_3$.

Fig. S7. Cross-sectional SEM images of PCM (a) and Fe$_3$O$_4$-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⁻¹). (b) Calculated magnetic-to-thermal energy storage efficiencies for the Fe₃O₄-GNS/PCM composites with different content of Fe₃O₄ in Fe₃O₄-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 Fe₃O₄-GNS/PCM composites with different content of Fe₃O₄ in Fe₃O₄-GNS.
Fig. S11. DSC curves of Fe$_3$O$_4$/PCM, GNS/PCM and Fe$_3$O$_4$-GNS/PCM with different content of Fe$_3$O$_4$ in Fe$_3$O$_4$-GNS.

Fig. S12. (a) Magnetic-to-thermal energy conversion curves of Fe$_3$O$_4$-GNS/PCM-2 (0.5g) before (black line) and after (red line) 100 cycles of magnetic heating (1.36 MHz, 550 A·m$^{-1}$), (b) Light-to-thermal energy conversion curves of Fe$_3$O$_4$-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$_3$O$_4$-GNS/PCM-2 before and after 100 magnetothermal and photothermal conversion and storage cycles.

Table S1. Phase change behavior of Fe$_3$O$_4$-GNS/PCM-2 before and after 100 magnetothermal and photothermal conversion and storage cycles.

<table>
<thead>
<tr>
<th>Cycle</th>
<th>ΔH (J/g)</th>
<th>T$_r$ (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Heating cycle</td>
<td>Cooling cycle</td>
</tr>
<tr>
<td>0</td>
<td>98.8</td>
<td>100.9</td>
</tr>
<tr>
<td>100 photothermal</td>
<td>101.6</td>
<td>102.1</td>
</tr>
<tr>
<td>100 magnetothermal</td>
<td>102.6</td>
<td>103.7</td>
</tr>
</tbody>
</table>

Fig. S14. Macroscopic morphology of the pure PEG, PCM, and Fe$_3$O$_4$-GNS/PCM nanocomposites at different temperatures.