

Supporting Information

PEG/3D graphene oxide network form-stable phase change materials with ultrahigh filling content

Jinjing Qiu,^a Xiaoqiao Fan,^a Yusheng Shi,^a Shufen Zhang,^a Xin Jin^b, Wentao Wang^c
and Bingtao Tang^{*a,b}

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

*b Eco-chemical Engineering Cooperative Innovation Center of Shandong, Qingdao University of Science &
Technology, Qingdao 266042, PR China*

*c Key Laboratory of Advanced Textile Materials and Manufacturing Technology, Ministry of Education, Zhejiang
Sci-Tech University, Hangzhou 310018, PR China*

Table S1. Components of PGCs.

Sample	PEG content (wt %)	Ca ²⁺ -GO content (wt %)
PGC0.5	99.5	0.5
PGC1.0	99.0	1.0
PGC2.0	98.0	2.0
PGC4.0	96.0	4.0
PGC6.0	94.0	6.0

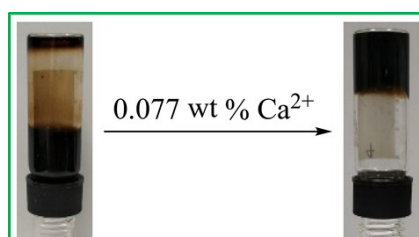


Fig. S1. Digital photographs of the reaction system for preparing GO gel before and after the Ca²⁺ cross-linking.

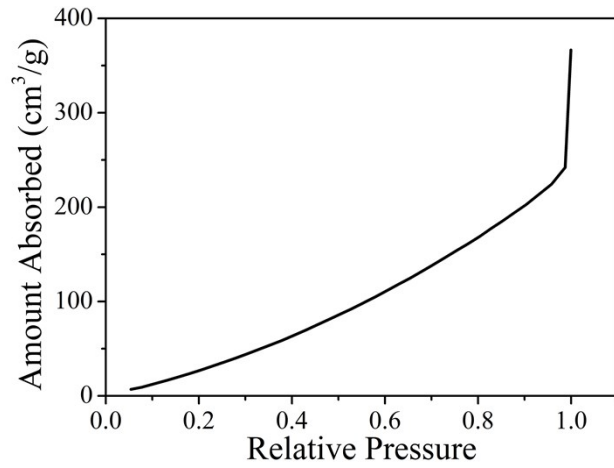


Fig. S2. Nitrogen sorption isotherm of GO aerogel cross-linked by Ca^{2+} .

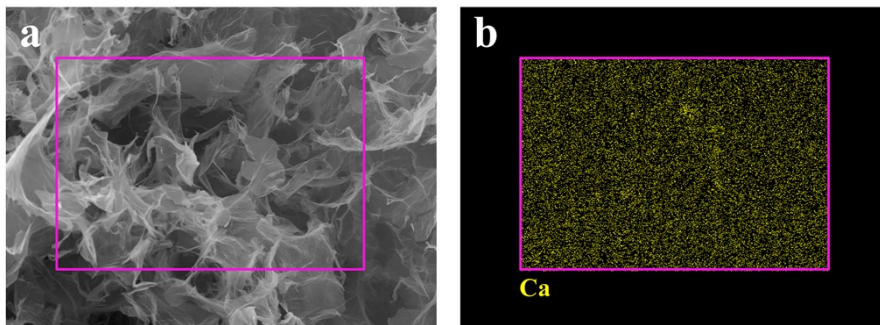


Fig. S3. EDS mapping of Ca on the surface of GO aerogel cross-linked by Ca^{2+} .

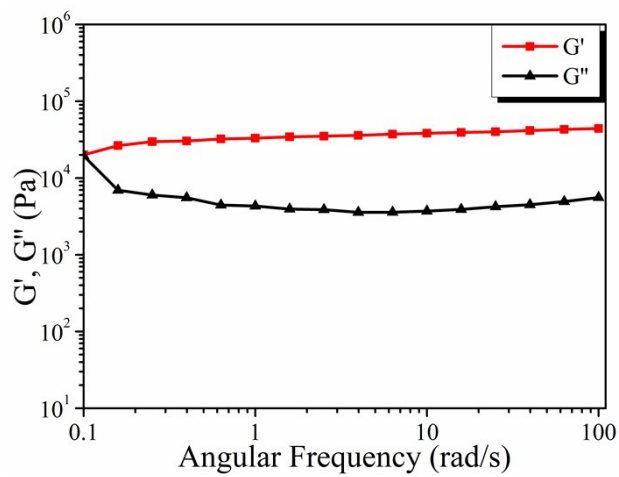


Fig. S4. Rheological property of the PGC1.0 gel.

Table S2. Phase change temperatures and enthalpies of PGCs.

Sample	Filling (wt %)	T _m (°C)	T _f (°C)	Δ H _m (J/g)	Δ H _f (J/g)
PGC6.0	94.0	60.5	40.8	189.7	178.2
PGC4.0	96.0	60.0	40.8	202.3	195.2
PGC2.0	98.0	61.2	41.7	212.4	207.2
PGC1.0	99.0	61.0	41.7	218.3	212.0
PGC0.5	99.5	62.2	42.7	218.9	213.2
PEG6000	--	62.4	42.1	221.7	217.8

Table S3. Thermal conductivities of PGCs and corresponding increasing ratio relative to PEG6000.

Sample	Filling (wt %)	Thermal conductivity (W·m ⁻¹ ·K ⁻¹)	Increasing ratio for thermal conductivity (%)
PGC6.0	94.0	0.396	87.7
PGC4.0	96.0	0.393	86.3
PGC2.0	98.0	0.384	82.0
PGC1.0	99.0	0.363	72.0
PGC0.5	99.5	0.344	63.0
Control sample	99.0	0.241	14.2
PEG6000	100.0	0.211	--

The leakage tests for the PGCs, pure PEG6000, and the control sample were conducted on a horizontal heating plate with a constant temperature for 20 min with varying temperatures, as shown in Fig. S5. The samples were prepared as 25 mm diameter disks with 5 mm thickness, and a digital camera was used to record the state of the samples. The control sample was produced by the direct mixing of a 99 wt % PEG6000 with GO (the control group in Fig. S5). The pure PEG6000 and the control sample started melting at 65 °C. However, the shapes of prepared PGCs were maintained without leakage at 65 °C, which is higher than the working temperature (the temperature range of phase transition is between 40.0 °C to 63.0 °C).

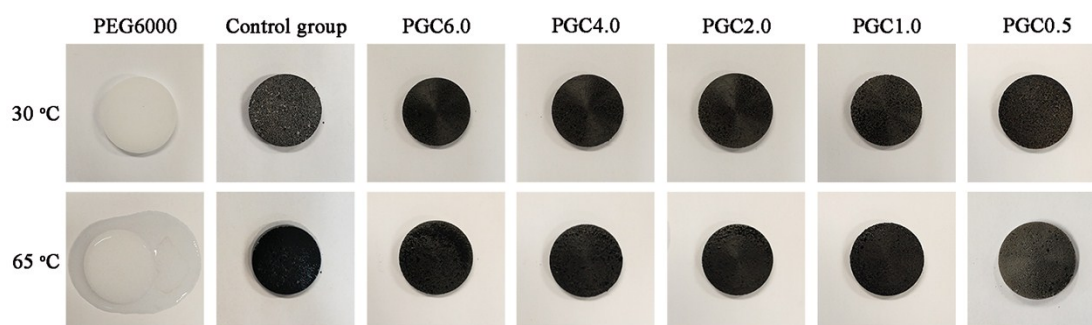


Fig. S5. Digital photographs for leakage test of the PGCs, PEG6000, and the control sample at different temperatures: 30 and 65 °C.

Table S4. Comparison of form-stable CPCMs prepared in this work and references.

Form-stable PCM	Preparation method	Melting enthalpy (J/g)	The mass fraction of supporting material (wt %)	Reference
PEG/GO/Boron nitride	Physical blending	107.4	34	1
PEG/GO	Physical blending	142.8	4	2
PEG/3D porous carbon	Physical blending and impregnation	160.3	15	3
PEG/Carbon nanotubes/Metal-organic frameworks	Physical blending and impregnation	83.1	30	4
PEG/Mesoporous silica	Blending and impregnation	88.2	30	5
PEG/Graphene aerogel fiber	Impregnation	124	17	6
PEG/GO/ Boron nitride	Vacuum Impregnation	145.6	19.0	7
PEG/Diatomite/Ag	Vacuum Impregnation	111.3	37	8
PEG/Ionic cross-linked GO	In situ filling by ionic cross-linking	218.9	0.5	This work

As shown in Table S4, in order to get stable shape, a large amount of supporting materials (higher than 4 wt %) is needed in physical blending or impregnation method reported by other references. Due to the influence of these supporting materials, the

melting enthalpies of the prepared CPCMs decrease below 160 J/g. Because of the excellent cross-linking ability of calcium ion, the 3D GO skeleton was rapidly formed to allow in situ filling more PEG in this study. The filling capacity reached to 99.5 wt %, and the melting enthalpy is 218.9 J/g, which is close enough to that of pure PEG. More importantly, the form-stable PGCs can be simply prepared through in situ filling by ionic cross-linking, which avoid re-impregnation.

Table S5. The thermal stability of the PGCs from TGA.

Sample	Filling (wt %)	5% weight loss temperature (°C)	Fast weight loss temperature (°C)	Char yield at 800 °C (wt %)
PGC6.0	94.0	355.4	405.2	5.8
PGC4.0	96.0	363.7	408.1	4.5
PGC2.0	98.0	366.2	406.9	2.0
PGC1.0	99.0	370.4	407.8	1.6
PGC0.5	99.5	371.5	408.9	1.4
PEG6000	--	387.2	414.2	1.9

References

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