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Supplementary Information

Preparation and Properties of a Novel Form-stable Phase Change Material Based on Gelator

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1. Synthesis of gelators:

Take *N, N′*-distearoyl-4, 4' -diaminodiphenylmethane (G18) as the example, the details were described as follows: First, synthetic stearoyl chloride: 22.72 g (0.08 mol) stearic acid and 30 ml THF was placed into a 250 ml round bottomed flask equipped with a magnetic stirrer, followed by drop 12.00 g (0.12 mol) thionyl chloride at room temperature. Then, the flask was equipped with a condenser and heated to reflux with an oil bath. After refluxing for 4 h, the excess thionyl chloride was removed by distillation under reduced pressure. The stearoyl chloride product was a light red liquid. Afterwards, a solution of 6.34 g (0.032 mol) 4, 4′-diaminodiphenylmethane and 8.50 g (0.082 mol) triethylamine in 30 ml THF was added dropwise to the stirred acid chloride at 40 °C. The mixture was stirred for 2 h and was subsequently poured into a large amount of acetone. A white precipitate was filtered off, and then washed with 5 wt% NaOH aqueous solution, 1 wt% HCl aqueous solution and pure THF two times, respectively. Yield: 90%. mp: 167.8 °C. ^{1}H NMR ($CDCl_3$, ppm, δ): 0.82-0.94 (h, 6H, -C*H*$_3$), 1.14-1.43 (g, 32H, -C*H*$_2$-), 1.66-1.78 (f, 4H, -C*H*$_2$-), 2.27-2.40 (e, 4H, -C*H*$_2$-), 3.87-3.93 (a, 2H, -C*H*$_2$-), 7.04 (d, 2H, -N*H*-CO-), 7.07-7.19 (b, 4H, Ar-*H*), 7.38-7.49(c, 4H, Ar-*H*). Mass Spectrometry (MS) (m/z) [M] Calcd for $C_{49}H_{82}N_2O_2$, 731.19; found, 731.751.

N, N′-hexadecanoyl-4, 4'-diaminodiphenylmethane (G16): Yield: 88%. mp: 176.2 °C. ^{1}H NMR ($CDCl_3$, ppm, δ): 0.80-0.92 (h, 6H, -C*H*$_3$), 1.13-1.45 (g, 48H, -C*H*$_2$-), 1.64-1.77 (f, 4H, -C*H*$_2$-), 2.25-2.40 (e, 4H, -C*H*$_2$-), 3.86-3.93 (a, 2H, -C*H*$_2$-), 7.04 (d, 2H, -N*H*-CO-), 7.07-7.19 (b, 4H, Ar-*H*), 7.38-7.49(c, 4H, Ar-*H*). Mass Spectrometry (MS) (m/z) [M] Calcd for $C_{45}H_{74}N_2O_2$, 675.08; found, 675.833.

N, N′-myristoyl-4, 4'-diaminodiphenylmethane (G14): Yield: 85%. mp: 178.3 °C. ^{1}H NMR ($CDCl_3$, ppm, δ): 0.81-0.94 (h, 6H, -C*H*$_3$), 1.13-1.43 (g, 40H, -C*H*$_2$-), 1.66-1.77 (f, 4H, -C*H*$_2$-), 2.25-2.40 (e, 4H, -C*H*$_2$-), 3.86-3.93 (a, 2H, -C*H*$_2$-), 7.04 (d, 2H, -N*H*-CO-), 7.07-7.19 (b, 4H, Ar-*H*), 7.38-7.49(c, 4H, Ar-*H*). Mass Spectrometry (MS) (m/z) [M] Calcd for $C_{41}H_{66}N_2O_2$, 618.97; found, 619.749.

N, N′-dodecanoyl-4, 4'-diaminodiphenylmethane (G12): Yield: 86%. mp: 183 – 180.8 °C. ^{1}H NMR ($CDCl_3$, ppm, δ): 0.80-0.97 (h, 6H, -C*H*$_3$), 1.08-1.47 (g, 32H, -C*H*$_2$-), 1.64-1.76 (f, 4H, -C*H*$_2$-), 2.27-2.39 (e, 4H, -C*H*$_2$-), 3.90 (a, 2H, -C*H*$_2$-), 7.03 (d, 2H, -N*H*-CO-), 7.05-7.16 (b, 4H, Ar-*H*), 7.45-7.85(c, 4H, Ar-*H*). Mass Spectrometry (MS) (m/z) [M] Calcd for $C_{37}H_{58}N_2O_2$, 562.87; found, 563.667.

N, N′-decanoyl-4, 4'-diaminodiphenylmethane (G10): Yield: 85%. mp: 185.3 °C. ^{1}H NMR ($CDCl_3$, ppm, δ): 0.82-0.93 (h, 6H, -C*H*$_3$), 1.18-1.47 (g, 24H, -C*H*$_2$-), 1.62-1.76 (f, 4H, -C*H*$_2$-), 2.25-2.39 (e, 4H, -C*H*$_2$-), 3.91 (a, 2H, -C*H*$_2$-), 7.04 (d, 2H, -N*H*-CO-), 7.06-7.14 (b, 4H, Ar-*H*), 7.41-7.75(c, 4H, Ar-*H*). Mass Spectrometry (MS) (m/z) [M] Calcd for $C_{33}H_{50}N_2O_2$, 506.76; found, 507.421.

N, N′-caprylyl-4, 4'-diaminodiphenylmethane (G8): Yield: 88%. mp: 189.3 °C. ^{1}H NMR ($CDCl_3$, ppm, δ): 0.89 (h, 6H, -C*H*$_3$), 1.20-1.38 (g, 16H, -C*H*$_2$-), 1.60-1.76 (f, 4H, -C*H*$_2$-), 2.26-2.40 (e, 4H, -C*H*$_2$-), 3.89 (a, 2H, -C*H*$_2$-), 7.05-7.15 (b, 4H, Ar-*H*), 7.04 (d, 2H, -N*H*-CO-), 7.35-7.49(c, 4H, Ar-*H*). Mass Spectrometry (MS) (m/z) [M] Calcd for $C_{29}H_{42}N_2O_2$, 450.66; found, 451.514.

N, N′-hexanoyl-4, 4'-diaminodiphenylmethane (G6): Yield: 90%. mp: 192.5 °C. ^{1}H NMR ($CDCl_3$, ppm, δ): 0.90 (h, 6H, -C*H*$_3$), 1.34 (g, 12H, -C*H*$_2$-), 1.62-1.81 (f, 4H, -C*H*$_2$-), 2.25-2.40 (e, 4H, -C*H*$_2$-), 3.89 (a, 2H, -C*H*$_2$-), 7.03-7.15 (b, 4H, Ar-*H*), 7.19 (d, 2H, -N*H*-CO-), 7.35-7.49(c, 4H, Ar-*H*). Mass Spectrometry (MS) (m/z) [M] Calcd for $C_{25}H_{34}N_2O_2$, 394.55; found, 395.419.

N, N′-butyryl-4, 4'-diaminodiphenylmethane (G4): Yield: 88%. mp: 202.8 °C. ^{1}H NMR ($CDCl_3$, ppm, δ): 0.99 (g, 6H, -C*H*$_3$), 1.72-1.86 (f, 4H, -C*H*$_2$-), 2.19-2.25 (e, 4H, -C*H*$_2$-), 3.89 (a, 2H, -C*H*$_2$-), 7.05-7.16 (b, 4H, Ar-*H*), 7.21 (d, 2H, -N*H*-CO-),7.45-7.85(c, 4H, Ar-*H*). Mass Spectrometry (MS) (m/z) [M] Calcd for $C_{21}H_{26}N_2O_2$, 338.44; found, 339.265.

N, N′-acetyl-4, 4'-diaminodiphenylmethane (G2): Yield: 85%. mp: 233.8 °C.^{1}H NMR ($CDCl_3$, ppm, δ):2.06 (e, 6H, -C*H*$_3$), 3.87 (a, 2H, -C*H*$_2$-), 7.24 (d, 2H, -N*H*-CO-), 7.07-7.19 (b, 4H, Ar-*H*), 7.38-7.49(c, 4H, Ar-*H*). Mass Spectrometry (MS) (m/z) [M] Calcd for $C_{17}H_{18}N_2O_2$, 282.34; found, 283.147.

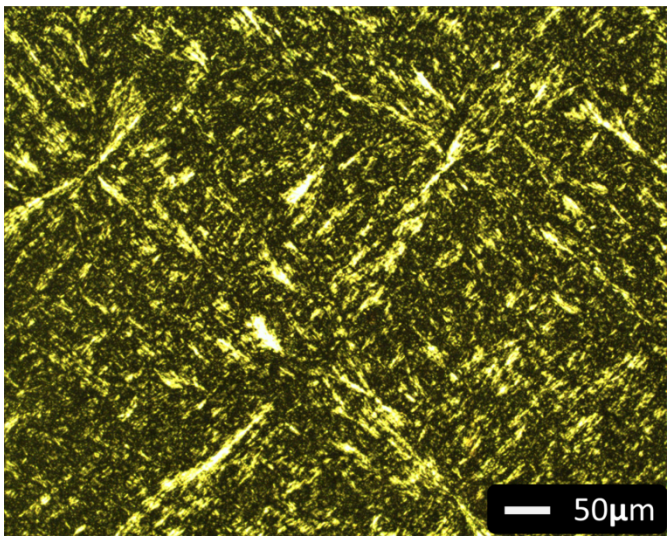

Fig.S1 POM images (×200) of neat G18.

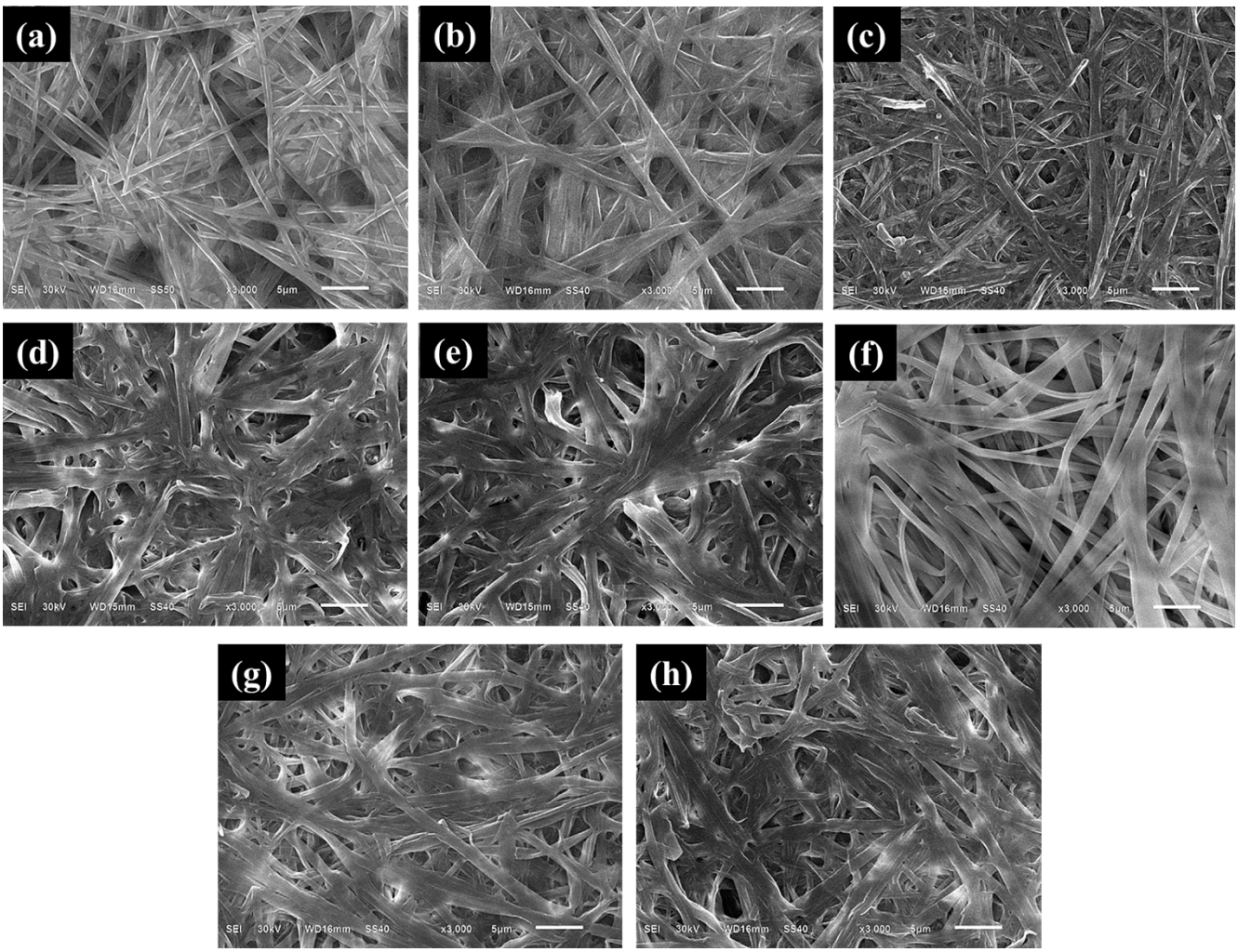

Fig.S2 SEM images of G18/paraffin composite xerogels at different concentration: (a) 0.5 wt%, (b) 1 wt%, (c) 2 wt%, (d) 3 wt%, (e) 4 wt%, (f) 6 wt%, (g) 8 wt%, (h) 10 wt%, respectively. Scale bar = 5 µm.

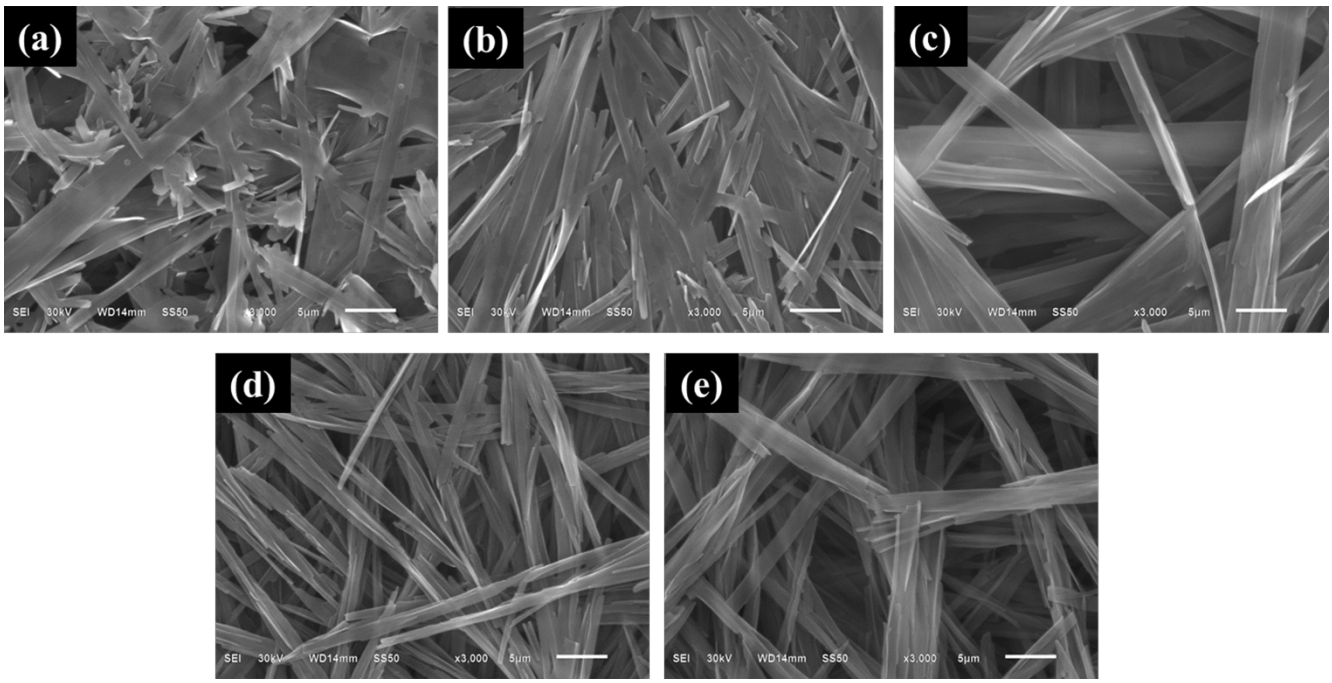

Fig.S3 SEM images Gm/paraffin composite xerogels at the same concentration: (a) G8/paraffin composite, (b) G10/paraffin composite, (c) G12/paraffin composite, (d) G14/paraffin composite, (e) G16/paraffin composite, respectively. Concentration = 3 wt%. Scale bar = 5 μm.

Table S1 The thermal characteristics of Gm/paraffin composites (m = 8, 10, 12, 14, 16).

	Melting			Freezing		
	T_m (^oC)[a]	ΔHobs m (J g^{-1})[a]	ΔHThe m (J g^{-1})[b]	T_f (^oC)[a]	ΔHobs f (J g^{-1})[a]	ΔHThe f (J g^{-1})[b]
3 wt%G8/paraffin	58.3	170.5	183.6	51.9	172.1	178.9
3 wt%G10/paraffin	58.5	180.1	183.6	51.9	181.8	178.9
3 wt%G12/paraffin	58.1	182.7	183.6	51.7	184.1	178.9
3 wt%G14/paraffin	58.8	172.8	183.6	51.1	172.8	178.9
3 wt%G16/paraffin	59.2	171.8	183.6	50.6	177.0	178.9

[a] Evaluated by DSC during the second heating process at a rate of 5 ^{o}C min^{-1} under nitrogen atmosphere.

[b] Calculated by multiplying the weight percentage of paraffin in the composite PCM by the melting or freezing enthalpies of pure paraffin.