Core–shell-like structured graphene aerogel encapsulating paraffin: shape-stable phase change material for thermal energy storage†

Shibing Ye, Qinglong Zhang, Dingding Hu and Jiachun Feng*

The development of energy storage materials is critical to the growth of sustainable energy infrastructures in the coming years. Here, a composite phase change material (PCM) based on graphene and paraffin was designed and prepared through a modified hydrothermal method. Graphene oxide sheets were reduced and self-assembled into three-dimensional graphene aerogels consisting of numerous hollow graphene cells, and paraffin was simultaneously encapsulated into the cells in the form of micrometer-scale droplets during the hydrothermal process. The resulting core–shell-like structured composite PCM exhibits a high encapsulation ratio of paraffin, large phase change enthalpy, and excellent cycling performance. Due to the unique encapsulated structure and continuous graphene network in the matrix, such a composite PCM holds a good shape-stable property, which prevents the leakage of paraffin above its melting point. In addition, it inherits the intrinsic thermally and electrically conductive nature of the embedded graphene, and thus shows enhanced thermal and electrical conductivity compared to pure paraffin. This novel composite PCM can realize efficient thermal energy storage and demonstrates the potential to be directly used as an actual thermal storage device without containers.

Introduction

To abate the energy crisis of fossil fuels, human beings have been focusing on the development of renewable energy resources. As a directly usable and ubiquitous form of energy, thermal energy is expected as a preeminent substitute for fossil fuels. One of the most promising ways to utilize thermal energy is the latent heat storage of phase change materials (PCMs). Organic solid–liquid PCMs (e.g. paraffin) have been studied extensively due to their large latent heat, wide melting temperatures for convenient use, chemical stability, and abundance in natural resources. To enhance the thermal and electrical conductivity of organic PCMs, many types of carbon materials, such as carbon nanofibers, carbon nanotubes (CNTs), graphite flakes, and graphene sheets, have been introduced. Generally, these dispersed fillers can constitute additional preferential transport paths within the organic matrix, and thus contribute to the heat transfer and electrical conduction. Composite PCMs with good thermal and electrical conductivity can directly realize solar and electroheat storage in many fields. Unfortunately, despite the thermal and electrical conductivity enhancement achieved by the addition of carbon materials, one critical problem lies in the fact that these dispersed fillers are usually unstable, which tend to precipitate when subjected to repeated thermal cycling. Recently, various efforts have been devoted for developing continuous fillers instead of dispersed fillers. Ruoff et al. and other researchers independently prepared graphene-based composite PCMs by backfilling organic PCMs into three-dimensional (3D) graphene sponges or graphene aerogels (GAs). Similarly, Zou et al. fabricated a multifunctional composite PCM by infiltrating paraffin into a porous, deformable CNTs sponge. In these composite PCMs, the immobilization of graphene or CNTs upon melting was realized by the preformed carbon networks, which possess unique inter-penetrated structures and considerable mechanical strength. In addition, practical applications of organic PCMs have been hindered by another substantial challenge: the leakage of the PCMs during repeated use, which is caused by the considerable volume expansion related to phase change. Unfortunately, the reported GA-based or CNT sponge-based composite PCMs always fail to solve the leakage problem because of the open-cell structure in the aerogels or sponges. The impregnation of high loading (beyond 10 wt%) graphite or graphene with PCMs can increase the shape stability to a certain extent, but it generally leads to significant decrease of the phase change enthalpy. Alternatively, microencapsulated PCMs can...
effectively prevent leakage throughout the phase change process. For example, Wang et al. reported the microencapsulation of PCMs with double-walled shells (polystyrene/graphene) and realized the protection of the core material from leakage and evaporation. However, the use of polymers as packaging materials usually suppresses the thermal conductivity of microencapsulated PCMs, which is inadequate for efficient energy storage and conversion. Taking all these questions into account, achieving a type of composite PCMs with both enhanced thermal and electrical conductivity and good shape-stable property, but without compromising the material enthalpy, is becoming an urgent challenge to be addressed.

To meet such a challenge, we focus on integrating the advantages of highly-conductive carbon nanomaterials, preformed continuous network, and microencapsulation technique into a single composite PCM. Due to the unique characteristics, including the superior thermal conductivity (≈5000 W m\(^{-1}\) K\(^{-1}\)), high electrical conductivity, and the attractive stability to self-assemble into a shell structure, two-dimensional graphene sheets attracted our attention as a promising thermal and electrical conductivity promoter and packaging material. In this work, we fabricated a core–shell-like structured graphene/paraffin composite PCM by a modified hydrothermal method proposed recently by Shi et al. Typically, graphene oxide (GO) sheets were reduced and self-assembled into 3D GAs consisting of numerous hollow graphene shells, and paraffin was simultaneously encapsulated into the shells in the form of micrometer-scale droplets. The obtained composite PCM exhibits a high encapsulation ratio of paraffin up to 97 wt%, large phase change enthalpy, and excellent cycling performance. The prevention of leakage above paraffin’s melting point is also achieved in such a composite PCM. In addition, it inherits the intrinsic thermally and electrically conductive nature of the embedded graphene, and shows enhanced thermal and electrical conductivity compared to pure paraffin. Therefore, this novel composite PCM has potential applications in renewable and clean energy sources.

Experimental section

Preparation of composite PCMs

GO was synthesized and purified by a modified Hummers method. The composite PCM was fabricated via a modified hydrothermal process proposed by Shi et al. Typically, 20 mL GO aqueous solution (5 mg mL\(^{-1}\)) was mixed with a 15 mL cyclohexane solution of paraffin (50, 100, and 200 mg mL\(^{-1}\)) followed by a violent shake to form homogeneous GO/paraffin emulsions. The emulsions were sealed in Teflon autoclaves and heated for 15 h at 160 °C to obtain graphene gels filled with cyclohexane/paraffin droplets. After cooling to room temperature, the gels were immersed in de-ionized water at 90 °C for 1 h to evaporate the cyclohexane, which was replaced by water. The obtained graphene/paraffin hydrogels were then subjected to freeze drying to form graphene/paraffin aerogels. In the following text, the composite aerogels are expressed as MH–GP\(n\), where MH represents modified hydrothermal process, GP represents graphene/paraffin, and \(n\) stands for the concentration of cyclohexane solutions of paraffin (mg mL\(^{-1}\)). The neat GAs prepared by the hydrothermal method and modified hydrothermal method were denoted as H–GA and MH–GA, respectively. For comparison, the composite PCMs based on H–GA and MH–GA were fabricated by immersing them into the melted paraffin, which were denoted as H–GA–P and MH–GA–P, respectively. It should be emphasized that the paraffin used in this study was experimentally verified to be thermally stable under the hydrothermal treatment (Fig. S1 and S2, ESI†).

Structure and morphology analysis

Optical microscopy images of GO/paraffin emulsions (4v/3v) were obtained on a polarizing microscope (Leica DM2500P, German). The morphologies of GAs and MH–GP\(n\) were characterized using a field-emission scanning electron microscope (SEM, Zeiss Ultra 55, German) operated at 5 kV. The content of paraffin in MH–GP\(n\) was determined using a thermogravimetric analyzer (TGA, Perkin-Elmer Pyris-1, USA). Each sample with a weight of ~5 mg was heated from 100 to 700 °C at a heating rate of 20 °C min\(^{-1}\) under nitrogen atmosphere. To determine the reduction of GO in the composite materials during the hydrothermal process, the reduced GO sample, which was isolated from the MH–GP\(n\) by repeatedly washing with excess cyclohexane, was characterized by X-ray photoelectron spectroscopy (XPS, Thermo ESCALAB 250XI, USA) with monochromatic 150 W Al K\(\alpha\) radiation.

Energy storage property measurements

The latent heat of the samples was tested on a differential scanning calorimeter (DSC, Mettler-Toledo DSC-821e, USA) under nitrogen atmosphere. Typically, samples (~5 mg) were first heated to 80 °C at 10 °C min\(^{-1}\) and kept for 5 min to erase the previous thermal history. Subsequently, the DSC measurements were performed between 0 and 80 °C with both a heating rate and cooling rate of 10 °C min\(^{-1}\). The thermal cycling experiments of MH–GP\(n\) were conducted by repeating the same heating/cooling steps 20 times.

Shape-stable property tests

The shape stability of paraffin, MH–GP\(n\), and MH–GA–P was monitored using a parallel plate rotary rheometer (Thermofisher, HAAKE MARS III, USA). Samples were cut into wafers with a thickness of ~2 mm for the rheological measurement, in this case, the dynamic temperature ramp test. Typically, it was performed at a frequency of 1 Hz with a heating rate of 5 °C min\(^{-1}\) from 35 to 120 °C, keeping the single shear strain of 1.0% for all the samples. In addition, the shape stability was also investigated by recording the states of the samples at different ambient temperatures with digital images. Samples were placed in an air-circulating oven in which the temperature increased by 10 °C each time and remained constant for 30 min at each temperature point.
Thermal and electrical conductivity measurements

The thermal conductivity of PCMs was measured using the transient hot wire method on a thermal conductivity meter (XIATECH TC3010, China). The electrical resistance was measured using a digital multimeter (VICTOR VC9808+, China). Samples were cut into cylinders with a height of ~1.5 cm and a diameter of ~1.3 cm. Two copper sheets were used as the current collectors contacting the top and bottom surfaces of the samples.

Results and discussion

Formation of MH–GPn

The prerequisite for the modified hydrothermal method is the formation of stable oil–water emulsions based on GO. Current models suggest that GO sheets bear hydrophilic carboxyl groups around their edges and a hydrophobic basal plane, which endow them with an attractive amphiphilic nature. Therefore, GO sheets can accumulate at oil–water interfaces and lower the surface or interfacial tension, acting like surfactants. In this work, cyclohexane, a good solvent for para, was selected as the organic phase. Based on the preliminary optimal experiment, the volume ratio of the GO solution (5 mg mL⁻¹) and cyclohexane was controlled at 4 : 3. By vigorously shaking the mixture, a serious emulsification phenomenon was observed, and the emulsion could keep stable, even after standing for 1 month. As shown by the optical microscope image in Fig. 1a, cyclohexane forms stable droplets in the GO solution (aqueous phase) with diameters in the range of several tens of micrometers to ~200 μm. Dissolving paraffin into cyclohexane does not change the emulsification behavior of GO sheets. When the volume ratio of the GO solution and organic solution was fixed at 4 : 3, the certain amount of GO sheets proved to be sufficient to stabilize the cyclohexane solutions containing paraffin. Even at the highest investigated paraffin concentration of cyclohexane solution (200 mg mL⁻¹), a homogeneous and stable emulsion could be formed just by vigorous shaking (inset in Fig. 1b). The GO-stabilized cyclohexane/paraffin/water emulsion has an organic phase with shapes and sizes similar to the GO-stabilized cyclohexane/water emulsion (Fig. 1b). It should be noticed that with higher paraffin concentration in the cyclohexane solution, the viscosity in the resulting emulsion is higher, which is beneficial for the stability of the oil–water emulsion.

In the traditional process to prepare GAs, GO sheets are gradually reduced and assembled into a 3D network with the reaction proceeding, the enhanced hydrophobicity and enlarged π-conjugated structures of the reduced GO sheets strengthen π–π stacking and increase the amount of cross-links, which result in a compact and continuous graphene network. Compared to the traditional hydrothermal method, the modified hydrothermal method involves a slightly different process for the formation of the aerogels. Still, the results of the systematic characterization showed that GO was reduced during the modified hydrothermal process (Fig. S3, ESIF). For example, the XPS data showed that the C/O ratio increased from 1.40 for raw GO to 6.97 for the reduced GO, indicating a significant reduction in the number of oxygen functional groups.

The modified hydrothermal process in our work could be schematically illustrated as Fig. 2. Upon hydrothermal treating, GO sheets are expected to be reduced and inclined to assemble into a 3D network. In practice, the reduced GO sheets are rejected from the cyclohexane droplets and subsequently accumulated at the surfaces of these soft templates (Fig. 2b). As a result, a gel including lots of organic droplets is formed whose structure is determined by the soft template. After the removal of cyclohexane and drying, a MH–GA constructed by a myriad of hollow, spherical cells, that almost maintain the shapes of the cyclohexane droplets, could be obtained. For the preparation of MH–GPn, the amorphous paraffin solution freezes into a crystallized spherical paraffin particle with the removal of cyclohexane. Thus, it is expected that a relatively compact graphene network embedded with many well-defined core–shell structures in the MH–GPn will be formed, where the closed-cell graphene cells act as shells, while the paraffin particles act as cores (Fig. 2c).

As revealed by the SEM images, a mass of hollow, spherical cells in size from dozens of micrometers to over 100 μm in diameter are distributed among the fracture surface of MH–GA (Fig. 3a), which is considerably different from the morphology of the H–GA fabricated by the traditional hydrothermal method (Fig. S4, ESIF). This structure is similar to that seen for the GAs prepared from GO and hexane mixture. Taking MH-GP200 as an example to examine the morphology of MH–GPn, it possesses a relatively compact morphology with uniformly distributed core paraffin of 100–200 μm in size (Fig. 3b). The cracked graphene shells and few voids on the fracture surfaces (Fig. 3c) must have been caused by the removal of the paraffin particles. The magnification of the encapsulated structures reveals that the shells with graphene walls of 100–200 nm in thickness (Fig. S5, ESIF) encapsulate the core paraffin (Fig. 3d). In addition, an obvious interspace between the graphene walls and spherical paraffin particles was observed, which should be due to the volume contraction related to the crystallization of paraffin. A fact that should be mentioned is that the graphene shells could be easily fractured during the peeling process because they are interconnected by the graphene network. Therefore, it is difficult for us to observe a complete core–shell

![Optical microscopy images of the emulsions of (a) cyclohexane/GO solution (5 mg mL⁻¹) (3v/4v) and (b) paraffin solution (200 mg mL⁻¹)/GO solution (5 mg mL⁻¹) (3v/4v). Inset shows the digital image of (b) emulsion.](image-url)
structure consisting of a paraffin core encapsulated by a continuous graphene shell from the SEM images. Still, based on these observations from the fracture surface, together with the formation mechanism of MH–GA proposed previously, it could be concluded that the paraffin particles in MH-GP200 are encapsulated in the closed-cell graphene shells. Such a unique core–shell-like structure endows MH–GA and MH–GPn with large volumes, as demonstrated in Fig. 4. The formed gels by the modified hydrothermal method exhibit negligible volume shrinkage after assembly, in contrast with the initial reactor (Fig. S6, ESI†), which is remarkably different from the graphene hydrogels prepared via other methods.45 All the gels show little volume shrinkage upon freeze drying (Fig. 4), indicating that the graphene cells are firm enough to maintain the shape of the aerogels during the drying process.

Latent heat of MH–GPn

For the composite PCMs, it is important for us to determine and control the filler content, which significantly affects the final latent heat of phase change. The weight loss percentage of paraffin in MH–GPn was measured by TGA, as shown in Fig. 5a. The only step occurring at a temperature range between 200 and 350 °C belongs to the thermal degradation of the paraffin molecular chains (Fig. S2, ESI†), and the remaining percentage could be attributed to the GA framework. By rough calculation, the paraffin mass percentages of MH-GP50, MH-GP100, and MH-GP200 were 88, 92, and 97 wt%, respectively. The progressive mass percentages suggest that the paraffin content can be readily controlled in certain range by adjusting the concentration of the paraffin solution. Note that the highest paraffin content of MH-GP200 obtained by the modified hydrothermal method is among the largest values reported for composite PCMs based on GAs or graphene sponges.18–20

The phase change behavior of the composite PCMs was determined using DSC. Fig. 5b shows the DSC curves of pure paraffin and the representative composite PCM sample, MH-GP200. It is found that both paraffin and MH-GP200 have the same onset point in the melting and freezing DSC curves, which suggests that the incorporation of graphene has no notable effects on the phase change temperature of paraffin. The melting and freezing temperatures of paraffin located on the peak position are 53 and 46 °C, but they shift to 57 and 41 °C for MH-GP200, respectively. This change is possibly a consequence of the influence of the GA framework, i.e., the embedded graphene network affects the surrounding organic molecules, making the phase change of paraffin delayed.21 Moreover, it has been reported that the crystallization of the encapsulated PCM droplets is generally confined, producing a serious supercooling phenomenon.24

For good PCMs, the high phase change enthalpy is a vital indicator. The phase change enthalpy was calculated from the DSC curves (Fig. S7, ESI†) and presented in Fig. 5c. For the MH–GPn samples, it is understandable that both the melting latent heat (ΔHm) and freezing latent heat (ΔHf) increase as the
paraffin content in the composite PCMs increases. Among the MH–GPn samples, MH-GP200 has a $\Delta H_m$ of 202.2 J g$^{-1}$ and a $\Delta H_f$ of 213 J g$^{-1}$, which are even higher than those of pure paraffin, 201.5 and 213 J g$^{-1}$, respectively. It is found that the H–GA–P exhibits a reduction effect, while the MH–GA–P exhibits an enhancement effect in the phase change enthalpy. Despite the fact that many studies have reported the enthalpy enhancement for composite PCMs, the mechanism for this difference has not been sufficiently clarified.$^{20,39}$ For these PCMs filled with graphene or CNTs, the additional enthalpy independent of the paraffin content is generally attributed to the interaction between paraffin and the fillers.$^{21,39}$

To prove the reversibility of MH–GPn, the solid–liquid phase transition cycling tests were performed on the DSC, and the results of the cycle test are shown in Fig. 5d. Compared with the latent heats before the cycles, the $\Delta H_m$ and $\Delta H_f$ of MH-GP200 were 202.1 and 212.8 J g$^{-1}$ after 20 cycles, respectively, showing only negligible reduction in the enthalpy. As shown in the inset of Fig. 5d, the DSC curves of the 20 cycles were virtually identical, which indicates the excellent reversibility of MH-GP200.

**Shape-stable properties of MH–GPn**

The shape stability of organic PCMs is crucial for preventing the leakage, storage and transport of materials. Here, the shape-stable properties of the composite PCMs were characterized through a leakage test. As shown in Fig. 6, the PCMs samples were heated to different temperatures, stored for 30 min, and observed via tracking photography using a digital camera. It can be seen that all the samples were solid with a definite shape at 40 °C, which is in agreement with the melting point of raw paraffin. When heated to 50 °C, the pure paraffin started to melt with obvious liquid leakage on the bottom. We noticed that H–GA–P and MH–GA–P also presented a small amount of leaked liquid, while the MH–GPn kept stable without a trace of moisture on the bottom. This indicates that the immersed samples (H–GA–P and MH–GA–P) cannot increase the shape-stable performance of paraffin, while the samples prepared via the modified hydrothermal method show a better ability to
prevent leakage. With the temperature increasing to 60 °C, lots of leaked liquid was observed in the pure paraffin, H–GA–P and MH–GA–P; however, no obvious liquid was found in MH–GPn. When the temperature was further increased to 70 °C, the pure paraffin completely melted into a liquid. Despite the fixed shape of H–GA–P and MH–GA–P, they were speculated to release most of the adsorbed paraffin and only leave the graphene framework. Amazingly, only a bit of moisture was found for MH-GP200 at such high temperature, and the MH-GP50 and MH-GP100 still maintained their shape without any leakage. This suggests that MH–GPn has an excellent shape-stable property, thereby extending its application in other fields.

Rheological measurement is a sensitive technique to detect the variation of modulus of polymer materials. Given the solid–liquid transition of organic PCMs, this technique was also used to monitor the leakage of PCMs and better understand the shape stability of the composite PCMs.40 Fig. 7 shows the temperature dependence of the storage modulus (G′) of pure paraffin, MH–GA–P, and MH–GPn at a heating rate of 5 °C min⁻¹. For all the samples, a plateau with a high G′ below 50 °C was found, indicating that the samples are in solid state below paraffin’s melting point. As the temperature increases above 50 °C, the G′ of pure paraffin rapidly decreases to be undetectable, suggesting the solid–liquid transition of paraffin, consistent with the observation in Fig. 6. Similarly, a dramatic drop in G′ also occurs for MH–GA–P, which implies a significant leakage of paraffin. It should be noted that MH–GA–P still has a plateau with a lower modulus even above the melting point of paraffin. We believe that the remaining modulus is independent of the solid paraffin and was contributed by the separate graphene framework. The results reveal that both pure paraffin and MH–GA–P rapidly change into liquid, leak heavily, and lose their shape stability when heated above paraffin’s melting point. However, MH–GPn exhibits a completely different rheological behavior. The plateau of G′ below 50 °C also exists for MH–GPn, but the G′ slowly weakens until constant, without observing obvious drops during the heating process. Combined with the observations in Fig. 6, we believe that the reservation of the modulus can be attributed to the packaging of paraffin by the sealed shells of MH–GPn.

As described before, the core paraffin is covered by graphene shells during the modified hydrothermal process, forming core–shell-like structures. The reserved space between the graphene shells and paraffin particles is believed to be large enough to allow the volume expansion during the phase change process of paraffin. Furthermore, the numerous micrometer-scale pores in the GAs collectively suck in the melted paraffin due to the capillary effect.41 In addition, the preformed GA network acts as a solid framework with a certain mechanical strength. Thus, although the encapsulated paraffin in each shell melts into a liquid above its melting point, it is still packaged in the graphene shell, acting like a solid-like gel with a high modulus. Therefore, MH–GPn shows a solid-like behavior in the rheological measurement, which is similar to polymer-supported PCMs.42 However, we note that MH–GA–P prepared by immersing MH–GA into melted paraffin has a weaker ability to package the paraffin. This may be because the sealed graphene cells are broken during the process of absorbing liquid paraffin, leading to the disability of the package above paraffin’s melting point. Based on abovementioned evidence, we conclude that the core–shell-like structures in the MH–GPn matrix endow the composite PCMs with good shape-stable properties.

**Thermal and electrical conductivity of MH–GPn**

One of the performance indicators of PCMs is the rate of energy storage and release, which highly depends on the thermal conductivity of the PCMs.43 As presented above, the hydrothermal process leads to an efficient removal of oxygenated groups and partial recovery of the conjugated structures of the GO sheets, which regains the intrinsic thermally conductive nature of graphene. Therefore, the presence of graphene networks should have a positive influence on the thermal conductivity of MH–GPn. As shown in Fig. 8a, the thermal conductivity of MH-GP200 is 0.274 W m⁻¹ K⁻¹, which constitutes an increase of 32% over that of pure paraffin (0.207 W m⁻¹ K⁻¹). The immersed MH–GA–P also shows an enhanced thermal conductivity (0.248 W m⁻¹ K⁻¹) compared with the paraffin. These results are consistent with the results in previous studies,18,42 which indicates that the addition of graphene into paraffin is exactly able to enhance its thermal conductivity. However, it should be noted that the enhancement of the thermal conductivity for MH-GP200 is limited in this work and is lower than some of the reported values.44 The low graphene content in MH-GP200 should be mainly responsible for the modest enhancement because the high thermal conductivity of the composite PCMs in most research was achieved by the addition of high load of graphene (beyond 10 wt%).14,19,42 Another possible reason that cannot be ignored is that different measurement techniques can also lead to remarkable differences.16,18,20

Previous studies found that the incorporation of polymers into graphene or CNT frameworks does not appreciably degrade their intrinsically high electrical conductivity because of the preformed, continuous, electrically conductive networks.45-47 It
is also expected, for the same reason, that MH–GPn maintains the satisfactory electrical conductivity of graphene. Fig. 8b presents the electrical resistance of MH–GA, MH–GPn, and MH–GA–P in the solid state. Pure paraffin is electrically insulating with high resistance beyond our instrumentation, while the cylindrical MH–GA with a height of 1.5 cm and a diameter of 1.3 cm shows a very low resistance of 0.25 kΩ. The electrical conducting property of MH–GA stems from the reduction and assembly of GO sheets during the hydrothermal process. When paraffin is packaged into the GAs, the resulting MH–GPn with the same size shows a slight increase in the resistance. The values are increased to 0.36, 0.46, and 0.87 kΩ for the MH-GP50, MH-GP100, and MH-GP200, respectively. This indicates that the MH–GPn has an appreciable retention of the electrical conductivity of GAs, which originates from the complete, interconnected, conductive graphene networks in the matrix. It should be noted that MH–GA–P shows a higher resistance of 1.18 kΩ than that of MH–GPn, which may be due to the fact that the graphene networks are partially broken during the absorbing process, producing defects and disruptions in the conduction pathway. These results suggest that such composite PCMs are also a promising route for achieving the thermal management of electro-heat storage and electronic devices.

Conclusions

In summary, a novel composite PCM based on graphene and paraffin was fabricated by a modified hydrothermal method. The resulting PCMs have unique structural features, including embedded continuous graphene networks with a low mass fraction of 3 wt%, and core–shell-like structure with a closed-cell graphene pore as the shell and the packaged paraffin as the core. Such composite PCMs demonstrate an excellent performance: large phase change enthalpy with a $\Delta H_m$ of 202.2 J g$^{-1}$ and a $\Delta H_f$ of 213 J g$^{-1}$, which is even higher than that of pure paraffin; outstanding cycling performance with 100% retention after 20 cycles; good shape stability that prevents the leakage above paraffin’s melting point; as well as enhanced thermal and electrical conductivity compared with pure paraffin. The peculiarity of our method and the materials provides a new platform for developing shape-stabilized PCMs, which can be widely applied to fields related to energy conversion and storage.

Acknowledgements

This work was financially supported by the Natural Science Foundation of China (51373042 and 21174032).

Notes and references
