Materials Advances

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PAPER

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Enhancing the phase change material properties by an energy-efficient one-step preparation method using organogelator–polyolefin composites†

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The synergistic combination of various sorbitol-based organogelators with polyolefins allows the preparation of porous support structures for immobilized phase change materials (PCMs). Using a PCM as a solvent for the preparation leads to dimensionally stable composite materials with extremely high loading rates and low leakage of PCMs. Detailed investigations were performed on the kind of polyolefin support and its mass fraction concentration in the PCM, the temperature-dependent softening and failure under superimposed load, the efficiency of heat transport and the retention capacity over several melting/solidification cycles in various measurement setups. In particular, paraffin wax in combination with 1,2,3-trideoxy-4,6:5,7-bis-*O*-[(4-propylphenyl)methylene]-nonitol (TBPMN) and ultrahigh molecular weight polyethylene (UHMWPE) showed the best results in terms of high dimensional stability, low leakage, excellent processability and competitive heat capacity. The herein-established one-step preparation method saves time and energy compared to the loading of pre-formed porous supports and improves application-related properties at the same time.

1. Introduction

Climate change and the resulting urgent need to change the entire energy sector impose challenges for all of us to use the available energy more responsibly. Worldwide one-third of the whole energy consumption is related to air conditioning; in Germany, this proportion is about 27%.[1,2] Typical insulation materials are organic polymers like polystyrene (PS) or polyurethane (PU) foams or glass and rock wool.[3] In terms of performance, aerogels are superior to conventional insulators, but their worse price/performance ratio turns them into niche products for special applications. Optimized insulators and latent heat storage systems are a highly relevant starting point for developments towards a more sustainable energy balance.

Phase change materials (PCMs) are a group of materials with high melting energy. This property makes them interesting for heat transport in the fluid state and heat absorption and storage in the immobilized solid state. Typical applications are solar energy storage,[4–7] heat recovery in industrial processes,[8] temperature control in electrical applications[9] and energy storage in buildings.[10] Both inorganic materials like hydrated salts[11] and organic components like fatty acids,[12] waxes[13] and polyethyleneglycol[14–16] are used. On the one hand, typical organic materials show a lower melting point and fewer problems with a phase separation over the melting and solidification cycles and are less corrosive. On the other hand, leakages can occur in the molten state connected with a potential material loss. In order to prevent leakage, such materials are often encapsulated and consequently immobilized using porous polymers,[17] polymer blends,[13,18] as well as aerogels based on graphene[12,19] or silicon dioxide,[16,20] graphite[21,22] and diatomaceous earth.[23,24] Gelators (*cf.* below) can also be used,[25,26] but they have shown a much worse retention capability.[26,27] Recently, Fan *et al.* described the preparation of a form-stable PCM using polynorbornene-based bottlebrush polymers as gelators and sulphur-treated nickel foams with interesting properties for application in solar technology. To avoid material loss, the PCM needs to show a high phase compatibility to the support material when molten, but should not dissolve the support.[28] Looking at the production costs of an immobilized PCM, both the synthesis of the support material and the subsequent loading of PCM, by "flooding" the support, have a high contribution.

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† Electronic supplementary information (ESI) available. See DOI: https://doi.org/10.1039/d2ma00578f

Aerogels are nanostructured open porous solids, which can also function as support materials for PCMs. Aerogels can be made *via* a sol–gel process from many inorganic materials such as silicon dioxide or titanium dioxide, which were the first documented ones.[29] Furthermore, the classical synthetic polymers (PU, PS),[30] biopolymers like cellulose[15,31] and starch[32] or carbon frameworks[33] can also be used as the solid phase.

Organogelators are molecules which, in a suited liquid, can build up three-dimensional networks by chemical or physical interactions. These are, for example, hydrogen bonds of OH- or NH-functionalities, π–π stacking, ionic interactions and van der Waals forces.[34–38] The specific network structure depends on the solvent or the molten porogen. Typical gelator material classes are amino acids,[38,39] isosorbide derivatives[35,37,40–43] or amines.[43–45] Despite their micro-scale structure, gelator networks are not applicable for aerogels because of their missing covalent bonds and the resulting low mechanical stability, which leads to a collapse of the structure after extraction of the solvent. Finally, compact bundles of associated gelator aggregate strings result. However, in 2016, Jamart-Grégoire *et al.* described an amino acid-based organogel that was supercritically dried to obtain small samples of monolithic aerogels.[46] However, these aerogels were very brittle and powdery and would therefore not be suitable as support for PCM.

In this work, an alternative and new approach for aerogel-immobilized PCM synthesis by using organogelators and cheap polyolefins in the PCM as a solvent is introduced. Recently, we demonstrated that by combining organogelators with suitable high molecular weight polymers, for example polyolefins, mechanically robust composite aerogels can be obtained, because a thin polymer film on the gelator fibers reinforces the network.[47] Here, we use such composites as support for PCMs such as paraffin or polyethyleneglycol. Importantly, we show that instead of subsequent loading of the support with PCMs, also a one-step fabrication directly in the PCM as solvent is possible and that robust materials at very high PCM/carrier ratios of up to 39 : 1 can be obtained. The main advantage of the composites obtained here compared to flooded PCM supports and gelator-stabilised PCMs is that they can be produced fast and easily while at the same time retaining very good material properties. The method is not limited to the system described and can also be adapted for various applications.

2. Experimental

2.1. Materials

The following support polymers were used in this work: Stamylan P, isotactic polypropylene (iPP), $M_w \sim$ 300 kg mol^{-1}, from Sabic; high-density polyethylene (HDPE), $M_w \sim$ 500 kg mol^{-1}, from Sabic; Stamylan UH, ultrahigh molecular weight polyethylene (UHMWPE), $M_w \sim$ 6.000 kg mol^{-1}, from DSM. Millad NX8000, 1,2,3-trideoxy-4,6:5,7-bis-*O*-[(4-propylphenyl)methylene]-nonitol (TBPMN) from Milliken, di-(4-ethylbenzylidene)sorbitol (DEBS), di-(2,4-dimethylbenzylidene)sorbitol (D2,4-DMBS) and di-(3,4-dimethylbenzylidene)sorbitol (D3,4-DMBS) (own synthesis, see
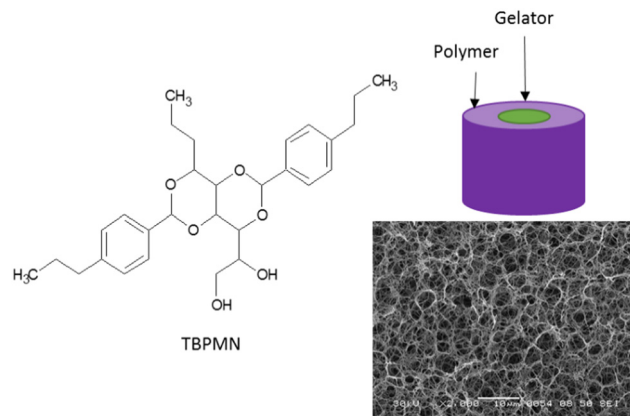

Fig. 1 Gelator structure, schematic drawing of the cross-section of a polymer-stabilized gelator fibril and SEM image of a freeze-dried TBPMN/polyolefin sample.

the ESI† 1) were used as gelators. The solvents used were 1,2,4-trichlorobenzene (TCB) ≥98% from AppliChem; benzene ≥99.7%, AnalaR NORMAPUR, from VWR; cyclohexane ≥99%, GPR RECTAPUR, from VWR. The PCMs used were 1-tetradecanole (TD), p.a., from Merck, paraffin wax from uncolored candles, and three polyethyleneglycoles (PEG), Polyglykol 2000S, 8000S, and 20000S from Clariant.

2.2. Synthesis of organogelator-based aerogels and PCM-composite materials

Herein, the synthesis of PCM composites by flooding a separate aerogel support and the one-step synthesis are described.

For the preparation of a polymer-based aerogel, defined amounts of polymer and gelator TBPMN (*e.g.* mass ratio 25 : 75 for iPP) are heated in a suitable high boiling solvent, *e.g.* TCB, under stirring, until a homogeneous solution is obtained. The stirring is stopped and the solution is cooled down to room temperature. During cooling, the gelator solidifies as fibrils first, followed by the crystallization of the polymer. Low concentrations of the polymer (≤0.5%) lead to a blueish shimmer caused by the gelator. At higher concentrations, it is covered by the white turbidity of the crystallizing polymer. Since freeze-drying from a high-boiling solvent is hardly possible, the solvent is exchanged to benzene. Therefore, the gel body is stored in five to ten times the amount of benzene for 16 to 20 hours on a shaker and then the solvent is refreshed. This is repeated at least three times. The freeze drying is done at −10 °C under vacuum (≤3 μbar). During this process, the gel body slightly shrinks (0–5 vol%). The obtained aerogel is then inserted into a molten PCM (at 90 °C) until mass constancy is reached. The PCM composite is freed from exceeding material and can now be used. Fig. 1 shows the structures of the preferred gelator used, an SEM image of the intermediate aerogel structure and a sketch of the cross-section of a support fibril.

For the one-step synthesis, the polymer and the gelator are directly solved in the hot PCM. Compared to aerogel formulations, it is necessary to increase the ratio of polymer to gelator (*e.g.* mass ratio 40 : 60 for iPP : TBPMN). After cooling, the PCM
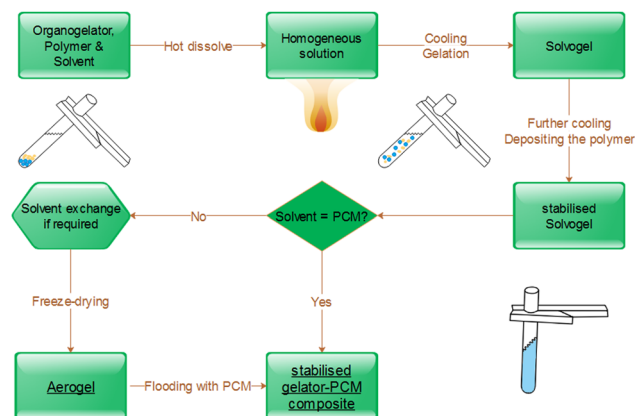
Fig. 2   Manufacturing scheme for PCM composites.

Fig. 4   SEM images of the support structure – left: 5% TBPMN/PP sample (60/40) obtained in paraffin after solvent exchange for cyclohexane and subsequent freeze-drying – right: 1.5% TBPMN/PP (75/25) aerogel (analogous to ref. 47).

composite is taken out of the synthesis pot under slight heating of the pot at the walls. The excess of PCMs is removed analogously to the process described above. Both manufacturing methods for aerogel-immobilized PCMs are illustrated in Fig. 2.

The direct path with PCM as a solvent is generally to be preferred, as this saves time, energy and material. The final samples can be cut into the desired shape and used according to their intended purpose. Furthermore, composites can be heated above the melting point of the PCM and their shape can easily be adjusted.

2.3.   Characterization

The thermal behavior was analyzed with a Mettler Toledo DSC 822 and Netzsch DSC 214 Polyma (see the ESI† for Temperature program). The morphological structure of the aerogels was investigated with a JEOL JSM-6460LV scanning electron microscope (SEM) (30 kV; high vacuum mode). All samples were cryogenically fractured in liquid nitrogen and gold coated. The mass loss of the samples was determined in an oven at 95 °C. Fig. 3 outlines the experimental setup.

The samples were placed on a grid. Different grids were used (for more details see the ESI† 2). All samples were heated for ≥ 1 hour per cycle, cooled to room temperature (RT) for another hour,
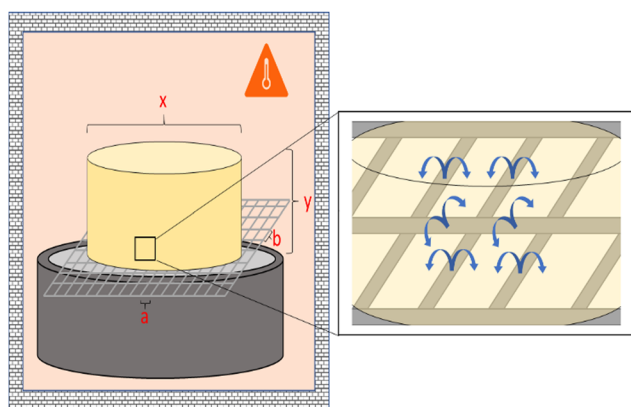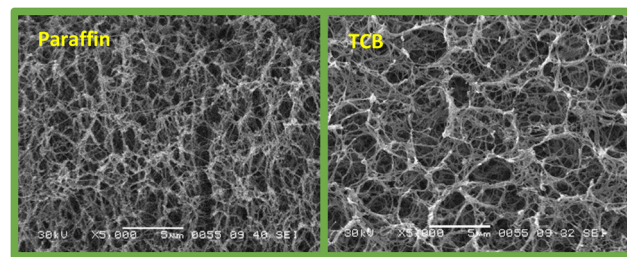
and then weighed on an analytical balance (Nimbus NBL 164i). Cylindrical specimens with a diameter of 21 mm and an initial weight of 7.5 g were used.

To measure the rate of temperature increase in the material, samples were fabricated with a centered temperature sensor and placed in a heating block designed for this purpose. The temperature sensor was kept centered even when the sample was melted, using a 3D-printed holder installed above the setup. During the measurement, the temperatures of the heating block, the sample's cores and the room were recorded simultaneously *via* an Arduino microcontroller. The samples used were 20 g cylindrical specimens with a diameter of 27 mm.

3. Results and discussion

Besides the intrinsic latent heat storage capability of a PCM, a high loading capacity of the support and its retention capacity over multiple melting and freezing cycles are central requirements of immobilized PCM composites. Therefore, the compatibility of the PCM with the support material and an open-pore structure with very high porosity at sufficient mechanical stability are necessary. Additionally, a low density is ideal to make high mass-based loading ratios possible. It is feasible to synthesize aerogels with TBPMN as a gelator and iPP or UHMWPE in TCB with densities of 0.03 to 0.25 g cm^{-3}.[47] Paraffin as a PCM was infiltrated as described in Section 2.2 and very well retained in the composite material. Depending on the support density, mass ratios PCM:support of up to 25 could be obtained.

Based on these results, the synthesis was done directly in the PCM in a single-step process. Although the structure of the support gets slightly rougher by the worse solvent (compared to TCB), PCM:support ratios of 39 : 1 with good retention capability are obtained. This can be attributed to the complete filling of the support structure, whereas when the aerogel is flooded, empty areas remain.

To prove that the network structure described in the previous work[47] is also formed in paraffin as the solvent, different TBPMN/polyolefin samples were synthesized and then exchanged against cyclohexane *via* Soxhlet extraction.[47] After subsequent freeze drying, the samples were analyzed by SEM. Fig. 4 shows the obtained aerogel structure of an extracted paraffin-derived sample compared to the structure obtained from TCB.
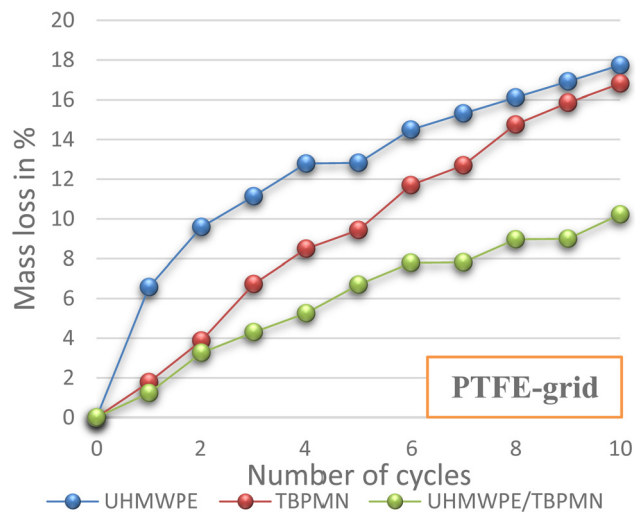

Fig. 3   Sketch of the measurement setup for determining the mass loss over several melting/solidification cycles with the sample on a grid.

Fig. 5 Mass loss of three different PCM samples (4 wt% support; composite 1 : 1 vs. 4 wt% individual components; PTFE grid).

The structures are very similar. Because paraffin is not the ideal solvent for the gelator-polyolefin system, the structures are slightly coarser, but still mesoporous. This observation is in agreement with the findings in previous work,[47] that using cyclohexane instead of aromatic solvents (like TCB) yielded coarser aerogels.

For the measurement of PCM loss from the composite material, the samples were placed on different grids in an oven and the weight loss over 20 melting/freezing cycles (RT to 95 °C to RT) was observed (cf. Section 2.3). It should be noted that no relevant change in mass could be detected when heating the sample just above the melting point, which is why the thermal stress on the specimens was increased by overheating to 95 °C. On the one hand, the higher temperature far above the melting point ensures a higher creeping capability of the PCM; on the other hand, this leads to a further volume increase of the PCM. If the support is too rigid, PCMs will be lost inevitably. Because

there is no standardized method for the measurement of PCM loss, it was tested using different setups (see the ESI† 2). Consequently, only values from the same setup are comparable. The mass losses on the PTFE grid are higher than those on, for example, a PE grid. The more demanding experimental setup was chosen at this point to show the difference between the samples more clearly. Under milder testing conditions (other grid than PTFE, lower overheating), the mass loss is nearly zero for many samples. However, the fact that overheating of the specimens is necessary in order to observe significant changes in the material at an early stage of the test sequence with a good measurement setup underlines the good cycling stability and thus performance of the composites established.

UHMWPE and TBPMN individually show a stabilizing effect for the PCM; hence, a comparative analysis against the polymer/gelator composite material was performed. Here, it was possible to show that the mixture shows significantly better PCM retention behavior than the single components. In particular after long thermal stress, it was observed that the composites creep less and show a better dimensional stability. In Fig. 5, the mass loss of polymer, gelator and polymer/gelator composite PCM samples on a PTFE grid is shown. The composite (UHMWPE/TBPMN) shows significantly better retention properties than the individual components (UHMWPE or TBPMN).

If HDPE or PP alone is used instead of the highly swellable UHMWPE, the samples already fail while the first melting. In contrast, gelator composites with PP or HDPE perform similarly to the UHMWPE-TBPMN composites.

Fig. 6 shows the mass loss of samples with different support fractions. The retention capability, presented as residual weight, initially increases with increasing weight fraction. Interestingly, an optimum is obtained at about 4 wt% of support material. Samples with more support show similar performance. This can possibly be explained by the structure of the support obtained in the PCM or by its lower flexibility when volume increases upon heating. The mass loss increments decrease with increasing cycle numbers; almost constant values are observed after
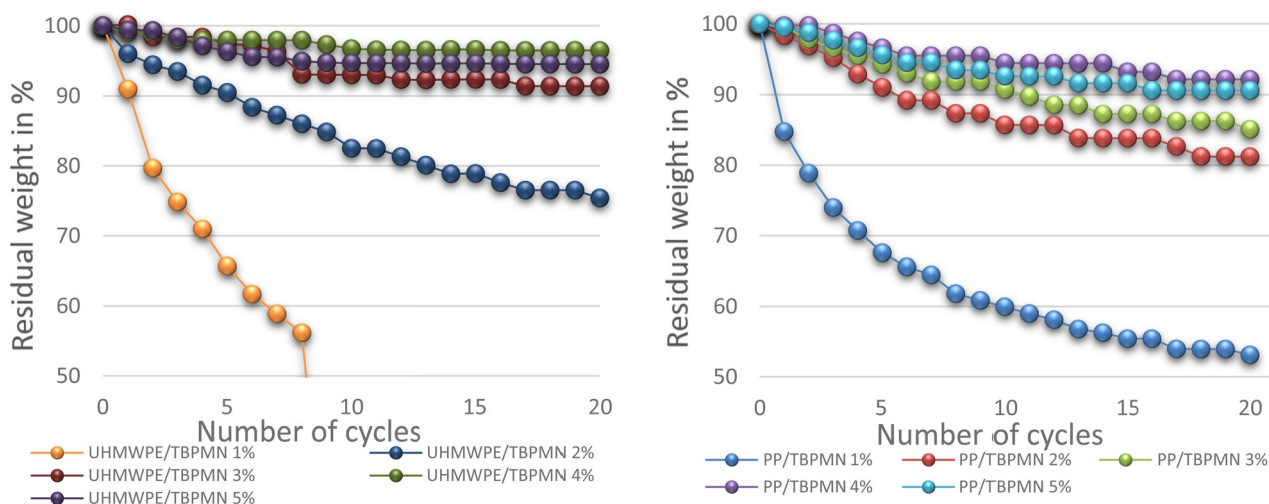

Fig. 6 Influence of the support mass fraction on the retention capacity (polymer/gelator 1 : 1) (PE grid).

Fig. 7 Shape stability after 20 melting/solidification cycles of PEG 8K samples with different gelators (2% composites; gelator/UHMWPE 70/30).

Fig. 8 Paraffin gelator/polyolefin composite before and after melting.

20 cycles. Exemplary measurements with 50 cycles and 24 h of thermal stress showed no further mass loss.

The composition of the aerogels described in the previous paper[47] does not necessarily represent the ideal one as PCM support; therefore, different formulations were evaluated. The requirement here is to obtain a stable and PCM-retaining network and not the finest porous structure possible, as it is required for thermal insulation applications. Experiments with different polymer/gelator ratios revealed that a significantly higher polymer content is advantageous compared to the aerogel mixtures; optima as PCM support were found for UHMWPE/TBPMN at 1 : 1 and PP/TBPMN at 2 : 3.

These interesting results have been verified with other systems. For the synthesis of the PCM composites, different sorbitol based gelators (themselves and UHMWPE stabilized) in the PCM materials paraffin, PEG (2 kg mol^{-1}, 8 kg mol^{-1}, 20 kg mol^{-1}) and 1-tetradecanole were evaluated. The best and most interesting results were observed with the paraffin/TBPMN/polyolefin system already described above. We propose that the optimum support material results from several synergistic contributions: TBPMN finely distributes the material, the crystalline fraction of the polyolefin stabilizes the gelator network especially at high temperatures and the amorphous and swellable part of the polyolefin improves the retention of the PCM. Finally, the specific properties (molecular weight, surface tension, polarity, viscosity) of the PCM also influence the retention capability.

An overview of all results obtained with various gelators, polyolefins and PCM is provided in the ESI† 3. Already in the preparation step, it is obvious that nonpolar gelators, exactly as polyolefins, show a better phase compatibility to PEG with increasing molecular weight. TBPMN can stabilize PEG even in low concentrations without any losses. The higher creeping tendency of samples with a lower support content and without polymer is optically visible. Long thermal stress increases this effect. While in paraffin as the PCM, the composite of gelator and polymer brings enhanced properties, in PEG, the amount of the gelator has the dominating effect on stability. The shape stability can be viewed as exemplary material in Fig. 7.

While the PCM composites with PEG show very good retention properties (mass loss after 20 cycles is close to zero for many of the PEG-containing specimens), their mechanics in the molten state are less good compared to the paraffin-containing materials. Most of the PEG-containing composites tend to creep and will fail in the long term. The solvogel "smears" easily on surfaces and does not transfer forces throughout the specimen. This may be due to the relatively hard crystallization of the PEG, destroying potentially cellular parts of the gel network. In the solid state, PEG samples behave in a brittle, fragile manner. The opposite is observed with paraffin. In the solid state, the composite can be easily cut and shaped. When the wax melts, the space-filling solvogel network provides high flexibility without losing material. Fig. 8 shows a paraffin composite before and after melting.

All the gelators show good retention properties for PEG with increasing molar mass, whereby the composites with higher form stability are preferred because of their longer lifetime. TBPMN is more suitable for paraffin than the other gelators. Tetradecanol, which can also be used as a PCM, has much lower performance in form stability tests, which was quite expectable. The gelators build up their network by π–π stacking and hydrogen bonds, which are disturbed by the hydroxyl groups of the long chain alcohol and the retention property is weakened. The stabilizing effect of the sorbitol derivatives for 1-tetradecanol reported in another study[26] could not be demonstrated in connection with the here used overheating of samples.

The gel stability was further tested temperature dependent with a modified ball drop test. For this, different samples of 2.5 g weight were loaded by a 5 g heavy stamp and successively heated. Samples with TBPMN and UHMWPE or PP were analyzed. The region of softening was defined as the temperature range from the first penetration of the stamp into the material and the point where the stamp fell through the gel sample. In Table 1, these softening regions are shown.

The experiments confirm that higher gelator fractions have a positive effect on the temperature stability. The polymer has a smaller influence, at least until the weight is increased. When the stamp weight is six times, the sample mass material failure appears earlier. In this case, the polymer concentration gets a higher influence, because the failure happens near the polymer melting temperature in the case of UHMWPE. For the sample with 1.5% TBPMN/UHMWPE (95/5), the region of failure shifts to 90–120 °C. Increasing the polymer proportion to 75/25, the region of failure widens to 90–140 °C. The same effect is visible in PP-containing samples. It is important that the measurements were all performed with paraffin as the PCM. If, for example, a solvogel with TCB is measured, the values decrease by 30–40 °C caused by the better solvent properties of TCB.

Since the PCM is immobilized in the PCM composites, this affects the heat transport in the material. While non-stabilized

Table 1 Softening range of paraffin composites with different gelator/polymer variations
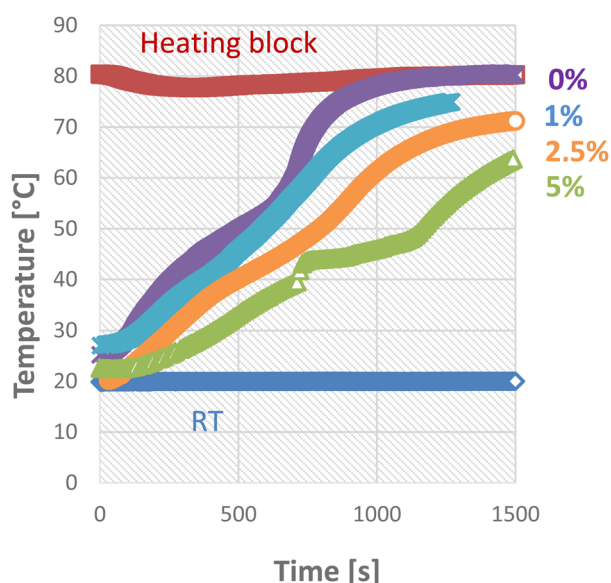
Sample	Softening temperature	Sample	Softening temperature
0.075% UHMWPE	Melting of the wax (60 °C)	0.375% PP	Melting of the wax (60 °C)
1.5% TBPMN/UHMWPE (95/5)	140–180 °C	1.5% TBPMN/PP 3 : 1	140–180 °C
3% TBPMN/UHMWPE (95/5)	160–200 °C	3% TBPMN/PP 3 : 1	160–190 °C
3% TBPMN/UHMWPE (50/50)	150–180 °C	2% TBPMN/PP 3 : 2	150–190 °C
1.425% TBPMN	170–190 °C	4% TBPMN/PP 3 : 2	180–190 °C

PCMs quickly reach a uniform temperature after melting by convection in the liquid, this is not the case with the materials described here. To illustrate this effect, cylindrical samples were heated from the outside by using a heating block, and the temperature was measured in their center. The influence of the support structure was determined using TBPMN in paraffin at various concentrations and plotted in Fig. 9.

In the case of pure paraffin, the change from solid to liquid state can be observed in the bending of the t (time) curve. Due to the gel network of the TBPMN, this effect disappears and the curve becomes largely linear. With an increasing fraction of the support structure, the heat transport slows down. This slower heat transport can possibly be a problem in applications where a lot of heat has to be dissipated in a short time. To counteract this, additives with high thermal conductivity such as carbon black or silver nanoparticles can be added.[48–51]

The same behavior can also be observed using DSC (see the ESI† 4). In addition to the broadening of the temperature range for melting and solidification processes, the DSC curves show the associated enthalpies. In Table 2, the obtained data of some selected materials are presented.

The PCM composites obtained have a high melting enthalpy, with PEG 8K composites between 156.9 and 166.7 J g^{-1}, PEG 20K composites between 152.6 and 154.3 J g^{-1} and paraffin composites between 172.6 and 182.0 J g^{-1}, in case of 4% support samples.

Table 2 Melting and solidification enthalpy of paraffin and paraffin-composites (measured by DSC)

	Solidification process			Melting process
Sample	T_s [°C]	ΔH_s [J g^{-1}]	T_m [°C]	ΔH_m [J g^{-1}]
Pure	36.4	186.3	59.7	180.3
4% TBPMN	36.7	184.1	66.3	182.0
4% TBPMN/UHMWPE (50/50)	36.4	177.5	66.9	172.6

In Table 3, the loading rates achieved in this work are compared with references from the literature.

Table 3 Loading ratios of different PCM composites

Classical PCM support composites		
Composite	Ref.	w% PCM (%)
EG/MA–PA–SA	52	93
EG/paraffin	22, 53	92
PP-aerogel/paraffin	17[a]	90–91
Gypsum/C18–C24	54	18
C-SiO$_2$-PA	20	61
C-SiO$_2$-OD	20	73
SiO$_2$/PEG	14	80
HDPE/paraffin	18	77
EPO/paraffin	55[b]	50
Diatomite/capric–lauric acid	24	62
Carbon/lauric acid	56	33
PCM-gelator-composit		
G18/paraffin	25	97
3,4-DMDBS/C18;C18-1ol	26[c]	97
DMDBS/EG-OD	27[c]	83–97
G18/C18	45	94
This work		
Isosorbide derivatives + UHMWPE/PEG		95–98
Isosorbide derivatives + UHMWPE/paraffin		

[a] The PP aerogel/paraffin system is chemically and structurally the most comparable to the composites shown in this work. Despite the lower loading rate and more moderate melting conditions, a PCM loss of 9.7% after 10 cycles and 14.9% after 20 cycles was observed in that study. The gelator/polyolefin composite established in the present work, with <5% PCM loss after 20 cycles, clearly shows better performance (or comparable performance in the suboptimal test setup (PTFE grid)). [b] For uncoated expanded perlite (EPW), a mass loss of 48% was found, for hydrophobic coated expanded perlite (EPO) 0%. [c] In these works, the loss of mass after 24 h in the molten state was observed. Depending on the PCM, the mass loss varied widely. Samples with 3% 3,4-DMDBS lost 13.6% of their mass with n-octadecane and 36.3% with 1-octadecanol. Xu et al.[27] describe a lower mass loss with 3,4-DMDBS and 1-octadecanol of ≤20% with 3% gelator. By adding 6% expanded graphite, the loss is reduced to 15.6%. This is also significantly worse than the results obtained in the present work. Long-term shape stability is not sufficiently given in all comparable works using organogelators.

Fig. 9 Melting behaviour of paraffin waxes stabilized with different amounts of TBPMN.

The literature comparison shows that the composites prepared in this work are in the range of the loading rates of gelator-stabilized composites, but show significantly better properties, especially in the case of the TBPMN + UHMWPE/paraffin composites. Of course, there are alternative PCM composites such as PP aerogel and EG stabilized materials, but these are much more complex and cost-intensive to produce. The stabilization of the gel network, which was shown in a previous work,[47] has a significant positive effect. This innovation gives the material a fundamental advantage over the established systems.

4. Conclusions

The most important accomplishment of this work is the massive simplification of the preparation step with parallel optimization of the product properties for the supported PCM composites. In particular, the separate production of porous support materials is time- and energy-consuming. For aerogels, the drying process with supercritical CO_2 or freeze drying is the limiting step; therefore, these materials only have limited applications. Because with the one-step method established in the present work, the fine structuring of the support is realized already in the PCM itself, the tedious procedures needed in traditional fabrication can be completely omitted. We also propose a rationale for the synergistic behavior of gelators and polymers in the composite structure. In other studies, composites with gelators and polymers have already been used, but without the synergetic effect of these two, which leads to problems with retention property, loading capacity, shape and thermal stability. In particular with paraffin as PCM, the composite of gelator and polyolefin shows a particularly strong improvement. In contrast, PCM immobilized in either only a gelator or only polyolefin showed a strong tendency to creep during prolonged overheating, resulting in shape and material loss.

In principle, all gelators that were studied are suitable for composite synthesis. However, the best results were obtained with the industrially established gelator TBPMN. The PCM loading rate of the supports can be set between 19:1 and 39:1, with a rate of 24:1 giving the best results, which in any case is extremely high. In this range, the amount of energy that can potentially be absorbed per mass of material is only marginally smaller compared with the plain PCM. In the case of a commercial application, the cost of the gelator is the decisive factor. The best composites obtained show a mass loss of 0–5% after 20 cycles. However, this usually occurs during the first few cycles, after which a mass constancy is observed; tests over 50 cycles and thermal stress over 24 h have confirmed the stability of the composites.

It can be assumed that such appropriately stabilized PCM composites have great potential for long-term and environmentally friendly energy savings or recycling. The composites themselves can be renewed by melting and can also be produced with recycled polyolefins. It is hoped that the knowledge gained in this work will help to provide solutions to mitigate problems caused by the climate and energy crisis.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

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