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Biobased phase change materials in energy storage and thermal management technologies

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ABSTRACT

Harnessing the potential of phase change materials can revolutionise thermal energy storage, addressing the discrepancy between energy generation and consumption. Phase change materials are renowned for their ability to absorb and release substantial heat during phase transformations and have proven invaluable in compact thermal energy storage technologies and thermal management applications. Present-day solutions mainly comprise of non-renewable phase change materials, where cyclability and sustainability concerns are increasingly being discussed. In pursuit of sustainable energy models, phase change material research has shifted towards biobased materials. This review explores the growing field of biobased phase change materials, aiming to identify prevailing trends, potential opportunities, and future applications. In addition, the persisting challenges of biobased phase change materials are discussed, where a critical dialogue on the current obstacles and research gaps is presented, offering a glimpse into the future of this rapidly evolving field. The authors furthermore present novel methods to enhance the integration of biobased phase change materials into thermal energy storage applications, ensuring their seamless adoption and maximum efficacy. With an analysis of 180 selected works, this review paints a vivid picture of the capabilities and promising prospects of biobased phase change materials, whilst highlighting the future research questions needing to be addressed.

1. Introduction

Developing and implementing fully sustainable energy storage systems to assist the incorporation of renewable energy sources remains a priority within the already emerged energy-conscious society. The motivation behind this is driven by economic growth, need for sustainable infrastructure development and our increasing awareness of the technological impacts on climate change. Transformation and contribution to sustainability is directly affecting market value of the companies as their consumers are demanding to see the reduction of environmental impacts caused by industries around the world. To achieve a sustainable future, market-dominating technologies cannot rely on rare, depletable, or toxic materials and must be cost-efficient. Therefore, environmentally friendly low-cost alternatives to energy storage in electrical batteries must be researched and developed. One major contribution to forming the sustainable future is to explore the opportunities for incorporation of biobased materials in currently used and newly developed energy storage systems. To evaluate the potential impact of such materials, not only their origin, but an assessment of the whole supply chain, needed chemical modification, stabilisation routes, compatibility, waste management, recyclability, environmental health, and societal impacts must be evaluated.

In the energy storage landscape, thermal energy storage (TES) can have an important role particularly in applications where the final energy demand is in the form of heating and cooling. TES systems allow heat and cold to be stored and released on demand through reversible physical and chemical processes [1]. The three existing types of TES technologies are based on different working principles: (i) sensible heat storage (SHS), latent heat storage (LHS), and thermochemical storage (TCS). SHS are generally low-cost but suffer from low energy density and are, therefore, unfit for applications requiring high storage capacity or involving urban areas as they generally require large storage spaces. In TCS, heat is stored/released during reversible endo/exothermic physical or chemical processes (e.g., water adsorption and absorption). While TCS can store high amounts of energy, the materials used are often expensive, corrosive, and pose health and environmental hazards. LHS exploits the latent heat of phase change whilst the storage medium (phase change material or PCM) undergoes a phase transition

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(solid-solid, solid-liquid, or liquid-gas). During this process, the PCM temperature remains nearly isothermal. The most common types of currently employed PCMs are those absorbing and releasing their melting/crystallization energy (latent heat) during solid-liquid phase changes. Solid-liquid PCMs are typically well-known materials, and can be divided into organic (e.g., paraffins, fatty acids, esters, and poly-alcohols) and inorganic (water, salt hydrates, salts, and metals) [1–4]. The most well-known PCM is water with its ability to undergo phase transitions from solid to liquid to gas at the temperature interval between 0 and 100 °C. For a PCM to be considered as a promising candidate for TES applications, it needs to present a phase change transition temperature relevant to the targeted application, high melting enthalpy (generally >150 J/g), low tendency to supercool, and long-term thermal stability (reproducible phase change) over consecutive heating and cooling cycles [3,5]. Additionally, the PCM should be non-corrosive to its surrounding materials and have minimum impacts on human health and ecosystems [3,6,7]. Thermal conductivity requirements might also vary depending on the application but generally a high thermal conductivity is desired. Supercooling, a phenomenon by which the PCM begins to crystallize well below its melting temperature leads to a temperature difference between the melting and crystallization point that can cause higher energy consumption or even a complete inability of the PCM to crystallize within the temperature range dictated by the system limitations [8]. Minimum supercooling ensures that the melting and solidification processes occur in a narrow temperature range, without the added need for extra energy for the solidification process [8]. Choosing the right PCM starts with a particular application. The selection criteria will, therefore, vary. Detailed guidelines on the selection criteria of typical PCMs were described in 1983 b y Lane et al. [4] and Abhat [9] amongst others. With the abovementioned criteria in mind, other important parameters discussed in a variety of studies include overall sustainability, process safety (low flammability), price and abundancy. Usually, one PCM is not capable of fulfilling all the requirements and a modification or functionalization process is needed to tailor it for a specific application. Another aspect is the increasing demand of carbon-neutral, compact, safe, and affordable storage concepts incorporating sustainable PCMs.

The majority of the commonly investigated and commercial PCMs (e.g., salt hydrates and paraffins) originate from non-renewable sources. Salt hydrates are generally extracted from the ground: for example, sodium sulphate decahydrate is mined as mirabilite [10]. Paraffins are products obtained from the process of crude oil distillation [11]. Considering the important role that TES is gaining in the context of energy turnaround, and the amounts of energy storage materials that will be required in the future, it is clear that developing PCMs from non-depletable resources will be crucial for guaranteeing the sustainable energy future [12–16]. Another important sustainability aspect is future waste management and recycling systems. Increased uptake of biodegradable and compostable PCMs could be a solution for even more sustainable TES. However, besides new opportunities, these materials may come with risks and challenges that must be carefully assessed, so that their properties can also be controlled and preserved in a sustainable manner (Fig. 1).

While the discussion on PCMs from biobased raw materials is relatively new, there are other sectors like the one of plastic production, where biobased materials are being developed and have been discussed for decades. Biobased plastics are typically compared to their fossil-based counterparts in terms of production and end-life sustainability, and it is clear that biobased plastics are not necessarily biodegradable or compostable. Biobased PCMs are made of organic components that can be partially or fully obtained from biomass, vegetable and tropical oils or animal/fish fats. Even though it is often assumed that they are biodegradable, this is a point that needs to be carefully studied for the different types of biobased PCMs. Biobased PCMs are generally non-toxic and can be used in relatively wide temperature ranges for low to high temperature applications (−80 to 275 °C). An overview of the thermophysical properties of the main types of biobased PCMs studied in terms of enthalpies of fusion versus melting temperature is given in

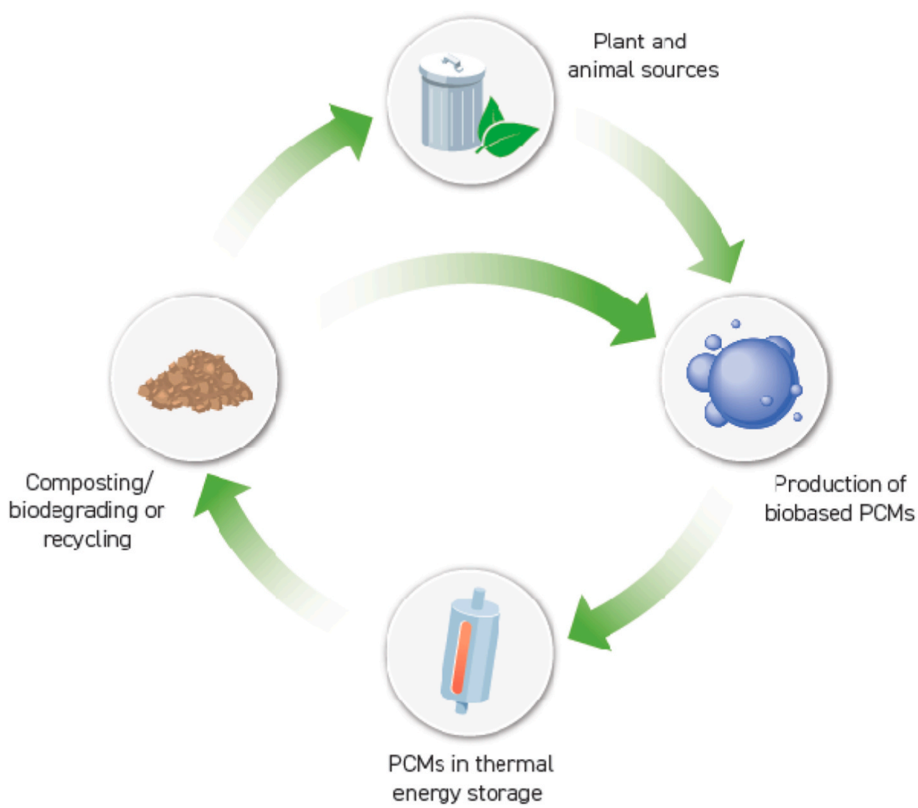

Fig. 1. Biobased PCM in thermal energy storage for a sustainable future.

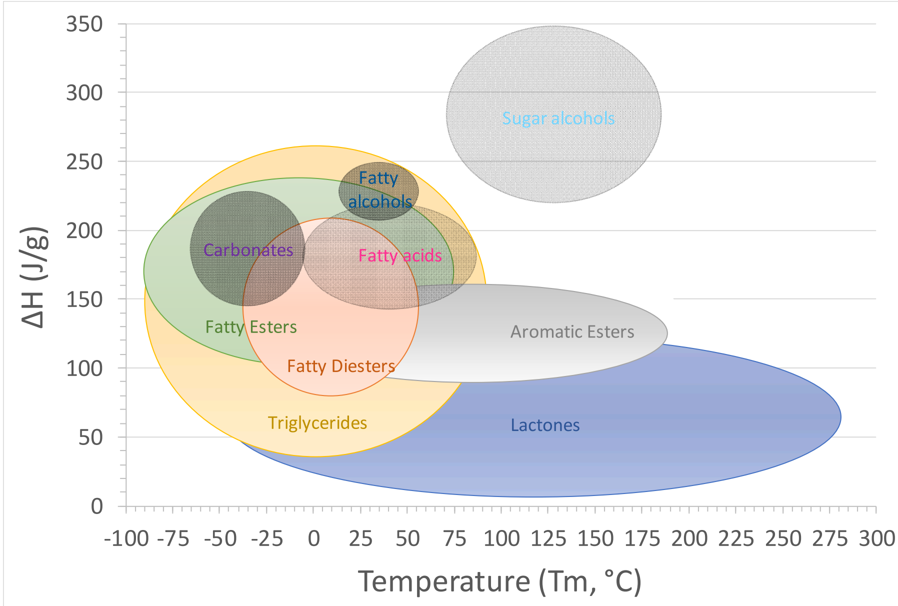

Fig. 2. Main classes of biobased materials with corresponding melting temperature ranges (°C) and enthalpies of fusion (J/g).

Fig. 2.

A large variety of the raw materials allows for PCM production from locally produced sources or even waste found all over the world. The most extensively studied biobased PCMs are esters [17], sugar alcohols [13,14], fatty acids, and their blends (either naturally occurring ones or mixtures produced in the lab) [18–20]. Many of the fatty acids, esters, alcohols, and natural mixtures are produced in large amounts worldwide and, especially at lower purities, could fulfil the abundancy and price requirements for a variety of applications. Earliest studies referring to the biobased PCMs date back to the 1980s and include works of Lane [1,4], Ahbat [9], and Achard et al. [18]. Even though the intrinsic importance of the development of renewable energy storage materials for sustainable future energy systems is widely accepted, relevant PCMs have only been presented in the form of individual studies with no attempt to summarize and compare the properties of the various materials from different chemical classes. The aim of this study is to bridge this gap by providing a comprehensive review of the performance, challenges, and opportunities related to the biobased PCMs that have been developed and investigated to date. Specific properties and limitations characteristic for the biobased PCMs are also discussed in this review.

2. Biobased PCMs and their mixtures

Hundreds of biobased PCMs have already been presented in literature and potentially several thousand exist that have not been investigated yet either in the general literature or specifically within the LHS framework. The majority of the existing studies focus on fatty acids (starting from early 1980s) and fatty acid esters (caught increasing attention since 2010s) as potential PCMs [21–27]. Sugar alcohols have also been the centre of research during the last ten years [12,13,28,29] and others such as alcohols [19]. Each of these classes of materials possesses different characteristics and thermophysical properties due to their overall chemical structures and thermodynamic stability. Section 2 summarizes the results of the published studies and discusses the most important characteristics of the different classes of biobased materials focusing on both pure PCMs and mixtures of pure PCMs without the inclusion of additives or other types of phases. An overview of the reviewed studies is presented in and discussed in the following paragraphs.

2.1. Alcohols

Alcohols are a chemical group of substances characterized by a free hydroxylic (-OH) group in their structure. Different types of alcohols exist, but the most common ones for LHS are fatty alcohols (alcohols from fat sources) and sugar alcohols (alcohols derived from sugars). Sugar alcohols (SA), also called hydrogenated carbohydrates and polyols are found naturally in various fruits and vegetables. SA have the general formula $HOCH_2(CHOH)_nCH_2OH$ (Fig. 3). Even though more than 900 S A are known, only a few of them are produced in large scale, namely sorbitol, mannitol, xylitol, lactitol, maltitol, erythritol, and isomalt. They are non-flammable, non-corrosive, non-toxic, economically efficient (<5 EUR/Kg) and are widely used in the food and pharmaceutical industries [30]. Despite being biobased, SA occur only in small percentages in nature (e.g., fungi, algae, plants), and therefore their extraction from natural raw material is rather difficult [21]. SA are normally produced industrially under strict reaction conditions through fermentation or the enzymatic hydrolysis of starch [22]. However, they are known to have high and stable supercooling (reproducible degrees of supercooling over consecutive cycles), slow crystallization rates and, most importantly, thermal stability issues. For example, D-mannitol, erythritol, xylitol, dulcitol, and galactitol show low chemically stability under thermal cycling [23–25,31,32]. This can constitute a major issue for long-term applications [26]. Despite those challenging traits, SA and th eir mixtures have been widely investigated in recent years for applications into medium temperature ranges of 75–150 °C due to their environmental friendliness and impressive latent heats of fusion between 219 and 340 J/g, making them one of the PCM classes with the highest energy densities [12,13,33,34,35] It should be noted that understanding the SA behaviour as PCM was significantly accelerated through a SAM.SSA project funded by the European Commission during 2012–2015. In this project, SA were investigated as cost-effective and

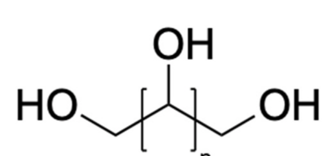

Fig. 3. Sugar alcohol.

environmentally friendly materials for compact seasonal TES applications. In this context, novel SA were characterized as PCMs [13] and thermal stability and kinetic crystallization behaviour of SA were studied in detail [23,28].

Despite having similar properties, fatty alcohols have been studied less than SA. This is presumably due to higher costs and a general lower availability compared to SA. The investigated alcohols are generally straight-chain primary alcohols with chain lengths varying from 2 to 26 carbon atoms. The most widely used fatty alcohols in LHS comprise of 1-hexadecanol and 1-tetradecanol [36]. Other fatty alcohols investigated include 1-hexadecanol and 1-octadecanol [36,35]. Overall fatty alcohols present melting ranges from 20 to 60 °C and enthalpies of fusion of 210–250 J/g. They have been shown to be highly biodegradable, which makes them interesting for applications in the food, cosmetic and drug-delivery sectors [33]. Nevertheless, many more remain currently untested, and therefore only limited information is available on this class of PCMs.

2.2. Fatty acids

Fatty acids are the first and only biobased PCMs mentioned by Abhat in his 1983 classification of the materials used in TES technologies [9]. Saturated and unsaturated fatty acids have been studied widely in the previous years. They are commonly found in natural oils and algae and are non-toxic [37]. Saturated fatty acids are described by the general formula $CH_3(CH_2)_nCOOH$ (Fig. 4). Fatty acids are usually produced industrially through the hydrolysis of fats (triglycerides) with high purities [38]. Fatty acids can also be efficiently extracted from dried biomass and algae, although in these cases, mixtures rather than single components will be obtained [39]. For LHS applications, fatty acids with carbon chain lengths ranging from 3 to 9 carbons have been mostly investigated with melting points between 16 and 70 °C and enthalpies of fusion of 140–220 J/g [17]. Even though most studies report that saturated fatty acids have small degrees of supercooling [40,41], it was recently reported that their supercooling degree depends strongly on their thermal history [42]. They are reported to be mildly corrosive. Yuan et al. [40] published a review on fatty acids as PCMs highlighting the applications, thermal properties and challenges connected to these materials in relation to the TES field. The authors found from studies in the literature that fatty acids do not undergo significant changes in either the melting points or the enthalpies of fusion after 1000 thermal cycles.

2.3. Esters

Esters, which commonly occur in nature in vegetable and animal fats, are compounds formed from the union of a carboxylic acid and alcohol. They have a general formula RCOOR', with R being a H atom, an alkyl group, or an aryl group, and R′ being an alkyl or an aryl group (Fig. 5). Since esters are comprised of carboxylic acids and alcohols, millions of them exist, each with their own specific thermophysical properties, making them an impressively large, mostly unexplored class of possible PCMs [34,35]. As well as being found in nature, esters can be synthesized with a catalyst (either an enzyme or an inorganic compound) through a process named Fischer esterification or alcoholysis [43,44]. Esters can be produced from a variety of sources, including waste (e.g., cooking oils, plastics) [45,46]. For example, a variety of promising alkyl threo-9, 10-dihydroxystearates for LHS applications

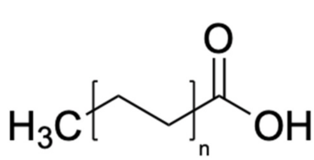

Fig. 4. Saturated fatty acid.

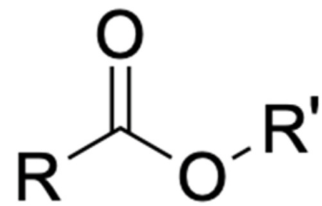

Fig. 5. Ester.

were synthesized from non-edible animal fat using a regeneratable *Candida antarctica Lipase-B* biocatalyst [47]. Alternatively, new esters can be produced from existing ones by simply exchanging the substituent attached to the carboxylic group (namely R') through a process called transesterification [48]. This is particularly advantageous when expensive esters are produced from cheap and easily available ones. Riemenschneider and Bolt [44], give an extensive review of the properties, production methods, and applications of esters.

Depending on the specific chemical moieties comprising their structures, esters can be further divided into subclasses. The most explored ones in terms of LHS applications are fatty esters (saturated), diesters (saturated), and triglycerides (saturated, unsaturated, symmetrical, and asymmetrical). Only limited studies have emerged on lactones, aromatic esters, and carbonates. Generally, esters show little to no supercooling, high chemical and thermal stability and no corrosiveness [49] and are considered only moderately flammable [50]. Compatibility of esters is ensured with all types of metals, but some limitations can be encountered with polymers, as long-chained esters are highly lipophilic. Stamatiou et al. [51] tested eleven commercially available unbranched, saturated carboxylic esters with molar masses between 88 and 509 g/mol and reported melting temperatures between −83 and 54 °C and fusion enthalpies in the range of 99–219 J/g. One of the main barriers for their investigation is the lack of high purity esters that can be commercially purchased and tested. Recent studies have bypassed this limitation by systematic synthesis of novel esters [15,52,53]. Ravotti et al. [52,53] synthesized carboxylic fatty esters and fatty diesters [54] with melting points in the range of 10–50 °C and −50 to 50°C respectively and found that they had relatively high enthalpies of fusion up to approximately 200 J/g. This was first discovered by Aydin et al. [,55–58] through a series of systematic experiments on synthesized fatty esters and diesters. Additionally, Ravotti et al. [53] discovered that higher thermal stabilities were found for esters with longer carbon chain length. Some trends correlating the melting points to the chemical structures were highlighted as well, which might open the way to future research on the prediction of melting points from structures alone. Generally, esters derived from odd-numbered alcohols were found to have lower melting points and enthalpies. This is supposedly due to lower degree of packing in the crystal structure. If this was the case, the same should be observed for unsaturated fatty esters as well. Unfortunately, studies on the thermal properties of unsaturated esters in the context of LHS have not been published to the best of the authors' knowledge.

Two classes of esters that were less investigated overall are those of lactones and aromatic esters. Ravotti et al. [15] tested three synthesized and two commercial biobased lactones and eight commercial aromatic esters but found that they are mostly unsuitable as PCMs due to high supercooling and extensive degradation at their respective melting temperatures. Only 2,6-dimethyl naphthalene dicarboxylate with densities >1 g/ml, enthalpy of 160 J/g and a melting point of 190 °C could be a suitable PCM for high temperature applications. Recently, Ravotti et al. [59] also discussed the ester class of triglycerides as promising for LHS applications on the basis of a literature review from properties reported mainly in the framework of food science-related studies. The subclass was recognized as very promising with melting points ranging from sub-zero to above 90 °C and with enthalpies of fusion in some cases reaching well above 200 J/g. However, the issue of polymorphism was recognized as a barrier that needs to be better understood and controlled for a successful implementation of triglycerides as PCMs [59]. A special class of esters belonging to both diesters and fatty esters is that of oleochemical carbonates, or simply carbonates. These are esters derived from carbonic acid and have been reported to possess interesting properties with enthalpies >200 J/g and melting points in the sub-zero region. Therefore, these compounds could be interesting for low temperature applications. Nevertheless, up to date, this class remains mostly undiscovered and further studies would be required.

From all the classes of esters that have been investigated, to date, saturated fatty acid esters [15,52] and saturated diesters [54,55,60] represent the most convenient ones in terms of range of melting temperatures, enthalpies of fusion, stability, costs and commercial availability. In particular, esters of myristic, palmitic, stearic, arachidic and behenic acids were shown to exhibit high enthalpies and low supercooling [52]. Stearic acid esters have been widely investigated for their potential application in building insulation [61]. They showed thermal stability for over 1000 cycles [61]. However, for these materials as well, the issue of polymorphism persists, although less prominently than for triglycerides [62,63]. An overview of the most frequently studied pure biobased PCMs (not mixtures) is presented in Table 1. The PCMs are sorted according to material classes reflecting their similar thermal behaviour.

2.4. Mixtures

Organic and non-organic PCMs alike can be used in combination with each other as well in binary, ternary, or even quaternary mixtures. Most often though, binary mixtures are produced to reach eutectic and peritectic points. While this is done more often with salt hydrates, mixtures of organic PCMs have been tested as well. PCM mixtures are useful to fine-tune the thermophysical properties and especially the melting points. Multiple studies from 1990s and early 2000s consider properties and potential applications of biobased PCM mixtures, mostly presented by the compositions made of fatty acids and esters [61,78,80,81] [82,83–85]. Binary and non-binary mixtures in general are quite attractive with regard to applications where the target melting point is not covered by pure substances. Yuan et al. [40] and Sari [82] have summarized the data from the main binary eutectic mixtures of fatty acids studied. In general, fatty acid binary mixtures show a range of melting points from 19 to 52 °C and enthalpies of fusion from 120 to 200 J/g. Fatty acids have been mixed with fatty alcohols as well to achieve different thermophysical properties. In particular, Zuo et al. [86] mixed caprylic acid with 1-dodecanol and lauric acid with 1-tetradecanol and achieved enthalpies of 170 to 160 J/g and melting points of 6.5 and 24.3 °C respectively, thus lowering the original melting points of the corresponding fatty acids. Rubio-Perez et al. [87] recently summarized the thermophysical properties of binary mixtures of fatty esters. Generally, the binary mixtures of fatty esters showed melting points ranging from −50 °C to 32 °C and enthalpies of fusion up to 200 J/g. Rubio-Perez et al. [87] showed that fatty esters have been commonly mixed with paraffins as well to give new PCMs with melting temperatures from −30 °C to 36 °C and enthalpies of maximum 210 J/g. Some of the mixtures of fatty acid esters with paraffins exhibit thermal stability for over 500 cycles [64]. While these are generally attractive properties, it must be kept in mind that paraffins are a by-product of fractionating columns from petrol refinement and as such are not considered biobased or sustainable.

2.5. Waste materials and multi-component substances

As esters, alcohols and fatty acids are organic biobased compounds, they occur naturally in a wide variety of substances, such as oils, waxes, animal and vegetable fats. However, all these substances are not characterized by precise, well-defined compositions, but rather by variable

Table 1
Overview of biobased PCMs and their thermophysical properties.

Material	Production route or material source	Melting temperature, °C	Melting enthalpy, J/g	References
Pure PCMs				
Fatty acids				
Fatty acids	Caprylic, capric, lauric, myristic, palmitic, stearic, oleic, pentadecane acids	0–88	140–220	[1,9,21,64–72]
Esters				
Fatty esters	Myristic, sebacic palmitic, stearic, arachidic and behenic acid esters, Threo-dihydroxystearic esters	−83–77	99–234	[19,34,47,52,,,58,61,73–76]
Lactones	Cyclic esters: ε-caprolactone, γ-valerolactone, 1,2-campholide, oxaadamantanone, and dibenzochromen-6-one	- 40–280	8–122	[15]
Fatty Diesters	Dimethyl oxalate, dipenthyl oxalate, dodecyl oxalate, dimethyl suberate, dipentyl suberate, dodecyl suberate, dimethyl sebacate, dipentyl sebacate, odecyl sebacate, dimethyl succinate, dipentyl succinate, odecyl succinate, didodecyl-1,8-octanedioate, ditetradecyl-1,8-octanedioate, dihexadecyl-1,8-octanedioate ditetradecyl-1,10-decanedioate, ditetradecyl-1,12-dodecanedioate, ditetradecyl-1,14-tetradecanedioate	−32–58	80–210	[62,77,78]
Triglycerides	Literature review for 25 saturated symmetrical triglycerides, 19 unsaturated symmetrical triglycerides, 15 saturated asymmetrical triglycerides	−84–93	36–261	[59]
Aromatic esters	Methyl-2-furoate, anysil acetate, benzyl benzoate, dimethyl terephthalate, dibenzyl oxalate, methyl-2-methoxybenzoate, phenyl benzoate, 2,6-dimethyl naphthalene dicarboxylate	−16–190	90–160	[16]
Oleochemical carbonates	Transesterification with dimethyl or diethyl carbonate in	−50–0	144–227	[77]

(continued on next page)

Table 1 (*continued*)

Material	Production route or material source	Melting temperature, °C	Melting enthalpy, J/g	References
	the presence of a catalyst			
Alcohols				
Fatty alcohols	1-tetradecanol, 1-dodecanol, 1-hexadecanol, 1-octadecanol	20–60	210–250	[19,36]
Sugar alcohols	Sorbitol, mannitol, xylitol, lactitol, maltitol, erythritol, erythrol, isomalt, galactitol, D-sorbitol	75–189	219–352	[12,13,28,79]

mixtures of multiple components, which are inevitably influenced by external parameters such as the environmental conditions. Olive oil for example consists of mainly oleic acid triglycerides together with lower amounts of various fatty acids (e.g., linoleic, palmitic, palmitoleic). The exact composition however strongly varies with altitude, extraction process, and weather conditions. Natural waxes are mixtures of various compounds, including fatty acids and their esters, primary and secondary alcohols, ketones, sterols, and aldehydes [65], but the exact composition is also influenced by the environment in which they are formed. While multi-component substances typically present broader melting ranges often characterized by several peaks, they can be extracted from waste materials, and therefore prove to be advantageous with respect to applications that don't require a precise melting range. Multi-component substances have only been limitedly studied with regards to TES applications. Nevertheless, a few studies on the matter have been published in recent years.

Gunasekara et al. [66] studied commercial-grade virgin olive oil from Spain as a possible PCM. The authors cycled the olive oil sample four times and analysed it through T-history to find that the melting points and enthalpies stayed relatively consistent. As expected, due to the presence of triglycerides, evidence of polymorphism was noticed. In general, the total melting and freezing enthalpies are 105 ± 11 and 97 ± 8 J/g over the temperature ranges −4.5 to 10.4 °C and −8 to −11.9 °C, respectively. This proves that part of the material does not crystallize, thus giving rise to a difference between the enthalpies of fusion and crystallization. Fabiani et al. [67] obtained a PCM from residual animal fat from slaughterhouses and analysed it by means of differential scanning calorimetry and thermogravimetric analysis. The PCM showed a multi-peak profile with melting occurring at approximately 20 °C and crystallization at 10 °C. However, the phase change was characterized by a very low enthalpy of fusion ranging between 5 and 23 J/g, which would make the material unsuitable for any LHS application. Silalahi et al. [68] and Alqahtani et al. [88] studied coconut oil as a possible PCM for buildings and room-temperature applications. Authors from both groups found that coconut oil melts over a wide range of temperatures and presents only limited supercooling, with peak freezing temperatures around 15–18 °C and melting temperatures of up to 22 °C. The enthalpy was found in both cases to be around 100 J/g. Although this is not considered high for LHS applications, Silalahi et al. claimed that this is enough to lower the temperature of a room facing south with coconut oil embedded in the walls and windows by approximately 23%. An overview of the most frequently studied biobased PCM mixtures is presented in Table 2. The PCMs are sorted according to material classes reflecting their similar thermal behaviour.

Table 2
Overview of biobased PCMs mixtures and their thermophysical properties.

Material	Production route or material source	Melting temp., °C	Melting enthalpy, J/g	References
Binary mixtures of fatty acid esters and fatty alcohols	1-dodecanol/methyl stearate, 1-dodecanol/methyl, palmitate, 1-tetradecanol/methyl stearate, 1-tetradecanol/methyl palmitate	20,3–32	202–224	[73]
Binary mixtures of oleochemical carbonates	Dodecyl carbonate, tetradecyl carbonate, hexadecyl carbonate, octadecyl carbonate	12–37	135–175	[89]
Binary mixtures of fatty acids	Capric/lauric (45/55 wt%), capric/lauric (65/35 mol %), capric/lauric (61.5/38.5 wt%), capric/myristic (73.5/26.5 wt%), capric/palmitic acid (75.2/24.8 wt %), capric/stearic (86.6/13.4 wt%), lauric/palmitic (64/36 wt%), lauric/palmitic (69/31 wt%), lauric/palmitic (77/23 wt%), lauric/palmitic (80/20 wt%), lauric acid/myristic (62.6/37.4 wt%), lauric/myristic (66/34 wt%), lauric/stearic (75.5/24.5 wt%), myristic/palmitic (51/49 wt %), myristic/palmitic (58/42 wt %), myristic/stearic acid (65.7/34.7 wt %), myristic/stearic acid (64/36 wt%), MA_2SA (myristic acid/stearic), MA_3PA_2 (myristic acid/palmitic), palmitic/stearic (64.9/35.1 wt%), palmitic/stearic (64.2/35.8 wt%), PeASA (pentadecane/stearic); PASA (palmitic/stearic)	18–21	143–148	[19,81,82,78,90–97]
Binary mixtures of fatty acid esters with paraffins	Ethyl oleate, ethyl linoleate, ethyl palmitate, ethyl myristate, ethyl stearate, methyl palmitate, methyl stearate, methyl nonadecanoate, eicosane, tetracosane, octacosane, hexadecane, decane, dodecane	−30–36	up to 210	[19,87,64]

(*continued on next page*)

Table 2 (*continued*)

Material	Production route or material source	Melting temp., °C	Melting enthalpy, J/g	References
Binary mixtures of fatty acid esters with other esters	ethyl palmitate, methyl palmitate, ethyl stearate, methyl stearate, methyl myristate, methyl oleate, methyl linoleate, methyl laurate, ethyl laurate, ethyl oleate, ethyl linoleate, ethyl caprate, ethyl caprylate, methyl heptadecanoate, methyl laurate	−50–32	up to 200	[87]
Binary mixtures of fatty acid esters	Methyl palmitate/methyl stearate (65–90/35–10 wt %)	22–25.5	120	[98]
Non-binary mixtures of fatty acids	Residual animal fat: 52% unsaturated and 42% saturated fatty acids; palmitic acid/stearic/other fatty acids (50/45.5/4.5 wt%); stearic/palmitic/other fatty acids (65/27.5/5.5 wt%); stearic/palmitic/other fatty acids acid (83/11/6 wt%)	2 and 25,51–66	29,180–206	[67,99]
Non-binary mixtures of fatty acid esters	Butyl palmitate/butyl stearate/other esters (49/48/3 wt %)	17	140	[100]

3. Enhanced biobased PCMs and biobased PCMs in encapsulation

3.1. Nanomaterial enhanced biobased PCMs

Several experimental and numerical studies focusing on enhancing thermal conductivity, reducing thermal resistances and reducing the supercooling in biobased PCMs through the addition of more conductive components, including various nanomaterials has been researched. Some of the methods include the dispersion of carbon nanotubes and graphite nanoplatelets in biobased PCMs, which have resulted in a 375% increase in thermal conductivity [83,84]. Additionally, graphite nanoplatelets combined with fumed silica for lightweight applications in buildings have been investigated [85]. One of the conclusions from those studies is that the nanomaterial fraction should be kept at 5–10 wt % to avoid undesirable changes in the thermophysical properties, such as increased supercooling and changes in melting enthalpies. The addition of CuO nanoparticles into a coconut oil PCM was reported to improve the melting process of the PCM in a cylindrical thermal energy storage system [101]. CuO nanoparticles were also shown to enhance the thermal conductivity of the coconut oil PCM by up to 7,5% and enhance the melting efficiency as the melting process proceeds [102,103]. Al_2O_3, CuO, Fe_3O_4, and TiO were shown to enhance melting process and energy storage rates of a coconut oil PCM by 57,5 and 1,2%, respectively [104].

3.2. Biobased PCMs in encapsulation

Encapsulation is typically used to prevent PCM leakage, improve heat transfer and add mechanical stability. The techniques can be classified as macro- and microencapsulation or matrix encapsulation, all significantly differing in price and applications. Emulsion-templated materials or encapsulation matrices are made of emulsions converted to polymeric structures [105]. These porous matrices can be used to accommodate other materials, including PCMs. Besides sphere encapsulation widely reported for PCMs from non-renewable sources, soybean wax PCM was encapsulated in the form of electro-spun fibers covered with a protective polyurethane shell. The PCM fraction varied from 10 to 60% and uniform fibers with good thermal stability and thermal resistance for over 100 thermal cycles were obtained (thermal storage and thermal shielding applications suggested) [106]. Butyl stearate was used as encapsulated PCM material in an emulsion-templated polysaccharide matrix made of biodegradable pullulan [107]. A biobased PCM with a melting point of 58°C was contained in aluminium bottles and studied for hot water usages by Mongibello et al. [108]. 1-Octadecanol and fatty acids with relevant melting temperatures are considered as promising PCM candidates for this application.

Microencapsulated coconut oil PCM with a melting enthalpy of 119 J/g was obtained through suspension polymerization with stearyl methacrylate and hydroxyethyl methacrylate [109]. Microcapsules with a core of capric acid PCM encapsulated in a polylactic acid shell were prepared by Skurkytė-Papievienė et al. [110]. Multiwall carbon nanotubes were used to improve thermal conductivity of the microcapsule core and shell. It was also registered that too high nanotube concentrations negatively affected enthalpies of the tested encapsulated PCM and PCM microcapsules.

4. Biobased PCM composites

4.1. Biobased PCM composites with non-biobased materials

Preparation of PCM composites with other materials is used to improve mechanical stability and thermal conductivity. This is typically done by incorporating a PCM into another material or vice versa. The majority of modification routes involve the use of non-biobased components that affect the biobased material's environmental benefits and overall embodied energy of the original material. One of the reported approaches is the addition of up to 10% of fatty acid PCM mixtures with desired melting enthalpies into concrete to be used for prefabricated building elements. The PCM can be either dispersed or added in a microencapsulated form [111,112]. The latter resulted in 13% energy savings over a 2-year testing period in test cabins. 56% of coconut fat was prepared by immersion into a mixture of clay and cellulose fibres. The resulting switchlike panels showed no PCM leakage as they were placed between protective steel sheets [113]. Hydrogenated palm kernel fat was used in a similar approach and where panels showed reduction in melting enthalpy from 74 to 40 J/g compared to the original PCM [114]. Form-stable fatty acid/polyethylene terephthalate composite fibres were obtained through electrospinning as increasing fatty acid content was shown to lead to increased latent heat of the composite fibres as well as increased fibre thickness. The phase transition temperature could be maintained for over 100 heating-cooling cycles [115].

4.2. Biobased PCM composites with other biobased materials

A completely biobased composite made of coconut oil PCM and impregnated in biochar showed promising properties for being used as an insulation material with a melting temperature of 24 °C and a melting enthalpy of 74.6 J/g [116]. Hardwood samples were impregnated with 10–50% of commercial microencapsulated PCM (biobased material from agricultural residues) with a melting point of 23 °C. A 77% enhancement of the heat storage capacity has been reported, which makes the PCM impregnated wood a promising material in wood flooring applications [117]. A full-scale test of wood panels impregnated with the same microencapsulated PCM showed a reduction of up to 41% during testing the heating consumption of a hut under seasonal variations [118]. Decorative wood panels impregnated with the microencapsulated PCM showed similar performance when compared to the panels impregnated with a mixture of capric and lauric acids [119]. Yoo et al. [120] produced PCM composites with spent coffee grounds as the supporting material and natural waxes as the energy storing material and found that broader melting ranges could be achieved. The composite materials presented melting peak ranges of 40 °C compared to 20–30 °C of the starting natural wax. However, the latent heats of fusion were found to significantly decrease from values > 100 J/g to values < 80 J/g. It was evident from their study that natural waxes do not crystallize completely as well, thus delivering a lower enthalpy of crystallization compared to that of fusion. All these aspects could be identified as challenges to the implementation of these kind of materials in LHS applications. Additionally, the authors reported a summary of the previous studies conducted on biocomposite PCM. Generally, these materials are characterized by enthalpies of fusion <150 J/g, which once again questions their applicability in LHS applications.

5. Biobased PCMs in dispersions

Oftentimes, it is useful to have PCM in the form a pumpable fluid, which is capable of both transporting and storing heat. Generally, these fluids are used in temperature stabilisation applications, such as within the machining industry, pharmaceuticals, and the cooling of electronics. More commonly, these fluids can be referred to as phase change dispersions (PCD) or phase change emulsions (PCE) and are mixtures of PCM and another immiscible fluid. Preventing phase separation in such systems is achieved through addition of surface-active compounds acting as stabilizers. Traditionally the PCM of choice in PCD is paraffins [121], however paraffins are derived from non-sustainable oil feed stocks, which subsequently removes PCD from contributing to a sustainable energy model. Despite this, in recent years, focus has been placed on using biobased PCMs for integration into PCD formulations. So far, few researchers have used biobased PCMs in PCD formulations, however from those that have, promising results have been obtained. Fischer et al. [122–124] used a combination of two fatty acid esters in a three-part investigation to be used in the cooling of High Voltage Direct Current (HVDC) components and found up to a 50% reduction in the heat sink surface temperature compared to standard industrial coolants under the constant pumping power assumption. Additionally, Zhang et al. [125] investigated fatty acid esters as potential PCMs for use in PCD due to their sustainability and non-toxicity. They found a wide range of eutectics which were suitable for numerous temperature stabilisation applications. Additionally, the PCD displayed highly favourable thermophysical properties, such as large specific heat capacities. However, when formulating PCD, numerous other materials are often enlisted to tackle issues with stability and crystallization, such as surfactants and nucleating agents. Currently, there is a very limited number of studies on the development of completely biobased PCD, however O'Neill et al. [126] formulated a PCD with fatty acid esters and with sucrose surfactant systems which led not only to a completely biobased PCD, but one with high stability, low viscosity, and high specific heat capacity.

Another type of PCM storage, which achieves high power, is direct contact latent heat storages, which constitute of the PCM being in direct contact with the heat transfer fluid. In these systems, the heat transfer fluid streams though the PCM ensuring efficient heat transfer between the immiscible phases, minimum stability is required to prevent emulsion build up in the system. Direct contact latent heat storages envisage to tackle the issues of PCM's inherently low thermal conductivities to allow fast charging and high thermal powers. In general, direct contact latent heat storage is a relatively understudied area of research, however some small laboratory set-ups have been developed and the main biobased material combinations that have been tested are erythritol/oil [127,128] and esters/water [129,130].

6. Current and future applications of biobased PCMs

Firstly, it is important to distinguish that different PCMs should be used in different applications based on temperature, weight and volume requirements and based on economic feasibility. For temperature requirements, four broad temperature ranges are considered and covered in this review. The temperature ranges are -20-6 °C, considered the low temperature range and covers applications such as commercial and domestic refrigeration and the transportation of temperature sensitive goods. The second temperature range to be considered is 6–40 °C and is classed as the medium-low temperature range covering passive and active heating and cooling in buildings, air conditioning systems and machine industry cooling. The third temperature range is 40–80 °C, which is the medium temperature range and encompasses applications such as domestic hot water systems, the cooling of electronic devices and solar air heaters. The fourth and final temperature range to be considered is 80–200 °C, named the high temperature range, and includes applications such as waste heat recovery and solar absorption cooling. Within the following sections, the temperature ranges and their subsequent applications are discussed in further detail. The specifications in regard to important parameters other than melting points, such as weight, volume restrictions and economic factors, are considered and discussed. Additionally, biobased PCMs that have already been investigated in specific applications are presented, and the authors present more biobased PCMs which fulfil the criteria of each application.

6.1. *Low temperature ranges: 20-6 °C*

Refrigeration transport is needed in the preservation of fresh and perishable products during transportation to minimize metabolic and microbial deterioration [131]. Biobased PCM solutions can be implemented into refrigeration trucks to replace the current expensive, noisy and polluting refrigeration technologies employed. The main considerations for implementation of PCM into refrigeration vans include the PCM having a high energy density to save weight in the transport vans and for the PCM to have the correct melting temperature. The melting temperature should be slightly below the lower temperature range of the application to minimize the required heat transfer area, but not be too low to prevent lower operational efficiency and energy consumption during charging [131]. Khan [132] gave a detailed summary of the requirements for PCM to be implemented into refrigeration technologies and these are split into, physical requirements, such as thermal cycling stability, large phase change enthalpy and suitable phase transition temperature, technical requirements such as; a low vapour pressure to reduce the requirements of mechanical stability, a small volume change upon phase change; compatibility of the PCM with the encapsulation material and surrounding materials in case of leaks and safety restraints such as, legislative restrictions such as using nontoxic, non-flammable materials. Additionally, Khan includes a list of economic requirements regarding the development of a marketable product such as a low price to be competitive and good recyclability of the PCM for environmental and marketing reasons [132]. Zhan et al. [133] investigated performance of biobased PCMs made by a mechanical agitation of edible additives (D-(+)-glucose, glycine and D-sorbitol) and a 1% NaCl solution. The new PCMs are suggested for applications within fruit and seafood refrigerated transportation systems (temperature ranges between −4 and -6 °C). In a 2022 review summarizing innovative PCM applications in cold-chain logistics for agricultural product storage, Zhao et al. [134] shortly mention capric-lauric acid mixtures as suitable PCMs. To date, there is no literature on the current implementation of biobased PCMs into refrigeration technologies, but given the requirements, mixtures of fatty acids, esters, diesters, and triglycerides could be promising candidates.

Typically, in domestic refrigeration applications, PCMs are incorporated onto the condenser side as heat storage, and on the evaporator side as cold storage to overall improve the operation performance of the refrigeration. For refrigeration technologies that require a phase change temperature close to 0°C, water with nucleating agents presents as great biobased PCM. Additionally, oleochemical carbonates are suggested as potentially high performing PCMs, with didecyl carbonate melting at 3°C and dioleyl carbonate melting at 0°C [135].

6.2. Medium-low temperature ranges: 6–40 °C

PCMs have been integrated into the passive and active cooling and heating of buildings and evaporative and radiative cooling systems to not only improve thermal comfort but to reduce the total energy consumption. Additionally, it has been stated by Cabeza et al. [136] that integrating biobased PCMs into building materials for passive cooling/heating can reduce the embodied energy of buildings. The requirements for integration of PCMs into building materials are for the PCM to melt in the correct temperature range, to be non-flammable and non-toxic and to have high enthalpies of fusion.

Vik et al. [137] used a commercial PCM called BioPCM Q23 M[51], with a melting point of 23 °C and integrated it into an office cubicle in the summer in Oslo, Norway, under aluminium ceiling plates and gypsum wallboards. The authors state that a significant passive cooling effect was achieved through using 17 m^2 of PCM-laden plates. Boussaba et al. [138] integrated coconut fat, recovered from underused feedstocks, with a melting point of 22.7 °C, into building envelopes to increase the thermal inertia. Eller et al. [139] also used coconut oil for passive cooling in 12 climates, where it was found that coconut oil provided year-round benefits in tropical and sub-tropical climates. Coconut oil has environmental advantages, wide availability and economic feasibility. Considering the requirements, butyl stearate/butyl palmitate and capric/lauric acid mixtures as well as propyl palmitate are reported as promising candidates for incorporation into wallboards by either direct incorporation through mixing or immersion [19]. Butyl stearate/butyl palmitate mixture, 1-tetradecanol and 1-dodecanol can be used in concrete blocks by direct incorporation, immersion, or microencapsulation [19]. Cellat et al. [112] used microencapsulated eutectic mixtures of capric/lauric acid fatty acids for passive solar storage energy. They monitored the passive experimental data for two years and 13% of energy savings were found. Fatty acids were found to be optimal due to their relatively high enthalpy of phase change, non-flammable behaviour and from their environmental inertness [140]. Compared to paraffins, fatty acids have smaller volume changes during phase transitions, higher latent heats per unit mass, availability at different melting temperature ranges and low flammability [141]. Wi et al. [142] compared different biobased PCMs; coconut oil, palm oil and commercial biobased PCMs for improving the thermal inertia in wallboards for passive cooling and heating.

For use in buildings, fatty acids are promising biobased PCMs because they are renewable, non-toxic, commercially available at low cost, and biodegradable, all which can help contribute to a sustainable energy model. Additionally, fatty acids have congruent melting, chemical and thermal stability and are therefore appropriate for low to medium temperature energy storage applications. However, normally the room temperature for summer and winter conditions are given to be in the range of 23–25.5°C and 21 to 23°C respectively and unfortunately, most fatty acids have melting points outside of this range, and so eutectics of fatty acids would work better. PCMs integrated into floorboards or underfloor heating have also been suggested for building heating applications. For example, the coupling of PCM layers with underfloor radiator heating systems is considered one of the most efficient active heating techniques, which requires PCM that melt in the range of 18 to 30°C for thermal comfort and optimal floor surface temperature. Additionally, the heat of fusion should be in the range of 120–160 J/g and with a thickness that must not exceed 2 cm, so the PCM must have a relatively high energy density. Huang et al. [143] added macroencapsulated capric acid into the floor in conjunction with solar water heating. Other functional materials that have been used for passive cooling/heating in buildings have come in the form of impregnated wood. Mathis et al. [117] placed biobased commercial Puretemp29 PC M (of agricultural origin) into the upper layer of wooden floorboards to increase the thermal mass of the floorboards which could store solar energy and thus improve building's energy efficiency. The 2000-m^2 ZEB (Zero Emission Building) Lab building in Trondheim (Norway) utilizes an integrated heat storage unit filled with an organic PCM/biowax (CrodaTherm 37) derived from plant-based feedstocks [144]. It is described as a crystalline wax when in solid state or oily liquid when liquified. The PCM was chosen based on the desired melting temperature of 34–37 °C, low supercooling, high flashpoint, and sustainability. The PCM unit is shown to successfully accumulate energy to be used as a heat source during colder periods [145].

Additionally, PCMs in this temperature range have been used for solar absorption chillers to improve the operational efficiency and furthermore, for air conditioning systems to move the daytime electricity load to night-time. Low-temperature solar devices typically operate in the 20–80 °C interval, and many biobased PCMs have been investigated for their potential application in solar thermal storage technologies. Khan et al. [146] suggested 1-Tetradecanol, a eutectic mixture of capric-lauric acid, dimethyl sebacate, a eutectic of myristic acid and capric acid, vinyl stearate and capric acid, which are all fatty acids, would be good candidates for storing the waste heat from solar absorption chillers.

Another potential application of biobased PCMs in the medium-low temperature range is in the cooling of electronic components using the biobased PCMs in the form of a pumpable fluid, e.g., a PCD. Fischer et al. stated that the requirements from the PCD is that for the cooling of electrical components, the PCM should have a low electric conductivity below 0.3 S cm^{-1}, have a high specific heat capacity within the desired melting range (generally stated as twice that of water at the same mass flow rate), have a low viscosity and pressure drop and have high heat transfer coefficients [122].

It has also been theorized that PCMs that melt in this temperature range can be used in thermoregulated fabrics. Paraffin based PCMs cannot be used in textiles due to their high flammability, low thermal conductivity, irritant behaviour and them not being eco-friendly. Skurkytė-Papievienė et al. [147] manufactured microcapsules for textile applications with a core of capric acid PCM encapsulated in a polylactic acid shell. Hsu et al. [148] synthesized fully biobased thermoplastic polyester PCM, to be used in textiles, medical support materials and coating applications with a melting point of 20°C. When designing biobased PCMs for integration into thermoregulated fabrics, the compatibility between the fabric material and the PCM needs to be carefully understood.

6.3. Medium temperature range: 40-80°C

One of the most studied applications for PCM technologies is within the domestic hot water market, by using PCMs as thermal energy stores to utilise domestic hot water duringF off-peak rates. Within domestic hot water systems, Abokersh et al. [149] stated there are four different types of requirements that PCM must fulfil, these are thermal properties, such as suitable operational temperature range, high latent heat per unit mass, high specific heat capacity and high thermal conductivity. The next criteria are physical properties such as high densities, low density variation upon phase change and small degrees of supercooling. Chemical properties such as being chemically stable, non-toxic and non-flammable were also listed as very important. Furthermore, there are economic factors to consider such as the PCM must be available in large quantities and be inexpensive. PCM can be integrated into domestic hot water systems to reduce off-peak power consumption, and to work during power outages. Mongibello et al. [108] used a commercial biobased PCM with a melting point of 58°C in aluminium cases inside a water tank for domestic hot water usages and found significant improvement in the energy stored. Other than this, few biobased PCM have been implemented for application into domestic hot water storages [150]. Here, the authors recommend that 1-octadecanol, with a melting point of 57°C and a high latent heat capacity of 252 J/g should be considered as a potential PCM candidate for use in domestic hot water storage. Additionally, fatty acids such as palmitic acid, with a melting point of 61°C and a latent heat of 211 J/g present as attractive candidates.

Additionally, in this temperature range, PCMs are integrated into solar air heaters, solar domestic hot water systems for heating purposes and in electronic devices for cooling and operational performance enhancement. For this temperature application, fatty acids are often implemented due to their suitable melting temperatures and relatively long lifetimes with not so much economic offset.

Behenic acid, with a melting temperature of 65°C, has been suggested to have potential applications in the storage of industrial waste heat, cooling of electronics, solar residential heating, the cooling of lithium-ion batteries and in automotive applications [141].

6.4. High temperature range: 80–200 °C

Within the high temperature range, PCMs are integrated into solar absorption cooling to achieve higher coefficients of performance (COP) and into water heat recovery systems into solar power plants to improve energy usage efficiencies and to subsequently reduce environmental pollutants. Approximately 60% of the overall wasted industrial heat is below 230 °C [151], which means that SA with melting temperatures of 75–189 °C and melting enthalpies of 219–352 J/g can act as highly energy dense waste heat recovery PCM. Kaizawa et al. [152] also investigated erythritol as a suitable PCM to supply hot water at a temperature of 50 °C for a waste heat transportation system. Furthermore, Wang et al. [128] also used erythritol in the same application and studied the charging and discharging process of mobilized TES with direct and indirect contact storage containers. Here it was found that the direct-contact storage container was better than the indirect-contact storage container with higher charging and discharging speeds of the erythritol.

Within this temperature range, solar absorption chillers and heaters are also considered. Agyenim et al. [153] used 100 L of erythritol and stated that it could store enough thermal energy for approximately 4.4 h of peak cooling loads based on a COP of 0.7 for absorption chiller applications to be applied in an 85 m^3 office building in Cardiff during a hot summer day. Papadimitratos et al. [154] integrated erythritol with a melting temperature of 118 °C within the evacuated solar tubes in solar water heaters and it was found that the efficiency of the system was improved by up to 26% under normal operational conditions and increased to 66% for stagnation operation conditions compared to a standard solar water heater without PCM. For such high temperature applications, the main class of biobased PCMs to be considered are sugar alcohols and the eutectics thereof.

Overall, compared to non-biobased PCMs, very few studies have been employed on utilizing biobased PCMs in many common thermal energy storage applications despite the plethora of biobased PCMs available with excellent thermophysical properties. Table 3 summarizes the four temperature ranges, the relevant applications to each temperature range, the PCMs that have previously been reportedly used for each application and suggestions by the authors of biobased PCMs that can potentially be beneficial for use at each temperature range.

Table 3
List of PCM temperature ranges and potential applications.

Temperature range, °C	Applications	Biobased PCMs used before	Potential biobased PCMs to be used
−20–6	Transport of temperature-sensitive goods Commercial/domestic refrigeration Commercial/domestic freezers	–	Tripalmitolein, trihexadecenoin, tripetroselinin, triolein, trioctadecenoin, tricaprin, dimethylamine, 1,2,3-Trihydroxybenzol, Diethyl sopropylmalonate, octadecanedioic acid, dodecanedioic acid, eicosanedioic acid, dioctyl carbonate, dinonyl carbonate, odecyl carbonate, dioleyl carbonate
7–40	Passive heating/cooling in buildings Active heating/cooling in buildings Air conditioning Machine industry cooling Thermo-regulated textiles	Coconut oil, palm oil, capric acid/lauric acid, capric acid, PureTemp29, Crodatherm 24	Butyl stearate/butyl palmitate, capric acid/lauric acid, propyl palmitate, 1-tetradecanol, 1-dodecanol
41–80	Domestic hot water Cooling of electronic devices Cooling of solar air heaters	Behenic acid	1-hexadecanol, 1-octadecanol, palmitic acid, myristic acid
81–200	Waste heat recovery Solar absorption cooling	Erthyritol	Erythritol, sorbitol, mannitol, xylitol, lactitol, maltitol, erythritol, erythrol, isomalt, galactitol, D-sorbitol

7. Life cycle assessments of biobased PCMs

As the planet is reaching its ecological limit, an increasing number of sectors are moving away from the linear “take, make, dispose” model and investing in more sustainable circular solutions with the support of governmental policies [155]. As a result, materials which can be regenerated in the biological cycle are the focus of many scientific fields (e.g., polymers, fuels) including the PCM field where an increasing number of studies on biobased PCMs has emerged in the last decade. However, biobased materials are not necessarily sustainable, if not extracted or disposed of in a sustainable manner. Issues related to the use of possibly harmful or unsustainable materials used for the extraction or synthesis of PCMs, as well as the impact on ecosystems through their destruction or fertiliser use and food security calls for detailed investigations on the impact of biobased solutions in comparison to their conventional counterparts, often in the form of life cycle analyses (LCAs). A review on the investigation of biobased PCM-related LCAs discussing these issues is presented hereby.

While LCAs of PCMs in general are still scarce, some reviews have emerged in the last couple of years. Many of these studies consider paraffins and salt hydrates as main PCMs, however paraffins are side products of the oil and gas industry and thus cannot be considered biobased or sustainable. Therefore, paraffins and salt hydrates will only be included in this review for comparison purposes. Only one publication considering solely biobased PCM aspects (not connected to specific applications) could be identified. Effects of palm-oil based PCM production routes were modelled and evaluated by Fabiani et al. [156]. Environmental impacts of PCMs were calculated in 24 impact categories and palm-oil production from food industry waste was shown to be most environmentally friendly.

A variety of comprehensive LCAs have been implemented in the construction field in the past two decades, reflecting specifically on the environmental contribution of the used materials. As it varies from building to building, the material production stage can be the highest contribution to embodied environmental impacts, reaching up to 90% [157]. To date, LCAs focusing on biobased PCMs have been almost exclusively performed with a focus on PCMs incorporated in buildings or construction materials. A review from Kylili et al. [158] in 2016 compares the impact of paraffins, salt hydrates, and esters of stearic acid incorporated in alveolar bricks and concrete, rammed earth matrix, tiles, and ventilated double skin facades. The study concludes that both esters and salt hydrates significantly reduce the impact on the environment in the production stage by 10.5 and 9% respectively, and thus esters represent the lowest impact PCMs. Cabeza et al. [159] and Sandak et al. [160] also showed that the partial substitution or inclusion of biobased materials in construction material helps to significantly reduce the CO_2 emissions and carbon footprint of the building as a whole, without necessarily impacting performance and longevity. Still, a review by Nazari et al. [161] highlights the challenges related to the definition of the system boundaries, and how they can significantly affect the results of the LCA. It is therefore still very difficult to assess the real impact of the biobased PCMs, especially given the limited number of studies and research performed on the topic up to date. In 2019, Heidari et al. [162] investigated the suitability of streamlined LCA (SLCA) tools and methods on wood based PCM for construction. The authors found that both SLCA and full LCA agree on the fact that the heating energy and the PCM itself were the biggest source of impact. While SLCA is less accurate than a full LCA, it provides smaller data sets and delivers a quick screening of a product environmental performance. Thus, it should be routinely performed by companies and developers alike to assess the impact of products launched on the market. The Impact of myristic acid 51 PC M integrated in a collector of a solar collector was evaluated through LCA in terms of the whole thermal system [163]. The PCM itself was shown to be most toxic of all materials incorporated in the system under selected boundary conditions, however, using more detailed data on its environmental performance could improve the evaluation results. Nöel et al. [164] conducted an interesting LCA on two PCMs from biobased dodecanoic acid from palm oil and ethyl hexadecanoate from algae for usage in wallboards. In particular, the authors focused on the energy input and carbon emissions associated with the large-scale production and their usage in thermal energy storage systems. The feasibility of the materials was evaluated in terms of energy and greenhouse gas emissions for both solar thermal domestic hot water heaters and solar thermal wall panels. In conclusions, the authors found that using palm oil to produce fatty acid PCM is a feasible solution, with less than 2 years to payback time and would reduce the CO_2 emissions of a typical American house by up to 16 t over 10 years. On the other hand, the usage of ethyl hexadecanoate from algae requires much more energy, and thus increases the payback time to longer than the overall lifetime of the building. This is due to the energy-intensive dewatering step. However, algae-based PCMs are considered efficient CO_2 sequesters, and thus the improvement of the dewatering step could bring significant changes to this forecast. Alvaro de Gracia et al. [165] also conducted an LCA on the environmental impact of including PCMs (a paraffin RT-27 and a salt hydrate SP25, both from Rubitherm GmbH) in Mediterranean buildings (Spain as a model country). Due to the paraffins being derived from fossil fuels, the impact of salt hydrates' manufacturing is about 75% lower. Therefore, when considering paraffins included in cubicles there is no significant variation since the lower energy impact is balanced out by the higher manufacturing impact. However, with the inclusion of salt hydrates the global impact of the cubicle is significantly reduced. Still, the authors estimate that PCM present a more significant contribution on the reduction of environmental impact over long periods of time (>25 years) and in areas where the climate tends to be more similar all year round. Similarly, Menoufi et al. [166] also investigated the impact of esters of stearic acid (possibly methyl stearate, but not specified) on the same experimental cubicles in Spain. Here the authors found that the esters produce the same or even slightly better results as salt hydrates on the environmental impact of the cubicles. This clearly shows that switching to fully sustainable, biobased materials can have a considerable impact on the whole impact of the technology. A 2022 review on LCAs of PCMs utilized in buildings highlights once more the challenges related to the lack of industrial data on PCM production and methodological differences limiting accuracy of the reviewed LCAs [167]. The review mentions an unpublished thesis by Allred from 2014 [168], where dodecanoid acid from the kernels of oil palms is evaluated from the energy, environmental and cost efficiency perspectives when used in a solar thermal energy storage system. Low embodied energy of the PCM is outweighed by that of the storage tanks and the environmental is significantly higher than for an electric water heater. It is, however, noted that potential energy savings could compensate for the embodied energy in 2 years of product use.

Although no LCA studies considering use of enhanced biobased PCMs or biobased PCM composites as building materials could be found, such materials are expected to increase the negative environmental impact as a result of the need to combine two or more materials, often nanoscale materials or certain types of graphite. This contributes not only to the increase of the material production impact (e.g., 1 kg of graphite production equals 18,3 kg CO2-eq. emissions [169]), but also makes the recycling more challenging. One study, however, argues that if encapsulated PCM is recycled and transformed into shape-stabilized PCM, the greenhouse emissions impact at the PCM end-of-life stage becomes small [170]. As the PCM type is not specified, the publication considers PCM-based cooling systems for power plants. Baldassarri et al. [171] performed material-level LCA of 7 polyols and 7 fatty acids to select best candidates for application in cement based composite panels. Methyl ester and glycerine produced from waste cooking oil give better performance in terms of latent heat capacity when compared to paraffins. The environmental impacts related to PCM incorporation into panels were suggested to improve by boosting energy efficiency of PCM production.

In conclusion, while some LCAs on PCMs have been performed, such studies remain scarce and still mostly focused on PCM in buildings. Additionally, biobased PCMs have only recently started to be investigated in LCAs, with the majority still considering paraffins and salt hydrates as the main PCMs. Nevertheless, from the limited studies performed, it emerges that while paraffins are unsuitable PCMs due to the high environmental impacts, salt hydrates and esters do provide a positive contribution to the impact reduction of the building. In particular, from preliminary studies it emerges that esters might be the most advantageous PCMs in terms of low environmental impact. Some barriers still exist (e.g., high energy costs of dewatering for algae) and thus overcoming such challenges will be crucial to the implementation of esters in LHS systems. Several LCA studies also highlight the importance of sustainable production routes (mostly energy efficiency) and recycling possibilities in relation to improvement of environmental impact of the biobased PCMs. In order to claim more conclusive statements, further LCAs on different scenarios and applications should be performed in the future. It is safe to say, LCAs should be one of the major focuses of the scientific community in the upcoming years.

8. Challenges and knowledge gaps

As it is clear from the state of the art presented till now, some challenges and knowledge gaps related to PCMs, and biobased PCMs in particular persist. One of the biggest challenges is the lack of LCAs on biobased PCMs in applications besides the incorporation in buildings or construction materials. Here a sort of paradox arises, as this is mostly due to the high amount of data required by LCAs, whilst however, biobased PCMs are characterized by an intrinsic lack of correct environmental data, including thermal performance, long-term stability, production and extraction routes, costs and general commercial availability. Still, more studies and lower data-intensive tools (e.g., SLCA) are now arising, and will hopefully bridge the existing gap in the upcoming years. Table 4 summarizes some of the important properties and technical challenges associated with the reviewed pure biobased PCMs. PCM mixtures and multi-component materials are not included here.

Lack of comprehensive data about oleochemical carbonates can be identified as one of the existing knowledge gaps, specifically interesting in the context of defining some of those as high performance PCMs for low temperature applications [135]. As for the other major knowledge gaps, among the reviewed biobased PCMs, default assumption of their biodegradability (due to the biobased nature), lack of comprehensive LCAs mentioned in the previous section and chemical compatibility with metals and plastics can the outlined. While cycling stability of fatty acids has been widely reported and accompanied by concrete numbers of thermal cycles, similar information about other biobased PCMs is somewhat limited.

A vast amount of literature and research projects with focus on understanding consumer perspectives on biobased products and biobased materials in general do not cover any specific examples of the biobased PCMs. Little is known about associated social impacts which vary demographically and may include poor working conditions in local environments and/or developing countries where PCMs are produced.

In addition to what is presented in Table 4, more technical challenges on the performance of PCMs remain, many associated with the intrinsic kinetics of these materials. Sugar alcohols for example, are characterized by high degrees of supercooling and very slow crystallization rates, which translates into slow power delivery in the LHS system. Most of the studies published in the past years deal with the supercooling aspect through the addition of nucleating agents or via mechanical induction, but no significant progress has been made on the slow rates of crystallization [13,172,173]. While being promising candidates, esters are also still characterized by several technical challenges, with polymorphism perhaps being the most prominent and least understood one. The formation of different crystalline forms, each with its own melting point and enthalpy of fusion, is detrimental to any LHS setup, not to mention difficulty to predict and contain. Although the polymorphism of esters has been known since the 1940s, effective methods to fine-tune it to reproducibly form the desired crystalline form directly from the melt over consecutive cycles have not been developed yet. Most methods developed crystallize the target form from the solution via seeding and templating [174–178]. The role and influence of thermal history on polymorphic transformations also remains poorly understood.

The common shortcoming of the majority of biobased PCMs is low thermal conductivity, which limits the application of biobased PCM storages in space heating and cooling application as they require a significant temperature difference between heat transfer fluid (HTF) and PCM[179]. On the other hand, incorporation of a PCM storage tank into a typical household could require quite a lot of space. The storage volume could be reduced by either increasing PCM conductivity or by using less conventional storage concepts, such as direct contact PCM storage, where the HTF of higher conductivity (e.g., water) bubble though a PCM [130].

Table 4
Important properties and technical challenges associated with biobased PCMs.

Property	Fatty acids	Esters	Alcohols
Thermal conductivity, W/mK	<0.2	<0.2	<1
Stability under cycling, number of cycles	up to 1000	over 1000	nonstable
Supercooling	little to no	little to no	yes
Polymorphism	yes	yes	yes
Volume changes during phase transitions	small	small	small
Volatility	some	yes	no
Density	<1 g/ml	<1 g/ml to >1 g/ml (aromatic esters)	>1 g/ml
Flammability	low	Moderate (short chained esters)	low
Toxicity	skin irritant	nontoxic	skin irritant

9. Conclusions

The main biobased materials investigated for their potential in energy storage applications have been summarized in this comprehensive review. Different classifications of biobased phase change materials, their respective thermophysical properties, underlining the advantages and disadvantages of each type have been presented. While some extensive research has been performed on certain material categories, such as fatty acids, fatty acid esters, alcohols and mixtures, significant fragmentation of the findings and variable material performance in applications with different system boundaries show the need for better systematization from both material characterisation and technology evaluation points of view. Overall, biobased PCMs represent a range of excellent properties compared to their conventional non-renewable alternatives and can be game-changing technologies shaping the future of our energy consumption. Production routes, improved durability and recyclability of those materials will play a key role in their implementation in maximum sustainability future energy storage technologies. As these materials can be successfully combined with other materials to improve their performance or contained through either micro- or macroencapsulation to benefit specific applications, potential environmental impacts and recycling routes of such material combinations must be thought through already at the developmental stages. Phase change dispersions, where phases are combined to obtain a pumpable fluid, on the other hand, can easily be separated into initial phases at the end of the lifetime. This allows for more efficient and environmentally friendly recycling routes.

Transformation and contribution to sustainability is already affecting market value of companies as their consumers are demanding to see the reduction of environmental impacts caused by industries around the world. Despite the lack of literature specific to public perception and barriers preventing greater use of biobased PCMs, these technologies currently have a better chance of winning social acceptance than conventional alternatives based on petroleum or non-renewable materials. Even if a new battery lasts for more than 20 years, durable biobased PCM storage might just be a more sustainable and safer product to save your energy.

Incorporation of biobased PCMs is an important topic in the thermal energy storage and within thermal management technologies. Some highlighted topics and knowledge gaps currently associated with biobased PCMs are.

- There is strong need for a comprehensive database governing existing biobased PCMs, their properties and associated environmental data.
- Reviewed LCAs lack clear conclusions about impacts of biobased PCM usage due to a lack of environmental data specific to each PCM as well comparisons with conventional energy storage technologies and often no service lifetime of the specific technologies.
- More research on biodegradability and compatibility of biobased PCMs with other components of TES systems is needed.
- Material functionalization needs to be systemized and the environmental impact of enhancement materials needs to be reflected together with benefits of the improved material performance.
- There is a lack of publications addressing and characterising biobased PCMs after extensive testing in operational environments (end of life characterisation).

- Biobased PCMs are excellent candidates for a variety of applications at the temperature ranges between −20 and 200 °C.

As for the innovative and potential future applications, biobased PCMs are excellent candidates to be used in thermoregulated fabrics, a technology commonly found on the market. An interesting future application already supported by ongoing NASA-funded research is use of PCMs in next-generation spacesuits. PCM and PCM dispersion-based concepts are also credited for their potential to be used in future exploration spacecraft and satellite thermal control systems. Even though biobased PCMs are not specifically mentioned in this context, their relevance could be uplifted in the view of resource availability in space. After all, plant cultivation in space exploration is another major research area in the respective scientific communities. A specific benefit of biobased PCMs can be mentioned in regard to medical applications, food and beverage storage, plant cultivation and associated transport technologies due to the importance of using non-toxic and potentially food safe or biocompatible materials. Multi-component substances are attractive as biobased PCMs due to their low costs and sustainable nature, but as can be observed, do not present superior thermophysical properties compared to pure PCMs or eutectic mixtures. Nevertheless, they might be valuable candidates for low power applications that do not require a precise melting temperature, but rather a broader range. Introducing programmability integrated on the material level to activate and change properties, such as phase transitions within the biobased PCMs, could also be an important research topic within TES community as it could open up for the new high-tech application areas.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Review article.

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