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Review

Review of Bioinspired Composites for Thermal Energy Storage: Preparation, Microstructures and Properties

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Abstract: Bioinspired composites for thermal energy storage have gained much attention all over the world. Bioinspired structures have several advantages as the skeleton for preparing thermal energy storage materials, including preventing leakage and improving thermal conductivity. Phase change materials (PCMs) play an important role in the development of energy storage materials because of their stable chemical/thermal properties and high latent heat storage capacity. However, their applications have been compromised, owing to low thermal conductivity and leakage. The plant-derived scaffolds (i.e., wood-derived SiC/Carbon) in the composites can not only provide higher thermal conductivity but also prevent leakage. In this paper, we review recent progress in the preparation, microstructures, properties and applications of bioinspired composites for thermal energy storage. Two methods are generally used for producing bioinspired composites, including the direct introduction of biomass-derived templates and the imitation of biological structures templates. Some of the key technologies for introducing PCMs into templates involves melting, vacuum impregnation, physical mixing, etc. Continuous and orderly channels inside the skeleton can improve the overall thermal conductivity, and the thermal conductivity of composites with biomass-derived, porous, silicon carbide skeleton can reach as high as 116 W/m*K. In addition, the tightly aligned microporous structure can cover the PCM well, resulting in good leakage resistance after up to 2500 hot and cold cycles. Currently, bioinspired composites for thermal energy storage hold the greatest promise for large-scale applications in the fields of building energy conservation and solar energy conversion/storage. This review provides guidance on the preparation methods, performance improvements and applications for the future research strategies of bioinspired composites for thermal energy storage.

Keywords: thermal energy storage; bioinspired composites; phase change materials (PCMs); preparation process; heat storage properties

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1. Introduction

The rapid development of the international economy and the continuous growth of energy demand has made the relationship between energy supply and demand lose balance.

Energy sources such as coal and oil are used as fuels, resulting in low efficiency. Energy sources such as solar and wind energy are unstable and difficult to store with existing technologies. In order to alleviate the energy crisis and achieve green development, research on environmentally friendly and clean new energy technologies is important. Thermal energy storage technology can improve energy efficiency and solve the contradiction of energy supply and demand mismatch in time and space. Thermal energy storage materials are the key to the practical application of this technology, and the exploration of new materials is the focus of current thermal energy storage technology research [1,2]. PCMs have been applied in different fields such as thermal energy storage utilization [3], battery thermal management [4,5], solar-thermal conversion [6] and building energy saving [7]. Two main problems of PCMs exist, including the incapability of maintaining a stable shape resulting in leakage [8] and a low thermal conductivity resulting in the low rate of heat filling and releasing [9]. This causes them to be unable to play their due roles in the field of waste heat recovery. Many researchers have been working on the preparation of shape-stabilized phase change composites to solve these problems by introducing a second material with high thermal conductivity, such as porous aerogels [10–13], expanded graphite [14,15] and porous silicon carbide scaffolds [16]. Although these composites have high photothermal transfer properties and improved thermal conductivity, the high price of raw materials and the complexity of some preparation processes are not conducive to large-scale applications. Compared with single organic materials (e.g., polyethylene glycol, paraffin [17], and stearic acid), inorganic materials [18], eutectic mixtures [19] and encapsulated PCMs [20], shape-stabilized composites based on bioinspired structures [21] are cheaper and easier to obtain. The diversity of natural materials can help improve the thermal conductivity and leakage resistance of composites in terms of energy storage.

Therefore, researchers study the excellent structure and characteristics of natural organisms, and apply their structural properties, energy transfer and information-conversion processes to the study of the energy storage field [22]. Bioinspired composites for thermal energy storage mimic the properties of living organisms, draw inspiration from natural materials, and study their functional principles to design and synthesize materials with the structural characteristics of biological materials. The application of natural structures is of great significance to promote research and progress in the field of energy storage. The skeleton structure of bioinspired composites for thermal energy storage is generally prepared by direct reference to organisms or simulated natural/biological structures [23]. Several methods have been used to introduce PCMs into the skeleton, including melt penetration (e.g., high-density-polyethylene and pentaerythritol as PCMs, at 150 °C for melting) [24], physical hybridization (e.g., expanded perlite/paraffin and graphite mixes at a 40:10:50 mass ratio, vacuum degree at −0.1 MPa) [25], and vacuum impregnation (e.g., hexadecanol as PCM into high porosity fly ash pebbles, at 100 °C for 10 h) [26]. In recent years, many scholars have conducted a lot of research on the preparation process, performance improvements, and applications of bioinspired composites for thermal energy storage.

Cheng et al. prepared a new nanocomposite material coated with sulfur from the carbon matrix of a mimosa by chemically modifying the mimosa clitoris and then coating it on the carbon of the mimosa [27]. He et al. used carbonization and freeze-dried (vacuum drying) water hyacinth as a biochar carrier and loaded PCM to make bioinspired composites for thermal energy storage. Compared with the PCMs, this composite can increase the thermal conductivity by 56.93%, and still maintain a phase change peak after 500 cycles, which exhibits good thermal stability [28]. Tang et al. used attapulgite (layer-chain porous structure) as a template, wrapped with glucose, carbonized at high temperature (800–1000 °C) to make a porous skeleton, and then used vacuum impregnation to penetrate stearic acid into the skeleton to make Stearic acid/Nano-porous carbon (SA/NPC) composite. The composites have more carbon defect sites, so their photothermal conversion efficiency is as high as 94.5% [29]. Sun et al. used gelatin as raw material for freeze-drying and carbonization to make porous aerogels, and then vacuum-impregnated epoxy to make the Eicosane/Fe-doped carbon aerogel composite PCM. There was no leakage after 1000 thermal cycles of testing, indicating that the composite materials exhibited excellent thermal reliability, and their photothermal conversion efficiency could reach up to 93.32% [30]. Wang et al. prepared mixed-clay sponges by solution polymerization, freeze-drying and carbonization to produce a loose skeleton, and then melt-impregnated hexadecylamine to make a shape-stable hexadecylamine/mixed clay sponge (HDA/MCS) composite, whose photothermal conversion efficiency can be as high as 75.6% [31]. Wen et al. carbonized sunflower straw as a support skeleton, and then used vacuum impregnation to impregnate stearic acid into a porous scaffold to prepare a stearic acid/carbonized sunflower straw (SA/CSS) composite. The thermal conductivity of the composite is 0.33 W/m*K, which is 106.3% higher than SA [32]. They also carbonized maize straw to form a porous skeleton, and then used vacuum impregnation to produce a stearic acid/carbonized maize straw (SA/CMS) composite. The thermal conductivity of this composite reaches 0.30 W/m*K, and the SA/CMS exhibits excellent thermal reliability after 200 thermal cycles without leakage [33]. Sarı et al. carbonized industrial waste sugar beet pulp, and then used vacuum impregnation to impregnate the capric-stearic acid eutectic mixture to prepare capric-stearic acid eutectic mixture/carbonized sugar beet pulp (CSEM/CSBP) composites. The combination of CSEM and CSBP can lead to a 79% increase in thermal conductivity, and the latent heat of the composite was reduced only less than 3% after 1000 thermal cycles [34]. Through the above studies, it is found that the biochar obtained by modifying natural materials has a larger specific surface area and better leakage prevention ability. Bioinspired composites for thermal energy storage can refine and modify PCMs from multiple angles, which greatly expands their application prospects in the field of energy storage. In this paper, the development of bioinspired composites for thermal energy storage is investigated, including the preparation, microstructures, and properties of composites, as well as prospects for the application of bioinspired composites in thermal energy storage. Compared with other reviews on phase change heat storage materials, we have introduced biological structures. This effectively improves the thermal conductivity of the materials and effectively solves the problem that phase change materials are prone to leakage [35,36].

2. Bioinspired Structures for Thermal Energy Storage

Natural materials are rich in resources and have diverse microstructures. There are many kinds of natural material structures, such as three-dimensional porous structures of bone, through-hole structures of wood [37–39], cell-like structures of pomegranate seeds, and layered structures of shells, etc. [40,41]. Bioinspired composites for thermal energy storage have been mainly developed by using bioinspired structures as the supporting skeleton, and then using some fusion methods to integrate PCMs into the skeleton. Since PCMs are used, the principle of the thermal energy storage is latent heat storage. The changes of substances during the phase change process are utilized to absorb or release thermal energy to achieve thermal energy storage. The inherent low thermal conductivity of PCMs affects fast energy transfer and efficient energy storage [42]. The three-dimensional connectivity structure can prevent leakage, and the tubular structure improves thermal conduction; these structures can compensate for the defects of the PCM. To better carry out heat conversion and thermal energy storage, natural porous structures are used as skeletons for producing thermal energy storage composites. Natural porous structures usually have the advantages of low density, high specific strength, high specific surface area, heat insulation and good permeability [43]. Our research group has conducted extensive research into the preparation of biomorphic wood-derived porous carbide materials (e.g., silicon carbide, zirconium carbide and titanium carbide) (Figure 1). The majority of the research lies in the preparation of biomorphic silicon carbide materials because introducing a Si source is more versatile and easier to prepare than introducing other metal sources (e.g., Ti, Zr, Nb, Ta) into biomass. To date, silicon carbide has been the most widely investigated material and has been actively explored for applications in a variety of fields.

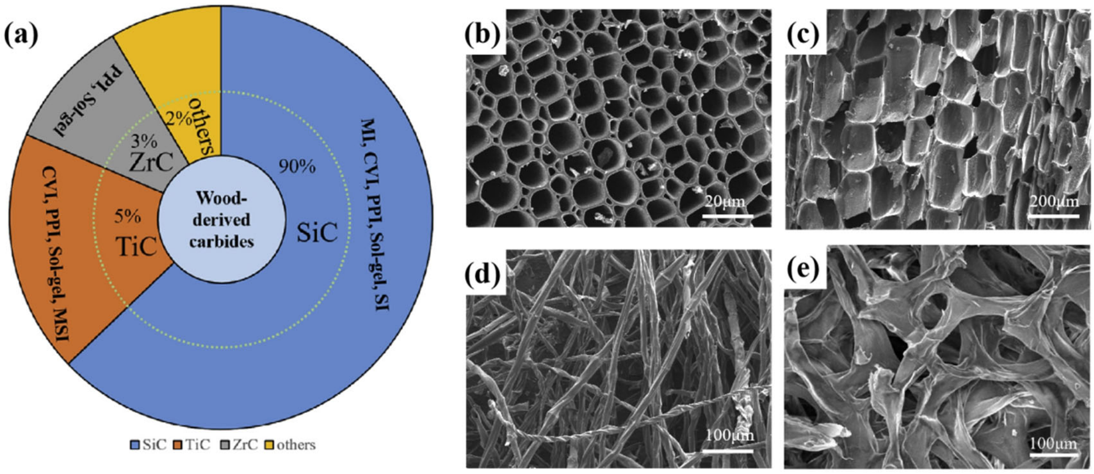

Figure 1. Summary of wood-derived carbides (**a**) and SEM images of microstructure of natural materials: (**b**) quercus laevis, (**c**) sugarcane, (**d**) sycamore fruit fiber and (**e**) grapefruit peel (adapted from Refs. [37,44]).

The wood-derived porous carbon is extremely attractive [44,45], the various applications reported for natural porous structures [46,47]. Since it is a sustainable and renewable carbon-rich material with high thermal conductivity [48], PCMs can be preserved in its pores [49], resulting in improving the thermal conductivity of composites [50]. So far, biomass-derived carbon materials have shown their attractive prospects in many areas such as energy conversion and storage. At present, there are two main types of PCMs used in bioinspired composites for thermal energy storage, including organic PCMs and inorganic PCMs. For PCMs embedded in the skeleton, organic PCMs have the advantages of preventing subcooling, phase separation and corrosion, reliable security and good cycle stability, and are mostly used for low-medium temperature phase change heat storage [51,52]. Inorganic materials have the characteristics of high latent heat, relatively high thermal conductivity, non-toxicity, non-flammability and low cost. But inorganic materials have problems such as leakage and corrosiveness. Compared with other materials (i.e., fatty acids, polymers and alloys), molten salt has a low cost, high operating temperature, good safety, large latent heat, high energy storage density and good thermal stability [53]. Today, most researchers choose organic materials, such as paraffin or stearic acid, as PCMs [54].

The methods of introducing bioinspired templates for thermal energy storage are mostly divided into two categories: one is to replicate natural structures (i.e., seashell, bone) using ceramic clay to produce a template, and the other is to directly using natural structures as a skeleton, as shown in Figure 2. The peel of pomegranate seeds is completely wrapped around the flesh, and mimicking this characteristic is beneficial to make a leak-proof skeleton material. The porous structure of wood exists in the form of conduits and fiber tubes, forming mesopores and micropores with different diameters and uniform pore distribution [55]. This structure can transmit water and nutrients quickly, and the wood can be modified by using this property to obtain skeleton materials with high thermal conductivity.

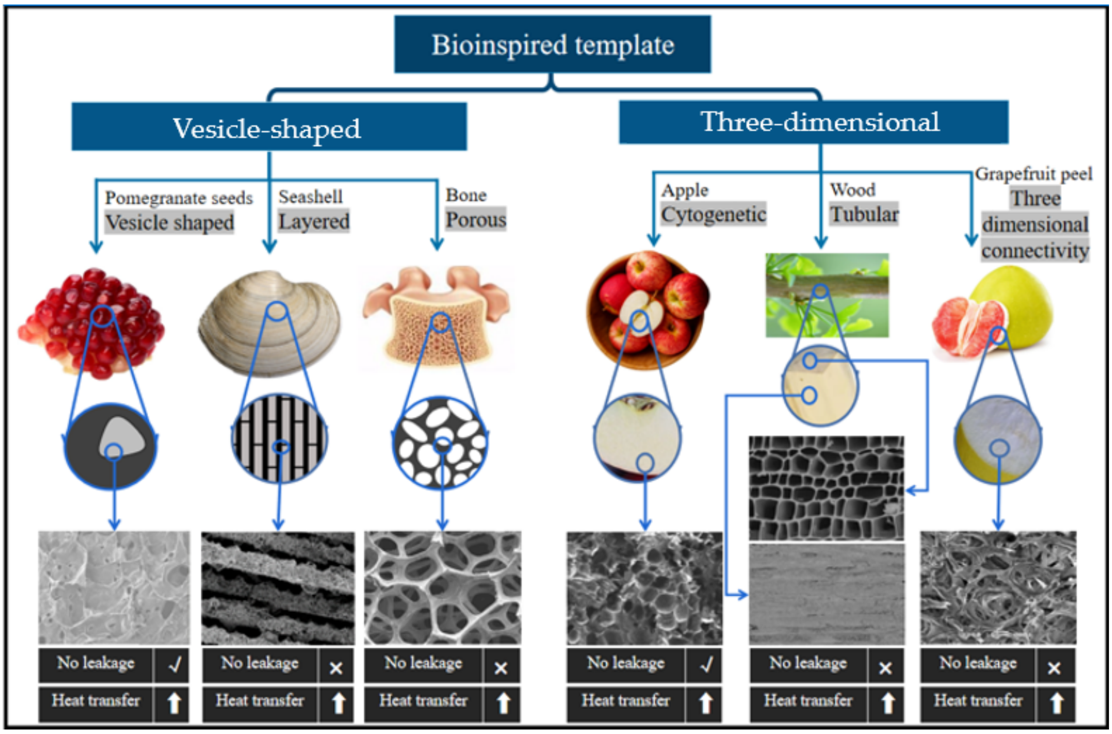

Figure 2. Bioinspired templates and microstructures for thermal energy storage composites.

3. Preparation Process of Bioinspired Composites for Thermal Energy Storage

Pure PCMs have their own limitations in applications, owing to leakage and low thermal conductivity. In order to solve these problems, PCMs and bioinspired skeletons can be compounded together to obtain bioinspired composites for thermal energy storage with excellent performance. The preparation process of bioinspired composites for thermal energy storage involves two main steps, including the preparation of skeleton materials and the preparation of composites for thermal energy storage.

The preparation process of skeletons mainly consists of three steps including modification, impregnation and sintering. Natural porous structures such as cell foramen, three dimensions interconnected pores or tubular pores are selected as raw materials. Chemical modification and thermal modification have been used to remove macromolecular compounds such as cellulose, hemicellulose and lignin from natural plants to improve impregnation efficiency. Different impregnation methods were used, including melt impregnation, polymer precursor impregnation, sol gel impregnation, slurry impregnation and physical vapor phase impregnation. Melt impregnation was more widely used, and physical vapor phase impregnation was still in the research stage due to higher process requirements, which makes it difficult to carry out mass production. Finally, the heat treatment at high temperatures (600–1800 °C) was carried out to obtain the porous skeleton.

The bioinspired composites for thermal energy storage were prepared by combining the prepared porous skeleton with PCMs by melt impregnation, physical hybridization, vacuum impregnation and other filling methods, as shown in Figure 3.

Our research group investigated a variety of infiltration technologies for preparing bioinspired skeletons (as shown in Figure 4), including polymer precursor impregnation, sol-gel impregnation, slurry impregnation and physical vapor-phase impregnation.

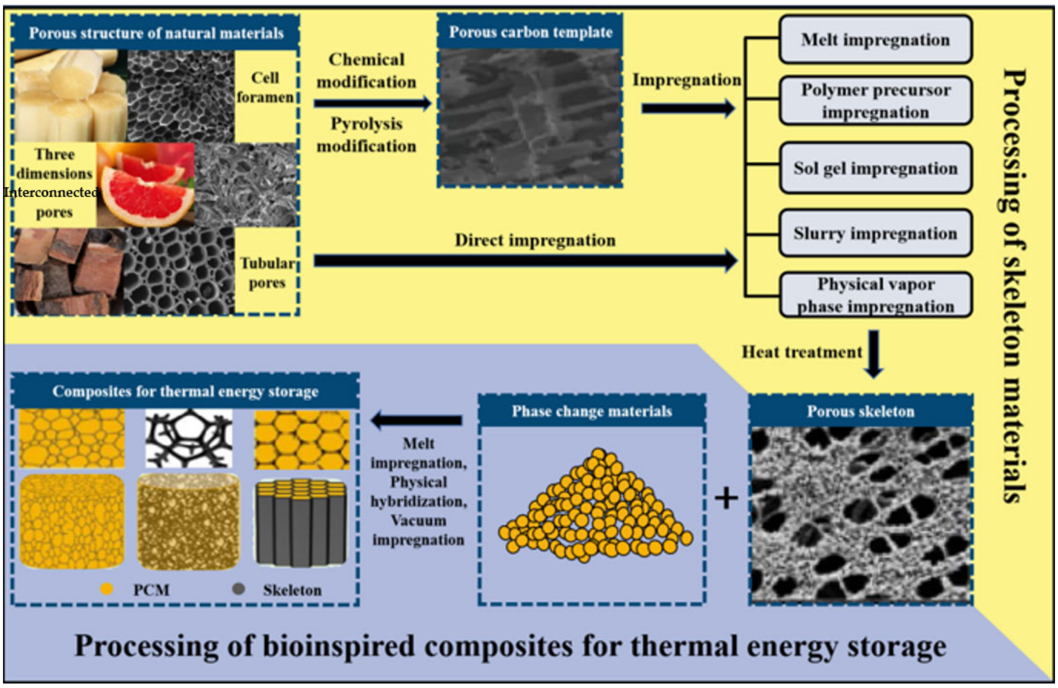

Figure 3. Simplified flow chart of processing bioinspired composites for thermal energy storage.

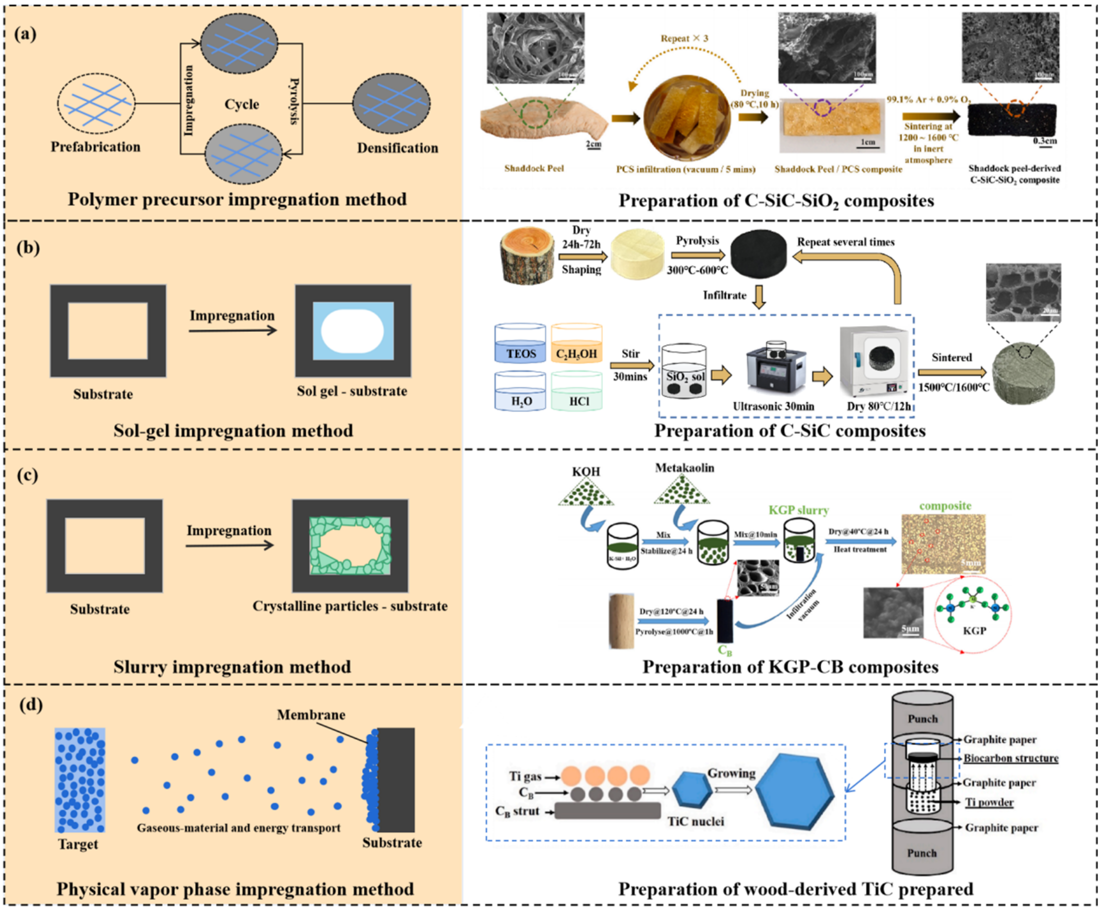

Figure 4. Summary of the impregnation methods for preparing bioinsipired ceramics/ceramic matrix composites in our research group: (**a**) polymer precursor impregnation, (**b**) sol-gel impregnation method, (**c**) slurry impregnation method, (**d**) physical vapor phase impregnation method (flowchart of material preparation inserted in Figure 4 adapted from references [41,44,45]).

3.1. Processing of Skeleton Materials

Skeleton templates are obtained in two ways (as shown in Figure 5), including mimicking biological structures and directly using natural templates. Directly using natural structures as templates for preparing thermal energy storage composites has been more widely used compared with mimicking biological structures. This might result from it being a simpler process, more homogeneous and having diverse microstructures with direct reference to the natural templates method. Considering the lightweight and large specific surface areas, porous materials with a porosity range from 60 to 90%, were chosen as the skeletons for preparing bioinspired thermal energy storage composites.

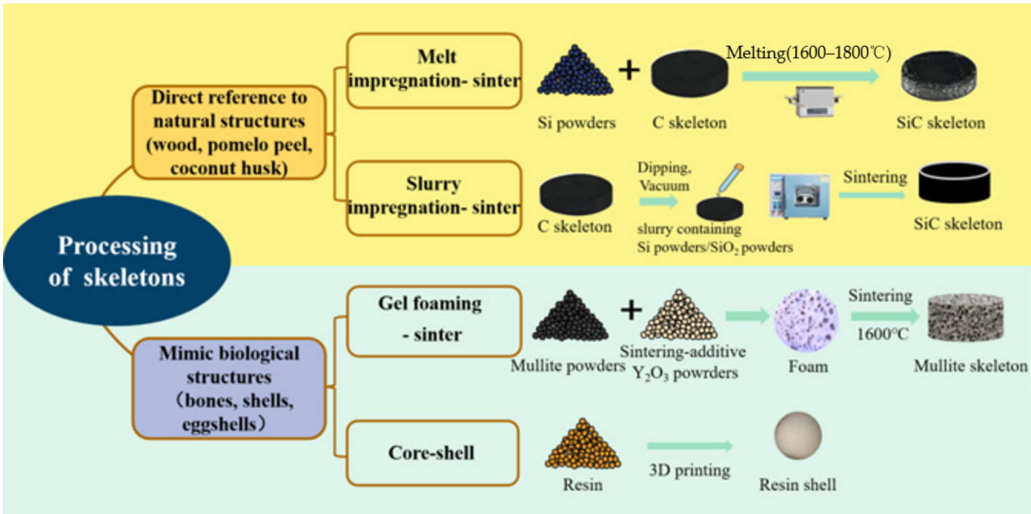

Figure 5. Processing of skeleton materials for bioinspired thermal energy storage composites.

For skeletons directly referred to natural structures, SiC materials are the most widely used for bioinspired thermal energy storage composites. This might result from their higher thermal conductivity (60–150 W/m*K), compared with C skeletons, and excellent chemical stability and corrosion resistance of SiC compared with Cu skeletons.

Based on the introduction of the silicon source, several methods have been used for processing SiC skeletons, including the melt impregnation-sintering method, and slurry impregnation-sintering method [56]. The SiC skeletons prepared by the melt impregnation method have been used as skeletons for preparing bioinspired thermal energy storage composites [57,58]. But those skeletons prepared by other methods (i.e., sol gel impregnation and physical vapor phase impregnation) have not been used for preparing bioinspired thermal energy storage composites. This might result from impurities in the skeletons, high cost and complicated preparation process of these methods.

The melt impregnation-sinter method is to immerse wood-derived C skeletons inside Si powders and subsequently heat them up to 1600–1800 °C, to prepare a bioinspired SiC skeleton. The slurry impregnation-sinter method is to disperse Si powders/SiO_2 powders in an appropriate amount of solvents (i.e., ethanol, water) to make a suspension. Then the carbon template is fully immersed in the slurry, and sintered in a horizontal tube furnace with an inert atmosphere to obtain a porous skeleton. Liu et al. immersed wood-derived carbon templates in the melt silicon powders at a high temperature (1600 °C) to induce the chemical reaction between C and Si to eventually form a porous SiC skeleton. The excess silicon can be physically removed by the escape of silicon vapor in a vacuum environment at 1800 °C to obtain a pure SiC skeleton [59,60]. Because the pores of wood are more uniform and micron-sized, the contact area is large and the silicon powders can easily combine with the carbon template to produce SiC. Xuan et al. introduced low-gluten batter into the pores of loofah sponges for carbonization (900 °C) to obtain porous carbon, and then there was a melt-silicon reaction and removal of excess silicon to obtain a porous SiC skeleton [57]. Because of the fine fibers and large pore size of loofah sponges, which are fragile after carbonization, low gluten batter was introduced to reduce the pore size and increase the structural strength. Nowadays, the melt impregnation–sintering method has been more widely used to prepare a uniform skeleton with a variety of structures, but it has a strict requirement for preparation conditions.

For skeletons from mimic biological structures, SiO_2 ceramics can provide high strength; resin materials can provide better sealing and AlN or SiC materials can provide better thermal conductivity.

Several methods have been used for mimicking natural structures, including the gel foaming-sinter method, and core-shell method [61]. The gel foaming-sinter method is to mix a slurry (i.e., containing sintering dispersant, Mullite powder and foaming agent) to form a foam by mechanical stirring and sinter at high temperatures (1600 °C) to obtain a mullite porous skeleton [62]. Qiu et al. mixed a slurry (containing sintering dispersant, AlN powders and foaming agent) with compressed air to form a foam by mechanical stirring and subsequently poured the foamed slurry into the mold and sinter at a high temperature (1950 °C) to obtain a porous AlN skeleton [63]. For porous skeletons of specific shapes (i.e., lamellar chains), they can be obtained by vacuum freeze-drying. Liu et al. added small amounts of SCMC, Y_2O_3, Al_2O_3 and deionized water to SiC powders to make a slurry, which was later ball-milled. It was degassed in a vacuum environment and then poured into molds for vacuum freezing (−40 °C) and drying. Finally, SCMC was removed by sinter at high temperature (1950 °C) to obtain L-SiC skeletons [64]. The core-shell method involves powdering the skeleton material into clay, placing it into a mold to set it and sintering to obtain a shell skeleton. When mimicking biological structures, the resin material can be used to directly fabricate the oval shell material through 3D printing [65]. Sun et al. mixed the ceramic powder with deionized water to grind the clay, which was then heated and dried to remove the water from the clay. The clay was placed in a mold and mechanical force was applied to prepare a hollow shell. Finally, the ceramic shell skeleton was obtained by cold sintering (200 °C) [66]. The pore size and uniformity of the gel foaming-sinter method are difficult to control, and the type of structure prepared by the core-shell method is relatively homogeneous, and some of the preparation methods are costly.

3.2. Processing of Bioinspired Composites for Thermal Energy Storage

Processing bioinspired composites for thermal energy storage involves impregnation of the phase change materials (PCMs) into wood-derived scaffolds. Several impregnation methods have been investigated including the vacuum impregnation method, physical hybridization method, and melt impregnation method [67] (as shown in Figure 6). The vacuum impregnation method has been most widely used. Using this impregnation, gases and water adsorbed in the pores of the material can be removed under vacuum conditions, leading to improved impregnation efficiency. The basic principle of the vacuum impregnation method has two aspects: one is the capillary pressure due to surface tension when the interface is in contact, and the other is the adsorption of components on the surface of the carrier. The melt impregnation method generally involves melting the PCM into the skeleton at high temperatures (>600 °C). For microporous structures, the vacuum impregnation method can lead to a more uniform distribution of PCM and easy access to the inside of the pores. Liu et al. used the vacuum impregnation method to impregnate melted paraffin wax into a porous bamboo-SiC (BSiC) skeleton, resulting in the porous BSiC skeleton tightly bonded to paraffin wax (96% filling rate) without any obvious gaps [57]. Porous carbon is a simple template structure into which PCM can be loaded by vacuum impregnation [68]. Li et al. used a vacuum impregnation method (under vacuum at

<5 Pa) to impregnate melted PEG (90 °C) into a three-dimensionally linked carbonized teak skin skeleton, which had strong anti-leakage properties and no significant mass loss after 200 cycles [69]. Vacuum impregnation achieves a high filling rate when the skeleton material is microporous. In addition, there is a vacuum adsorption method in which the PCM is covered with a porous material surface in a solid state, and then sequentially vacuum-insulated and pressure-retained. However, the preparation effect of this method is poor, and there are fewer application studies.

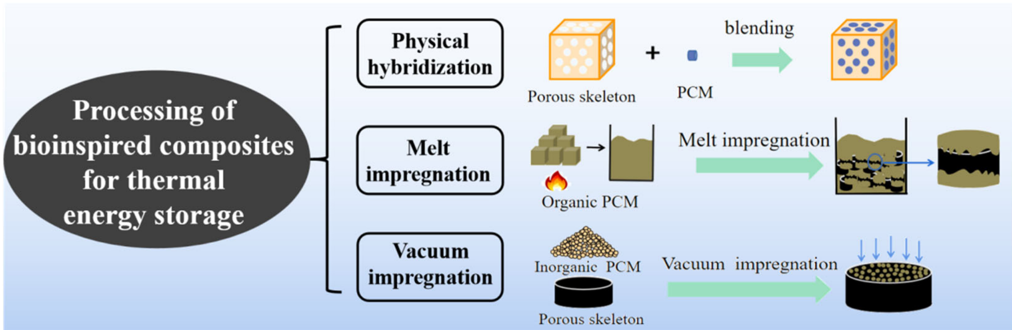

Figure 6. Processing of bioinspired composites for thermal energy storage.

Moreover, PCMs can be impregnated into templates by the physical hybridization method which involves a two-step aqueous solution or mechanical mixing. The two-step aqueous solution first oscillates by using ultrasound to make the skeleton material evenly dispersed in the PCM and then it heats and evaporates the obtained composites. Shi et al. used a two-step method by first mixing nanoparticles in titanium (IV) solution with stirring, transferring them into an autoclave (48 h at 180 °C) and then stirring them vigorously with liquid paraffin (5 wt.%) to obtain hierarchical mimetic porous phase change materials [70]. The advantage of this method is that the composite construction is homogeneous, but the high temperature stability is poor. Mechanical mixing comprises first mixing the skeleton material with the PCM by mechanical force, and then performing high temperature sintering or cooling curing. Sun et al. added SiO_2 nanoparticles within the molten salt, mechanically stirred well and then cooled to room temperature to obtain the composite material. The composites prepared by this method can be used stably for a long time at high temperatures.

4. Properties of Bioinspired Composites for Thermal Energy Storage

The price of raw materials, preparation process and properties (i.e., heat storage density, thermal conductivity, leakage prevention and cycle times) are key factors which affect the large-scale industrial applications of bioinspired composites for thermal energy storage. In recent years, research on this material has focused on improving thermal conductivity and enhancing leakage resistance in order to obtain excellent overall properties.

The heat storage density was mainly affected by the PCMs, and the NaCl-NaF/loofah-derived porous SiC composites had the highest heat storage density up to 424 kJ/kg. The thermal conductivity was mainly affected by the skeleton, and the NaCl-KCl/wood-derived porous SiC composites had the highest thermal conductivity up to 116 W/m*K. Leakage prevention is related to the skeleton structure, with cellular pores and three-dimensional interconnecting pores being able to wrap the PCMs to reduce leakage in the elliptical capsules as the skeleton can wrap the PCMs to prevent leaking, and methyl palmitate/walnut shell-derived carbon composites will not leak after 2500 cycles. The composition and structure, preparation process, performance statistics and application prospects of bioinspired composites for thermal energy storage over the last five years are summarized in Table 1.

Table 1. The detailed information of preparation process, performance and application of bioinspired composites for thermal energy storage.

First Author, Publication Year	Materials and Structures (Porosity, Pore Size)	Preparation Process	Performance				Application Temperature (°C)	Application System	Geometric Dimensions (mm)
			Heat Storage Density (kJ/kg):	Thermal Conductivity (W/m*K):	Leakage:	Number of Cycles:			
Dong, Yan 2022 [65]	n-octadecane Oval-shaped capsules	3D printing	243.5	0.1505	No	/	/	solar thermal chemical reactions	Oval a = 50 b = 40 c = 30
Shi, Lei 2020 [70]	paraffin@magneticTiO, paraffin@magneticFe3O4	two-step method	353.2 (PMF) 377.6 (PMT)	>0.55 (PMF) >0.18 (PMT)	/	/	88.6 (PMF) 94.2 (PMT)	solar direct absorption collectors	Globosity D = (1~2) × 10^{-4}
Feng, Guangpeng 2022 [58]	Li2CO3–K2CO3, porous aluminum nitride biomorphic porous (55%)	Gel foaming method	342	13.6	No	/	/	/	Disc-shaped D = 13 H = 5
Li, Shaowei 2022 [70]	polyethylene glycol (PEG), Carbonized grapefruit peel [100 μm(pipeline), 3.91 nm(Mesoporous)]	Vacuum impregnation	162.4	/	No	>200	40.4~ 61.3	solar thermal conversion and thermoelectric conversion	Cuboid
Tan, Yunzhi 2020 [71]	polyethylene glycol (PEG) spherulite crystals, Crosslinked polymer (CPA)	In situ polymerization	188.8	/	No	>100	60.2	Energy-saving and insulation of buildings and waste heat utilization of factories	Vesicle-like
Qiu, Lin 2021 [63]	polyethylene glycol (PEG), aluminium nitride (AlN) ceramic (<500 μm)	Gel foaming method	88.73	17.16	/	/	54.75	solar power stations, industrial waste heat recovery	Disc-shaped D = 12
Zhang, Hongyun 2021 [72]	Paraffin, copper foam, carbon material (graphene oxide and reduced graphene oxide) (95%, 100~300 μm)	Vacuum impregnation and physical blending	111.53	1.04	No	/	57.96~ 59.5	Solar energy absorption and storage	Disc-shaped D = 12
Wang, Jie 2019 [73]	oxalic acid dihydrate/glycolic acid, hydrothermal carbon	Physical blending	318.8	1.3867	No	>101	72	low temperature architecturalthermal applications	/
Xu, Qiao 2022 [57]	NaCl-NaF, porous SiC ceramics (64–87%)	Molten silicon, Melting impregnation	424	20.7	No	>1000	≈700	harvesting solar thermal energy	Disc-shaped
Xu, Q. 2021 [60]	NaCl-KCl molten salts, wood-like biomorphic porous SiC skeleton	Melting impregnation	157	116	/	/	/	harvest solar and thermal energy simultaneously	Disc-shaped D = 12.7 H = 3
Liu, Xianglei 2022 [59]	LiOH–LiF, Porous Bamboo SiC ceramics (66~77%)	Vacuum impregnation	309	35.0	No	>2500	435	High performance solar thermal conversion and storage	Disc-sedhap D = 18

Table 1. *Cont.*

First Author, Publication Year	Materials and Structures (Porosity, Pore Size)	Preparation Process	Performance				Application Temperature (°C)	Application System	Geometric Dimensions (mm)
			Heat Storage Density (kJ/kg):	Thermal Conductivity (W/m*K):	Leakage:	Number of Cycles:			
Liu, Xianglei 2021 [64]	Erythritol-TiN composite powder, L-SiC ceramic skeletons	Vacuum impregnation	157.93	25.63	No	/	≈120	Fast and efficient solar energy harvesting and thermal energy storage	Layered
Zhu, C 2022 [74]	Paraffin, 3D porous carbon scaffolds consisted of SiC-wrapped biomass carbon fibers	Vacuum impregnation	186	0.61	No	100	27.1~ 72.3	storage systems and advanced thermal management	Disc-shaped D = 20 H = 10
Xu, Qiao 2023 [75]	PEG, porous SiC (80%) >100 µm	Molten silicon, Melting impregnation	106.67	31.2	No	>50	52~60	environmentally friendly, and scalable route for efficient solar and thermal energy storage	Disc-shaped D = 12.7 H = 3
Lin, Xianxian 2023 [76]	Polyurethane (PEG monomers:isophorone diisocyanate cross-linker 1:2)	Chemical modification, Vacuum impregnation	116.1	/	No	/	32.4~ 54.4	Directional load-bearing projects for energy conservation and temperature regulation in the automotive and building sectors	/
Wen, Ruilong 2021 [33]	Stearic acid Carbonized maize straw	Vacuum impregnation	160.74	0.3	No	200	65.3~ 67.9	Solar heat energy storage system and energy-efficient buildings	/
Wen, Ruilong 2018 [32]	Stearic acid Carbonized sunflower straw	Vacuum impregnation	186.1	0.33	/	/	65.9~ 66.4	Solar heat energy storage system and energy conservation buildings.	Disc-shaped
Tang, Yili 2022 [29]	Stearic acid Nano-porous carbon	Vacuum impregnation	166.5	0.41	No	200	68~71.9	Solar energy collection and storage	Disc-shaped
Sarı,Ahmet 2022 [34]	Capric-stearic acid eutectic Sugar beet pulp	Vacuum impregnation	117	0.34	No	1000	23~24.5	Temperature controlling of buildings	Diamonds
Song, Jiayin 2022 [77]	PEG, loofah sponge (3.9~4.7 nm)	Vacuum-assisted impregnation	137.6	/	No	>100	40.4~ 52.4	Thermal management systems (intelligent and thermoregulated textiles and infrared stealth of military target)	Cylinder
Yue, Xianfeng 2023 [78]	Paraffin, porous bamboo-derived carbon (34%)	Chemical modification, Vacuum impregnation	116.5	0.522	No	>100	26~29	Building temperature regulation	Disc-shaped
Hekimoğlu, Gökhan 2021 [79]	Methyl palmitate, walnut shell carbon	Chemical modification, Vacuum impregnation	108.32	0.72	No	1000	26.27	Solar thermal controlling of buildings	Disc-shaped

As the main factor affecting the application field, the phase transition temperature of bioinspired composites for thermal energy storage has received a lot of research and attention. The type of PCM, different skeleton structures and material composition will affect the phase change temperature of the composites. Among these, the type of PCM plays an important role in influencing the phase change temperature of the composites. According to the different application temperature, the bioinspired composites for thermal energy storage are divided into low-temperature thermal energy storage materials (<120 °C) [80], medium-temperature thermal energy storage materials (120~400 °C) [81] and high-temperature thermal energy storage materials (400~1000 °C). Due to the low melting point of organic PCMs, such as polyethylene glycol, paraffin and stearic acid, most of the composites composed of these materials operate at temperatures below 120 °C and belong to the category of low-temperature thermal energy storage materials. Inorganic PCMs represented by binary and ternary molten salts have higher melting points, and the composites composed of them basically operating at temperatures ranging from 400 °C to 700 °C are mainly medium and high-temperature thermal energy storage materials [82]. The operating temperatures of several typical materials are summarized in Figure 7.

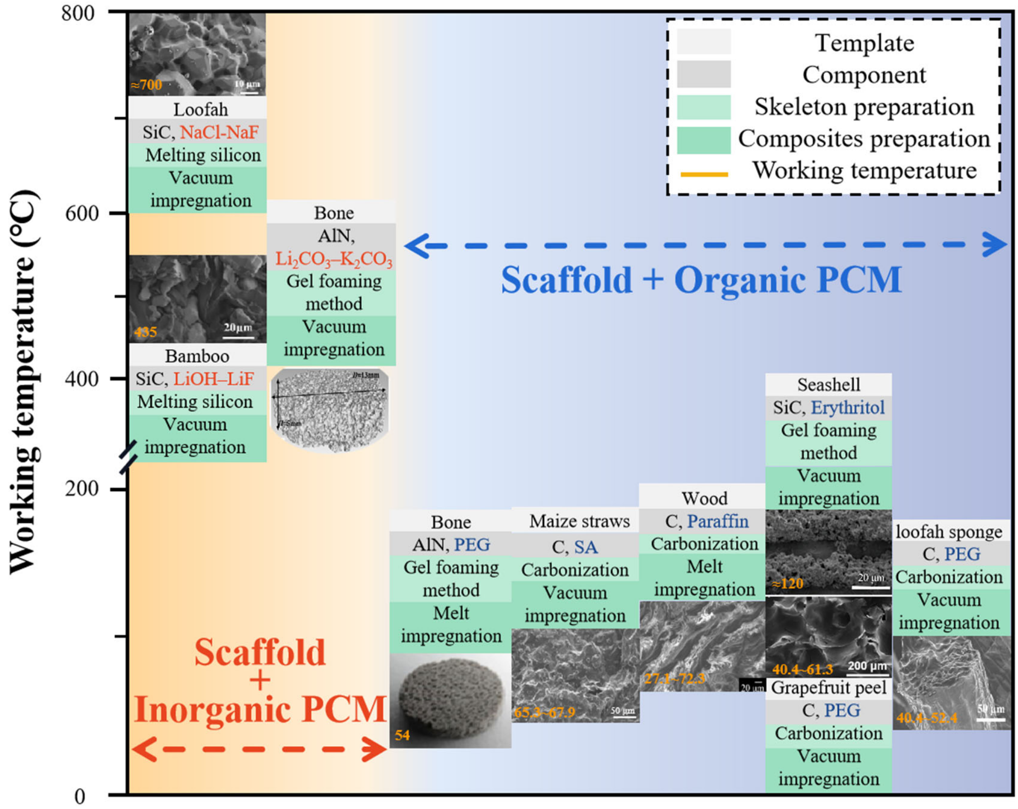

Figure 7. Working temperature diagram of bioinspired composites for thermal energy storage. (Inserted SEM images in Figure 7 are adapted from [33,57,59,62,63,69,70,74,77]).

4.1. Thermal Conductivity Mechanism of Materials

There are three basic ways of heat transfer (as shown in Figure 8), namely: heat conduction, convective heat transfer and heat radiation [83]. Heat conduction is due to temperature differences within or between materials; the movement of microscopic particles is relied upon to produce the process of heat energy transfer. The mechanism is microscopic vibration, displacement and collision of molecules, atoms and electrons inside the material to transfer energy. Convective heat transfer occurs only in a fluid, and heat energy is transmitted through the particles of the flowing medium. The mechanism is the transfer of thermal energy from one place to another due to the relative displacement and mixing of particles in the fluid. Heat radiation refers to the phenomenon of emitting radiation due to heat. The mechanism is that after the material is heated; the atoms or molecules inside will spontaneously go from the excited state to the low-energy state and finally emit energy in the form of electromagnetic waves. Materials are constantly radiating outward and absorbing inward, transferring heat between material in the form of radiation.

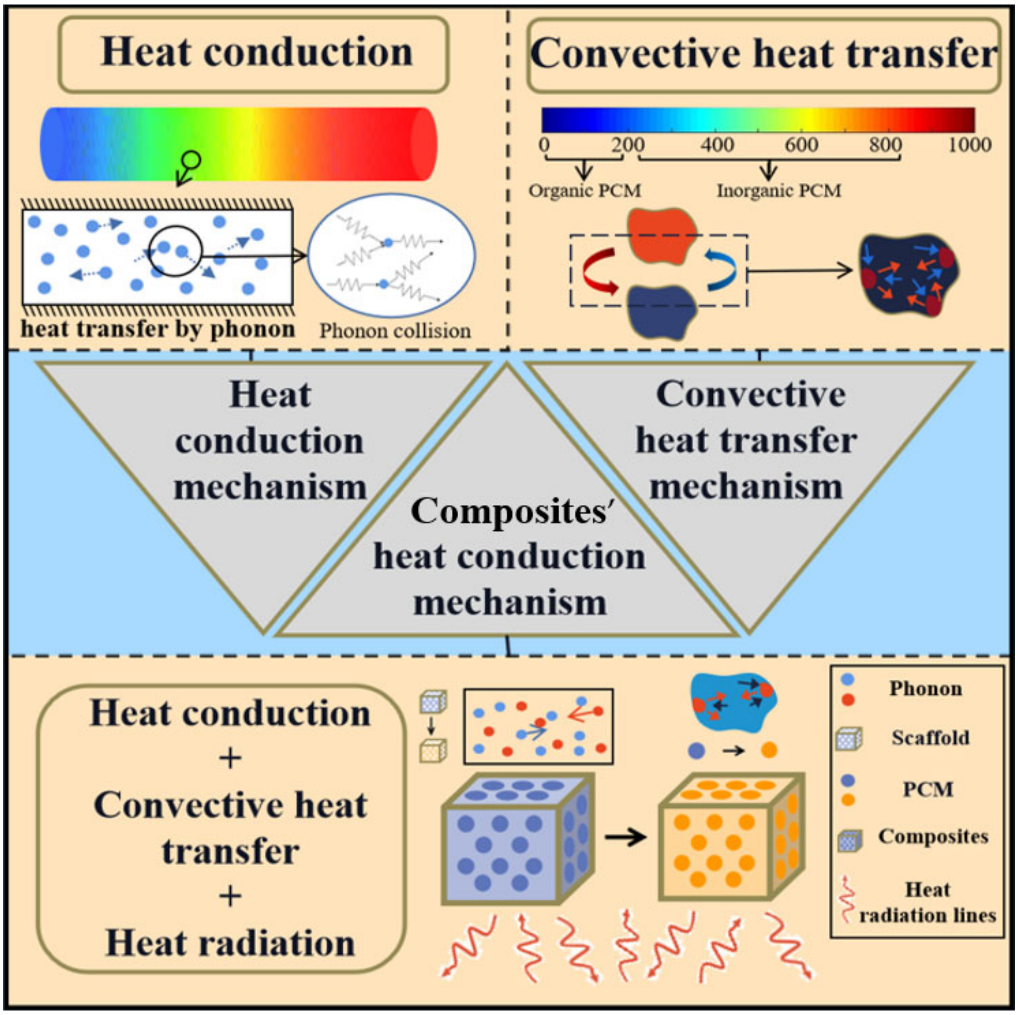

Figure 8. Heat transfer mechanism of bioinspired composites.

The thermal conductivity of bioinspired composites for thermal energy storage is determined by the thermal conductivity of PCMs and the thermal conductivity of skeletons. The thermal conductivity of different components in bioinspired composites is analyzed to study the overall thermal conductivity of composites. Take, for example, that paraffin has a low thermal conductivity. When paraffin with porous silicon carbide forms a composite, paraffin is dispersed into fine components in this composite, and the skeleton material plays a leading role in the thermal conductivity of energy storage composites. The effects of insufficient thermal conductivity of paraffin are compensated for by size effects [84] and thermal conductivity networks. Compared with pure PCM, the composite system improves the thermal energy storage rate of the system. The theoretical thermal conductivity of porous silicon carbide is high, but its thermal conductivity path will be greatly expanded in composites. The thermal conduction between the molecular layers of silicon carbide will transform into the transfer between macroscopic particles, compared with pure porous silicon carbide, and the thermal conductivity will decrease significantly. The porous skeleton structures can be used as a thermal-conductivity enhancer to envelop the PCM. There are a large number of heat transfer interfaces in the composite, and doping excites the vibration of low-frequency atoms, and the increase of overlapping energy makes the phonon vibration match the composite better, and the overall thermal conductivity is improved [85]. In composite systems, the dominant role of convective heat transfer in PCMs will increase with the increase of porosity [86]. In addition, the thermal energy storage characteristics, the encapsulation capacity and the thermal energy transfer performance of the composite can be effectively improved by adjusting the surface properties and pore structures of the skeleton material [87,88]. Bioinspired composites for thermal energy storage use high thermal conductivity skeleton materials, which can significantly improve the thermal conductivity of the overall composites.

4.2. Thermal Energy Storage Performance

The quality of bioinspired composites for thermal energy storage should be judged by their thermal energy storage performance (i.e., heat storage density, thermal conductivity, leakage prevention and cycle times).

The heat storage density of bioinspired composites for thermal energy storage mainly results from the PCMs in the composites (as shown in Figure 9), including organic PCMs (paraffin, PEG, stearic acid, etc.) and inorganic PCMs (molten salt). Heat storage density of composites using organic PCMs are mostly in the range of 100 kJ/kg to 200 kJ/kg, while that of composites using inorganic PCMs is much higher, with most of them reaching more than 300 kJ/kg. The thermal conductivity of composites for thermal energy storage is influenced by the skeleton and the PCM, with above 90% of the thermal conductivity results from the skeleton (SiC skeleton, C skeleton and AlN skeleton). Most C skeleton composites have a thermal conductivity of 1 W/m*K or less, while SiC skeleton composites have a thermal conductivity of 20 W/m*K or more. The heat storage density determines the storage capacity of the material, and the thermal conductivity determines the rate of heat exchange between the material and outside.

Loofah-derived SiC skeleton/NaCl-NaF composites exhibited the highest heat storage density of 424 kJ/kg, which was attributed to the large porosity of the SiC skeleton combined with the large energy density of NaCl-NaF. The thermal conductivity is affected by the large porosity, with the well-connected and continuous SiC skeleton acting as a heat transfer channel, which exhibits a thermal conductivity of up to 20.7 W/m*K [59]. Oak-derived SiC skeleton/NaCl-KCl composites exhibited the highest thermal conductivity of 116 W/m*K, which was attributed to the high thermal conductivity of the SiC skeleton and moderate porosity, resulting in a thermal storage density of 157 kJ/kg [58]. For porous AlN skeleton/Li_2CO_3–K_2CO_3 composites, the thermal conductivity of the material is 13.6 W/m*K due to the lower thermal conductivity of AlN than SiC, and Li_2CO_3–K_2CO_3 has a large energy storage density so that the material's thermal storage density can reach 342 kJ/kg [69]. Bamboo-derived SiC skeleton/LiOH-LiF composites exhibited a thermal conductivity of 35 W/m*K and a thermal storage density of 309 kJ/kg, and there was no significant change in the thermal conductivity and energy storage density after 2500 cycles [57]. Furthermore, the TiN nanoparticles are loaded on the surface of BSiC which can significantly improve the solar energy absorption rates.

Among the bioinspired inorganic skeleton-organic PCMs, oval-shaped resin/n-octadecane composite has the highest thermal storage density of 243.5 kJ/kg and a thermal conductivity of only 0.1505 W/m*K [65]. However, its completely closed structure ensures that the composite does not leak. When SiC and AlN are used as the skeleton, the vertical and compact grains act as heat transfer channels so that the composites exhibit better thermal conductivity (31.2 W/m*K, 17.16 W/m*K, 25.63 W/m*K) [62,63,75]. In addition, with the help of porous carbon scaffolds, the carbon one-dimensional arrangement forming a layered pore structure has a high specific surface area, which can improve the heat transfer to increase the thermal conductivity to 0.61 W/m*K, with no leakage after 100 thermal cycles, and the heat storage density reaches 186 kJ/kg [74].

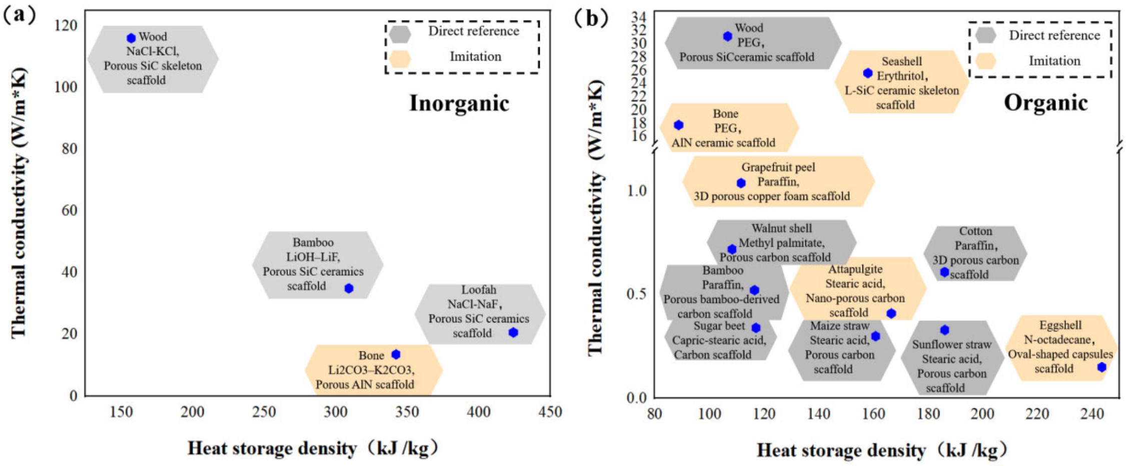

Figure 9. Heat storage performance diagram of bioinspired composites for thermal energy storage: (**a**) Summary of heat storage properties of (bioinspired inorganic skeleton-inorganic PCMs) [55–57,67]. (**b**) Summary of heat storage properties of (bioinspired inorganic skeleton-organic PCMs) [29,32–34,60,61,63,70,72,73,76,77].

5. Applications

Bioinspired composites for thermal energy storage have become the key direction of thermal energy storage technology development because of their low cost, high degree of technology, high energy density, stable heat release process and wide application prospects. Because of the leakage and corrosion of PCMs, most researchers have chosen leakage-proof and corrosion-resistant inorganic scaffolds to improve the thermal conductivity of the composites, providing a basis for the controllable regulation of the thermal energy storage (heat storage density, thermal conductivity and leakage prevention, etc.). Due to its capillary action, the PCM can remain in the carrier material after a hot melt without spillage, and the material has the advantages of large energy storage density, controllable heat release and long life. Bioinspired composites for thermal energy storage can absorb/release a large amount of heat through phase change while keeping the temperature constant, so they can be widely used in energy efficiency in buildings [89,90], waste heat recovery [62], cold chain transportation [91,92], solar–thermal energy conversion/storage [93], battery thermal management and other fields.

Nowadays, the most popular research is applied to the field of building energy efficiency and solar-thermal energy conversion/storage. In the application of energy efficiency in buildings, bioinspired composites for thermal energy storage can effectively use solar energy to store heat or electricity during the trough of power load to store heat or cold so that the heat fluctuation amplitude between the indoor and outdoor buildings is weakened and the action time is delayed, thereby reducing indoor temperature fluctuations, improving comfort and saving energy consumption. In the application of solar–thermal energy conversion/storage, bioinspired composites for thermal energy storage have high photothermal conversion efficiency, which can quickly convert light energy into heat energy, reduce energy consumption, store excess solar energy and release it when it is insufficient to meet the needs of continuous and stable supply of energy for production and life. Liu et al. discuss bioinspired porous BSiC ceramics prepared from bamboo by gene-assisted methods, which exhibit a high heat storage density of 309 (kJ/kg) and a thermal conductivity of up to 35 W/m*K for high-temperature applications [57]. Moreover, the TiN nanoparticles loaded on the surface of BSiC ceramics have extremely high solar absorption rates. He et al. developed a magnetically accelerated solar thermal energy storage method that can effectively integrate the magnetic and high thermal conductivity characteristics of nanomaterials, which is conducive to solar-direct absorption collectors and energy-storage technologies [68]. Currently, the application in this aspect is still at the laboratory-exploration stage. It is necessary to explore effective methods to solve problems such as its service life and cost.

In the application of waste heat recovery, industrial waste heat resources are abundant, but intermittent discharge and recycling are difficult. Bioinspired composites used for thermal energy storage have a large energy storage density and the output temperature and energy are quite stable, which is conducive to improving energy utilization. In the application of cold chain transportation, the most important factors affecting the quality of freeze-sensitive products are temperature changes and fluctuations during product storage and transportation, which will lead to a decrease in food quality and a shorter shelf life. Bioinspired composites for thermal energy storage have an approximately constant application temperature, which can slow down temperature changes and fluctuations. In the application of batteries, they can serve as temperature-controlled energy storage for batteries to adapt to high temperatures and cold environments, maintain the optimal charging and discharging temperature of the batteries and increase the battery life. In terms of lithium-ion batteries, Liu et al. prepared biomorphic straw-like Co-doped Fe_2O_3 (SCF) beams by the hydrothermal process, which has ultra-high initial discharge specific capacity and cycle stability [94]. The unique straw, bundle-like structure accelerates the diffusion of lithium ions and mitigates huge volume expansion during cycling. In terms of lithium-sulfur batteries, Cui et al. prepared cabbage-like nitrogen-doped graphene/sulfur complexes (NG/S) [95]. This novel cabbage-like morphology has a three-dimensional connecting structure with unique fine grooves and folds that can counteract defects in non-conductive sulfur species and store sulfur with high cyclic stability.

In summary, the application areas of these five areas can be divided into two parts: those that are immediately applied to mass production and those that are still under research and have large applications, as shown in Figure 10. In the field of building energy saving, bioinspired composites can be made into insulation layers for walls and floors, and materials with fast photothermal conversion energy storage can be used in the production of new solar water heaters.

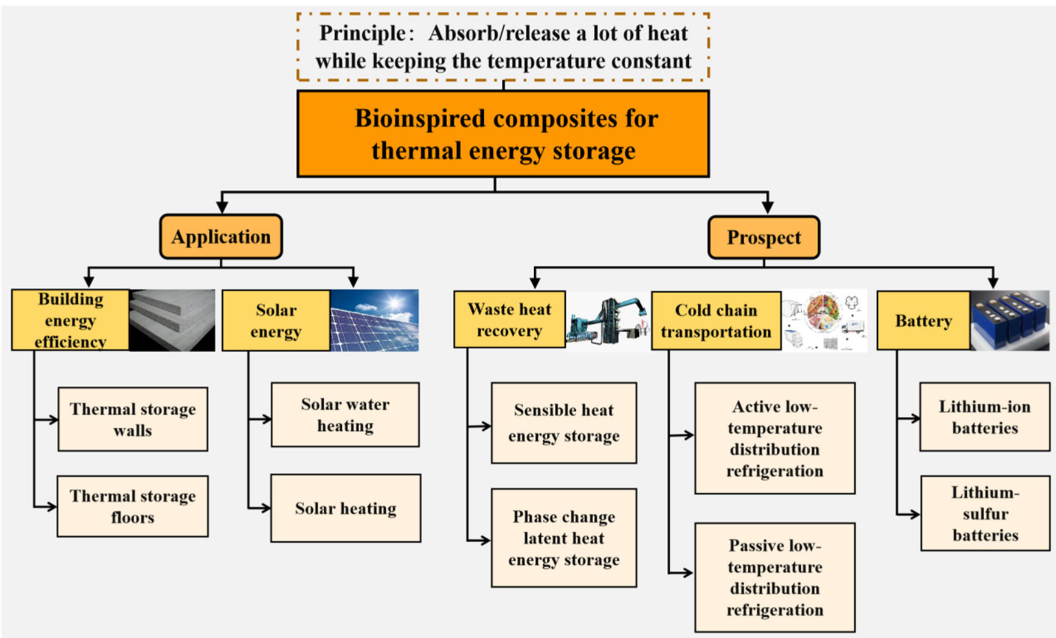

Figure 10. Bioinspired composites for thermal energy storage application system diagram.

6. Conclusions and Outlook

Firstly, the origin of the development of bioinspired composites for thermal energy storage was introduced and the processing methods, properties improvement and enhancement of bioinspired composites for thermal energy storage were comprehensively reviewed. The bioinspired composites for thermal energy storage can effectively improve the overall thermal conductivity and leakage resistance, owing to the high thermal conductivity of the skeleton and the effective encapsulation of the PCM. Taking the carbide sunflower straw (CSS) skeleton as an example, the thermal conductivity of the SA/CSS composite is increased by 106.3% compared with pure SA [32]. Then, the preparation methods of the bioinspired energy storage composites were introduced in detail, including the main preparation methods of the skeleton and the composites. The skeleton is prepared by using the natural structure as a template. Its preparation process mainly includes modification, impregnation and sintering. The composite material is prepared by impregnating the phase change material into the wood-derived scaffold through the impregnation method. In recent work, the performance of composites (thermal conductivity, heat storage density and leakage resistance) has been summarized. The thermal conductivity of the composite (inorganic NaCl-KCl/Wood-like porous SiC) can reach as high as 116 W/m*K, and the composite (organic PEG/Wood-like porous SiC) can reach as high as 31.2 W/m*K. The heat-storage density of the composite (inorganic NaCl-NaF/Wood-like porous SiC) can reach as high as 424 kJ/kg, and the composite (organic n-octadecane/Oval-shaped capsules) can reach as high as 243.5 kJ/kg. The leakage resistance of the composite (inorganic LiOH-LiF/Bamboo-like porous SiC) can reach up to 2500 cycles without leakage, and the composite (organic Methyl palmitate/walnut shell-like carbon) can reach up to 2500 cycles without leakage. There are already many methods that can improve the performance of PCMs, but there is still a lack of effective ways to simplify the preparation process and reduce the production cost. The large-scale application of PCMs in the future needs to address these issues.

In conclusion, bioinspired composites for thermal energy storage have diverse microstructures, simple preparation procedures, excellent heat storage properties and wide application prospects. At present, the development of bioinspired composites for thermal energy storage is influenced by the following factors:

1. The skeleton preparation process of these bioinspired composites for thermal energy storage is meticulous, and how to simplify the preparation process so that it can be used on a large scale still needs to be explored.
2. Most bioinspired composites for thermal energy storage cannot meet the rapid energy transfer and efficient energy storage demands at the same time.
3. Nowadays, most bioinspired thermal energy storage composites are small disc types or elliptical spheres, while large-sized materials with good performance need to be further investigated.
4. The research of bioinspired composites for thermal energy storage is at early stage, and their applications are mostly in the stage of laboratory verification. Energy storage efficiency, lifespan, cost and technology maturity still need to be further verified and improved.

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