

Perspective

Phase change material-based thermal energy storage

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SUMMARY

Phase change materials (PCMs) having a large latent heat during solid-liquid phase transition are promising for thermal energy storage applications. However, the relatively low thermal conductivity of the majority of promising PCMs ($<10\text{ W}/(\text{m}\cdot\text{K})$) limits the power density and overall storage efficiency. Developing pure or composite PCMs with high heat capacity and cooling power, engineering effective thermal storage devices, and optimizing system integration have long been desired. Our perspective outlines the needs for better understanding of multi-physics phase change phenomena, engineering PCMs for better overall transport and thermodynamic properties, co-optimizing device design, and integrating PCMs with potential applications. We start by covering the heat transfer fundamentals of PCMs. We then discuss PCM property characterization and need for materials design. We conclude by discussing higher-level device design and integration principles, as well as emerging applications and requirements. We also identify future research opportunities for PCM in thermal energy storage.

INTRODUCTION

Solid-liquid phase change materials (PCMs) have been studied for decades, with application to thermal management and energy storage due to the large latent heat with a relatively low temperature or volume change. Recent advances and challenges associated with electrification (photovoltaics and wind), high-power-density electronic devices and machines, electrified transportation, energy conversion, and building air conditioning have re-invigorated interest in PCM thermal storage.^{1–3} Thermal storage using a PCM can buffer transient heat loads, balance generation and demand of renewable energy, store grid-scale energy, recover waste heat,⁴ and help achieve carbon neutrality.⁵ Compared with other energy storage methods such as electrochemical batteries, PCMs are attractive for their relatively low cost and ease of integration with readily available energy resources such as solar power.^{6,7}

Although the large latent heat of pure PCMs enables the storage of thermal energy, the cooling capacity and storage efficiency are limited by the relatively low thermal conductivity ($\sim 1\text{ W}/(\text{m}\cdot\text{K})$) when compared to metals ($\sim 100\text{ W}/(\text{m}\cdot\text{K})$).^{8,9} To achieve both high energy density and cooling capacity, PCMs having both high latent heat and high thermal conductivity are required. One method to increase the thermal conductivity of a PCM is to mix the PCM with a high thermal conductivity material, forming a composite structure via the inclusion of fins, matrices, or particles.^{10–12} However, the introduction of thermal conductors reduces energy density due to their low heat capacity ($\sim 1\text{ kJ}/(\text{kg}\cdot\text{K})$) when compared to the typical PCM latent heat ($\sim 100\text{ kJ/kg}$). The tradeoff between energy density and power density must be considered and evaluated using rigorous figures of merit that allow for

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the comparison of cooling capacity for different materials. However, cooling capacity is not an intrinsic material property, and depends highly on geometry, boundary conditions, and temperature cycling. The analysis therefore requires careful evaluation and understanding of heat transfer and phase change phenomena at molecular, device, and system levels. Computational methods such as the finite element method (FEM), the finite volume method (FVM), machine learning (ML), and topology optimization (TO), when coupled with experiments, represent promising tools for optimal device development and integration. Critical to understanding heat transfer in PCMs is the lack of standard experimental procedures for materials characterization and testing, which prevents the collection of consistent data related to material properties and device performance.

As we consider heat transfer from the molecular length scale to the application length scale, emerging applications of thermal storage come into view. These include personal cooling, consumer electronics, building thermal energy storage, and biomedical devices.^{13,14} In real applications, the benefits derived from PCM thermal storage must be considered at the systems level. In addition to energy and power density, the cost, safety, and reliability represent the most important factors. For example, organic PCMs have been used for transient cooling of Si-based microprocessors due to their large latent heat and high dielectric breakdown strength.¹⁵ However, organic PCMs are not applicable to high-temperature wide-band-gap (WBG) power electronics due to their flammability and degradation with extreme temperature cycling. To enable the efficient analysis of the numerous factors related to systems-level implementation, artificial intelligence- (AI) and ML-enabled tools could provide effective selection methodologies. In addition, manufacturing scale-up and automation represent vitally important criteria for thermal storage implementation at the industrial scale. Recent advancements in polymer and metal additive manufacturing, especially when coupled with TO, can unlock novel geometries and complex structures co-designed for specific applications.

In this perspective, we focus on PCM-based thermal energy storage, starting from heat transfer fundamentals and demands to motivate research needs. We discuss key challenges to the tailoring of PCM thermophysical properties and figures of merit. We then progress to engineering challenges related to PCM device design and system integration. We end our perspective by summarizing emerging opportunities and applications in the thermal storage and heat transfer communities. We provide a road map to guide the community toward what we believe are fundamental and applied needs to overcome the challenges of PCM implementation.

Heat transfer fundamentals of PCM thermal storage

Melting and solidification have been studied for centuries, forming the cornerstones of PCM thermal storage for peak load shifting and temperature stabilization. **Figure 1A** shows a conceptual phase diagram of ice-water phase change. At the melting temperature T_m , a large amount of thermal energy is stored by latent heat ΔH due to the phase transition of the hydrogen-bond network shown by the inset image. The solidifying temperature T_s can be tens of degrees Celsius lower than T_m due to supercooling caused by a lack of nucleation sites.¹⁶ Usually, supercooling is not preferred due to the uncontrollability of phase transition as well as variability in the onset temperature. By controlling the temperature of phase transition, thermal energy can be stored in or released from the PCM efficiently. **Figure 1B** is a schematic of a PCM storing heat from a heat source and transferring heat to a heat sink. The PCM consists of a composite Field's metal having a large volumetric latent heat ($\approx 315 \text{ MJ/m}^3$) and a copper (Cu) conductor having a high thermal conductivity

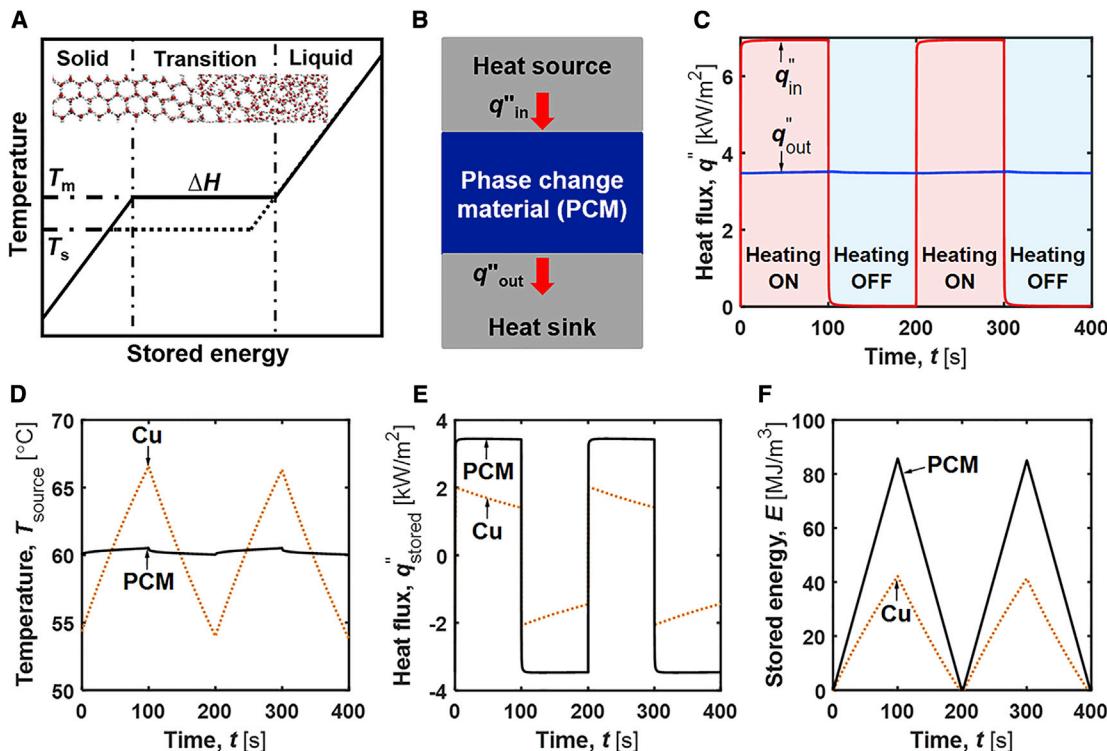


Figure 1. Phase change material (PCM) thermal storage behavior under transient heat loads

(A) Conceptual PCM phase diagram showing temperature as a function of stored energy including sensible heat and latent heat (ΔH) during phase transition. The solidification temperature (T_s) is lower than the melting temperature (T_m) due to supercooling. Inset: molecular structure of the ice-water phase transition.

(B) Schematic of a PCM (blue color) absorbing a transient heat flux (q''_{in}) from a volumetric heat source (top gray color) and conducting heat (q''_{out}) to a heat sink (bottom gray color). The heat source generates 28 W for 100 s and 0 W for a following 100 s. The heat sink is cooled by forced convection with a heat transfer coefficient of 100 W/(m² K) and a far-field temperature of 25°C. The heat source and heat sink are 4-mm wide and 2-mm high copper (Cu) blocks. The PCM is a composite material consisting of a Cu foam (13% by volume) embedded in a Field's metal.

(C–F) Transient heat flux of absorption (q''_{in}) and conduction (q''_{out}) by the PCM as a function of time (t) (C). Thermal storage behavior of the PCM is compared with pure Cu for (D) heat source temperature (T_{source}), (E) stored heat flux (q''_{stored}), and (F) stored energy (E).

($\approx 384 \text{ W}/(\text{m} \cdot \text{K})$), to enable both high energy density and cooling power. When the heat source generates a transient heat load during heating in the ON state, the PCM absorbs the heat q''_{in} , stores $\sim 50\%$ of the q''_{in} , and conducts the remaining q''_{out} to the heat sink shown in Figure 1C. During the heating OFF state, the stored heat within the PCM is released and conducted out. The analysis shows that the PCM buffers heat spikes from the heat source and reduces the transient peak heat loads by 50% for the heat rejection system. Compared with a solid Cu thermal spreader, the PCM reduces heat source temperature swings by $>90\%$ (Figure 1D) and stores twice the transient heat flux q''_{stored} (Figure 1E) and thermal energy E (Figure 1F). The calculations of energy absorption, storage, and conduction show that composite PCMs having high cooling capacity can buffer transient heat loads and enhance temperature stability. Note S1 and Figure S1 show the full details of the modeling.

The temperatures and zones at which melting or solidification occur are key parameters for PCMs. Superheating rarely occurs in PCMs. For pure, alloy, or mixture PCMs, supercooling can be large, which affects the solidification process, microstructure evolution, and thermal energy release. Supercooling of composite PCMs is smaller than pure PCMs because the presence of a high thermal conductivity phase increases the available nucleation sites and facilitates heterogeneous

nucleation.^{17–19} The asymmetry of the phase transition can be attributed to different nucleation behavior, structure evolution, thermodynamics, and kinetics between the melting and solidification processes.²⁰ The microstructure evolution influenced by melting is particularly important for metal alloy PCMs. For example, re-melting significantly influences structure evolution, which can occur in transient casting processes even if the overall enthalpy decreases. Convection-induced re-melting can occur for large volumes and long timescales, which affects thermal storage and heat transfer. For non-eutectic mixtures, melting and solidification can happen simultaneously in the mushy zone, which can cause dendrite arm coarsening and liquid film migration.²¹ Thermophysical properties such as thermal conductivity exhibit a discontinuity while approaching the phase transition mushy zone. The structure evolution, phase composition, solute diffusion, and heat transfer kinetics are poorly understood in the mushy zone, and there is a need for additional experimental and modeling efforts.

To model phase change and heat transfer from a multi-physics perspective, various methods have been studied, such as the temperature method, apparent heat capacity method, effective capacity method, heat integration method, source-based method, and enthalpy method.²² The temperature method solves for the moving boundary during phase change in pure or eutectic materials. The heat capacity and effective capacity methods use equivalent latent heat capacity within a finite temperature interval, which is applicable for mushy zone phase change. The simple heat integration method is a post-iterative method and can be easily applied to multidimensional problems. However, prediction of the moving boundary is often inaccurate. The source-based method treats the latent heat as a heat source or sink directly related to the grid point temperature, which can adapt in existing numerical codes and provide fairly good accuracy and computational efficiency. The enthalpy method relates the enthalpy with temperature to account for the latent heat and avoid nonlinearity in solving the heat transfer problem. However, the enthalpy method requires linearization when the thermal conductivity is a function of temperature.²³ For high heat fluxes transferred into the PCM at short timescales, rapid phase change with local non-equilibrium heat transfer needs to be resolved using the finite heat propagation velocity or thermal relaxation approximation.^{24,25}

Phase change material properties

PCMs can be classified as organics, hydrates, molten salts, and metal alloys. For thermal storage, the melting temperature, latent heat, and thermal conductivity of the PCM are important thermophysical parameters. The melting temperature determines the temperature range for which the PCM thermal storage is effective. The latent heat indicates the energy density of the PCM during store or release cycles. The thermal conductivity governs the charge or discharge rate of thermal energy, sometimes labeled as the cooling power. Figure 2A shows the volumetric latent heat and thermal conductivity of different PCMs in the solid phase. Compared with organic PCMs, metal alloy PCMs have higher melting temperature, volumetric latent heat, as well as thermal conductivity. This makes metal alloys more promising for high temperature and high heat flux applications. However, for applications requiring high specific power density, organics and salts with higher specific latent heat are appropriate (Figure 2B). The molar latent heat ΔH strongly depends on the melting temperature T_m by the thermodynamic correlation of $\Delta H = T_m \cdot \Delta S$, where the molar entropy change during phase change (ΔS) is $<4.5R$ for salts, $<3R$ for semiconductors, and $<1.5R$ for metals where R is the ideal gas constant (8.314 J/(mol · K)).^{26,27} The entropy change is difficult to predict accurately due to contributions from multiple factors such as configurational, volumetric, rotational, mixing,

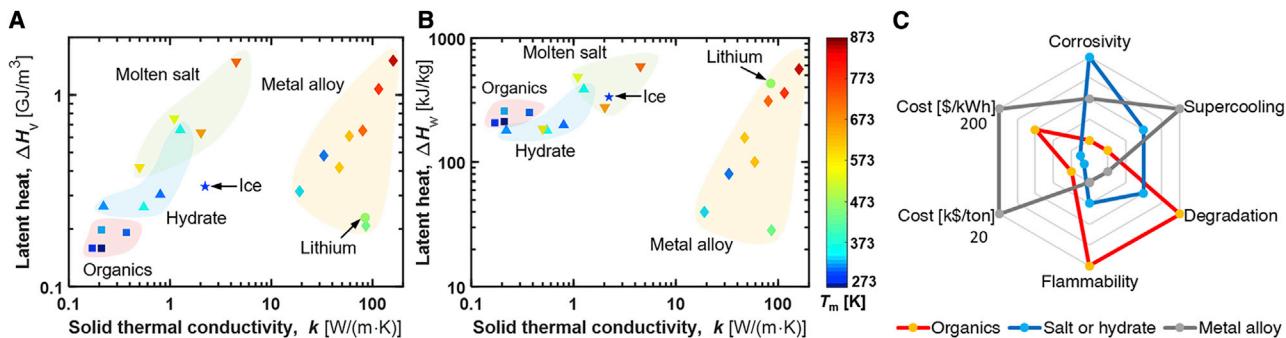


Figure 2. Thermophysical properties of PCMs

(A and B) Volumetric (A) and specific (B) latent heat of different PCMs as a function of thermal conductivity in the solid phase. The melting temperature (T_m) is indicated by the color map.

(C) Characteristics of PCMs. Metrics and values are summarized in Note S2 and Table S1.

The error bars in (A) and (B) are smaller than the symbol size and are not shown for clarity.

and electronic entropic terms associated with the melting process.²⁸ The entropy change of polymers can be predicted using the Clapeyron equation by $\Delta S = \Delta V / (dT/dP)$ near the melting temperature.²⁹ Several methods have been proposed to predict the entropy change of salt mixtures or metal alloys by taking the summation of individual entropy change components along with the entropy of mixing.^{30,31} However, these prediction methods either require extensive experimental data or suffer from large errors.

One of the main challenges to predicting PCM thermophysical properties is the lack of understanding of the relationships between constituent properties and molecular and microstructural compositions. At the molecular or atomic levels, quantum mechanics-based calculations are used to solve for force fields and energy parameters via approximations of the Schrodinger equation using the principles of thermodynamic hypothesis, additivity, and transferability. The quality of the results depends critically on the quality of the approximations used. More rigorous computational approaches using *ab initio* calculations are more computationally demanding than classical semi-empirical approaches. Among these approximations, density functional theory (DFT) relying on the Born-Oppenheimer approximation provides good accuracy and requires fair computational resources especially for large systems such as polymer PCMs.³² For example, some work has considered the relationship between molecular structure and thermophysical properties of organic PCMs, revealing the links between molecular functional groups, orientation, thermal conductivity, melting temperature, and phase change.^{33–35} At the larger scale, all engineering materials have certain types of microstructure. Designing new materials is largely dependent on controlling these structural features during materials processing and phase transformations. Enormous efforts have been made to develop microstructure models, of which the phase field method based on a diffuse-interface description for the thermodynamics of inhomogeneous systems has received tremendous attention. However, such continuum simulations are based on approximate models not derived from quantum mechanics. The underlying model assumptions should be carefully checked for applicability to a given application such as phase field simulations of solidification and statistical theory of grain growth.³⁶ Overall, the links between molecule, microstructure, and properties have motivated research on computational methods for property prediction and materials design. First-principle methods such as DFT have been developed to predict the properties of PCMs.^{37,38} Empirical methods such as the computer coupling of phase diagrams

and thermochemistry (CALPHAD) method were developed to predict properties based on experimental data.^{39,40} However, in real applications, fast heat transfer rates induce the metastability of phase change, which requires kinetic phase diagrams for PCM design by assessing thermodynamic stability relationships. Future research will likely combine computational tools with AI to find novel PCMs with optimal structures and properties.

Within the context of applications, the economic cost, safety, durability, and supercooling form additional selection criteria for PCMs. Figure 2C shows tradeoffs between different types of PCMs. Note S2 and Table S1 show full details of the PCM metrics and values. For example, organics have the lowest supercooling and also tend to degrade and are flammable. Salts have the lowest economic cost; however, they are the most corrosive to containers and suffer from phase separation, which requires expensive auxiliary components. Metal alloys are the most stable and the most expensive. Understanding tradeoffs between different performance parameters are a necessity in future engineering research. To reduce the economic cost of materials, PCM blends are applied. Congruent PCMs whose solid and liquid phases have the same composition are ideal PCMs without phase separation even under supercooling. However, phase separation may exist in eutectic PCMs when one of the solid phases supercools.⁴¹ To design ideal PCMs such as congruent or eutectic melting systems, phase diagrams and Tamman plots are used to determine material compositions.⁴²

A tradeoff exists between the energy density (latent heat) and power density (thermal conductivity) for optimal PCM design. Figure 3A shows the transient boundary heat flux ($q'' = f(t)$) absorbed by solid-liquid phase change as a function of time (t) when the left boundary superheat reaches 10 K for various boundary conditions. By solving the one-dimensional (1D) Neumann-Stefan problem in a semi-infinite solid initially at the melting temperature,^{43,44} the constant temperature ($T_w = T_m + 10$ K) boundary condition generates the analytical solution for the transient boundary heat flux $q'' = (k \cdot \Delta H_V)^{1/2} \cdot (T_w - T_m)^{1/2} \cdot (\pi t)^{-1/2}$, where the Stefan number $\text{Ste} = C_p \cdot (T_w - T_m)/\Delta H_V < 0.5$, which forms the lower limit of shaded bands in Figure 3Ai. The numerical solution of the phase change problem having a constant heat flux boundary ($q'' = \text{constant}$) as a function of time when the boundary superheat reaches $T_w - T_m = 10$ K forms the upper limit of the shaded bands. Figure 3Ai shows that the cooling capacity figure of merit (FOM) $\eta = (k \cdot \Delta H_V)^{1/2}$ of metallic PCMs outperforms salts and organics due to the high thermal conductivity and large volumetric latent heat. Figure 3Aii shows the cooling capacity of a 5-mm-thick solid PCM with a constant heat flux left boundary and a convection right boundary, with the heat transfer coefficient h ranging from 0 to 10 kW/(m² · K) at the cooling temperature $T_c = T_m - 20$ K, common to real applications. Convective cooling enhances the long-term cooling performance but has less influence at short times, depending on the thermal effusivity of the PCM. For thermal storage of highly transient heat flux spikes, the PCM cooling capacity is more important than the convectively cooled heat sink in short times. However, in longer times after PCM exhaustion, the PCM cooling capacity is limited by its low intrinsic thermal conductivity when compared to an all-metal heat sink. To enhance the cooling capacity for high heat flux storage, both high latent heat and thermal conductivity are desired for PCMs.

The cooling capacity of a PCM can be increased by adding a high thermal conductivity material such as nanoparticles, carbon fiber, or metal fins to form a heat conducting matrix.⁴⁵⁻⁵⁰ Figure 3B shows the effective cooling capacity η_{eff} and effective thermal conductivity k_{eff} of composite PCMs enhanced via the addition of diamond or Cu thermal

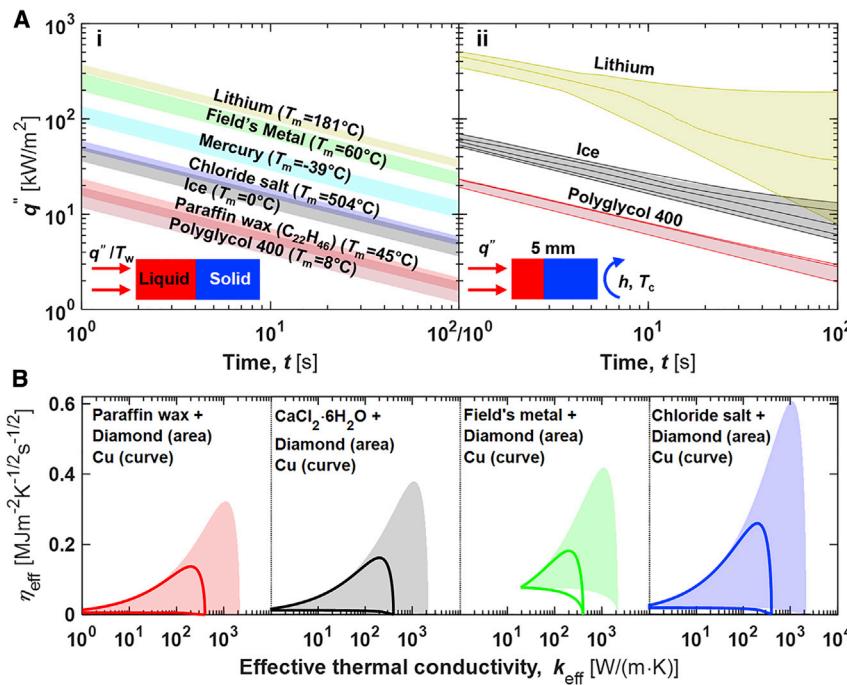


Figure 3. Cooling capacity figures of merit and composite PCMs properties

(A) Heat flux (q'') absorbed by different melting boundary conditions as a function of time (t) for (i) semi-infinite solid PCM and (ii) 5-mm-thick solid PCM cooled by forced convection, when the wall superheat reaches 10 K. The shaded bands represent the cooling capacity range for different boundary conditions. Insets of (i) and (ii) are schematics of the simulation domain for each respective boundary condition.

(B) Effective cooling capacity (η_{eff}) and effective thermal conductivity (k_{eff}) for PCM composites having different volume fractions of thermal conductors (diamond or Cu). Diamond-enhanced figures of merit are shown by shaded areas. Cu-enhanced figures of merit are shown by solid lines.

conductors to a variety of PCMs. Note S3 and Figure S2 show the full details of the PCM composite geometry and modeling conducted here. The results show that k_{eff} increases with conductor volume fraction ϕ , showing an upper bound of $\phi \cdot k_{\text{cond}} + (1 - \phi) \cdot k_{\text{PCM}}$ parallel and a lower bound of $[\phi/k_{\text{cond}} + (1 - \phi)/k_{\text{PCM}}]^{-1}$ series thermal transport.⁵¹ While the introduction of a thermal conductor enhances heat conduction, it reduces the thermal energy that can be stored due to the negligible heat capacity of the conductors when compared to the PCM latent heat. The effective volumetric latent heat decreases with ϕ as $\Delta H_{V,\text{eff}} = (1 - \phi) \cdot \Delta H_V$. The local maxima of cooling capacity FOM $\eta_{\text{eff}} = (k_{\text{eff}} \cdot \Delta H_{V,\text{eff}})^{1/2}$ in Figure 3B indicate that an appropriately designed composite PCM with a high- k thermal conductor and optimal volume fraction can buffer temperature spikers better than either a thermal conductor or PCM. However, adding conductors can stymie natural convection in liquid PCMs induced by the buoyancy force, which is dependent on material properties and application context and should be rigorously evaluated by future research. Research challenges exist in achieving conductor TO for a given application as well as materials selection and compatibility for the minimum economic cost.

More research opportunities emerge in modifying PCMs with additional materials or structures for novel functions. Some work has considered organic PCMs with dispersed nanoparticles or graphene sheets to enhance the thermal conductivity and latent heat simultaneously.^{35,52,53} Nanomaterials (<100 nm) can change the microstructure of the PCM and enhance the crystallinity of organics, which increases

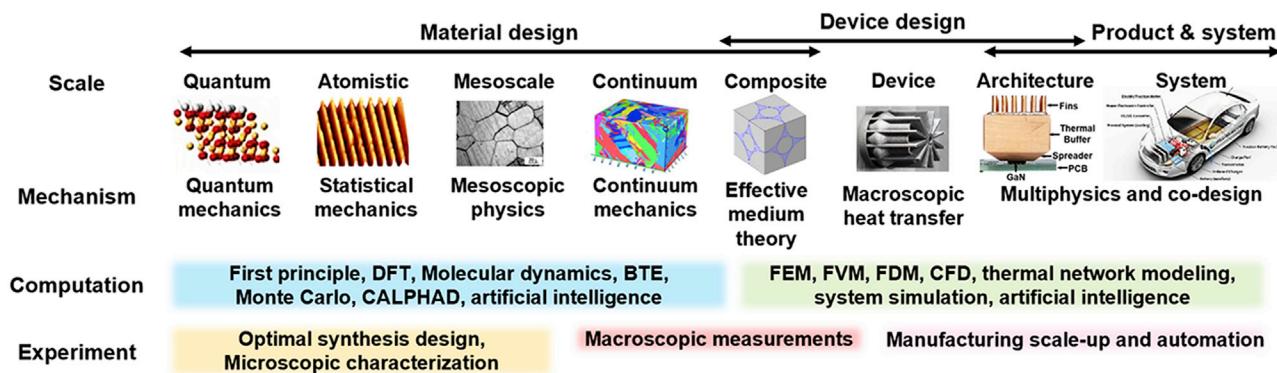


Figure 4. Length scales and associated modeling, design, and test methods for PCMs, devices, and systems

Multiphysics modeling of PCMs at different length scales is governed by different scale-dependent mechanisms. Based on these mechanisms and empirical parameters, computational methods are developed to predict, design, and optimize materials, devices, and systems. Experimental methods provide approaches that enable material synthesis and characterization, property measurements, and manufacturing and prototype testing. BTE, Boltzmann transport equation; CFD, computational fluid dynamics; FDM, finite difference method.

the latent heat of fusion. However, the improvement is relatively small and requires further investigation. Such measurements require care with respect to procedures used and sample sizes characterized, as small changes in the sample heating rate can result in different latent heat values.^{54,55} Standards from lab to lab and round-robin measurements need to be established for consistency. Dispersion of multi-functional materials has received attention in the exploitation of multi-functional composite PCMs. Core-shell encapsulation using metal oxides has been shown to reduce supercooling and form shape-stable PCMs.⁵⁶ Solar-thermal energy storage can be accelerated by the dynamic tuning of Fe₃O₄/graphene optical absorbers within PCMs using magnetic fields.¹ Latent heat storage or release can be controlled by electrical triggering of photoisomer-decorated organic PCMs or mechanical triggering of molecular switches dispersed within PCMs.¹⁶ However, further research is needed in aspects of both mechanisms and applications toward multi-functional and controllable PCMs.

PCM thermal storage device design and system integration

Thermal storage with PCMs requires multiple disciplines, including materials engineering, device design, and product and system integration. Figure 4 shows mechanisms, computational methods, and experimental efforts at different length scales within PCM thermal storage research and development. Understanding the fundamentals and interconnection of these length scales is important to develop novel materials, design high-performance devices, and integrate PCMs with real systems. For example, nanoscale-to-microscale material syntheses create novel microstructures from which macroscopic properties can be characterized. The properties govern the device design and system performance. The integration of experimental data and analysis tools across length scales is required for materials innovation and manufacturing scale-up. For example, to develop a thermal storage technique for the transient cooling of electric vehicle fast charging, we must first obtain both theoretical and experimental data of transient heat generation within the battery and associated components. Then, modeling approaches such as reduced-order thermal networks can be used to determine the appropriate architecture and PCM cooling capacity as well as properties such as the required melting temperature T_m . Then, composite PCMs and devices can be designed to meet the cooling capacity requirements, during which computational methods can predict thermal performance.

Engineering the properties of PCMs likely requires materials design at atomic and molecular scales, and then synthesis and validation at larger scales. Atomistic or molecular-level modeling should be calibrated with experimental data, such as CALPHAD models for metal alloy phase analysis, because understanding of equilibria or kinetic mechanisms at small length scales is still relatively immature. With the developed materials, prototype devices and integrated architectures, along with prototype testing, are necessary to evaluate product performance and to obtain feedback for iterative improvement. Co-design approaches can be used to develop thermal storage devices tailored for specific applications having optimal multi-physics performance. With high-fidelity database development in full integration of various length scales and their interconnections, AI stands to play an important role to realize the full potential of design and optimization.

Here, we focus on thermal storage device design and integration due to the significant need to bridge fundamental materials-level PCM research with applications. Although device designs are application dependent, general design principles for improved thermal storage do exist. First, the charging or discharging rate for thermal energy storage or release should be maximized to enhance efficiency and avoid superheat. One of the challenges to enhance power density is the low thermal conductivity of organics (<1 W/(m · K)), salts (<10 W/(m · K)), or metal alloys (<30 W/(m · K)).⁵⁷ Enhancement by dispersing high thermal conductivity materials is limited by the volume fraction constraint, potential for component separation, and lack of full dispersal leading to randomly distributed structures.¹¹ One solution is to design devices with heterogeneous fin structures optimized for hotspots,⁵⁸ heat exchangers,⁵⁹ or plane heat sources.^{60,61} A second solution is to integrate PCMs with heat-spreading devices such as heat pipes and vapor chambers.^{62,63} To maintain high energy density, computational methods such as genetic algorithms can be used to predict thermal performance and optimize device design. Complex boundary conditions, natural convection in the liquid PCM, and non-equilibrium heat transfer between different components that affect prediction accuracy need to be accounted for within the multi-physics simulations. Second, thermal storage device design should consider the specific architecture and integration challenges associated with the heat source as well as the heat rejection system. Some work has shown that it may be more efficient to buffer heat spikes by integrating a PCM heat sink in closer proximity to the heat source with respect to available heat transfer pathways.⁶⁴ The geometry of one or multiple heterogeneous heat sources and different operating conditions also influence the performance of the thermal storage device.^{13,65–67} Considering customized architecture integration, reduced-order simulation tools such as thermal resistance-capacitance circuits have been developed to enable rapid analysis and iterative design methods. Third, the device design should consider the manufacturability and cost associated with manufacturing, integration, and maintenance. In this context, additive manufacturing presents a promising method to unlock new geometries such as lattice structures that can reduce the weight and volume of thermal storage devices having high thermal performance.⁶⁸ Challenges and opportunities remain associated with the processing of high thermal conductivity materials such as metals, fabricating PCM devices having accurate microgeometries, and controlling surface roughness.

Emerging applications of PCM thermal storage

Thermal storage using PCMs has a wide range of applications, ranging from small-scale electronic devices (~1 mm), to medium-scale building energy thermal storage (~1 m), to large-scale concentrated solar power generation (~100 m). Figure 5 shows ranges for power and temperature for applications of PCM thermal storage,

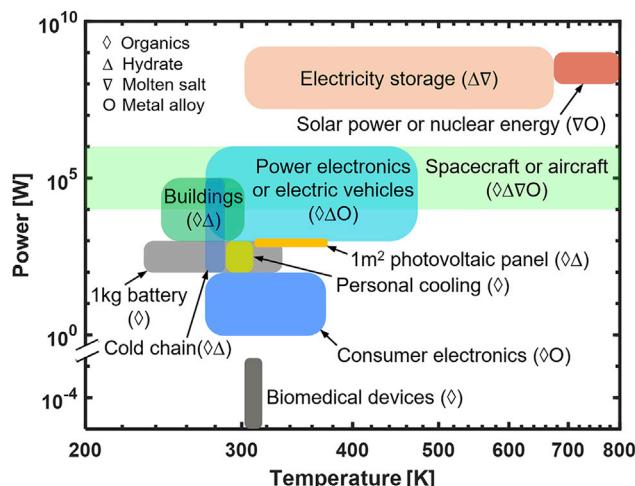


Figure 5. Emerging applications of PCM thermal storage

Power (electrical or thermal) as a function of temperature for applications in which PCM thermal storage is desired. For each application, appropriate PCMs are recommended based on melting temperature, cooling capacity, safety, and economic cost. To select a specific PCM for a real application, environmental cost and local policy need additional consideration. Applications of nuclear energy and aerospace craft can experience higher temperatures than 800 K, which are not shown for clarity.

along with the corresponding PCMs. High-temperature thermal storage has been widely investigated in power plants for load shifting, in which thermal storage allows for operation at a constant power level even as demand varies.^{69,70} Thermal storage is used to help regulate fluctuations in the electricity grid, either with sensible heat storage methods (rocks) or mechanical storage methods (compressed gases), and PCMs could be an alternative to these traditional methods.⁷¹ Spacecraft or aircraft can experience transient thermal shock during takeoff, which could be buffered by a PCM.⁷² The recent development of low-emission and lightweight electric aircraft having aggressive power dissipation profiles makes passive PCM solutions highly attractive. Vehicles for space exploration also require thermal buffering for protection from low-temperature limits, which is a key driver of failures in these systems.⁷³ At even larger length scales, cold chains equipped with PCM thermal storage could extend drug or food storage life and reduce logistics costs. PCMs integrated with building walls could provide energy savings by storing or releasing heat near the comfortable room temperature setting.^{74–76} Applying PCMs to photovoltaic (PV) panels helps keep PV cells cool and efficient by absorbing incident solar energy that is not converted to electricity.^{77,78} Personal cooling via the integration of PCMs with clothing textiles can reduce the risk of dehydration and fatigue by maintaining a comfortable body temperature.^{79,80} Transient cooling of power electronics, thermal buffering of electric vehicle fast charging, and temperature regulation for battery protection are applications of PCM thermal storage due to rapid proliferation of our electrified society.^{3,81–83} Applying a PCM layer between chips and heat pipes in consumer electronics can absorb excess heat and buffer heat spikes to improve performance.¹⁵ For biological applications, thermal storage using PCMs can protect biomedical devices from overheating or tissues from thermal injury during device failure.⁸⁴

In the context of application temperature range, low-temperature applications (<400 K) typically use organics, salt hydrates, and low-melting-temperature metal alloys. For medium temperatures (400–500 K), far fewer PCMs have been developed,

with only a few high-melting-point paraffin waxes, fatty acids, and hydrates. The low thermal conductivity of these materials requires good thermal conductors and smart device designs to handle power demands for these applications. For higher-temperature applications (>500 K), PCMs are almost entirely composed of molten salts and metal alloys, whose strong corrosivity requires careful container material selection and safety precautions.

While numerous theoretical and lab-scale studies have analyzed the feasibility of thermal storage within these applications, PCM devices are still not commonly used. We suggest that there are many systems-level issues that still must be addressed. For example, thermal storage using PCMs reduces temperature swings of electronics and improves long-term reliability. However, the added mass and volume of the PCM may reduce the specific and volumetric power density in some systems. For other potential applications such as ice storage in buildings, capital cost and payback period are much more sensitive criteria than power density. Tradeoffs between these various application-dependent requirements motivate the need for comprehensive systems engineering. Quantitative assessment methodologies based on techno-economic performance indicators present one approach to enable the comparative analysis of different thermal storage solutions. The techno-economic analysis for solar thermal power applications indicates that the energy consumption and maintenance of auxiliary storage equipment and the cost of PCM feedstock are the most important factors of the system capital cost.⁸⁵ In addition to cost, another obstacle is the long-term durability and performance of PCMs in real applications. For example, metal matrices are used to enhance the thermal conductivity of molten salts in power plants. However, after multiple cycles of high-temperature melting and solidification, metal matrices are corroded, reducing thermal performance significantly. For power electronics, organics are not used because of flammability concerns. To overcome this obstacle, material research for improved durability and advanced techniques to protect devices from PCM failures are desired to elevate the technology readiness level. Overall, technical issues remain and prevent the transition of lab-scale research to real applications, bringing numerous research opportunities as well.

Summary and outlook

PCM thermal storage is a flourishing research field and offers numerous opportunities to address the challenges of electrification and renewable energy. PCMs have extensive application potential, including the passive thermal management of electronics, battery protection, short- and long-term energy storage, and energy conversion. In this work, we presented a comprehensive overview of PCM thermal storage at the multi-physics fundamental level, materials level, device level, and systems level. Challenges and opportunities exist for researchers to develop PCM thermal storage techniques that are both more energy dense and more efficient. Communities of physicists, thermal scientists, mechanical engineers, chemical engineers, materials engineers, electrical engineers, and computational scientists, can make contributions to better understanding the multi-physics mechanisms governing phase change, exploring novel high-FOM materials, developing high-performance devices, and reducing integration and systems cost. We encourage potential research directions in the future as follows:

- (1) Thermal storage requires the selection of PCMs having an optimal melting point, latent heat, and thermal conductivity based on the temperature range and heat-generation characteristics of the application. Metal alloys have received very limited attention within the PCM community when compared

with organics and salts due to the emphasis on specific latent heat. The high volumetric latent heat and thermal conductivity of metal alloys has the potential to develop much higher energy density at the systems level. The need for medium-temperature PCMs (350–500 K) presents an outstanding opportunity to develop safe and nontoxic metal alloys with melting points <500 K.

- (2) Multi-scale computational and modeling approaches should be developed that can enable materials development, multi-physics simulation, device design, and system simulation. Integrated computational materials engineering (ICME) methods can reduce cost and time to develop new materials. Accurately modeling the relationship between material structures and properties under static or dynamic conditions remains to be achieved. Many opportunities exist to develop tools that can predict material properties by analyzing internal structures and tailor materials to specific industrial applications. Although multi-physics simulation and device design tools based on FEM and FVM do exist, these tools consume large computational resources and must be calibrated with experiments. ML and AI integrated with physics-based principles have the potential for rapid performance prediction and design optimization.
- (3) To enable high-fidelity thermal storage component development, the consolidation of PCM datasets is necessary. The current status quo places little emphasis on data collection or cataloging, with PCMs data dispersed over the literature with different levels of detail. In fact, liquid-phase thermal conductivity measurements are lacking for most PCMs, despite the fact that this parameter is the most important factor in cooling capacity due to its role in heat transfer from the heat source to the melting front. Significant variation exists in the measurement methodologies used to characterize PCM melting temperature, latent heat, and solid/liquid thermal conductivity. Standard measurement techniques should be established and used for property measurements such as differential scanning calorimetry (DSC) or temperature-history methods. The establishment of a central handbook or database of PCM thermodynamic and transport information presents an opportunity to provide correct and consistent data for all communities of interest.
- (4) Manufacturing is a vital step linking device design and product development. Geometry and material constraints from legacy manufacturing methods limit the design freedom and reduce potential product performance. Additive manufacturing coupled with TO can unlock new geometries and next-generation thermal storage products having novel structures. Current and future research needs to continue the utilization of additive manufacturing to enable rapid iteration design principles and to produce customized PCM devices optimized for specific applications.
- (5) Economic and environmental characteristics of thermal storage systems are important for the deployment of these technologies in real applications. For example, in large concentrated solar power plants, although many PCMs have been proposed, latent heat storage systems are not yet available due to high maintenance costs stemming from materials degradation, corrosivity, solid deposition on heat transfer surfaces (heat exchangers), or volumetric expansion during phase change. The quantification of system-level costs and benefits using thermo-economic analysis has the potential to promote PCM thermal storage techniques to a variety of broad applications. Moreover, the investigation of energy and environment policy in a country or region has the potential to avoid risks or to cater to local thermal storage development.
- (6) PCM recycling has not yet been considered either in the cyclic utilization of devices or in recovering high-value materials. Opportunities and challenges will arise as the proliferation of PCMs increases. Analogous to lithium-ion

battery technologies, PCM devices can be characterized by a state of health (SoH) determined by several parameters such as cooling capacity, total latent heat charged and discharged, thermal resistance, phase separation, and melting temperature. Considering the economic cost and environmental burden, one PCM device retired from a critical application after a critical number of cycles can be reused for another application having lower SoH requirements. The associated challenges are identifying the SoH of PCM devices and sorting them for appropriate applications automatically. Research on developing processes to optimize productivity, yield, and cost of advanced materials recovery is desired to make PCM recycling profitable.

- (7) Systems-level thermal control strategies using PCM thermal storage should consider more realistic heat inputs. The majority of prior work on PCM thermal storage focused on canonical thermal loads (step functions, constant ramp functions, steady heating). However, heat generation in real systems is complex, often coupled with other forms of energy or power as well as additional components that add to the thermal inertia of the problem. For example, combined heat and power (CHP) systems for recovering and using waste heat can synchronously generate electricity and heat.⁸⁶ To regulate the heat load from the CHP system, a dynamic thermal storage strategy is desired to enable an enhancement by considering the transient waste heat and dynamic electricity generation. Systems-level PCM thermal storage with dynamic control and integration with complex heat rejection systems remains a promising opportunity for multidisciplinary research.

SUPPLEMENTAL INFORMATION

Supplemental information can be found online at <https://doi.org/10.1016/j.xcsp.2021.100540>.

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AUTHOR CONTRIBUTIONS

T.Y., W.P.K., and N.M. contributed equally to the conception, writing, and revision of this perspective.

DECLARATION OF INTERESTS

The authors declare no competing interests.

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