

Review

Nano-engineered pathways for advanced thermal energy storage systems

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SUMMARY

Nearly half of the global energy consumption goes toward the heating and cooling of buildings and processes. This quantity could be considerably reduced through the addition of advanced thermal energy storage systems. One emerging pathway for thermal energy storage is through nano-engineered phase change materials, which have very high energy densities and enable several degrees of design freedom in selecting their composition and morphology. Although the literature has indicated that these advanced materials provide a clear thermodynamic boost for thermal energy storage, they are subject to much more complex multiscale governing phenomena (e.g., non-uniform temperatures across the medium). This review highlights the most promising configurations that have been proposed for improved heat transfer along with the critical future needs in this field. We conclude that significant effort is still required to move up the technological readiness scale and to create commercially viable novel nano-engineered phase change systems.

INTRODUCTION

Most people are familiar with phase transition processes from day-to-day experiences with boiling of water and freezing/melting of ice. From these examples, it can be understood that thermal energy and phase transitions are inextricably linked. Although the term “phase transition” refers to the full change of matter from one phase to another, the process occurs over time and involves statistical physics.

In a phase change process, physical matter goes from one state to another by adding or removing heat. When matter transitions from one phase to another through heat addition, the process is referred to as a high-energy state-phase transition. When phase transitions occur through removal of heat, they are called low-energy phase transitions. Figure 1 illustrates examples of the classic phase transitions of water. As shown in Figure 1, the energy level of matter increases when it absorbs heat.

During a phase change process, thermodynamic variables, such as pressure, temperature, and enthalpy, play a crucial role in determining the specifics of the process. In the case of a solid, the intermolecular forces between molecules are greater compared with those in a fluid, which, in turn, are greater than in a gas. Thus, the phase transition process can be viewed as the summative change in the intermolecular forces of the medium.¹

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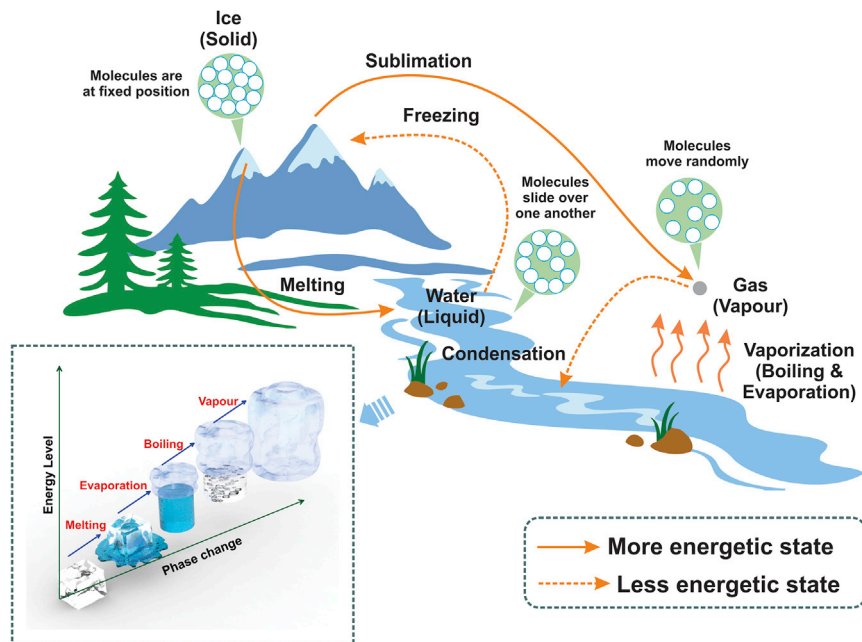


Figure 1. Schematic of phase transitions of water

Shown is a representation of phase transitions of water, depicting the molecule spacing for different energetic states.

Many engineering processes and natural phenomena involve phase transitions, such as freezing (solidification), melting, boiling, evaporation, condensation, deposition, and sublimation. For applications where thermal storage or thermal energy transfer (i.e., under specified conditions) is required, it is possible to utilize the solid-to-liquid phase transition because the material's density is similar in both phases. It is possible to select materials with a high latent heat of phase change (i.e., a physical property measured in J per kg) for relevant applications in solar-thermal systems, building heat and cooling systems, industrial processes, micro-electronics, and battery thermal management.^{2–7}

One potential pathway toward enhancing/controlling the underlying thermophysical properties and/or the mechanisms involved in phase transitions is through addition of nanoparticles. The term “nanoparticles” refers to particles with at least one dimension ranging from 1–100 nm.⁸ Nanoparticle synthesis methods have matured over the last few decades. They can now be designed from any material and can even have heterogeneous structures; e.g., including a surface layer (to functionalize the nanoparticle), a shell layer, and a core.⁹ Nanoparticles can also be synthesized in any imaginable shape, such as spheres, discs, platelets, rods, tubes, cubes, stars, and many others. As expected, the properties of such nanoparticles (physical and chemical) vary in response to their morphology (shape, size, and structure).¹⁰ Decreasing the size of nanoparticles increases their specific surface area and, thus, their surface energy. Their small size makes them suitable for adding to liquids and surfaces, thereby elevating a conventional material to a nano-engineered material to suit specific applications. For example, by adding nanoparticles to a base fluid (e.g., water), a large body of research has shown that it is possible to improve the effective thermophysical and optical properties of the fluid and, consequently, the efficiency of nanofluid-based energy systems.^{11,12}

Most industrial processes involve use of thermal energy. If thermal energy can be added or extracted from a process at faster rates, then most industrial processes will reap benefits in terms of saved time and productivity. If thermal processes can be made more efficient, then it is also possible to save energy and increase the working life of equipment. Numerous studies have been conducted to understand and develop methods to improve the rate of heat transfer in such systems. Several of these studies have reported that addition of nanoparticles to the base fluid can enhance heat transfer efficiency. However, an in-depth understanding of strategic niches in phase change heat transfer using nanoparticles is still needed if we hope to improve heating, cooling, or energy recovery from a diverse range of industrial processes.

Motivated by the complex physics of phase change processes and the potential for enhancing systems employing these processes with use of nanoparticles, this perspective provides a qualitative discussion of strategies for optimizing the performance of nano-engineered materials for phase change heat transfer and energy storage applications.

STRATEGIC NICHES FOR NANOPARTICLE-ENHANCED PHASE CHANGE HEAT TRANSFER

Nanoparticle-enhanced boiling heat transfer characteristics

Boiling occurs when the liquid temperature is increased to the saturation temperature at a given pressure. The boiling process is similar to evaporation, but there is a clear distinction between the two processes. Evaporation occurs when the vapor pressure is lower than the liquid's saturation pressure at the liquid-vapor interface. On the other hand, boiling occurs at the solid-liquid interface when the liquid makes contact with the surface having a temperature above the saturation temperature of the liquid.^{13,14} The boiling process does not occur under equilibrium conditions because the vapor temperature and pressure in the bubbles are different from those of the liquid bulk.^{15,16} During this process, surface tension at the liquid-vapor interface acts to counterbalance the pressure difference, and the difference in temperature between vapor bubbles and the surrounding liquid acts as a driving factor for boiling heat transfer. Numerous studies have reported the effects of adding nanoparticles under various operating conditions and boiling types on heat transfer (Figure 2).

The boiling heat transfer coefficient increases with increasing nanoparticle volume concentration up to the optimum level and then decreases.²⁴ This reflects the fact that there is an optimum mass concentration that corresponds to the highest heat transfer enhancement. However, heat transfer can also be directly proportional to the concentration of nanoparticles at each mass flux (especially in the case of flow boiling).²⁵ Formation of a molecular adsorption layer and disturbance of nanoparticles can result in a decrease in boundary layer height, which can lead to an increase in boiling heat transfer.²⁵ The higher the particle concentration, the lower the effect of pressure on the boiling heat transfer coefficient.²⁶ With the increase in concentration, the rate of boiling decreases significantly as the nanoparticles become adsorbed onto the surface, decreasing its roughness because of the reduction in the number of nucleation sites.^{17,27,28}

Surface tension has a dominant effect on boiling heat transfer.²⁹ The presence of surfactant in a nanofluid typically decreases the boiling performance by increasing its dynamic viscosity. When a surfactant is added above the optimum quantity to a nanofluid, a drastic decline in boiling heat transfer occurs.³⁰ For every surfactant

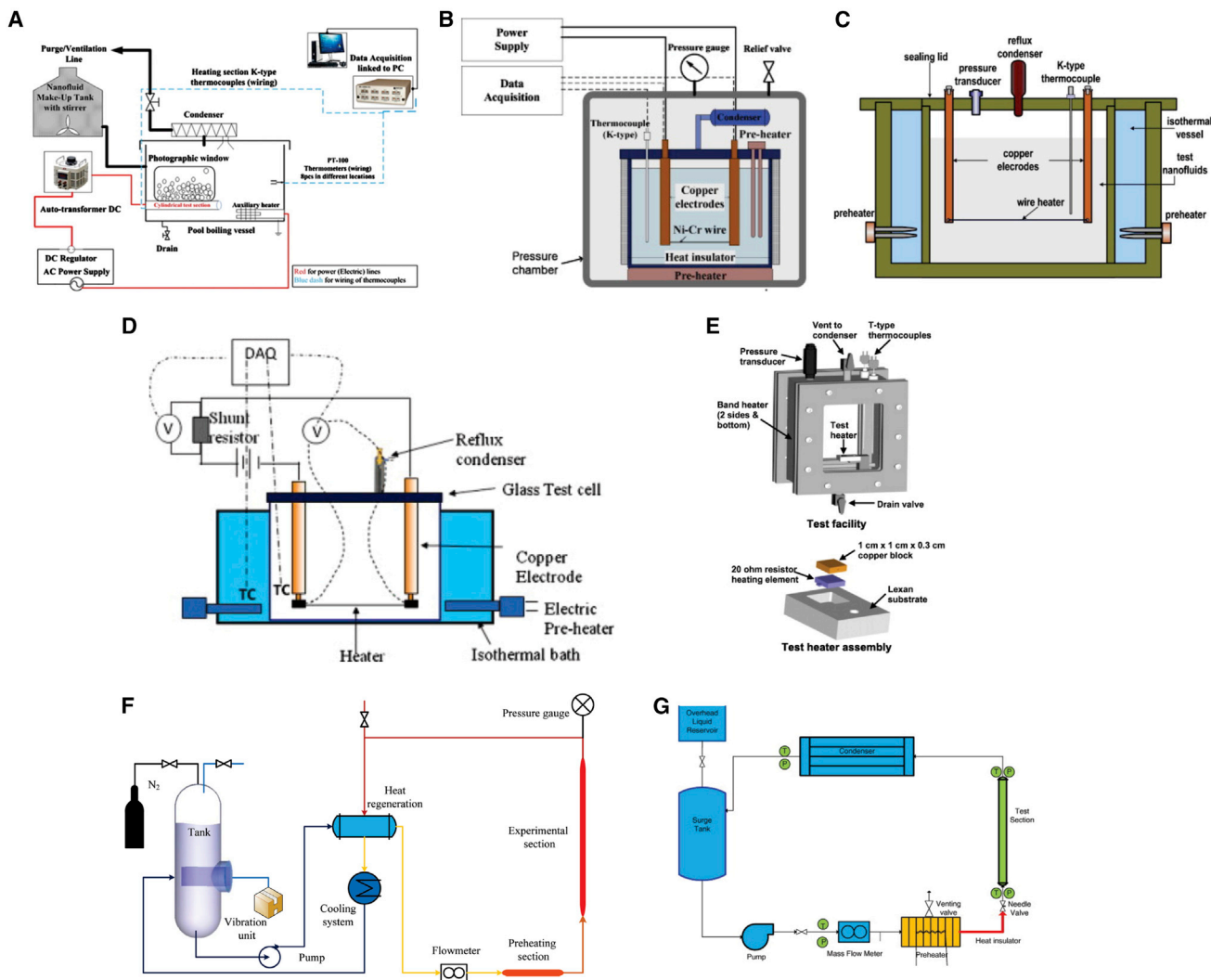


Figure 2. Schematics of the experimental setup developed for nano-enhanced boiling heat transfer

- (A) nucleate pool boiling setup. Reproduced with permission.¹⁷ Copyright Elsevier, 2014.
 (B) Pool boiling apparatus. Reproduced with permission.¹⁸ Copyright Elsevier, 2013.
 (C) Resistance heating wire pool boiling setup. Reproduced with permission.¹⁹ Copyright Elsevier, 2007.
 (D) Pool boiling critical heat flux facility. Reproduced with permission.²⁰ Copyright Elsevier, 2010.
 (E) Pool boiling facility and heater assembly. Reproduced with permission.²¹ Copyright Elsevier, 2010.
 (F) Flow boiling setup. Reproduced with permission.²² Copyright Elsevier, 2020.
 (G) Flow boiling critical heat flux experimental loop. Reproduced with permission.²³ Copyright Elsevier, 2011.

used, an increase of surfactant concentration increases the surfactant enhancement ratio and then decreases as the concentration exceeds a threshold value.³¹

In the case of smooth surfaces, the particles deposited on the surface increase the surface roughness, resulting in an increase in nucleate boiling heat transfer. In the case of rough surfaces, there is not much change in surface roughness, resulting in little or, in some cases, no change in boiling.³² The porosity of the surface also affects the size of the bubbles formed.³³ Deposition of nanoparticles can also increase the heat transfer coefficient of smooth surfaces by increasing surface roughness.^{34,35} These deposited nanoparticles decrease the contact angle, resulting in an increase in surface wettability, which, in turn, leads to an increase in boiling crisis or critical

heat flux values. At higher heat fluxes, the bubble coalescence phenomenon increases, which, in turn, increases the critical heat flux as well.³⁶ In cases where agglomeration does not occur, it may even lead to deposition of particles on the heating surface and critical heat flux enhancement.³⁷ As the nanoparticles continuously deposit on the surface, a change in static surface contact angle occurs, which results in an increase in critical heat flux in nanofluids.³⁸

Flow velocity and changes in surface wettability because of the layer formed by nanoparticles deposited on the surface affect the critical heat flux enhancement in nanofluids.³⁹ Increasing the nanoparticle wettability increases the solid/liquid interaction, which increases the heat transfer; by employing varied wettability nanoparticles, it is possible to increase the heat transfer of explosive boiling.⁴⁰ The wettability of nanoparticles affects the structure of the layer of nanoparticles deposited on the heater surface, and it plays an important role in vapor bubble coalescence in the case of pool boiling. Nanofluids that contain moderately hydrophilic nanoparticles are most effective in increasing boiling heat transfer.⁴¹ Nanofluid containing moderately hydrophilic nanoparticles increases the bubble coalescence time because of adsorption of nanoparticles at vapor bubble interfaces, which, in turn, results in smaller bubble size.⁴² In pool boiling and flow boiling, the enhancement mechanisms of heat transfer are the higher frequency of bubble nucleation and the improved surface wettability.⁴³

Brownian motion is enhanced at higher bulk nanoparticle concentrations or for smaller nanoparticles sizes. For smaller nanoparticles, the thermophoresis strength decreases, so that the particles can deposit on the heated wall and increase the rate of heat transfer. However, nanoparticles are not equally distributed in the film because of thermophoresis; rather, the lowest concentration is at the heated wall, and the highest concentration of nanoparticles is at the vapor-liquid interface.⁴⁴

The following points can be made from the literature in relation to boiling:

- Nanoparticle concentration has a remarkable influence on the boiling heat transfer coefficient and critical heat flux.
- The larger the nanoparticle size, the better the boiling heat transfer performance. Conversely, smaller nanoparticle sizes can enhance the significance of nanoparticle Brownian motion-induced heat removal.
- Surface tension predominantly acts to lower the heat transfer coefficient of nanoparticle dispersed fluids. Thus, addition of a surfactant above an optimal level causes a drastic deterioration in nucleate boiling heat transfer.
- Deposition of particles influences surface roughness; thus, the boiling heat transfer and the heat transfer coefficient are typically reduced.
- The thermophoresis strength decreases for smaller nanoparticles, which results in increased deposition on the heated wall and an increase the rate of heat transfer.

Nanoparticle-enhanced evaporation of liquid droplets

In the previous section, the basic differences between boiling and evaporation were discussed. The evaporation process is a liquid-to-vapor phase change process. Unlike boiling, bubble formation and bubble motion are not seen in the evaporation process.¹⁴ By using the latent energy of its molecules, the evaporating droplet loses heat until it changes into a vapor phase. Instead of using water as a fluid for heat removal, use of advanced thermal fluids, such as nanoparticle dispersed fluids, can help remove heat at higher rates. Schematics of the experimental setup and

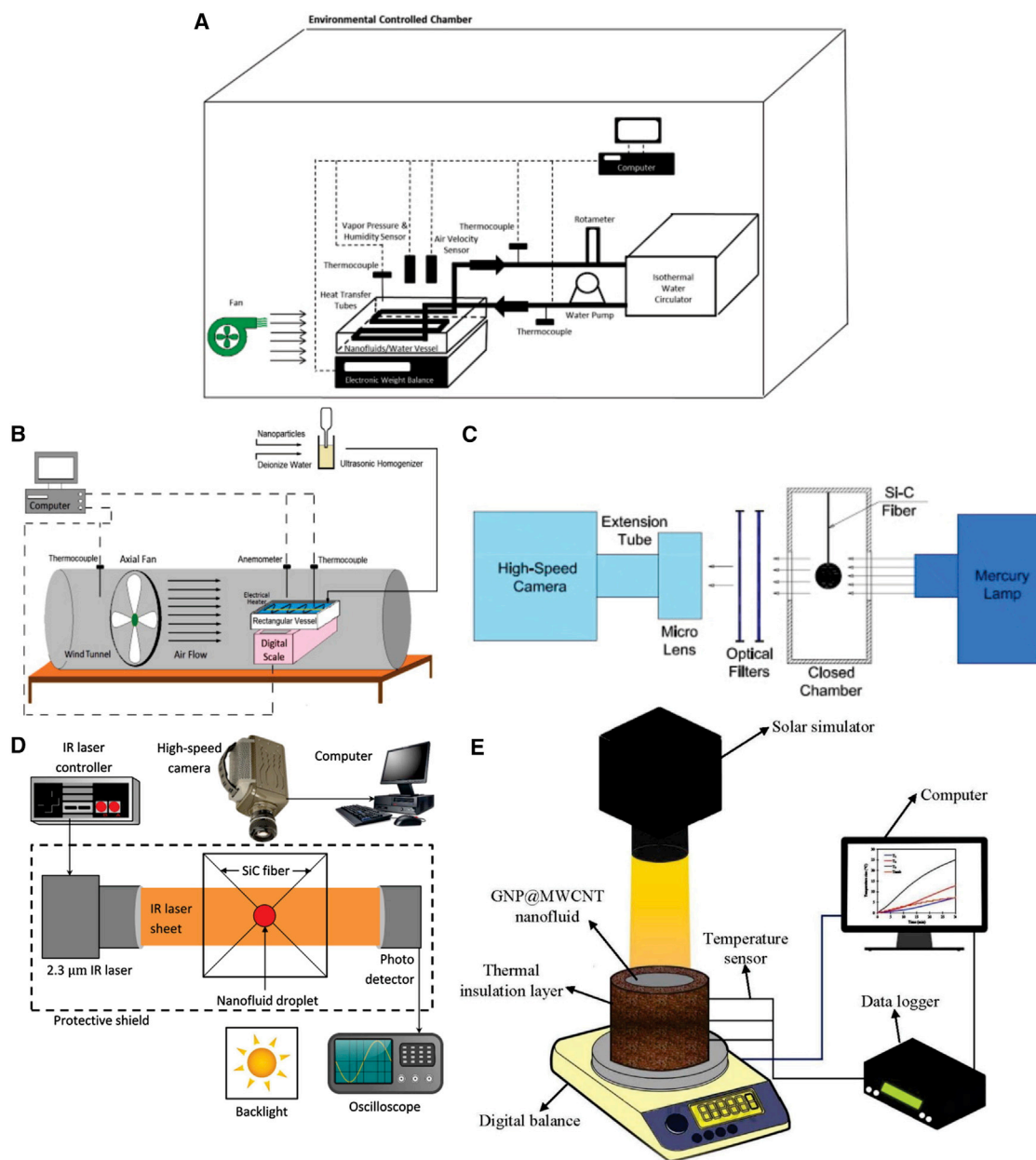


Figure 3. Schematics of the experimental setup for the nano-enhanced evaporation process

(A) Setup for estimating the evaporation rate of aqueous nanofluids. Reproduced with permission.⁴⁹ Copyright Elsevier, 2015.

(B) Apparatus for nanofluid evaporation measurements. Reproduced with permission.⁵⁰ Copyright Elsevier, 2013.

(C) Setup for nanofluid droplet evaporation under radiation. Reproduced with permission.⁵¹ Copyright Elsevier, 2012.

(D) Setup to study the effects of near-infrared (IR) radiation on the evaporation characteristics. Reproduced with permission.⁵² Copyright Elsevier, 2017.

(E) Setup for solar steam generation using nanofluids. Reproduced with permission.⁵³ Copyright Elsevier, 2020.

special techniques to study the evaporation of nanofluids are depicted in Figure 3. Numerous variables influence the rate of evaporation.⁴⁵ For instance, surface tension, Marangoni forces, wettability, pinning/depinning effects, and nanoparticle type, size, and concentration all affect the evaporation dynamics of a sessile droplet.^{45–47} Suspension of nanomaterials in the base fluid will not improve sessile droplet evaporation when there is thermal equilibrium of phases.⁴⁸

With an increase in nanoparticle concentration, most nanofluids exhibit a lower saturated vapor pressure and lower evaporation rate than pure water, with a few exceptions for nanofluids with low particle concentrations that show values of saturated vapor pressure and an evaporation rate slightly greater than that of water.⁴⁹ In free convection turbulent evaporation, hidden complex cavities appear that are dependent on self-assembly parameters (e.g., surface hydrophobicity).⁵⁴ For problems involving radiant heat transfer over a liquid-vapor interface, cases with high nanoparticle concentration and/or high incident solar radiation result in trapping of energy in a thin layer of nanofluid near the interface, resulting in higher interface temperatures and higher evaporation rates.⁵⁵ A direct relation exists between temperature and variation in concentration of nanoparticles. The increase in nanoparticle concentration increases the temperature.⁵⁶ Droplets of higher particle concentration have greater surface tension, and so they require a greater amount of heat for vaporization; therefore, disintegration is adversely affected because of a delay in the vaporization process caused by an increase in nanoparticle concentration.⁵⁷

Variations in the nanoparticle size greatly influence the enthalpy of evaporation of nanofluids. Liquid-dominant evaporation is influenced by nanoparticle size and occurs in a fashion similar to that of pure water with little effect on the total heat and mass transfer by the suspended nanoparticles. The dry-out process describes the later stages of evaporation, when the nanoparticle effect on water is decreased. Discontinuous spikes of temperature and heat flux because of the high thermal conductivity of the nanoparticles quickly improve the dry heater condition, with the formation of nanoparticle stain time being dependent on the size of the nanoparticle.⁵⁸

Drag influences the motion of a single particle suspended in liquid in the early stages of evaporation. Most particles lie on the substrate at the occurrence of the contact line recession and partially immerse themselves in the liquid layer, and the surface tension force gains direct control of the movement of the single particle.⁵⁹

When nanofluid droplets are over their corresponding wetted residues, the interfacial region and droplet evaporation rate can be increased.⁴⁵ In the case of lyophilic surfaces, with the increase in nanoparticle energy coefficient (which represents nanoparticle wettability; i.e., the higher the nanoparticle energy coefficient, the better the nanoparticle wettability), the contact angle of sessile nanofluid nanodroplets increases, and, therefore, total evaporation decreases. Thus, surface wettability relates the effects on the wetting and evaporation behaviors of sessile nanofluid nanodroplets. The greater the nanoparticle volume percentage, the greater the contact angle and, therefore, the lesser the rate of evaporation for a lyophilic surface. In the case of neutral and lyophobic surfaces, a rise in concentration decreases evaporation.⁶⁰

Stresses because of the Marangoni effect (surface tension gradient on a liquid) play a significant role in sustaining internal circulation as the temperature of the superheated vapor increases. The Marangoni effect influences nanoparticle stability.⁶¹ Convection dominates particle distribution and exhibits twin peaks with minimum concentration at the non-dimensional location from the center. This phenomenon can be observed because of strong internal circulation.⁶² Evaporation-induced pore water flow has been shown to contribute to deposition of suspended nanoparticles even in cases where interaction of nanomaterial on the surface is not beneficial.⁶³

Sonication (i.e., the process of applying sound energy to agitate particles) can have a significant effect on the evaporation rate. Considering the effects of sonication in

free convection with an increase in the concentration of a nanofluid, the surface temperature of the nanofluid increases, whereas changes in the temperature of the bulk fluid are negligible.⁵³

The following points can be made about nanoparticle-enhanced evaporation:

- Increasing the nanoparticle concentration increases the evaporation rate.
- The nanoparticle size decides the average breakup droplet diameter.
- A higher initial concentration of nanoparticles leads to larger residual deposits, and accumulation of particles on the surface of the droplet minimize the efficient area ratio and inhibit the evaporation rate.
- A temperature rise can occur because of absorption of radiation by the nanoparticles, which enables the evaporation rate to increase.
- Microphysical mechanisms are critical for improving the evaporation rate and mitigating stability issues.

Nanoparticle-enhanced condensation heat transfer

Condensation occurs when the vapor temperature is brought below the saturation temperature. This is typically accomplished by bringing a vapor into contact with a solid surface whose temperature is below the vapor's saturation temperature. To understand the effects of nanoparticles/nanofluids on heat and mass transfer enhancement during condensation, a better understanding of nanomaterials on the condensation phase transition process is needed. A schematic representation of condensation heat transfer in nano-enhanced refrigeration cycles is shown in Figure 4.

The type of nanoparticle used plays a major role in phase transition condensation phenomena.⁶⁹ The thermophysical characteristics of dilute gas-particle flows are key factors affecting condensation heat transfer. Various factors, such as nanoparticle disturbance and reduction in the thickness of boundary layers or of Brownian motion effects can improve condensation heat transfer.⁶⁵ Superheat is also a primary factor affecting condensation heat transfer. An increase in nanoparticle dosage increases the heat transfer coefficient and superheat temperature, but at low mass flux, the increase in condensation heat transfer is greater.⁶⁷ By dispersion of nanoparticles, the condensation of the humid air increases, and the effect of nanoparticle mass concentration in cases with higher relative humidity can be even more crucial.⁶⁹ An interlaced wettability pattern can alter the nanoparticle surface and lead to better condensation heat transfer performance, particularly in situations without support of the droplet-sweeping effect by the gravity force.⁷⁰

Thermophoretic diffusion enhances heat transfer, whereas Brownian diffusion deteriorates heat transfer.⁶⁸ Migration of nanoparticles toward a cold wall intensifies the film's local viscosity and thermal conductivity. As the Brownian-to-thermophoretic diffusivity ratio decreases, nanoparticle migration is substantially improved. Overall, smaller nanoparticles make the distribution of nanoparticles more uniform and increase the condensation heat transfer rate.⁷¹

The following concluding remarks can be made about nanoparticle-engineered condensation mechanisms:

- Adding nanoparticles can improve the condensation heat transfer coefficient, whereas the optimum nanoparticle concentration is highly dependent on the mass flux of the fluid.

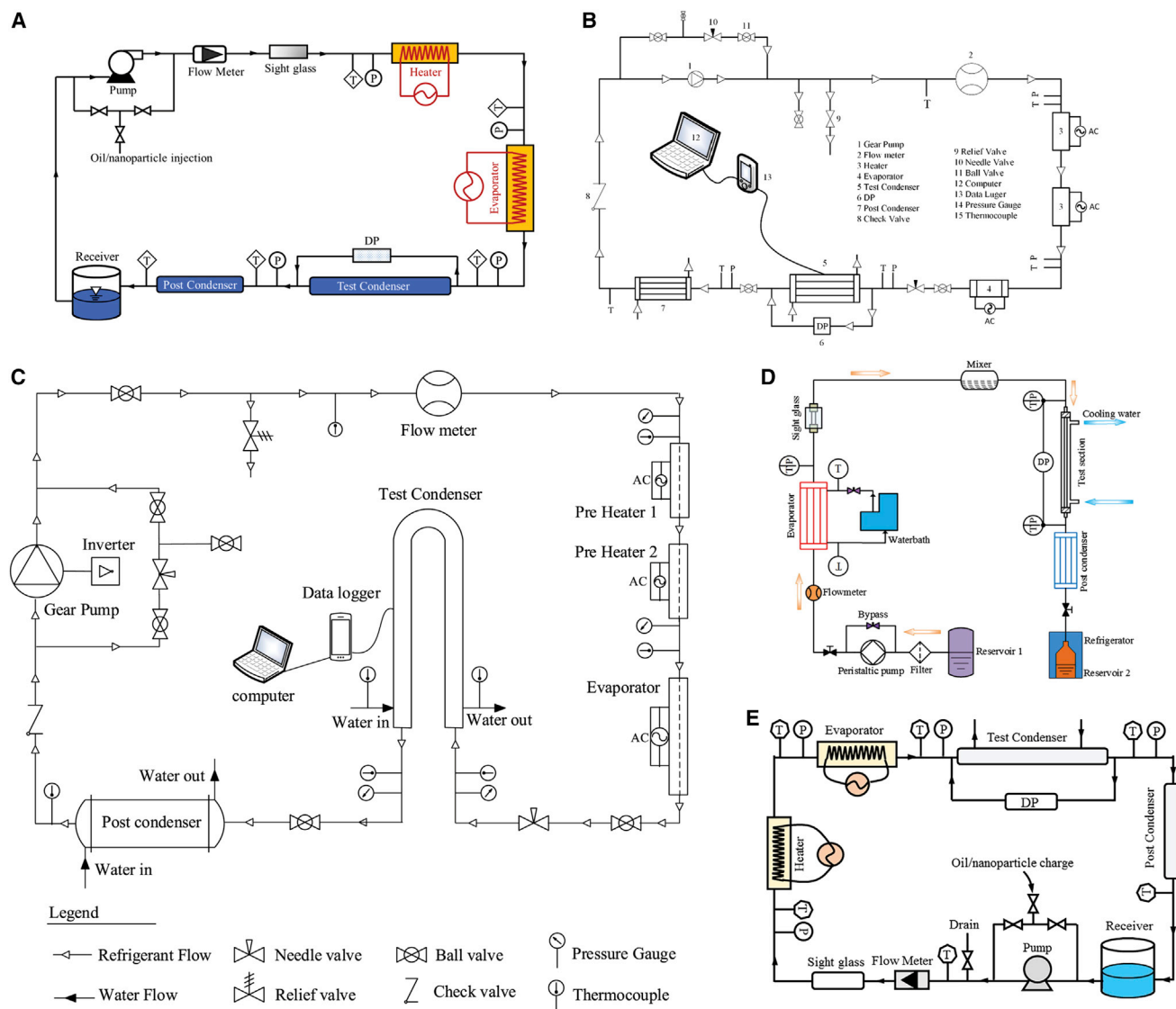


Figure 4. Schematics of the experimental setup developed to study condensation heat transfer in nano-enhanced refrigeration cycles

- (A) Counterflow pipe setup. Reproduced with permission.⁶⁴ Copyright Elsevier, 2020.
 (B) Round and flattened tube setup. Reproduced with permission.⁶⁵ Copyright Elsevier, 2017.
 (C) Horizontal U-shaped tube setup. Reproduced with permission.⁶⁶ Copyright Elsevier, 2015.
 (D) Vertical circular tube setup. Reproduced with permission.⁶⁷ Copyright Elsevier, 2018.
 (E) Shell and tube counterflow heat exchanger setup. Reproduced with permission.⁶⁸ Copyright Elsevier, 2018.

- The diameter of the nanoparticle has a significant effect on the thermal conductivity of a nanofluid and, thus, the condensation rate.
- Microphysical mechanisms have a significant influence on movement of the nanoparticles and, thus, the condensation heat transfer rate.

STRATEGIC NICHES FOR NANOPARTICLE ENHANCEMENT OF LATENT HEAT THERMAL STORAGE

A wide variety of thermal systems have been integrated with solid-to-liquid phase change materials (PCMs) to improve performance and operation. For example, PCMs have been integrated with heat pumps to provide continuous heating without

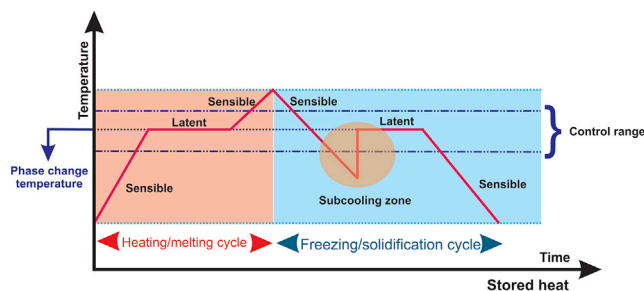


Figure 5. General representation of heating/melting and solidification/freezing phenomena in temperature cycles of the phase change process

drawing on the electrical grid during peak load periods.^{3,72,73} Investigators have also demonstrated that integration of PCMs with the ground heat exchangers of ground-source heat pumps can smooth the thermal shock to the ground as the operating conditions of the heat pump are varied, resulting in higher coefficient of performance (COP) values.⁷⁴ Use of PCMs has also been shown to be effective in building cooling applications, either in an entirely passive mode or in conjunction with air conditioning systems, depending on the ambient temperature fluctuation range.^{3,75} Energy storage of recovered waste heat using PCMs has been used by numerous investigators to improve the efficiency of various systems.^{76,77} In all of these examples, the underlying thermophysical properties and fundamental physical characteristics of the configuration (e.g., the location and number of nucleation sites) can significantly influence the overall performance of the PCMs in these applications.

In latent heat energy storage systems, a solid-liquid phase transition process can be nano-engineered to improve the latent heat of phase change or increase the heat transfer rate in either state.^{78,79} Material compatibility, thermal stability, and chemical stability of PCM usually determine its life span.⁸⁰ Particularly, it is desirable to assure the thermal stability of PCM for its long-term performance.⁸⁰ The heat conductivity of solid paraffin wax usually increases with the number of thermal cycles.⁸¹ The effects of heating and solidification phenomena on temperature cycles of phase change process are depicted in Figure 5. Adding nanoparticles to PCM reduces the fluctuations in melting temperature and latent heat of samples throughout multiple thermal cycles.⁸² Including nanoparticles raises the energy level of PCMs (Figure 6).

Nanoparticle-enhanced PCM solidification

In liquids, the freezing/solidification process occurs at a uniform temperature when the temperature is brought below the freezing point. This phenomenon results in formation of a layer of solid material at nucleation sites. In freezing, the heat that is released during the process is usually carried across the solid phase via thermal conduction, whereas convection heat transfer typically dominates in the liquid phase. Both heat transfer modes act to transport heat through the system and across the moving solid-liquid interface. In a situation where the temperature of the cold wall stays below the freezing point, the thickness of the frozen layer increases with time.^{83,84}

In latent heat thermal energy storage (LHTES) systems using PCMs, the heat flux through the material typically reduces when the solidification process starts because convective heat transfer is suppressed. The reduction in heat transfer rate during solidification can increase the duration of energy storage and, thus, reduce the system's efficiency. The reduction in the heat transfer rate during solidification can be

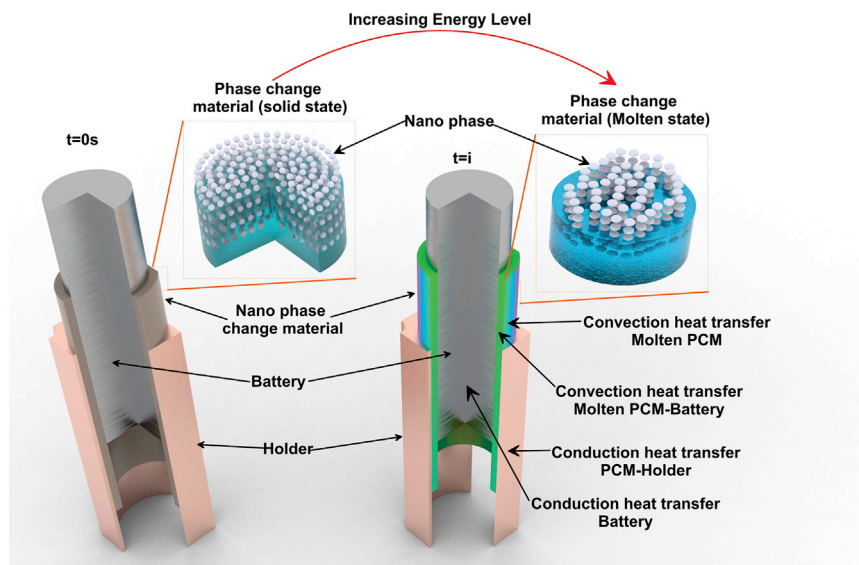


Figure 6. Nano-enhanced phase change energy for battery cooling, illustrating increasing energy levels

addressed using various heat transfer improvement methods such as addition of nanomaterials or using fins, heat pipes, or Lessing rings, among others.⁷⁸ Among different heat transfer improvement techniques, the most widely used method is addition of nanomaterials to PCMs to enhance the heat transfer rate and improve the performance of LHTES systems. Here we present a variety of important considerations when attempting to improve the energy storage density and rate of nano-particle-enhanced solidification processes.

Changes in the size, shape, structure, and concentration of nanoparticles can affect the material solidification rate. In particular, the size of nanoparticles appears to be inversely proportional to the solidification rate.⁸⁵ When the volume fraction of nano-fluid is increased, thermal conductivity can be improved, and the solidification rate can be accelerated.⁸⁵ Studies have observed that the presence of platelets leads to higher solidification rates (up to 14%),⁸⁶ amplification of the shape factor,⁸⁷ and increased thermal penetration depths.⁸⁸ When nanomaterials that are highly conductive are added to a less conductive fluid, they lead to an increase in thermal conductivity, promoting an increased solidification rate by 14% for a 0.5 wt % nano-particle-enhanced PCM.⁸⁹

Considering the presence of surfactants on the solidification process, mixtures with quantities of surfactant can reduce the surface tension of liquids.⁹⁰ The mass concentration of surfactant affects the supercooling degree and freezing time.⁹¹ The major undercooling comes from solute constitutional supercooling because of the additives, whereas the minor undercooling comes from particulate constitutional supercooling.⁹² Pertaining to the temperature effects in the liquid phase, loading of nanoparticles increases the thermal conductivity, whereas in the solid phase, thermal conductivity is linearly proportional to the temperature.⁹³ When the inlet temperature of the heat transfer fluid is decreased, there is more discharge of sensible heat from nano-enhanced PCMs (NePCMs).⁹⁴ The effect of dilution on freezing rate can lead to formation of a nanofluid layer, and the thickness of this layer increases as the solidification proceeds; the presence of this layer also increases the thermal resistance.⁹⁵

The effects of magnetic/electric fields on solidification can pave the way for heterogeneous nucleation.⁹⁶ The magnetic lift or squeezing conditions influence the time it takes for the droplet to freeze.⁹⁷ Inclusion of Lorentz forces (the combined force experienced by a point charge because of electric and magnetic fields) can also enhance the rate of flow or discharge, whereas nanoparticles decrease the discharge time.^{98,99} Although a magnetic field leads to quicker solidification in fluids that are non-electrically conductive as well as in electrically conductive ferrofluid, the ferrohydrodynamics and magnetohydrodynamics have a greater effect on electrically conductive ferrofluid, leading to shorter solidification time.¹⁰⁰

In addition to the above, microphysical mechanisms like Brownian motion can improve the solidification rate. The high thermal conductivity of the nanoparticles improves thermal conduction through the medium, while increasing the phase transition rate promotes convection.¹⁰¹ On the contrary, particle rejection from the mushy region in solidification and development of a particle-rich layer in the surrounding fluid are key physical phenomena that are uniquely associated with particles in solidification regions. Nanoparticles influence phase change through these important phenomena. According to a study,¹⁰² when particle rejection is taken into account, the solid-liquid interface is decelerated, and with a decrease in particle size, this deceleration becomes apparent. Another study¹⁰³ found that the frozen layer formed by dendritic growth includes regions of depleted concentration because of size-dependent rejection of nanoparticles. A force balance model can provide an indirect measure of the repelling mean thermomolecular pressure over a particle entering the front.¹⁰⁴ However, the presence of multiple particles can increase the thermomolecular pressure after the thickness of the thin liquid film that separates the particles and reduce the front.¹⁰⁴ These observations provide an important foundation for understanding many complicated solidification situations in engineering, where several objects communicate with the front and influence the solidification patterns.

Nanoparticles adsorb surfactants, resulting in an increase in the nucleation effect and in attraction of intermolecular forces between molecules (van der Waals forces), leading to formation of agglomerates of particles on the order of nanometers.¹⁰⁵ Thus, the emulsion volume fraction and size of the droplet should be considered carefully because the microstructure of the agglomerate depends on them. Likewise, natural convection currents¹⁰⁶ and use of micro-nano bubbles (minute bubbles of micrometer and nanoscale dimensions)¹⁰⁷ can effectively increase the rate of freezing of solutions. Natural convection affects the initial stages of solidification, whereas the last stages are controlled by conduction because of the absence of molten PCM. Micro-nano bubbles enhance the rate of nucleation as well as crystal growth. Thus, applying micro-nano bubbles significantly reduces the lagging time and phase change time.

The following points can be highlighted from the literature about NePCM solidification:

- Increasing nanoparticle concentration can accelerate the solidification rate because of improved thermal conductivity. However, using bigger nanoparticles can have the reverse effect on solidification.
- Addition of nanoparticles increases the thermal conductivity of the liquid phase, and the thermal conductivity is linearly proportional to the temperature in the solid phase.
- The surfactant mass concentration affects the supercooling degree and the freezing time for nano-dispersed fluids.

- When the nanoparticle diameter is large, settling is faster, so the supercooling degree and freezing time of larger particles dispersed in fluids are closer to those of deionized water.
- Applying Lorentz forces can promote the phase change solidification process.
- Microphysical mechanisms, such as Brownian motion, have a predominant effect on the solidification process and promote higher solidification rates.

NePCM melting

The physical melting process (or fusion) occurs when a solid substance becomes liquid. Typically, this occurs when the internal energy of a solid increases by absorption of heat, which raises the temperature of substances to their melting point.

Melting incongruent PCMs is a significant challenge when commercializing latent heat storage systems. The melting process cannot proceed as expected; therefore, the system consumes less energy at the desired rate.⁸⁰ To increase the melting rate, nanoparticles can be incorporated into the phase change process.⁷⁹ In the following section, we examine the effect of NePCMs on melting rates under various operating conditions.

Pure PCMs melt faster when coated with nanoparticles dispersed throughout.¹⁰⁸ On the other hand, particle size is associated with critical thermodynamic properties of integral melting.¹⁰⁹ Nanomaterials have been shown to increase melting rates,^{110,111} but heat transfer into the melted region tends to degrade as the mass fraction increases.¹¹² Convection and conduction are reduced by incorporating nanomaterials into enclosures.¹¹³ During NePCM melting, some effects, such as non-monotonic deviation¹¹⁴ and rapid interface advancement in the shrinking solid regimen,¹¹⁵ can also be observed. Increasing the inclination angle of the PCM above the desired value in a heat exchanger along with adding nanoparticles decreases the melting rate.¹¹⁶

PCM melting can be facilitated by magnetic fields.¹¹⁷ The melting rate decreases with increasing Hartmann numbers (electromagnetic force/viscous force).¹⁰⁰ Dimensionless velocity profiles can be improved and temperature effects reduced when a melting parameter is increased in conjunction with a high Hartmann number and a high nanoparticle dosage.¹¹⁸ Because of their higher thermal conductivity,¹¹⁹ magnetic nanoparticles can be used in conjunction with or without a magnetic field to reduce thermal gradients and speed up the melting process.

The charging process of the heat storage unit can be improved by extending the external or internal surfaces of pipes with extended surfaces such as fins.¹²⁰ The longer the fin, the better the heat absorption and performance, and the thinner the fin, the better the melting rate. Addition of fins decreases the charging time. By increasing the emulsification of nanoparticles, the melt rate can be increased. Subcooling also negatively affects the melting rate.¹²¹

The following points can be made:

- The melting time is shortened by nanoparticle dispersion, and this effect increases with increasing nanoparticle concentration.
- As a result of nanoparticle addition, the melting process is accelerated. As the volume fraction of the nanoparticle increases, the melt fraction and energy storage capacity of the nanoparticle also increase.

- In terms of conduction heat transfer, an increase in viscosity will cancel out or even diminish the benefit of nanoparticle-enhanced melting processes.
- In the case of fins used to transfer heat during melting, the thinner the fins, the faster they melt.

It is recommended that nanoengineered materials be used primarily to recover waste energy despite the impending commercialization of thermal heat storage. Recovering and using waste heat can increase economic benefits, reduce greenhouse gas emissions, and conserve energy.¹²²

ROADMAP FOR NANO-ENGINEERED MATERIALS COMMERCIALIZATION

As discussed above, nano-enhanced phase change heat transfer processes depend on several criteria. Although use of nanofluids for the phase change process has gained attention in the last few years, there are several challenges associated with nanofluid preparation techniques and their properties. Biosynthesized nanoparticles are promising because they pose little environmental risks and are biocompatible. The green synthesis method is an environmentally friendly, simple, affordable, and repeatable development pathway for producing metallic nanoparticles. Regarding nanofluid preparation techniques, a cost-effective technique for large-scale production is currently needed. The consensus in the literature is that one-step techniques are superior to two-step techniques to produce nanofluids and that cost-effective routes are available for commercial nanofluid preparation. Regarding nanofluid properties, further investigations are still needed to develop higher-heat-capacity coolants and analyze nanofluids at higher temperatures (greater than 100°C) and under cryogenic conditions. Nanofluids for heat transfer applications have a critical challenge of fouling on the surface. The nanoparticles deposit on the surface and change the surface morphology and wettability. The morphology and mechanism of nanoparticles in phase change heat transfer processes need to be explored in depth.

Simple models have been developed to simplify the phase change problem. However, real-world experience with these simplified models is limited. Robust numerical models need to be developed, with advanced numerical simulations to extend the application domains of nanoparticle-phase change heat transfer processes. Nanofluids for waste heat recovery have been widely implemented in heat-to-heat waste heat recovery applications. On the other hand, in the heat-to-work waste heat recovery strategy, the applications are limited and are mostly utilized indirectly to improve heat transfer from/to the major waste heat source/sink, as in the case of the organic Rankine cycle.¹²³ Heat-to-work is a very promising waste heat recovery technique because it can convert heat into electrical energy using a pyroelectric generator. However, currently this technique has a low energy conversion efficiency of less than 5% and is not cost efficient.¹²³ The pyroelectric generator unit may be developed to better use external vibration to achieve waste heat recovery without an auxiliary device, which has tremendous potential and application value in the field of waste heat recovery.¹²⁴

There is also a pressing need for advanced waste energy recovery for CO₂ emission reduction. Figure 7 shows the top 10 countries and funding sponsors in “waste heat recovery” research according to the Scopus database (as of January 22, 2022). It can be inferred that funding organizations in China significantly support waste heat recovery research. The most documents with funding sponsors are from the National

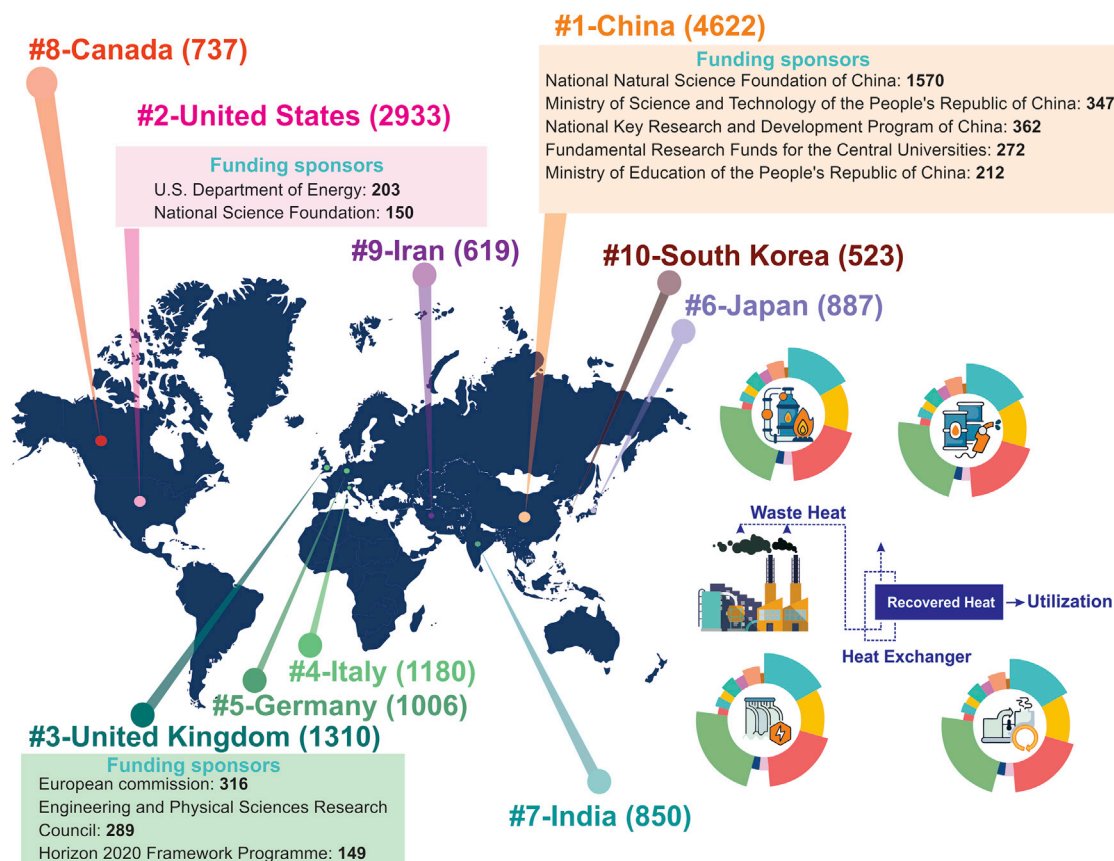


Figure 7. Top 10 countries and funding sponsors in waste heat recovery research

Natural Science Foundation of China (>1,550 documents). Five of the top 10 funding organizations are from China.

The conflicting effects of post-coronavirus disease 2019 (COVID-19) investments in fossil fuel-based infrastructure and the recent strengthening of climate target levels can potentially be tackled by deployment of technologies that re-use waste heat and other sustainable policies to still achieve a net drop in global emissions.^{125,126} For example, it is estimated that about 40 TWh of energy can be saved annually through waste heat recovery in United Kingdom industries.¹²⁷ By saving such a large amount of energy, about 10 million metric tons per year of CO₂ emissions would be reduced.¹²⁸ Therefore, developing advanced waste energy recovery systems must be prioritized, which will be helpful for reducing energy consumption and CO₂ emissions.

For technology like micro-electronics, thermal management systems, global power systems, additive manufacturing, and implementation of Industry 4.0 concepts, any nano-enhanced materials will need to be carefully considered in the context of the specific needs of the system. We suggest that the future scope for development can be divided into five categories: nanoparticle synthesis, nano-dispersion preparation methods, nano-enhanced property evaluation, modeling, and technology, as depicted in Figure 8.

SUMMARY AND OUTLOOK

From a review of the literature, it emerges that the introduction of nanoengineered materials holds great promise in terms of enhancing the performance of a wide

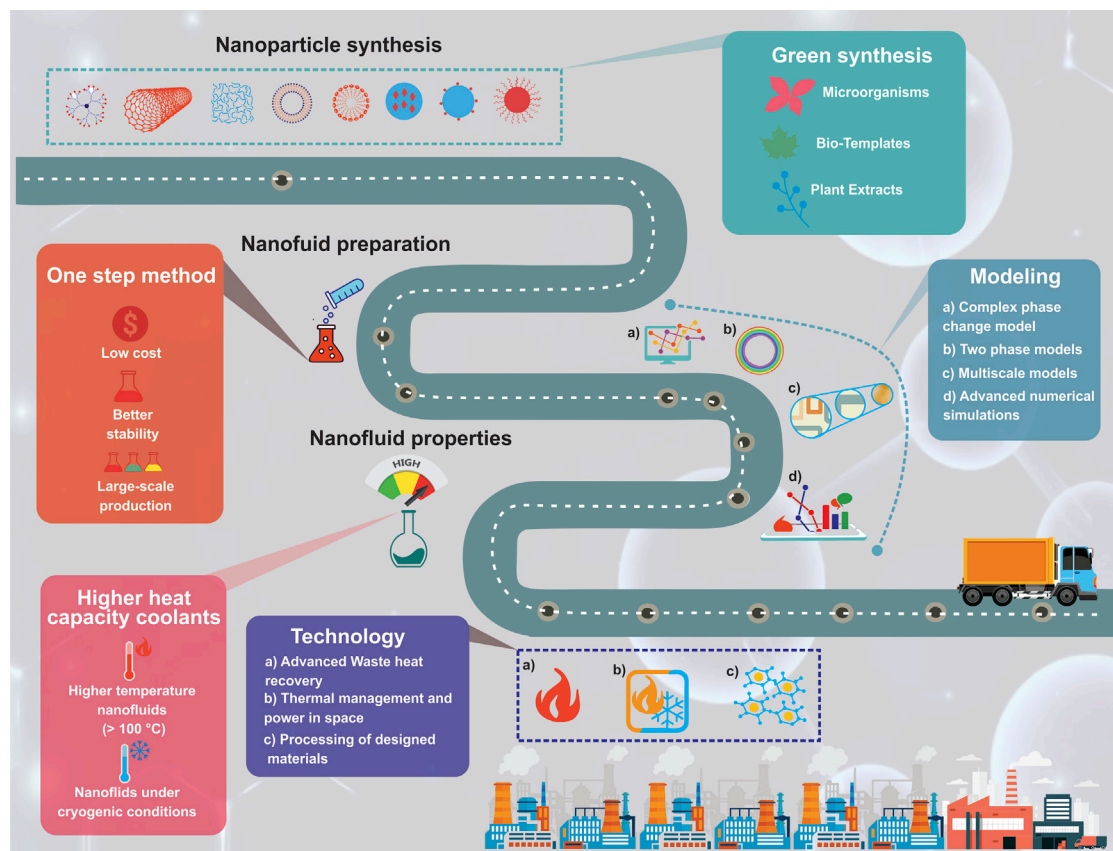


Figure 8. Road-map for nano-engineered heat transfer and energy storage systems

Shown is a roadmap depicting five categories: nanoparticle synthesis, nano-dispersion preparation methods, nano-enhanced property evaluation, modeling, and technology. Recently, there has been interest in using nanoparticles to control the speed of phase transition processes. This review presents different strategies for optimizing the performance of nano-engineered materials for phase change heat transfer and energy storage applications. A roadmap for potential research needs in nano-engineered materials commercialization is also outlined.

variety of thermal systems. However, further research is needed in this area to understand the key physical phenomena that govern such improvements to the performance of conventional heat transfer systems.

In thermal systems where boiling is a key process, high critical heat fluxes can be realized by enhancing surface wettability. This can be achieved by the employment of nanoparticles, leading to enhanced heat transfer coefficients during the boiling process. For condensation, a significant effort has been made to investigate film condensation, so future studies may benefit from focusing on improving our understanding and the performance of drop condensation phenomena in relevant systems and applications. One specific pathway to improve the performance of processes that rely on drop condensation is to identify a viable route for addressing and overcoming the detrimental effects of non-condensable gases on heat transfer performance during this process in real systems.

Turning to the phase transition processes involved in melting, the melting rate typically increases at higher nanoparticle concentrations. However, in the case of solidification, an increase in the solidification rate is observed only up to a certain optimal nanoparticle concentration. To enhance the heat transfer rates and improve the energy storage density of relevant systems in real applications, in-depth studies of the

interactions of nanoparticles in latent heat thermal storage systems employing nanoparticles is required. Issues related to sedimentation and aggregation of particles also need more attention.

The exploitation of nanoengineered materials for phase change heat transfer needs further study, and there are significant opportunities for exploration in this area. Although a variety of new nanoscale materials are being developed continuously, there remains a gap in our understanding of how these can promote phase change heat transfer and energy storage, thus improving overall cost effectiveness in real applications. Of importance in relation to this point is the proposal of robust routes for improved heat transfer rates and reliable thermal energy storage, leading to an improved ability to design materials with optimized properties and characteristics. The quantification of the benefits at the industrial scale remains a particular challenge.

Conventional physicochemical approaches have led to heightened environmental concerns because of reduction of metal ions preceded by surface modification and generation of toxic and/or otherwise harmful by-products. In this regard, microorganisms, bio-templates, and plant extracts can be used as biodegradable agent materials. For nanofluid preparation, one-step methods may be a viable option for developing cost-effective and stable nanofluids for large-scale uptake; this makes it easier to manage particle size and to eliminate particle agglomeration.

To realize market uptake of nanofluids, it is necessary to produce high-heat-capacity nanofluids appropriate for use in long-term energy storage applications. Validated models with higher prediction accuracy, reliability, and reduced computational cost are required for the evaluation of key material properties and, by extension, the performance of nanofluid-based systems. Recent developments in artificial intelligence and machine learning approaches offer a promising avenue for development of such prediction models and tools. Finally, efforts should be made to develop nanofluids with unique properties tailored to specific applications, with a focus on cost cutting and reducing the environmental impact of their end-of-life disposal.

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AUTHOR CONTRIBUTIONS

Conceptualization, L.Y., O.M., L.Y., and C.N.M.; writing – original draft, A.A.; writing – review & editing, L.Y., Y.D., J.S.M., M.M., S.W., M.M.R., R.A.T., M.S., L.-P.W., and C.N.M.; supervision, O.M., R.A.T., and C.N.M.; project administration, O.M. and C.N.M.

DECLARATION OF INTERESTS

The authors declare no competing interests.

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