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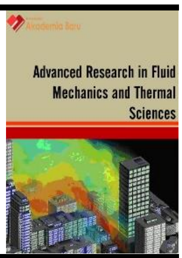

A Review on Nano Enhanced Phase Change Materials: An Enhancement in Thermal Properties and Specific Heat Capacity

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Nurfatihah Jamil[1,*], Jesbains Kaur[1], AK. Pandey[1], Syed Shahabuddin[1], Samir Hassani[1], Rahman Saidur[1], Roshafima Rasit Ali[2], Nor Azwadi Che Sidik[2], Mohd Naim[2]

[1] Research Centre for Nano-Materials and Energy Technology, School of Science and Technology, Sunway University, Bandar Sunway, 47500 Subang Jaya, Selangor, Malaysia
[2] Malaysia-Japan International Institute of Technology, Universiti Teknologi Malaysia, Jalan Sultan Yahya Petra, 54100 Kuala Lumpur, Malaysia

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ABSTRACT

Phase change materials (PCM) has gain attention for years as a suitable medium in thermal energy storage system. Previous studies reported the use of PCM on PV panel increases the efficiency of heat storage system. One of the most common method to increase the thermal conductivity and improving the thermos physical properties of PCM is by adding nanoparticles making it as nano enhanced PCM. This review paper focuses on categories of PCM including organic, inorganic and eutectics. The enhancement of thermos physical properties such as latent heat and thermal conductivity of phase change materials is discussed. However, there are very few studies reported on specific heat capacity of nano-enhanced PCM (NEPCM). Thus the current study focuses on the enhancement of specific heat capacity for NEPCM. On the other hand, some PCM has disadvantages such as supercooling and combustible that needs to be consider for further improvement is determined.

Keywords:
Nano enhanced phase change materials, thermo-physical properties, specific heat capacity

1. Introduction

The sun has always been a free and limitless source of energy. Besides the constant hike of price of fuels, the rapid depleting of fossil fuels also has become a major reason for researchers to focus on solar energy as a promising alternative. There are approximately 5 x 10^4 exajoules (EJ) of solar energy that can be yield freely to supply world's energy consumption. Current advanced technologies are employed to enable this collected irradiations during daytime so that it can be used later [1-3]. Domanski and Jaworski[4] concluded that there are various systems to store this energy including:

- Mechanical: hydro-pumped-storage facilities and compress air (CAES)

* *Corresponding author.*
E-mail address: nurfatihahj@sunway.edu.my (Nurfatihah Jamil)

- Thermal: heat storage system (PCM, solid, liquid), fuel production and chemical reaction heat storage
- Electrochemical: Batteries, galvanic cell and converters
- Biochemical storage

Above all, latent heat storage is suggested as an effective energy storage system because of its capacity to store bulk energy where an extensive amount of energy can be kept within a small volume. According to Mishra *et al.,* [5], latent heat can be divided into two types, latent heat of fusion and latent heat of vaporization. Any changes from solid to liquid or liquid to solid is considered as latent heat of fusion and vaporization of solid or liquid and condensation of vapour is a latent heat of vaporization. Figure 1 shows the classification of thermal energy storage and examples of latent heat storage.

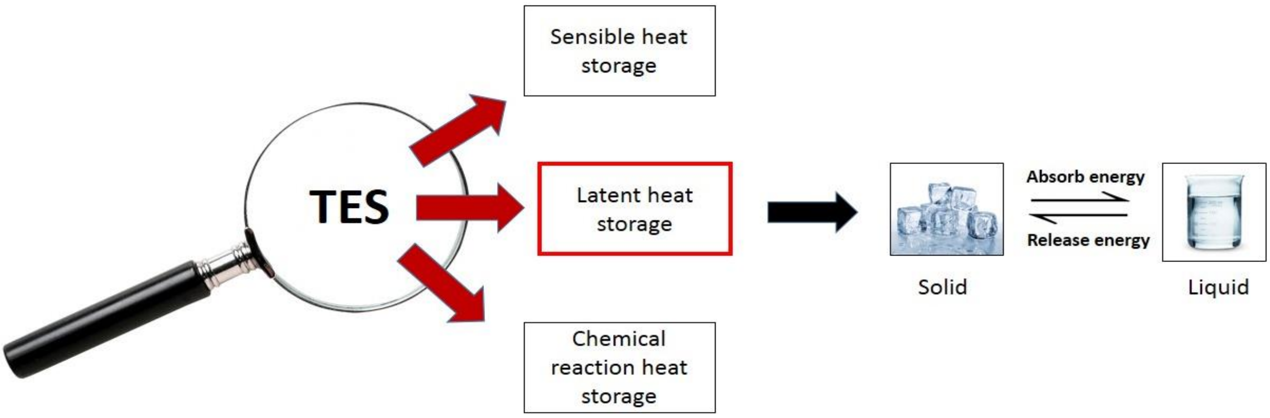


Fig. 1. Classification of TES and Example of LHS

2. Types of Phase Change Materials (PCM)

Phase change material (PCM) is a component that has high heat of fusion where it can absorb and release huge amount of energy during its solidification and liquidation. As an example, during daylight, solid-liquid PCM in LHS absorbs energy from sunlight and it will gradually melt. The heat energy remain stored until the PCM solidify again in the night where the temperature is lower. During solidification, it release the energy it stored thus generate electricity for home use. According to Zalba *et al.,*[6], the PCM is generally grouped into three types; organic, inorganic and eutectics. The classification of PCM is shown in Figure 2 [1].

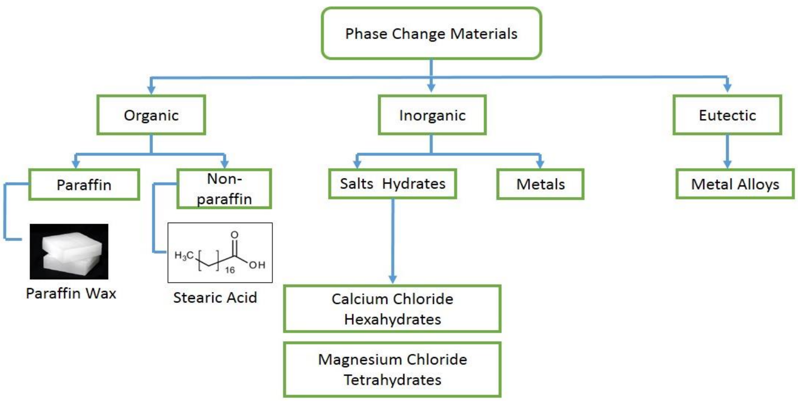


Fig. 2. Classification of PCM [1]

2.1 Organic PCM

Organic PCM consists of paraffin and non-paraffin materials. It has been broadly used in thermal energy storage due to its excellent characteristic such as non-corrosive, almost no super cooling and shows a consistent melting with no phase separation [7,8]. In addition, organic group includes paraffin wax, stearic acid, palmitic acid and oleic acid. Chung *et al.,*[9] utilized octadecane and bioPCM as phase change materials in his experiment and reported that both of the organic PCM demonstrate thermal stability under room condition. Dheep and Sreekumar [10] reported that glutaric acid was a promising phase change materials due to its minimal changes after 2000 thermal cycles and less corrosive to stainless steel container.

Instead of its extensive benefits, organic PCMs also has its limit. Pandey *et al.,* [7] has pointed out that organic PCMs are mostly expensive, combustible and possess a low thermal conductivity during solid state condition.

2.2 Inorganic PCM

Inorganic PCM gained a lot of attention from researchers as a material to store energy. It is much cheaper than organic PCM, abundance, high of volumetric latent heat density, high in thermal conductivity and high operating temperatures [1][8][11]. Salts hydrates and molten salts are grouped as inorganic phase change materials including $CaCl_2.6H_2O$, $MgCl_2.6H_2O$, $AlCl_3$ and KNO_3.

Calcium chloride hexahydrates (CCH) was found to have a stable thermal cycle by Tyagi and Buddhi [12]. They did a thermal cycling test on CCH where process of melting-freezing were carried out up to 60°C with temperature change rate of 7°C per minute. The findings showed that there were no noticeable changes in its thermal properties. Pilar *et al.,* [13] also have investigated the effect of thermal cycling on $MgCl_2.6H_2O$ by 50 cycles and results no significant changes. A study from Wu and Wang [14] also revealed that after 100 cycles, the composite of hydrated salts-expanded graphite has a negligible loss of enthalpy hence proved its reliability.

Despite its stable thermal cycling, salts hydrates has supercooling issues. Pilar *et al.,* [13] reported supercooling problems and introduced a nucleating agent $SrCO_3$, $Sr(OH)_2$ and $Mg(OH)_2$ into $MgCl_2.6H_2O$ mixture to overcome this problem. Li *et al.,* [15] also suggested to add nucleating agent such as strontium chloride in order to minimize supercooling. In terms of corrosion, Ren *et al.*, [16] has conducted a corrosion test of $CaCl_2.6H_2O$ on four different metal which are copper, carbon steel, aluminium 6061 (Al 6061) and aluminium 5086 (Al 5086). All four metals were dipped in a sealed-liquid $CaCl_2.6H_2O$ with different operating temperatures (30, 50 and 80°C) for a time period up to 16 weeks. As a results, carbon steel showed excellent performance with a reduction of corrosion as time increased. It is observed that a layer of oxide was formed on the metal as a protective layer hence reduced the corrosion rate. There are also studies from Oravcová *et al.,*[17] where they focused on metal corrosion when exposed to PCM; $CaCl_2.6H_2O$ and Zn(NO3)2.6H2O respectively. Aluminium, copper and carbon steel were immersed in both PCM at 55°C for 9 weeks. From the analysis, $CaCl_2.6H_2O$ caused very much lower corrosive effects on all metals, especially copper with a rate of corrosion 4.24 mg/cm^2 a year and advocated for long term use. Table 1 shows the advantages, disadvantages and examples for some organic and inorganic PCM's. In addition, Table 2 shows the potential of salts and salts hydrates PCM.

Table 1
Comparison of organic and inorganic PCM [1]

	Advantages	Disadvantages	Example
Organics	Non-corrosive Almost no supercooling Chemical and thermal stability	Lower phase change enthalpy Low thermal conductivity Inflammability	Paraffin wax Bees wax Palmitic acids
Inorganics	Greater phase change enthalpy	Supercooling	Calcium Chloride hexahydrates
		Corrosion	Magnesium chloride hexahydrates
		Phase isolation	KNO_3
		Phase segregation, lack of thermal stability	

Table 2
Salts and salts hydrates as potential PCM [1,42]

Compound	Melting Temperatures (°C)	Heat of fusion (kJ/kg)
Salts		
$AlCl_3$	192	280
$LiNO_3$	250	370
$NaNO_3$	307	172
KNO_3	333	266
KOH	380	150
$KClO_4$	527	1253
LiH	699	2678
$MgCl_2$	714	452
Salts Hydrates		
$Na_2P_2O_7.10H_2O$	70	184
$Ba(OH)_2.8H_20$	78	266
$(NH_4)Al(SO_4)_2.12H_2O$	95	269
$MgCl_2.6H_2O$	117	169
$Mg(NO_3)_2.6H_2O$	89.3	150

2.3 Eutectics

Combination of salts and salts hydrates will produce eutectics PCM. Eutectics PCM usually will solve the supercooling problems as it has different melting point from two different compositions. Going through the melting and freezing cycles, eutectics does not separate because they are freeze at well amalgam of crystals [1]. Thaib *et al.,* [18] prepared an organic composite phase change material (CPCM) consists of myristic acid/palmitic acid/sodium myristate (MA/PA/SM) and myristic acid/palmitic acid/sodium laurate (MP/PA/SL). This eutectic CPCM then integrated with damar gum, also known as Shorea Javanica (SJ) in order to boost its thermal conductivity. As a result, MA/PA/SL and MA/PA/SM shows an increment of thermal conductivity with 3 wt% and 2 wt% of SJ and has least degradation of latent heat. This also indicate that SJ is a suitable porous substance to be introduce in CPCM.

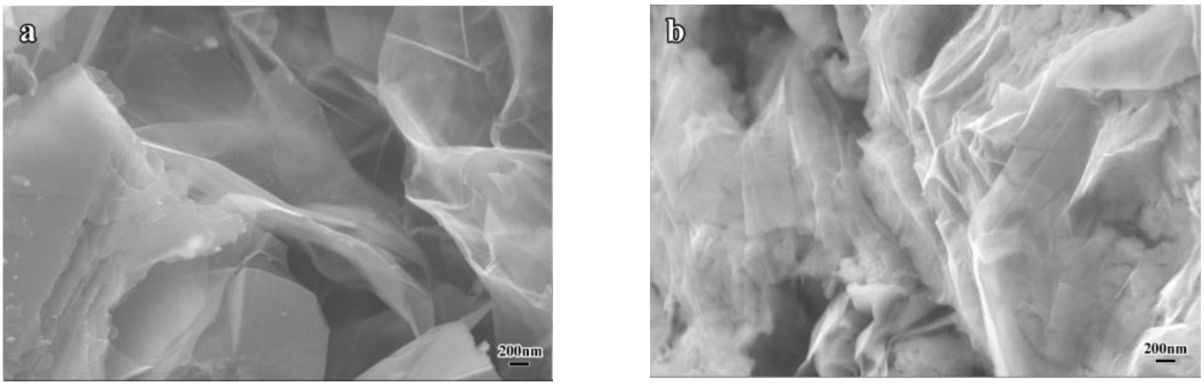
Salts hydrates also has bright potential as eutectic PCM. A study by He *et al.,* [19] where they synthesized a binary hydrated salts eutectic PCM consists of $CaCl_2.6H_2O$-$MgCl_2.6H_2O$. The modified eutectic PCM then tested with 100 thermal cycles and shows a thermal stability with an outcomes of latent heat 120.62 J/g with a slight deviation of 1.9 J/g.

3. Enhancement of Thermo Physicals Properties (Thermal Conductivity, Specific Heat Capacity, Latent Heat)

3.1 Thermal Conductivity

Heat transfer has been an important feature in thermal energy storage. Thermal conductivity can be define as the capability of a materials to conduct heat. If it can conduct heat faster, it will make it as a good medium to store energy. By increasing thermal conductivity, time taken for PCM to completely charge and discharge will be effected. Organic PCM is known for low thermal conductivity hence researchers used additive such as nanomaterials to boost its ability to conduct heat.

Li *et al.,* [20] reported a huge increment of thermal conductivity in his work. Montmorillonite (OMMT)/Paraffin was used as PCM and it was dispersed ultrasonically with multi-walled carbon nanotubes (MWCNT) that act as a nanomaterial. It was recorded the thermal conductivity of OMMT/paraffin/MWCNT has risen about 34% and 65% higher than OMMT/paraffin and paraffin only. Nanomaterials like Copper (Cu) are well known to have high thermal conductivity and it helps organic PCM like paraffin and fatty acids to achieve better thermal conductivity. This is proved by Wu *et al.,* [21] where paraffin was mixed with Cu nanoparticle by two step method. The results shows Cu/paraffin of 2 wt% Cu gave an increment up to 14.2% (in solid state) and 18.1% (in liquid state) of thermal conductivity. Although inorganic PCM possesses higher thermal conductivity compared to organic PCM, but it is still not sufficient to store solar energy unless with additives such as expanded graphite, TiO_2 and MWCNT. Figure 3 shows nanocomposites of modified expanded graphite (MEG)/MgCl2.6H2O (MCH) was prepared by Zhou *et al.,* [22] in order to investigate the thermal conductivity enhancement compared to pure MCH. From the results, it is confirmed that MEG/MCH offers 7.7 times higher thermal conductivity than pure MCH. Furthermore, to overcome the limitation of salt hydrates such as corrosiveness, the EG has been modified by adding a surfactant to increase EG's hydrophilicity. As shown in Figure 3, the MCH was adsorbed on pores of MEG. They are bond with hydrogen bond hence, there are no chances for melted MCH to flow out from MEG pores. Saeed *et al.,* [23] prepared a eutectic PCM methyl palmitate (MP) – lauric acid (LA) (MP/LA) where nano graphene platelet (NGP) was used as nanoparticles to enhance thermal conductivity. It is shown in Figure 4 that integration of NGP into MP/LA has boosted the thermal conductivity up to 110% in solid state.



(a) (b)

Fig. 3. SEM images of (a) MCH and (b) MOG/MCH composites to increase thermal conductivity and reduce corrosiveness [22]

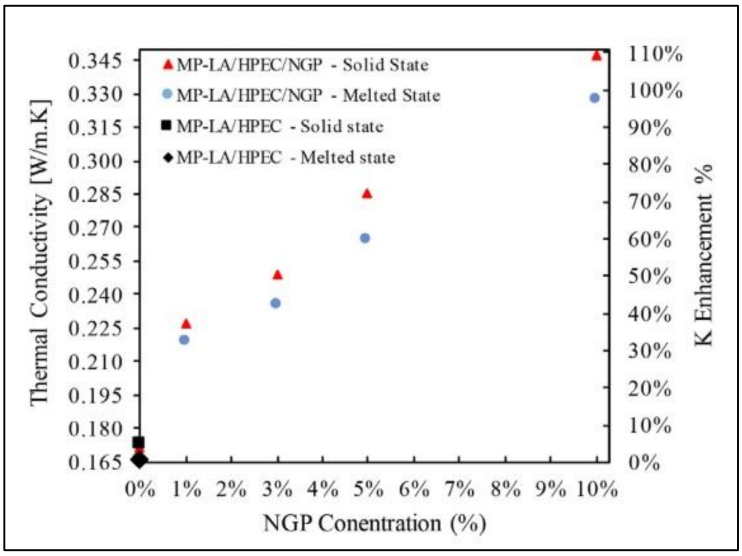


Fig. 4. Thermal conductivity enhancement of MP/LA/NGP [23]

3.2 Specific Heat Capacity

Specific heat capacity (C_p) in a simple explanation is the amount of energy needed to increase the temperature of a material by one degree. Due to under focused topic and less covered in literature, Ferrer *et al.,* [24] recently reported three methods to measure specific heat capacity by using DSC for thermal energy storage which is areas method, isostep method and dynamic method. Water, KNO_3 and slate were used as tested material and from the study, it is suggested that areas method gives the best result of specific heat capacity with the least error which is <3% from the theoretical C_p. It also shows areas method does not have sensitivity problem just like dynamic and isostep method where they produce continuous signal and deviate far from theoretical values of C_p.

In another paper, Ferrer *et al.,* [25] suggested 5 empirical equations to calculate the C_p of organic PCM; fatty acids and paraffin. By using the DSC areas method, they derived two empirical equations to calculate viscosity and C_p for all paraffin PCM group and the other three for fatty acid PCM group. However, this empirical equations may results 4% of error if subjected to another group of PCM thus showing that there are still lacking research for this topics. Tukimon *et al.,* [26] synthesized a quaternary nitrate salts which consists of $LiNO_3$, KNO_2, KNO_3 and $NaNO_2$ and determined their thermal properties including specific heat capacity. The molten salts were mixed with different composition to produce 3 different samples. The C_p of the 3 samples were determined by using DSC testing's where the average value of specific heat capacity was taken. As reported, sample 1 has the highest average specific heat capacity compared to the other 2 sample.

There are also studies where nanoparticles has been used to improve C_p for thermal energy storage. Shin and Banerjee [27] add silica nanoparticles into a eutectic solution, lithium carbonate-potassium carbonate. They managed to obtain an increment in C_p of the nanofluid by 19-24% with uncertainty less than 5%. By using the same eutectic salts as stated above, Byeongnam and Banerjee [28] investigated the effect of multi walled carbon nanotube (MWCNT) dispersion into solid and liquid PCM. As a results, with addition of 5 wt% of MWCNT, the C_p of nanocomposites (solid phase) shows 12% enhancement while in nanofluid (liquid phase) the C_p boost to 15%. Zhou *et al.,* [29] also reported a 6% increment of C_p of ethylene glycol when integrated with CuO nanoparticles. 50% enhancement of Cp was reported by Nelson *et al.,* [30] when they add exfoliated graphite nanoparticles into polyalphaolefin that has 0.3 and 0.6% mass concentration. As stated above, many studies focusing on nanocomposites and nanofluids from eutectic and organic group as they produce excellent results. However, inorganic group is less highlighted and has potential to be discovered in terms of enhancement of specific heat capacity.

3.3 Latent Heat

A melting-freezing of matter plays a significant role in thermal energy storage. Latent heat of fusion is a process where solidification and melting cycles will determine the energy stored by PCM. PCM will melt when it absorbs greater energy from the sun and the energy will be released once the temperature is reduced. Different materials definitely will have different latent heat value and the key in selecting PCM is always by looking for a high latent heat value. High latent heat value often possessed by inorganic PCM compared to organic PCM. This can be proven in a several recent studies reported by Mohamed *et al.,* [1] where most of the inorganic PCM showed better thermo physical properties (Table 3).

According to Alva *et al.,* [8] latent heat thermal energy storage have a high energy density compared to sensible heat. They also have melting temperature around desired operating temperature and almost no subcooling. Figure 5 shows a latent heat value between an organic and inorganic PCM [15,31].

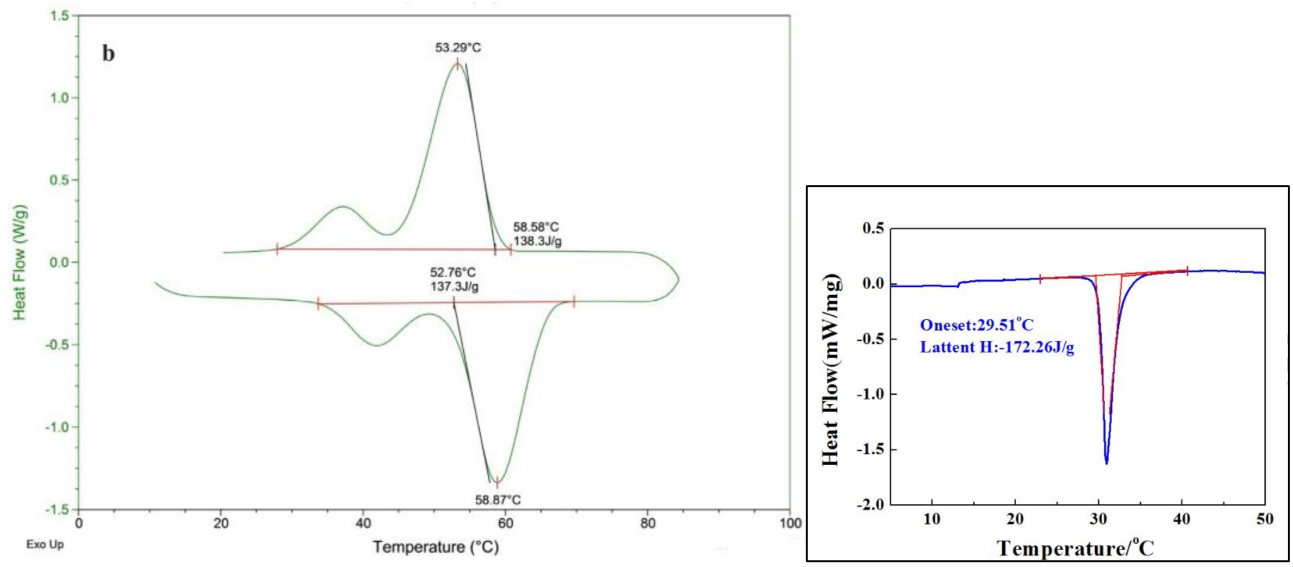


Fig. 5. Latent heat difference in (left) organic PCM and (right) inorganic PCM [15,31]

Table 3
Thermo physical properties of some inorganic PCMs [1]

PCM	wt%	Melting Temp, °C	Latent heat, kJ/kg	Cp kJ/kgK	Thermal conductivity, w/mK
$KCL/LiNO_3$	50/50	165.6	201.7	1.1 (s) 1.87 (l)	1.749 0.3315 (powder)
$NaNO_3/KNO_3$	6:04	223.2	142.2	2.351 (s)	2.27201 (s)
$NaNO_3$		306	172	1.1 (s)	0.5 (s)
KNO_3/KCl	95.5/4.5	320	74	0.953	0.5 (s)
KOH		360	134	1.34	0.5 (s)
$MgCl_2/KCL/NaCl$	60/20.4/19.6	380	400	0.96	
Cu	1083		205	0.495 (l)	180.4 (l) 386.44 (s)
Fe			272	795 (l)	27.2 (l)
Al		661	288	0.90 (s&l)	

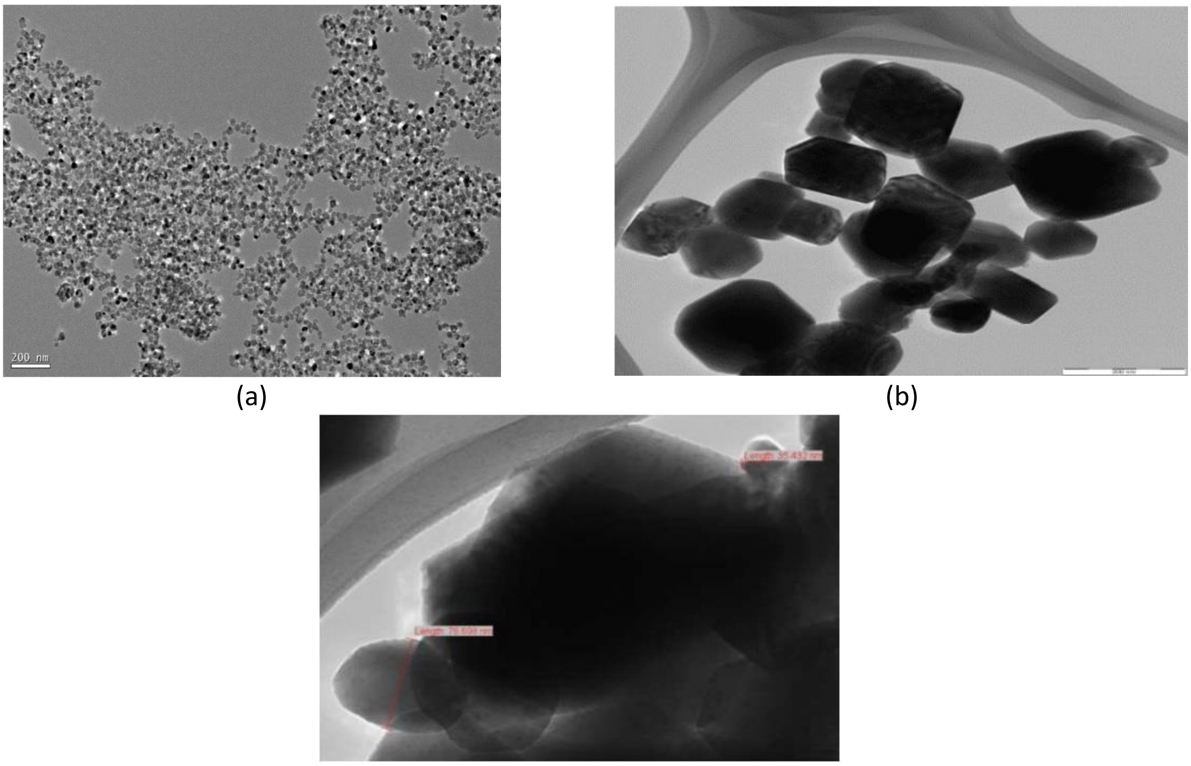
4. Nano Enhanced PCM

In order to enhance the performance of PCM in thermal energy storage, a few methods were suggested in the literatures including impregnation, encapsulation of PCM and also adding porous metals or introducing high conductivity nanomaterials into the PCM. Several studies reported the use of nanomaterials has increased thermal conductivity of the PCM as shown in Table 4.

Colla *et al.,* [32] recorded a 25% improvement of thermal conductivity when integrated a carbon black nanoparticles with PCM. Besides lowering the degree of supercooling, Cui *et al.,* [33] also reported a 51.36% increment of thermal conductivity of PCM when integrated with 1.5 wt% of graphene nanoplatelets. An increment of thermal conductivity also reported by Saeed *et al.,* [23] when a eutectic PCM methyl palmitate (MP) – lauric acid (LA) was mixed with nano-graphene platelet (NGP) together with gelling agent HPEC. With 10 wt% of NGP, MP-LA/HPEC/NGP boosted the thermal conductivity up to 102.2% in solid form and 97% in liquid. Furthermore, specific heat capacity also improved to 52% in solid form. Table 4 summarised the NEPCM observation from literatures. Figure 6 shows TEM images on the shape and morphology of copper and aluminium oxide nanoparticles which was induced in the PCM.

Table 4
Summary of nanomaterials used with PCM for performance enhancement

PCM Used	Nanomaterials used	Fraction	Method preparation	Observation
Paraffin [21]	Copper	2wt%	Two-step method	14.2% increase in thermal conductivity in solid state and 18% in liquid state
Paraffin [34]	Alumina, titania, silica	1,2,3 wt%	Direct synthesis	Titania most efficient in enhancement of heat conduction and thermal storage of paraffin
Erythritol [35]	Zinc oxide Expanded graphite	1,2,3 wt% 15 vol%	In-situ	EG has increased the thermal conductivity of PCM by 640%.
Barium Chloride [36]	Titania	0.07, 0.13, 0.25, 0.50 wt%	Two step method	With 0.50 wt%, 12.76% increment of thermal conductivity occur under the temperature of -5°C
1-dodecanol [37]	Multi-walled carbon nanotubes	1, 2 wt%	Dispersion of NP into PCM	Thermal conductivity enhanced by 4.6% and 11.0% with introduction of 1 and 2 wt% of MWCNT into the PCM.
Stearic acid [38]	Titania	0.05, 0.1, 0.15, 0.2, 0.25, 0.3 wt%	Sol-gel method	Thermal conductivity of the mixture escalated by 21.05-70.53% when titania was added.
$CaCl_2.6H_2O$ [39]	Expanded graphite	NA	UV curable resin system	The nanocomposite has high latent heat. Supercooling decreased to 7°C.
$CaCl_2.6H_2O$ [15]	Oxidation expanded graphite	0.2, 0.4, 0.6, 0.8, 1.0, 1.2 wt%	Emulsion by ultrasonic dispersion	Thermal conductivity of the nanocomposites has boost to 1.832 W/m.k with addition of 1 wt% of EGO.



(a) (b)

(c)

Fig. 6. (a) TEM image of α-Al_2O_3 [40] (b) TEM image of shape of the copper nanoparticles and (c) Image of size of the nanoparticles [41]

5. Conclusion

This paper highlighted the important studies in thermal energy storage systems where phase change materials is chosen to be the medium of heat storage. The potential of PCM makes it a suitable alternative to store energy efficiently. Although it has a few drawbacks, there are ways to overcome it such as adding nucleating agent for supercooling issues, introducing nanomaterials with high thermal conductivity into PCM to increase thermal conductivity, encapsulates PCM to avoid loss to surrounding and make eutectics to solve phase separation during melting- freezing cycles. This paper also presents the thermos physical characteristics enhancement of PCM such as latent heat and thermal conductivity. List of materials with their thermos physical properties was also tabulated.

6. Recommendation

PCM performance can always be enhanced by using the methods mentioned in this paper. This enhancement are significant because it can results in better thermal energy storage. However, there are only a handful of researches whom has studied on the specific heat capacity. Indeed specific heat capacity plays an important role in selecting the suitable PCM.

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