Limitations of using phase change materials for thermal energy storage

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Abstract. The use of a phase change material (PCMs) is a very promising technology for thermal energy storage where it can absorb and release a large amount of latent heat during the phase transition process. The issues that have restricted the use of latent heat storage include the thermal stability of the storage materials and the limitation of the container size. The study of the influence of thermal cycling on the properties of PCMs, such as melting temperature and latent heat, is important. It is found that the paraffin wax and fatty acids (e.g., lauric acid, myristic acid, palmitic acid, and stearic acid) have good thermal stability and can be used for solar thermal energy storage applications. However, Calcium chloride hexahydrate (CaCl₂·6H₂O) is a good PCM in building applications.

1. Introduction

We must not overlook the optimization of current energy usage, even though renewable energy has been developed by researchers to replace traditional fossil-based energy resources. It is important to study how to conserve and improve energy usages such as using thermal energy storage. The different ways of storing energy are mechanical, electrical, thermal, and thermochemical energy storage [1-3].

The basic concept of the thermal energy storage system is to store the thermal energy within the PCM and restore it later when needed. Three kinds of thermal energy storage can be used and these include the sensible heat, latent heat, and thermal chemical storage system. However, the storage density of the latent heat is higher than the sensible heat. Depending on the application, the time of the storage cycle varies from a few hours storage to several months are needed for others[1-3]. This paper presents the influence of thermal cycling tests on the thermophysical properties of phase change materials, namely, melting point temperature and latent heat of fusion.

2. The selection of phase change heat storage materials

The main principles that control the choice of phase change materials are [4]:

- Having a melting temperature in the required working temperature range.
- Having a high latent heat of fusion per unit mass in order to minimize the required amount of material.
- High specific heat to increase the effect of sensible heat storage.
- High thermal conductivity to reduce the time of charging and discharging.
- The volumetric expansion and shrinking coefficients are required to be small through the phase transition.
- Having a little or no subcooling during solidification.
Having chemical stability and corrosion resistance to the material of Containers and Heat exchangers. Free from any toxic, flammable, or explosive elements/compounds. Available in massive quantities at low cost.

3. Long term stability
The selection of a latent heat storage material is primarily based on the thermophysical properties, namely, melting point temperature, latent heat of fusion, specific heat, and thermal conductivity. However, the most significant parameters that have restricted the use of latent heat storage are the number of cycles with no change in the properties of the material and the useful working life of PCM–container systems. So the issue that has restricted the use of latent heat storage is that the long term stability of the storage materials and containers is not enough. Insufficient stability is due to changes in the material properties, which resulted from thermal cycling [5,6].

3.1 Stability of the PCM–container system
The physical and thermal stability of PCM can be demonstrated by developing PCM containers. Hadjieva used three paraffin mixtures; the effect of the thermal cycling on the properties of paraffin is verified to be minimal. The PCM, based on the selected paraffin mixtures, is found to be unchanged after many thermal cycles. The aim of the thermal cycles tests is to identity which PCM is more suitable for the thermal energy storage applications [6].

Thermal cycles
PCM has to be qualified to undergo many thermal cycles. Some materials exhibit changes within a relatively short time. It is significant to study the influence of thermal cycling on the melting temperature, latent heat of fusion due to their effect on the performance of the thermal energy storage system [7]. The details of the effect of thermal cycling on melting point and latent heat of fusion of Latent Heat Storage Materials (LHSMs), including: organic, inorganic, and eutectic, are the following:

Sharma et al. [8] conducted a test on urea. The change in melting point temperature and latent heat of fusion is found to be -23.6 °C and -21% after 30 thermal cycles tests. It is observed that urea after 50 cycles did not melt. They suggested that it should not be used as a PCM. Sharma et al. [9] carried out 300 thermal cycle tests for paraffin wax, stearic acid, and acetamide. The used device to measure the latent heat of fusion and melting point is Differential Scanning Calorimeter (DSC). It is found that commercial-grade PCMs do not show variation in melting point temperature but the latent heat was observed to vary by -10.3%, 2.8%, and 1.9%, respectively.

Organic
Sharma et al. [10] found that the range of change in melting point temperature for stearic acid, acetamide, and paraffin wax are 1, 2, and -3 °C respectively after 1500 thermal cycles. The change in latent heat of fusion is -20.6%, -1.14%, and -26%, respectively. They observed that commercial-grade PCMs have shown no regular declination in their melting points after 1500 thermal cycles. It is observed that acetamide and paraffin wax have good stability and they can be considered as promising PCMs. Stearic acid melts within a range of temperature. Thermal cycle testing was carried out for the mixture of technical-grade paraffin by Hadjieva et al [11]. It is observed that the change in melting point temperature and latent heat of fusion are about 0.7 °C and 1.8%, respectively after 900 heating/cooling cycles. It is found that the reduction in latent heat of paraffin is small.

Gibbs and Hasan [12] reported that paraffin shows excellent thermal stability after a number of thermal cycles. Shukla et al. [13] observed that the change in melting point temperature for Paraffin wax (58–60) and Paraffin wax (60–62) is -2.73 °C and 0.22 °C, respectively. The change in latent heat of fusion is -21.5% and -16.1%, respectively after 600 heating/cooling cycles. It is found that paraffin is the most suitable LHSM in terms of stability in melting point temperature and the latent heat of fusion. The change in melting point temperature and latent heat of fusion for Erythritol is 2 °C and 10% respectively. A maximum supercooling of 14 °C is observed in the 9th cycle. Erythritol has a very high
energy density; therefore, it can be a promising PCM for higher temperature thermal energy storage purposes. The change in melting point temperature and latent heat of fusion for paraffin/polypropylene (PP) composite (70/30 w/w %) are 0.75 °C and 0.3%, respectively [14]. The results indicate that PCM had good thermal reliability.

It is found that the change in melting point temperature for myristic, palmitic, and stearic acids is about -0.6 °C, -0.1 °C, and 1 °C, respectively after 450 cycles. It is observed that there is little reduction in the latent heat. The change in latent heat of fusion is -13.6 %, -9.2 %, and -11.7 %, respectively. The reduction in latent heat is due to the chemical degradation of fatty acids[15].

Table 1. Influence of thermal cycling on melting point and latent heat of fusion of organic materials

| Sr. No. | Phase change material( PCM) | Melting temperature (°C) | Latent heat of fusion (kJ/kg) | No. of cycles | References |
|--------|-----------------------------|--------------------------|-----------------------------|---------------|------------|
|        |                             | Initial | Final | Initial | Final                          |            |
| 1      | Paraffin (C22H44.1) (technical grade) | 47.1    | 46.6  | 166     | 163                           | 900        | Hadjieva et al. [11] |
|        | Paraffin (C23H48.4) (technical grade) | 57.1    | 57.8  | 220     | 224                           | 900        | [11] |
|        | Paraffin wax 53 (commercial grade) | 53      | 53    | 184     | 165                           | 300        | Hadjieva et al. [11] |
|        | Paraffin wax 53 (commercial grade) | 53      | 50    | 184     | 136                           | 1500       | Hadjieva et al. [11] |
|        | Paraffin wax 58–60            | 58.27   | 55    | 129.8   | 102                           | 600        | Sharma et al. [9] |
|        | Paraffin wax 60–62            | 57.78   | 59    | 129.7   | 109                           | 600        | Sharma et al. [9] |
|        | Paraffin (70wt %) + Polypropylene (30 wt. %) | 44.77   | 45.52 | 136.16  | 136.59                        | 3000       | Sharma et al. [10] |

|        | Acetamide                    | 82      | 82    | 263     | 268                           | 300        | Sharma et al. [9] |
|        |                             | 82      | 84    | 263     | 260                           | 1500       | Sharma et al. [10] |

| 3      | Erythritol, C4H6O4          | 117     | 119   | 339     | 305                           | 1000       | Shukla et al. [13] |

| 4      | Lauric acid                 | 42.6    | 41.3  | 176.6   | 156.6                         | 1200       | Sari [16] |
|        |                             | 42.6    | 39.5–44.1 | 211.6 | 132.8                         | 910        | Sari and Kaygusuz [17] |
| 5      | Myristic acid               | 50.4    | 49.8  | 189.4   | 163.5                         | 450        | Hasan and Sayigh [15] |
|        |                             | 52.99   | 46.21 | 181.0   | 159.1                         | 1200       | Sari [16] |
|        |                             | 53.8    | 45.3–52.2 | 192.0 | 159.1                         | 910        | Sari and Kaygusuz [17] |
| 6      | Palmitic acid               | 57.8    | 57.7  | 201.2   | 184.4                         | 450        | Hasan and Sayigh [15] |
|        |                             | 61.31   | 55.47 | 197.9   | 172.4                         | 1200       | Sari [15] |
|        |                             | 60.9    | 55.5–62.2 | 197.9 | 162.9                         | 910        | Sari and Kaygusuz [17] |
| 7      | Stearic acid                | 65.2    | 65.9  | 209.9   | 185.3                         | 450        | Hasan and Sayigh [15] |
|        |                             | 62.59   | 63    | 154.63  | 159                            | 300        | Sharma et al. [9] |
|        |                             | 54.7    | 46.83 | 159.3   | 157.7                         | 1200       | Sharma et al. [9] |
|        |                             | 53.8    | 46.9–50.2 | 174.6 | 118.9                         | 910        | Sari [16] |
|        |                             | 63      | 64    | 155     | 123                           | 1500       | Sari and Kaygusuz [17] |
| 8      | Urea                        | 133.9   | 110.3 | 195     | 154                           | 30         | Sharma et al. [8] |
|        |                             | 133.9   | -     | 195     | -                             | 50         | Sari et al. [20] |
| 9      | Myristic acid + glycerol    | 31.96   | 31.22 | 154.3   | 151.8                         | 1000       | Sari et al. [20] |
| 10     | Palmitic acid + glycerol    | 58.50   | 57.45 | 185.9   | 175.8                         | 1000       | Sari et al. [20] |

The range of change in melting point temperature and latent heat of fusion for stearic acid, palmitic acid, myristic acid, and lauric acids after 1200 thermal cycles are -7.87 °C, -5.84 °C, -6.78 °C, and -1.3
°C, respectively and -1%, -12.8%, -12.1%, and -11.3%, respectively [16]. The chosen fatty acids presented good thermal stability [16]. The latent heat for stearic acid, palmitic acid, myristic acid, and lauric acid after 910 cycles is reduced to 31.9%, 17.8%, 17.1%, and 37.2%, respectively [17]. It is observed that the range of change in melting point temperature is about -6.9 to 3.6 °C, -5.4 to 1.3 °C, -6.9 to -3.6 °C, and -6.9 to -3.6 °C respectively. It is found that the fatty acids chosen have good thermal stability. Palmitic and myristic acid may be considered more appropriate than the other PCMs for solar thermal applications [17].

Sari et al. [20] found that the range of change in melting point temperature for myristic, palmitic, and stearic acids with glycerol is -0.74°C, -1.05°C, and -0.38 °C, respectively, after 1000 thermal cycles. It is observed that the range of change in latent heat of fusion is -1.6%, -5.4%, and 2.2% respectively. It is found that the esters as PCMs had good thermal reliability. The details about the melting point and latent heat of fusion of organic materials are given in Table 1.

Inorganic

Tyagi and Buddhi [18] found that there is no alteration in melting point and latent heat for calcium chloride hexahydrate (CaCl₂, 6H₂O) after 1000 thermal cycles. So Calcium chloride hexahydrate is considered to be a promising PCM for the heating and cooling process in building applications. Sebaii et al. [19] observed the range of change in melting point temperature and latent heats of fusion for magnesium chloride hexahydrate MgCl₂.6H₂O are 12.6 °C and -45.2%, respectively after 500 thermal cycles. It is found to be MgCl₂.6H₂O not suitable as a PCM inside solar cookers for cooking indoors.

It is found the range of change in melting point temperature for sodium hydroxide. Di-sodium borate, and ferric nitrate is 2 °C, 4 °C, and -8 °C, respectively after one thermal cycle. Barium hydroxide does not get melted when subjected to very higher temperatures. After the second cycle, the Di-sodium tetraborate is remained solid even temperature up to 100 °C and the sodium hydroxide does not get melted. The Ferric nitrate does not solidify after keeping it for sufficient time. So, it is observed that the tested sodium hydroxide, Di-sodium borate, ferric nitrate, and barium hydroxide are not qualified as a good latent heat storage material (LHSM)[13].

Marks [24] found that the change of the latent heat of fusion for Glauber's salt is -73.5% after 40 cycles and for the thickened mixture is -48% after 200 cycles. It is clear that a significant improvement in the performance of the thickened mixture over that of the Glauber's salt and borax mixture. However, the thickened mixture exhibit a drop in performance with thermal cycling. The results show that the pure Glauber salt and the thickened mixture are unsuitable for long-term use as latent heat storage materials. Wada et al. [25] found that the change in latent heat of fusion for guaranteed grade and technical grade of sodium acetate trihydrate are -21.25% and -9.2%, respectively after 30 cycles and are -37% and -22.7%, respectively after 400 cycles. When the Sodium pyrophosphate decahydrate (Na₃P₂O₁₀,10H₂O) is used as a nucleating agent, the technical grade sample's heat storage capacity decreased more slowly than the guaranteed grade sample. The performance of thickened material during the thermal cycle test indicated little deterioration. Table 2 provides the details of the melting point and latent heat of fusion of inorganic material.
Table 2. Influence of thermal cycling on melting point and latent heat of fusion of inorganic materials

| Sr. No | PCMs                                      | Melting point (°C) | Latent heat of fusion (kJ/kg) | No. of cycles |
|--------|-------------------------------------------|--------------------|------------------------------|---------------|
|        |                                           | Initial | Final | Initial | Final |               |               |
| 1      | Calcium chloride hexahydrate              | 23.26   | 23.26 | 125.4   | 125.4 | 1000           | Tyagi and Budhi [18] |
| 2      | Sodium hydroxide, NaOH                    | 64      | 66    | 227     | 227   | 1              | Shukla et al. [13] |
| 3      | Di-sodium borate, Na₂B₄O₇·10H₂O           | 68      | 72    | 138     | 138   | 1              | Shukla et al. [13] |
| 4      | Ferric nitrate, Fe(NO₃)₃·6H₂O             | 60      | 52    | 99      | 99.23 | 1              | Shukla et al. [13] |
| 5      | Barium hydroxide, Ba(OH)₂·8H₂O           | 78      | -     | 265     | -     | 1              | Shukla et al. [13] |
| 6      | Magnesium chloride hexahydrate (MgCl₂·6H₂O) | 111.5   | 124.12 | 155.11 | 85    | 500           | Sebaii et al. [19] |
| 7      | Glauber’s salt(Na₂SO₄·10H₂O)             | 32.4    | -     | 238     | 63    | 40            | Marks [24] |
|        | Glauber’s salt(Na₂SO₄·10H₂O)+ attapulgite clay as thickener + borax (Na₂B₄O₇·10H₂O) as nucleating agent | - | - | 202 | 105 | 200 | Marks [24] |
| 8      | Guaranteed grade Sodium acetate trihydrate (NaCH₂COO·3H₂O) + Na₃P₂O₅·10H₂O as nucleating agent | 58 | - | 254 | 200 | 30 | Wada et al. [25] |
|        | Technical grade Sodium acetate trihydrate (NaCH₂COO·3H₂O) + Na₃P₂O₅·10H₂O as nucleating agent | 58 | - | 254 | 160 | 400 | Wada et al. [25] |

Eutectic

Zhang et al. [21] noted that the change of melting point for the eutectic mixture of lauric acid and palmitic acid is about -0.51% and the latent heat in reduced by 10% after 100 thermal cycles. It is found that binary systems of fatty acids can be used in solar energy storage. Sari et al. [22] conducted 360 thermal cycles test for Lauric acid (LA 75.5 wt %) + Stearic acid (SA 24.5 wt %), Myristic acid (MA 58 wt. %)+ palmitic acid (PA 42 wt. %), and Palmitic acid (PA 64.2 wt. %) + Stearic acid (SA 35.8 wt. %) form eutectic mixtures. It is observed that the range of change in melting point temperature is 0.36 °C, -0.2 °C, and 1.28 °C for LA–SA, MA–PA, and PA–SA, respectively. The range of change in latent heats of fusion is 0.2%, 2.8%, and 2.8% for LA–SA, MA–PA, and PA–SA, respectively. These materials have good thermal reliability with thermal cycling for about a one year utility period. By increasing the number of thermal cycles, the changes in the melting temperatures and latent heats of fusion of the studied eutectic mixtures are not uniform. These materials can be used for heat storage in passive solar heating applications [22].

Meiži He et al [23] studied and prepared binary eutectic hydrated salts inorganic phase change materials CaCl₂·6H₂O-20wt% MgCl₂·6H₂O with adding nucleating agent 1 wt% SrCl₂·6H₂O and thickening agent 0.5 wt% carboxymethyl cellulose (CMC). It is observed the change of melting temperature for the cooling and heating process are -0.18°C and 0.1°C after 100 thermal cycles test. The change in latent heats is 4.2 % and the average supercooling and superheat degree are 0.59°C and 0.49°C respectively. Their results showed that the modified inorganic CaCl₂·6H₂O-20wt% MgCl₂·6H₂O-1wt% SrCl₂·6H₂O-0.5wt% CMC could stay excellent circulation stability within 100 cycles, and providing reference value in practical use.

Sari [26] observed that the change in the melting temperatures and the latent heats of fusion are in the range of 0.9 °C, 0.23°C, and -0.2°C and -0.9%, 2.3%, and -1.1%, respectively for (LA–MA) Lauric acid (66.0 wt %) + Myristic acid (34.0 wt. %), (LA–PA) Lauric acid (69 wt. %) + palmitic acid (31 wt. %).
%), and (MA–SA) Myristic acid (64 wt. %) + Stearic acid (36 wt. %) after 360 thermal cycles. After 1460 thermal cycles, the change in the melting temperatures and the latent heats of fusion are in the range of 0.31 °C, -0.4 °C, and -1.1°C and 2.4%, 1.5%, and 1 %, respectively for LA–MA, LA–PA, and MA–SA. It is shown that the tested PCMs have good thermal stability for a storage period of four years. Table 3 provides the details of the melting point and latent heat of fusion of eutectic material.

Table 3. Influence of thermal cycling on melting point and latent heat of fusion of eutectic materials.

| Sr. No. | PCMs | Melting point (°C) | Latent heat of fusion (kJ/kg) | No. of cycles | Reference |
|---------|------|-------------------|-------------------------------|---------------|-----------|
|         |      | Initial | Final | Initial | Final | | |
| 1       | Lauric acid (77.05 wt %) + palmitic acid (22.95 wt%) | 33.09 | 32.92 | 150.6 | 165.69 | 100 | Zhang et al. [21] |
| 2       | Lauric acid (LA 75.5 wt %) + Searic acid (SA 24.5 wt %) | 37 | 37.36 | 182.7 | 183.1 | 360 | Sari et al. [22] |
| 3       | Myristic acid (MA 58 wt. %) + palmitic acid (PA 42 wt %) | 42.6 | 42.4 | 169.7 | 174.6 | 360 | Sari et al. [22] |
| 4       | Palmitic acid (PA 64.2 wt %) + Stearic acid (SA 35.8 wt %) | 52.3 | 53.58 | 181.7 | 186.3 | 360 | Sari et al. [22] |
| 5       | CaCl2·6H2O+20wt%MgCl2·6H2O nucleating agent 1wt% SrCl2·6H2O and thickening agent 0.5wt% carboxyl methyl cellulose (CMC) | 27.29–25.88 | 27.39–25.7 | 123.13 | 117.88 | 100 | Meizhi He et al. [23] |
| 6       | Lauric acid (66wt %) + myristic acid (34 wt %) | 34.2 | 34.12 | 166.8 | 168.3 | 360 | Sari [26] |
| 7       | Lauric acid (69wt %) + palmitic acid (31wt %) | 35.2 | 35.43 | 166.3 | 170.2 | 360 | Sari [26] |
| 8       | Myristic acid (64wt %) + stearic acid (36wt %) | 44.1 | 43.9 | 182.4 | 180.4 | 1460 | Sari [26] |

4. Conclusion
The selection of a latent heat storage material is primarily based on the thermophysical properties, namely, melting point temperature, latent heat of fusion, specific heat, and thermal conductivity. The most significant standards that have restricted utilization of latent heat storage are the number of cycles with no change in the properties of a material and the helpful working life of PCM–container systems. The selection of a latent heat storage material is primarily based on the thermophysical properties, namely, melting point temperature, latent heat of fusion, specific heat, and thermal conductivity. However, the most significant parameters that have restricted the use of latent heat storage are the number of cycles with no change in the properties of the material and the useful working life of PCM–container systems.

The effects of thermal cycling on the melting temperature and latent heat of commercial-grade phase change materials (PCMs) fusion is found to be important in the selection of the proper PCMs. It is observed that the melting temperature is in the range of 30–80 °C and latent heat of fusion is in the range of 140–260 kJ/kg for most organic and inorganic PCMs in the literature. The most eutectic PCMs studied in literature have the melting point in the range of 25–55 °C and its latent heat of fusion is in the range of 125–190 kJ/kg.

The effect of thermal cycling on various PCM have been studied in details. It is found that the majority of organics PCMs such as; paraffin and fatty acids (lauric acid, myristic acid, palmitic acid, and stearic acid) have good thermal stability and can be utilized for solar thermal energy storage applications. However, Urea is found to be not suitable to be used as a PCM.

Most of the tested inorganic PCMs are not suitable after some cycles and not qualified as a good latent heat storage material. Calcium chloride hexahydrate is considered to be a promising PCM in building applications. Using the pure Glauber salt and the thickened mixture for long-term is not suitable as LHSM. The MgCl2·6H2O is not adequate as an LHSM inside solar cookers.
It is found that fatty acids can be used in solar energy storage. The eutectic mixtures of Lauric acid (66.0 wt. %) + Myristic acid (34.0 wt. %), Lauric acid (69 wt. %) + palmitic acid (31 wt. %), and Myristic acid (64 wt. %) + Stearic acid (36 wt. %) form eutectic mixtures as PCMs and they have good thermal stability for a storage period of four years.

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