

The present work is specifically focused on the form stability of paraffin as a phase change material (PCM) through the addition of high-density polyethylene (HDPE). The aim of adding HDPE is to obtain a stable form of paraffin during the phase transition. Moreover, improving the performance of PCM leads to an advanced operation of the latent heat storage unit with an excellent charging duration and response time. The study uses HDPE at a ratio of 5 wt %, 10 wt % and 15 wt %. The results indicate significant differences between the form-stable PCM (FSPCM) and pure paraffin. For instance, the supercooling degree is decreased with the addition of HDPE, where paraffin has a supercooling degree of 8.01 °C while FSPCM with 15 wt % HDPE has a supercooling degree of 3.73 °C. The latent heat of fusion by adding 10 wt % and 15 wt % HDPE is slightly decreased by 1.85 %, which is much lower compared to adding 5 wt % HDPE, which reduces the latent heat of fusion by about 6.02 %. Adding HDPE leads to a faster charging process and a better response time during the discharging process. The charging rate is increased significantly by adding 15 wt % HDPE with a substantial increment of around 40 % with an average charging rate of 2.39 °C/min. The heat release during the discharging process is increased for FSPCM with 5 wt % HDPE where the temperature drops by more than 70 °C within 20 minutes. The findings indicate that adding HDPE contributed positively to reducing the supercooling degree and providing a steady phase transition. Thus, the heat exchange process of paraffin is more favorable, which improves the performance of the latent heat storage unit. Furthermore, the operation can be improved significantly by providing a faster charging and discharging process

Keywords: charging, discharging, latent heat, paraffin, PCM, polymer, supercooling, thermal storage

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ANALYSIS OF THE THERMAL CHARACTERISTICS OF THE PARAFFIN WAX /HIGH-DENSITY POLYETHYLENE (HDPE) COMPOSITE AS A FORM-STABLE PHASE CHANGE MATERIAL (FSPCM) FOR THERMAL ENERGY STORAGE

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1. Introduction

Energy plays a critical role as a major economic driving force and is taken as a fundamental need of modern civilization. The exploration of new fossil fuel sources is accompanied by the massive utilization of alternative fuels and renewable energy. This can be done by installing a heat exchanger for the dewatering process reducing the water content to less than <0.1 % [1]. Another effort can also be made by improving the

system's efficiency supporting renewable energy technology as a serious effort for the transition to low-carbon power systems significantly reducing greenhouse gas emissions where a critical aspect is the feasibility [2] of the components and dispatchability of the system. Furthermore, designing a new configuration for energy harvesting equipment such as using a specific design to improve the blade design for the Savonius rotor [3] is expected to produce more energy from renewable sources. As an intermittent energy source, the produced energy

must be stored to ensure continuous supply to the system. Thus, massive improvement and innovation in the energy storage system are expected to boost the renewable energy system [4].

One convenient method to store energy is through a thermal energy storage (TES) system. It works by utilizing a storage material to store heat during the peak season and release it when required. Heat can be supplied from renewable sources, such as solar energy and industrial processes, such as waste heat. The total global capacity of TES is tremendous and is expected to be more than 800 GWh in 2030 [5]. Sensible TES is the most common technology in TES systems with the highest technology level and is widely applied. This reduces the cost of the system. However, it has a serious drawback due to low energy density in both gravimetric and volumetric terms per unit of energy. This limits its application in large mobile systems. Another type of TES is latent heat storage using phase change material (PCM) as a storage medium [6]. It has a better storage capacity than a sensible system and provides a higher technology level than a thermochemical system. Thus, it is considered an ideal TES for the system and has been continuously developed recently.

TES also can be coupled into a biomass burner where residual heat from the combustion process can be extracted and stored in the PCM using a mantle burner [7]. To accommodate the storage capacity, various phase change materials are potentially applied in latent heat storage systems, including salt hydrates, organic and inorganic PCM [8]. Among various options of PCM, paraffin wax is the most common material for latent heat storage, especially for low-temperature systems. Factors that make paraffin an ideal candidate are better storage density, economic feasibility, and excellent compatibility when using different storage tanks due to the nontoxic nature of paraffin. It can be adjusted to numerous applications and is suitable for stationary and mobile systems [9]. The outstanding advantage of paraffin as PCM accelerates the exploration and improvement of the latent heat storage system.

The paraffinic latent heat storage improvement is focused on overcoming unstable operation during the phase transition. It is highly relevant for the current development to reduce the unstable operation of the paraffinic latent heat system to enhance the heat storage system. Therefore, research to solve the given problem related to unsteady phase change is urgent and is further developed to provide a proper solution.

2. Literature review and problem statement

The application of paraffin as a storage material for thermal energy storage is very attractive. Paraffin provides an optimal storage system, particularly in terms of storage density compared to a sensible system. Paraffin has been used for numerous applications and can be further developed in environmentally friendly applications, such as solar dryers [10], where the heat storage unit allows the system to operate at night and reduce the drying time effectively. However, an extended achievement is required to improve the system response. For example, adding volcanic ash [11] is expected to improve the thermal conductivity of paraffin up to $19.598 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$. However, local agglomeration is observed with a higher ash ratio (up to 50 wt %), which severely reduces the latent heat capacity. An alternative approach can be taken by adding a nano-additive to improve thermal conductivity. The study [12] uses nano- Al_2O_3 and nano-graphite as thermal conductivity enrichment to im-

prove the heat exchange process in the storage unit. Despite that, it is limited to high-density polyethylene (HDPE) as a supporting matrix for myristic acid as the primary PCM.

Using HDPE as a supporting matrix is convenient to ensure the storage material's thermal stability since it has excellent thermal behavior. The thermal cycling study [13] takes up to 10,000 actual thermal cycles and demonstrates a significant decrement in paraffin's thermal capacity due to repeating the thermal process. This causes the lifetime of paraffin to be relatively low, which is not ideal for long-term operation. As discussed in [14], an advanced study is carried out by adding HDPE to paraffin for lifetime assessment. It is shown that the paraffin/HDPE composite can withstand large thermal cycles and keep the outstanding performance of paraffin, the capacity of which decreases by only 5.5 %. However, the study focuses on the charging variation by using one specific ratio of HDPE as a matrix for the paraffin/HDPE composite.

Several polymers are considered to be composited with paraffin as a supporting matrix in order to stabilize the phase transition. The review [15] discusses in detail the reference to using styrene-butadiene-styrene (SBS) as a supporting matrix to develop a stable phase transition of paraffinic latent heat storage. The application of SBS shows good thermal stability for the paraffin/SBS composite, as reported in [16]. The study uses 20 wt % SBS added to paraffin and indicates that the supercooling degree of the composite decreases notably with less than 1°C . However, adding SBS reduces the thermal conductivity of the composite to $0.202 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$. HDPE is still an ideal alternative to form stable paraffin during the phase transition since it has a better thermal conductivity than SBS with an average value of $0.415\text{--}0.278 \text{ W}/\text{m}\cdot\text{K}$ at a temperature of $30\text{--}150^\circ\text{C}$.

The use of HDPE for the paraffin composite is shown as a positive outcome, which allows the composite to be used for large-scale production as discussed broadly in the work [17], which focuses on the simulation of the extrusion method for the PCM and paraffin composite. The study demonstrates the potential of large-scale production of the paraffin and HDPE composite, specifically intended as a thermal energy storage material. The main challenge for the paraffin composite is the proper ratio of HDPE to be used. The paper [18] shows that the thermal capacity based on the latent heat of fusion is decreased for the paraffin/HDPE composite. The upper limit is taken using 20 wt % HDPE, which still indicates a significant decrease in the latent heat of fusion by 29.7 % as a direct impact of the HDPE presence in the composite [19]. This reduces the total thermal capacity of the storage material and requires further adjustment to maintain the same capacity compared to pure paraffin.

A stable phase transition can be achieved by using HDPE, which is the main advantage of using HDPE as a form stabilizer while ensuring the thermal capacity should also be considered. However, this requires more exploration and study using different HDPE ratios and observing the effect on the thermal characteristics and performance of paraffinic latent heat storage. Using the proper ratio is expected to maintain a suitable thermal capacity while ensuring a steady phase transition for the existing system.

3. The aim and objectives of the study

The study aims to form a steady phase transition of paraffin by adding HDPE as a composite form-stable phase

change material (FSPCM). This will provide steady characteristics of the paraffin and HDPE composite during the latent heat storage system operation.

To achieve the aim, the following objectives were set:

- to determine the change in the melting and freezing temperature of the paraffin and HDPE composite;
- to evaluate the effect of the HDPE ratio on the change in the latent heat of fusion of paraffin;
- to observe the influence of the HDPE ratio on the thermal performance of the paraffin and HDPE composite.

4. Materials and methods of research

The object of the study was the phase change material (PCM), which was intended to be applied as latent heat storage material. The key point is to provide a stable phase transition of the PCM by adding a supporting matrix without making significant changes to the latent heat of fusion of the composite. Therefore, paraffin and HDPE (commercial grade) were the main materials for the present study. The maximum limit of HDPE is set at 20 wt %. Therefore, the present study uses three different ratios of HDPE with an interval of 5 wt % (Table 1). The composite made of paraffin and HDPE was designed to produce form-stable phase change material (FSPCM). The composite was processed carefully by pouring molten HDPE steadily into molten paraffin. The mixture was kept stirred slowly at a temperature of 180 °C. The mixing process took 1 hour to ensure the homogeneity of the composite. After that, the sample was cooled for further characterization and performance assessment.

Table 1

Prepared samples

Sample	Paraffin (wt %)	HDPE (wt %)	Identifier
Paraffin	100	0	P
Paraffin/HDPE composite	95	5	FSPCM ₁
	90	10	FSPCM ₂
	85	15	FSPCM ₃

The thermal characteristics of the samples (Table 1) were evaluated through differential scanning calorimetry (DSC). The evaluation provided the melting and freezing temperatures of the samples. Moreover, it also provided the latent heat of fusion of the given sample. The performance of the samples based on charging and discharging was characterized through the static test using an external heat source (Fig. 1, a). According to the recommendation [20], the test was intended based on the active test. The charging process was described as a heat intake process (endothermic), which means that the oil temperature in the oil bath should be higher than that of the sample. Therefore, the oil temperature was kept constant at 180 °C. The charging process was carried out using thermal oil and an electric heater, which acts as an external heat source. The process continued until the sample reached a temperature of 150 °C.

The discharging process was exothermic, where the stored heat is released to the environment (Fig. 1, b). It was conducted by placing the high-temperature samples in the cooling bath using water as a cooling medium. The water

temperature was kept constant at 15 °C. The measurement was conducted until the sample reached a temperature of 40 °C. The final aspect of thermal performance is evaluating the sample’s ability to maintain its temperature in the storing mode. The test was taken using the same storage container (aluminum cylinder), which makes the storage container’s effect negligible during the test. The storing test was conducted by storing the container at room temperature (30 °C). The test was specifically intended to understand the effect of HDPE on the thermal characteristics and performance of the paraffin and HDPE composite as latent heat storage material.

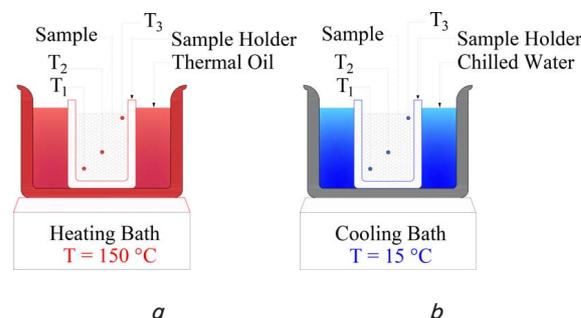


Fig. 1. Schematic design for experimental test: a – static charging test; b – static discharging test

5. Results of the study of the paraffin and high-density polyethylene composite as form-stable phase change material

5.1. Results of the melting and freezing temperature of paraffin and form-stable phase change material

The melting and freezing temperatures for each sample are plotted in Fig. 2. The highest melting temperature is obtained at 60.22 °C. The value is suitable for low to medium-temperature latent heat storage system applications. The addition of 5 wt % HDPE for the FSPCM₁ sample reduces the melting temperature insignificantly. Higher ratios of 10 wt % and 15 wt % for FSPCM₂ and FSPCM₃ also indicate a decrease in melting temperature. This implies that adding HDPE at the given ratio contributes to a lower melting point of the FSPCM composite. Even though the melting temperature of HDPE is higher than that of pure paraffin [21], the higher ratio of paraffin makes most HDPE melt together during the phase transition of paraffin.

Adding HDPE also alters the freezing temperature of paraffin. Paraffin has a freezing temperature of 68.23 °C, generally higher than its melting temperature. Interestingly, adding HDPE at different ratios changes the freezing temperature of FSPCM. Adding 5 wt % HDPE leads to a decrease in the freezing temperature of the composite by 1.11 °C. At a higher ratio of HDPE, FSPCM₂ and FSPCM₃ have a higher decrement in freezing temperature by 3.39 °C and 6.08 °C, respectively. This significantly changes the composite’s freezing temperature due to a higher ratio of HDPE. FSPCM₂ and FSPCM₃, which use 10 wt % and 15 wt % HDPE, promote slower solidification, which decreases the composite’s freezing temperature significantly.

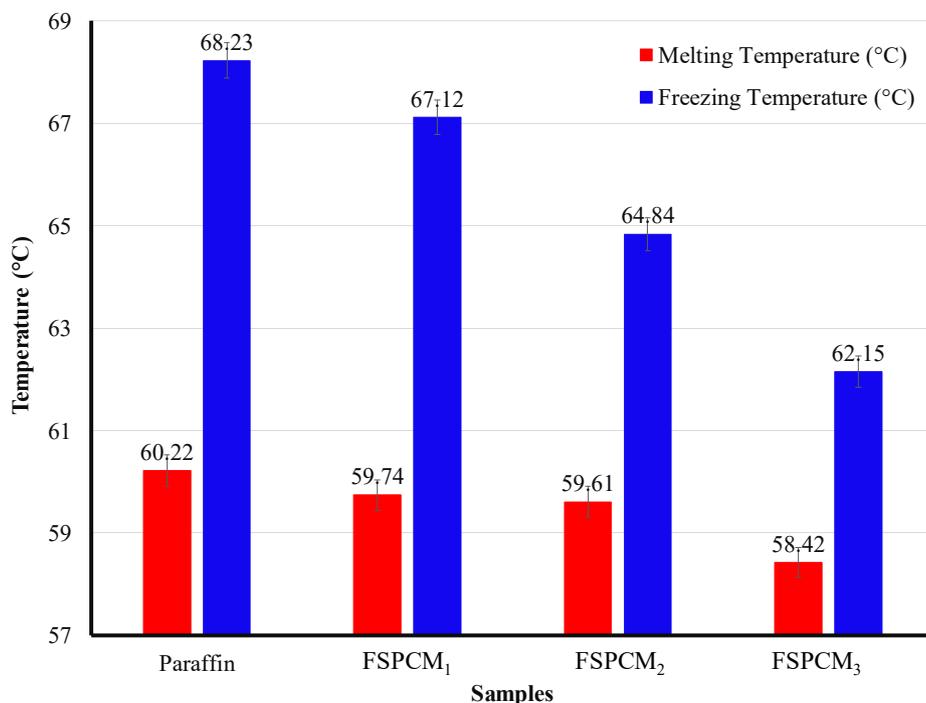


Fig. 2. Melting and freezing temperature of paraffin and form-stable phase change material

The supercooling degree is a critical aspect of melting and freezing temperature change. It is an essential aspect of latent heat storage material since it relates to the solidification process during discharging, which commonly occurs faster than its melting [22]. The decreasing melting temperature of the FSPCM composite increases the supercooling degree. However, a lower melting temperature of the FSPCM composite is accompanied by a decrement in the freezing temperature, which reduces the severe effect of the supercooling phenomenon. For instance, pure paraffin has a higher supercooling degree by 8.01 °C, which is too high for the latent heat storage system. This is disadvantageous for controlling the operation of the working fluid, particularly for a thermal management system that highly depends on the temperature of the storage material [23]. Adding HDPE supports reducing the supercooling degree with the lowest value in the FSPCM₃ composite, which uses 15 wt % HDPE.

5. 2. Results of the latent heat of fusion of paraffin and form-stable phase change material

The latent heat of fusion is an essential indicator in a latent heat storage system since the storage capacity is highly dependent on it. Fig. 3

presents the latent heat of fusion for paraffin and FSPCM. Paraffin has the highest latent heat of fusion amounting to 145.95 J·g⁻¹. The high latent heat of fusion is a major reason for using the paraffin-based PCM as a storage material in the latent heat storage system. The effect of HDPE on the FSPCM composite can be observed as a decrement in the latent heat of fusion for the paraffin and HDPE composite. As seen in Fig. 3, the latent heat of fusion for each FSPCM tends to decrease and become higher with an increment in the HDPE ratio in the composite. Thus, adding HDPE correlates with a decrement in latent heat of fusion for the FSPCM composite.

The latent heat of fusion for the samples with 5 wt % and 10 wt % HDPE (FSPCM₁ and FSPCM₂) indicates a consid-

erable decrement of latent heat. Both samples show a decrement of 5.1 % and 10.8 %. Even though the trend implies that the addition of HDPE reduces the latent heat of fusion with a higher ratio of HDPE, the latent heat of fusion for FSPCM₃ with 15 wt % HDPE tends to reduce slightly compared to FSPCM₂. The percentage of decrement for FSPCM₃ is only 12.5 %. This indicates that the change in latent heat of fusion differs for each FSPCM and depends on the specific ratio of HDPE in the composite.

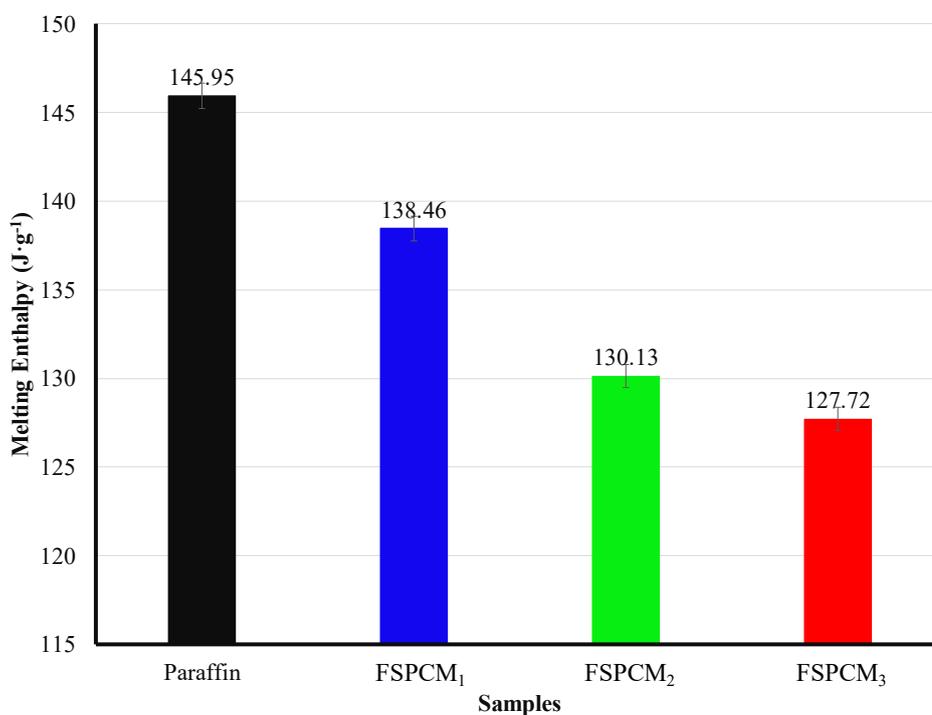


Fig. 3. Latent heat of fusion of paraffin and form-stable phase change material

5.3. Results of the thermal performance of paraffin and form-stable phase change material

The profile of temperature increment during the charging process as part of the thermal performance test is plotted in Fig. 4. The temperature increment for the samples demonstrates different profiles, which implies the effect of HDPE on the sample during the charging process of the composite. As can be observed in Fig. 4, the charging duration for paraffin is the longest, which takes 72 minutes to reach the target temperature. The endothermic process for paraffin is relatively slow due to low thermal conductivity [24]. This makes the heat interaction during the process occur at a slower rate. The addition of HDPE seems to improve the charging behavior of paraffin, where a faster charging duration for the FPSCM composite is indicated. The charging duration for the FPSCM ranges from 65 to 48 minutes. The fastest charging process is observed for the sample with 15 wt % HDPE (FSPCM₃). The FSPCM₁ and FSPCM₂ composites still demonstrate a slow temperature increment that occurs unsteadily related to the charging profile of pure paraffin. However, both samples exhibit a better charging process where the charging duration can be faster by 7 and 15 minutes compared to pure paraffin.

The temperature decrement during the discharging process requires more time for all samples compared to the charging process. As seen in Fig. 5, the samples take more time to achieve the target temperature during the discharging process. Pure paraffin demonstrates the longest duration, which takes about 76 minutes to achieve a temperature of 40 °C. FSPCM₁ and FSPCM₂ have a slightly shorter discharging duration compared to pure paraffin. It is demonstrated that the slow heat release process of paraffin still affects the paraffin and HDPE composite at a ratio of 5 wt % and 10 wt %. The discharge profile of the sample with

15 wt % HDPE (FSPCM₃) implies a faster process that takes a shorter discharge time. The pattern indicates that the heat release process for the sample occurs effectively, which is related to the charging process. This suggests the consistency of the sample since the charging and discharging profiles exhibit an identical pattern with suitable duration.

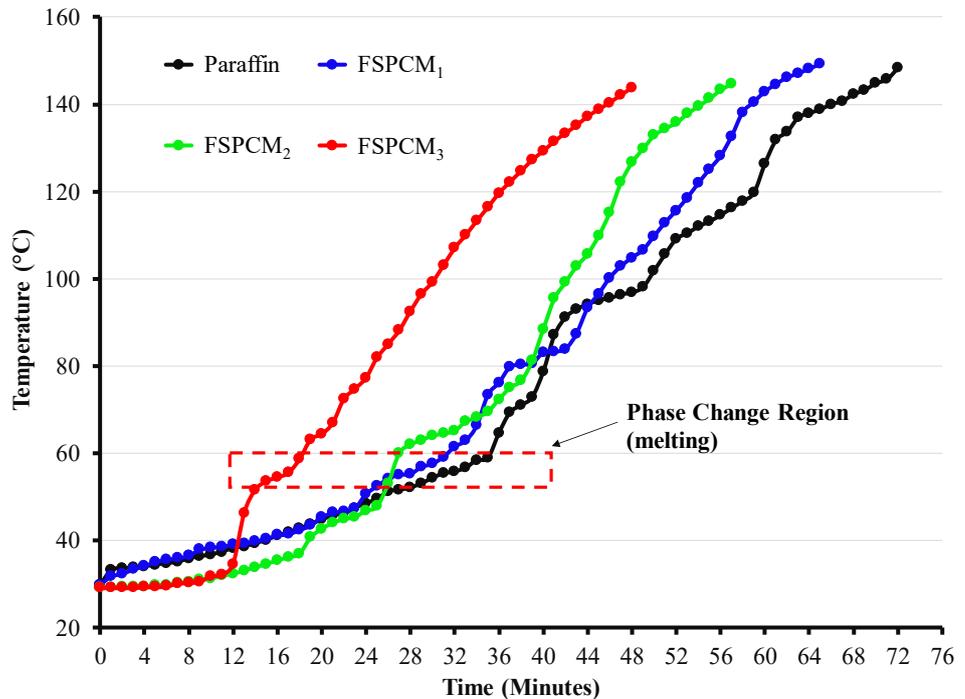


Fig. 4. Temperature increments in the charging process for paraffin and form-stable phase change material

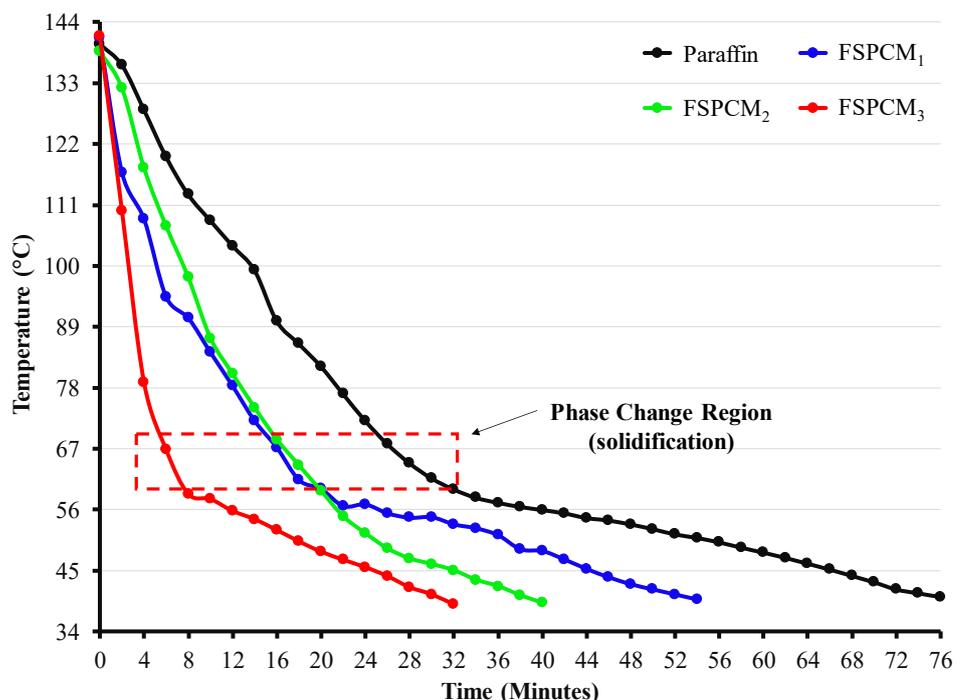


Fig. 5. Temperature decrements in the discharging process for paraffin and form-stable phase change material

A significant factor that distinguishes the effect of HDPE content in the composite can be observed during the storing process. As plotted in Fig. 6, the pattern of temperature decrement for paraffin, FSPCM₂ and FSPCM₃ is identical with a slow temperature decrement at temperatures of 115–65 °C. This affects the sample's heat losses, which occur at a slower rate at the given temperature.

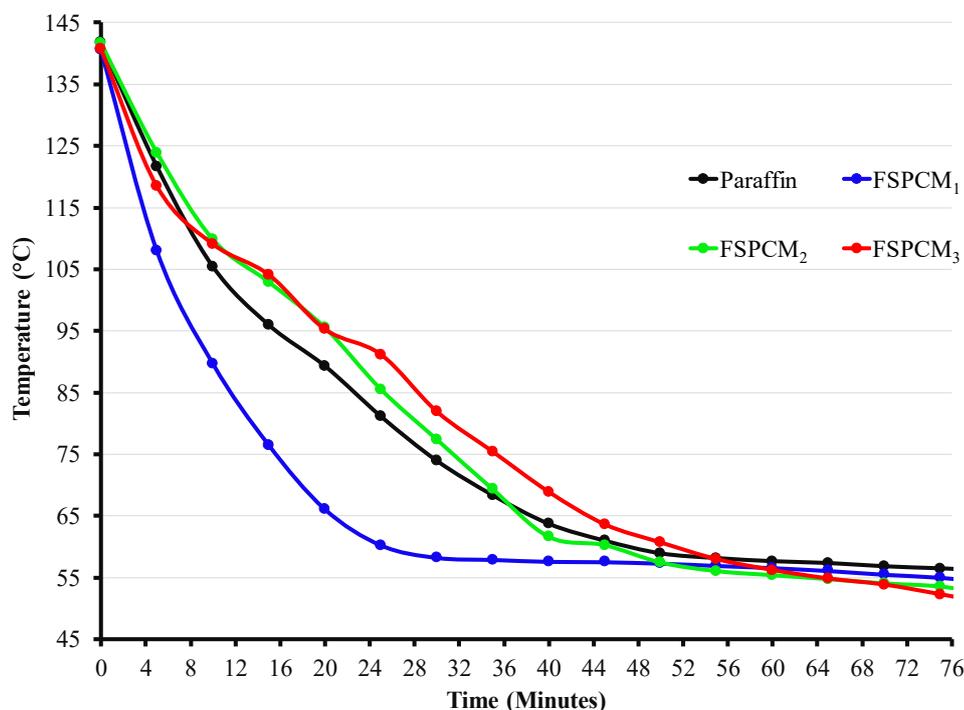


Fig. 6. Temperature decrements in the storing test for paraffin and form-stable phase change material

In contrast, FSPCM₁ with 5 wt % HDPE implies extreme temperature drops until the sample reaches a temperature of 58.2 °C. The heat release process during the storing test involves natural convection, where air works as a cooling medium. FSPCM₁ shows an accelerated temperature drop during the test. Thus, the sample is more likely to experience heat losses during the actual process than pure paraffin and FSPCM at a higher HDPE content (10 wt % and 15 wt %).

6. Discussion of the results of the thermal properties and performance of form-stable phase change material and paraffin

Thermal properties related to the melting and freezing temperature of PCM are taken as an important factor in the latent heat storage system. They describe the operating characteristics, specifically the supercooling degree, which affects the operation of the latent heat storage unit. Adding HDPE to paraffin to form the FSPCM composite contributes directly to the change in the composite's melting and freezing temperatures. Fig. 2 shows the change in the melting and freezing temperatures of the composite as a direct effect of the presence of HDPE. FSPCM demonstrates a lower supercooling degree compared to pure paraffin. The supercooling degree of paraffin is obtained at a temperature of 8.01 °C while it decreases for FSPCM. The presence of 15 wt % HDPE in the paraffin and HDPE composite leads to a significant decrement in the

supercooling degree, which occurs only by 3.73 °C. This signifies a noticeable effect on the stability of the phase transition of the paraffin and HDPE composite, where the melting and freezing occur at rather close temperatures.

The change in melting and freezing temperatures is also followed by the change in latent heat of fusion as the impact of HDPE presence in the composite. As presented in Fig. 3, the latent

heat of fusion for paraffin is 145.95 J·g⁻¹, which is generally high for the latent heat storage system. Adding HDPE causes a decrement in the latent heat of fusion, which can be taken as a side effect of HDPE addition. This affects the reduction of latent heat of fusion as the HDPE content increases. Moreover, the percentage of the decrement varies, where the latent heat of fusion for FSPCM₁ and FSPCM₂ has a significant decrement. However, compared to the decrement for FSPCM₁ and FSPCM₂, a decrease for FSPCM₃ is only 1.7 %. It should be considered that the decrement in latent heat of fusion does not occur linearly. Therefore, a suitable HDPE content in the composite can maintain sufficient latent heat of fusion of the composite.

The change in thermal properties affects the thermal performance of the storage material. As presented in Fig. 4, the temperature increment during the charging process varies between paraffin and FSPCM. This mainly occurs after the phase change region, which causes the melting transition in the samples. The charging rate of pure paraffin is obtained at 1.63 °C/min, while all FSPCM demonstrate a higher charging rate. The profile pattern of FSPCM₁ and FSPCM₂ indicates an unsteady temperature increment, which can be observed as the intercept area from 58 °C to 80 °C. The charging rate for both samples is obtained at 1.85 °C/min and 2.04 °C/min, respectively. The decreasing melting temperature for both samples is unsubstantial, which takes place around 59 °C (Fig. 2) due to a disruption of heat uptake in the phase transition region. FSPCM₃ gets a better charging rate with an average value of 2.39 °C/min. The temperature profile for the sample demonstrates an outstanding performance during the phase transition, promoting a better charging rate for the composite.

The heat release and discharge processes express a faster temperature decrement for FSPCM (Fig. 5). Paraffin tends to have a slow temperature decrement, which shows a lower discharge rate by 1.29 °C/min. The optimal outcome is achieved by FSPCM, which has a higher discharging rate ranging from 1.88 °C/min to 3.2 °C/min. Since the freezing temperature of the samples is reduced, this contributes directly to the acceleration of discharging rate for FSPCM. In more detail, FSPCM₁ and FSPCM₂ indicate a similar

pattern with a significant temperature drop for 24 minutes of the discharging process, where its temperature drops to 56 °C. In contrast, FSPCM₃ demonstrates an even faster discharging process where its temperature drops significantly for 10 minutes. As a result, it has the highest discharging rate with an outstanding value of up to 3.2 °C/min, which is almost twice as much as that of pure paraffin. This is advantageous for the actual latent heat storage unit since it promotes a better response time and delivers a higher power rate during the operation [25]. Furthermore, the fast temperature decrement during the early stage of the discharging process makes the system more favorable since it promotes a better response time as commonly designed for recuperative heat exchangers.

The side effect of fast heat release can be observed during storing test (Fig. 6). All FSPCM show a fast temperature drop compared to paraffin. This is unfavorable since the system experiences a faster heat loss in the storing mode during the operation. The heat loss rate for paraffin is obtained at a value of 0.59 °C/min, much lower than for FSPCM, with an average heat loss rate of around 0.96 °C/min (FSPCM₃). However, FSPCM₂ and FSPCM₃ have a more stable temperature drop than FSPCM₁. It can be observed that the temperature profile for FSPCM₁ during the storing test dropped faster for 20 minutes with a temperature gradient of about 76 °C. This leads to the fact that the system needs an extensive insulator to minimize the effect of heat loss in the storing mode during the operation.

The change in the thermal properties and performance of paraffin indicates a significant effect of HDPE addition. Adding HDPE at a ratio of 15 wt % reduces the supercooling degree substantially, implying that a steady phase transition can be obtained with a minimum temperature gradient during the operation. This promotes the sample's ability to interact with the heat during the charging/discharging, which promotes a faster charging/discharging process. As a result, the system can reach the maximum state of charge (SoC) for less time and its response time increases during the discharging mode. This is highly advantageous since the supercooling degree can be minimized while maintaining superior performance, which accelerates the charging and discharging rate by adding 15 wt % HDPE to paraffin.

The main consequence of adding HDPE to paraffin is a decrease in the latent heat of fusion since most HDPE melts together with paraffin during the endothermic process. This should be taken into account to adjust the storage capacity in order to maintain the actual capacity of the latent heat storage tank. It also can be adjusted by raising the operating temperature of the tank since certain parts of HDPE also melt at a temperature around 110 °C. The second consideration should also be taken by using a suitable thermal insulator to reduce heat loss in the storing mode. The adjustment can be used to maintain the system's overall performance by using FSPCM made of paraffin and HDPE.

Nevertheless, there are still limitations in the study. The presented results indicate transient temperature improve-

ment for FSPCM. Although the temperature gradient is much lower than for pure paraffin, this condition should be addressed carefully determining the state of charge for the actual application. Thus, further tests are required to observe the influence of adding HDPE through the testing apparatus, which uses a heat exchanger in the storage tank. Moreover, an in-depth analysis of void formation during the operation is required to observe the effect of HDPE on paraffin in more detail. Therefore, further development is essentially focused on performance evaluation by using a heat exchanger and phase transition in an internal storage tank related to void formation.

7. Conclusions

1. Adding HDPE causes a decrease in the melting and freezing temperature of paraffin, where the lowest temperature is obtained by adding 15 wt % at 62.15 °C (melting) and 58.42 °C (freezing).

2. The latent heat of fusion for the paraffin and HDPE composite is decreased by 7.49 J·g⁻¹ (5 wt % HDPE), 15.82 J·g⁻¹ (10 wt % HDPE) and 18.23 J·g⁻¹ (15 wt % HDPE).

3. The thermal performance of the paraffin and HDPE composite is improved with the highest charging rate of 2.38 °C/min (charging) and discharging rate of 3.2 °C/min, while the heat loss rate in the storing mode is obtained at a temperature of 0.96 °C/min.

Conflict of interest

The authors declare that they have no conflict of interest in relation to this research, whether financial, personal, authorship or otherwise, that could affect the research and its results presented in this paper.

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Data availability

Data will be made available on reasonable request.

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