

# Synthesis and Characterization of Encapsulated PCM

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**Abstract:** The purpose of this research was to develop and examine the TES properties of a unique organic PCM that has been encapsulated (Encapsulated PCM). The methyl palmitate PCM core and the melamine-formaldehyde shell of the microcapsules were synthesized by a simple in-situ polymerization process. According to the results of the microstructural examination, it can be assumed that the as-prepared microcapsules were of spherical shape, and the presence of carbon, oxygen, and nitrogen elements proves that the encapsulated PCM was successfully formed. The improvements in chemical stability, surface roughness, and average particle size (490.2 nm) have allowed for greater storage stability. Having a high latent heat of fusion of 70 kJ/kg, the microcapsules have a high thermal storage capacity of 88%. The operational temperature range of the pure PCM was significantly lower than the microcapsules' thermal stability of 130 °C. Moreover, the Micro PCM met the thermal insulation standards due to its low observed thermal conductivity of 0.1587 W/m K. During the thermal cycling test, microcapsules showed incredible chemical stability (up to 200 heating/cooling cycles) and thermal reliability (reliability index = 92.9%). The results of the tests suggest that the encapsulated PCM with the improved characteristics can be a good candidate for low temperature thermal energy storage.

**Index Terms:** PCM, Encapsulation.

## I. INTRODUCTION

In recent decades, researchers have paid more attention to the process of microencapsulation, in which a functional component is enclosed in a membrane or polymeric shell material. Cell transplantation, food processing, drug delivery, pharmaceutical products, cosmetics, adhesives, self-healing coatings, fermentation, pesticides, textile printing and dyeing, energy storage, and so on are just some of the major application domains where microencapsulation is well established and being implemented.

With its energy storage qualities by virtue of their solid-liquid phase transition behaviour, phase change materials (PCMs) have become highly desirable in recent years for use in thermal energy storage (TES). The organic PCMs in the form of microcapsules are chosen over the inorganic PCMs and the eutectic PCMs for use in thermal energy storage (TES).

This is because phase change materials (PCMs) can undergo the process of phase transition within the shell material without any leakage. Also, the relatively small size of the microcapsules would greatly enhance the influence of surface energy. In addition, the PCM's thermal-physical and physico-chemical characteristics can be shielded from disturbances caused by physical, chemical, and sudden

changes in the process/environmental conditions by microencapsulation.

On the other hand, organic PCM has some drawbacks, such as poor heat conductivity, problems with handling and leakage during phase transformation, and interactions with other materials, etc. Thermo-physical property improvements in PCMs have been examined in the past with the goal of improving their TES capacities.

PCMs, on the other hand, would degrade if left unprotected from the elements. In a similar manner, extended exposure to extreme temperatures may affect the thermo-physical properties of Encapsulated PCMs. In this regard, the cycling stability test of a storage unit is essential for ensuring its long-term functioning. In a standard thermal cycling test, the latent heat storage system goes via at least one melt-freeze cycle per day [1].

Melt-freeze cycles of Encapsulated PCM, however, are performed in a lab setting under controlled conditions during an accelerated thermal cycle test. If the PCM is not well encapsulated, it could escape and undergo heat and/or chemical interaction with the surrounding matrix materials, losing its essential properties in the process.

Methyl Palmitate was used as the PCM and melamine-formaldehyde was used as the shell material in the reported fabrication of encapsulated PCM. When the MEPCM was put through 100 thermal cycles, there was reportedly no substantial thermal drop in enthalpy or leakage of the PCM from shell material.

Notably, melamine's great mechanical strength makes it a viable candidate as a shell material to prevent leakage problems in the PCM. Melamine also extinguishes itself if it catches fire. Furthermore, melamine has a propensity to decrease water content, which can improve the fluidity and workability of the surrounding materials in specific contexts [2].

In this context, a variety of synthesis methods are available for the manufacture of the MPCMs, and they can be roughly divided into:

Each of the aforementioned approaches to synthesis has advantages and potential uses in certain contexts. The in-situ polymerization approach for manufacturing MPCMs has gained traction in recent years, expanding their use across the board of TES applications. Because in-situ polymerization is one of the most efficient techniques because the reactants are not mixed in with the PCM and the whole process occurs in a single, unbroken stream.

Numerous recent studies have demonstrated the encapsulation of various PCMs using different shell

materials, giving credence to the effectiveness of the in situ polymerization technique.

Urea formaldehyde (UF) and melamine formaldehyde are two of the most common candidate shell materials (MF). Cross-linked microcapsules consisting of a PCM core and a UF or MF shell can be manufactured with ease using a condensation polymerization procedure.

Microcapsules with interesting compositions, such as paraffin/melamine-UF, Methyl Palmitate/poly(styrene-co-divinylbenzene-co) acrylamide, n-icosane with a poly(methyl methacrylate-co-methacrylic acid), and n-dodecane/MF shell, have been produced and effectively used to store.

The ratio of the mass of the microcapsules after encapsulation to the mass of the PCM before encapsulation is a fundamental determinant of the quality of any microencapsulation procedure. Furthermore, the quality of the microencapsulation is determined by crucial factors such as (a) the mean diameter of the particle, (b) the thickness of the shell, (c) the type of the precursors, surfactants, reagents, solvents, pH regulators, etc., and their proportions, (d) the reaction time, and so on.

Although there have been many studies on the synthesis of MPCMs with respectable encapsulation quality and properties, these studies have been restricted to medium and palmitate. The white powder was then separated from the supernatant and collected. After being washed twice with acetone, this powder was used to create the final encapsulated PCM.

## II. EXPERIMENTAL PROCEDURE

### 2.1. Preparation of Encapsulated PCM

Encapsulated PCM was made by encapsulating Methyl palmitate PCM in a melamine formaldehyde shell using a simple in-situ polymerization technique. Different methods of PCM encapsulation have been tested. Trials revealed that adjusting the cross-linking agent concentration influenced microcapsule formation. Furthermore, the production of microcapsules was greatly aided by the process taking place in acidic conditions.

Although it was not possible to completely prevent agglomeration/coalescence, good control over encapsulated PCM formation was achieved by lowering the copolymer percentage, fine-tuning the hydrophilic-lipophilic balance (HLB) scale of surfactant molecules, and adjusting the pH value for the reaction. As a result of these measurements, the response was adjusted as follows: Equal parts of Tween 20 and Span 60 were used as surfactants in the O/W mix. An additional 50 ml of Methyl palmitate PCM was added, and the whole thing was mixed at a speed of 8000 rpm to 8500 rpm in a high-speed shear mixer.

Oil in water emulsion had a pH value between 5.8 and 7. The shell precursor, on the other hand, was made by dissolving 30 mm of formaldehyde solution into 100 milliliters of dimethyl-dimethyl-siloxane (DDW).

high temperature energy storage applications. Even fewer studies have been done on the topic of encapsulated organic PCMs with MF or UF shell material for low temperature energy storage applications [2].

This study considered Methyl palmitate, or lauryl alcohol, a saturated 12-carbon organic fatty alcohol that can be prepared from the fatty acids found in coconut oil. Methyl palmitate is used in a wide variety of industries, from the cosmetics and lubricant oil industries to flavor enhancers, perfumes, construction, agriculture, and the pharmaceutical industry. Despite Methyl palmitate's widespread application, our research shows that its capacity as a storage medium for energy is still relatively unexplored. Consequently, this research is unique in its investigation of the energy-storage characteristics of Methyl Palmitate as PCM for use at low temperatures.

To be more specific, this study aimed to develop a simple method for synthesis of encapsulated PCM with Methyl palmitate in the center and melamine-formaldehyde on the exterior. Following synthesis, the microcapsules were studied for microstructural, chemical, and thermal energy storage capabilities. The effects of 200 thermal cycles on the chemical stability and thermal reliability of encapsulated PCM were also investigated. The findings are shown and explained in detail.

## III. RESULTS AND DISCUSSION

### 3.1. FESEM Results of the encapsulated PCM

The as-synthesized microcapsules' surface morphology was examined using FESEM with an EDS detector. The FESEM images in Fig. 1 demonstrate that the microcapsules were roughly spherical in shape and displayed a modest degree of surface roughness. Encapsulated PCM capsules were primarily formed due to interfacial tension between the PCM, polymer, and surfactant.

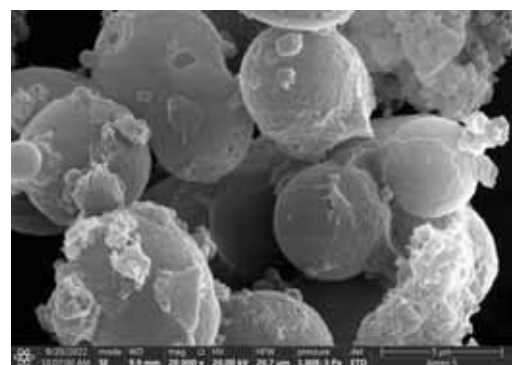


Figure1. FESEM images of Encapsulated PCM

### 3.2. Particle size distribution of the Encapsulated PCM

From Fig. 2, the microcapsules ranged in size from far under 60 nm to well over 980 nm. Capsules with a first peak averaged 119.6 nm in diameter and a second peak averaged 550.1 nm in diameter at 65.9% and 34.1% intensity, respectively. To be precise, we calculated a Z-average microcapsule diameter of 380 nm across both peaks. Furthermore, this provided strong backing for the study's simple synthesis of Encapsulated PCM, which formed at the micro/nano scale. In addition, as shown in Fig. 1 (b) and because of the smaller particle size, it was predicted that the

surface energy would increase as the radius of curvature between two adjacent particles was decreased [4].

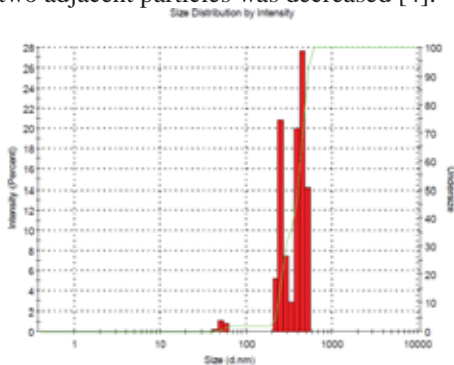


Figure 2. Particle size distribution of the Encapsulated PCM.

### 3.3. Crystal structure of the Encapsulated PCM

In Fig. 3, we see the XRD pattern of the Encapsulated PCM, which exhibits a broad and massive peak at  $21.24^\circ$ , indicating that the material is mostly amorphous. This might be because of the Encapsulated PCM's disordered molecular structure, which caused the lattice parameter to shift. Physical interaction between the PCM and the shell material or a lack of incident electrons diffracted along that lattice direction of the Encapsulated PCM may explain the lack of a highly crystalline structure [5].

Encapsulated PCM's lack of crystalline ability and weak peak intensity may have been caused, at least in part, by the sample powder's coarse grain structure. Encapsulated PCM's amorphous structure was predicted to have a high internal energy, and the powdered form of the microcapsules being manufactured showed a promising dispersal performance. Additionally, the shell stability and resistance against cracking were characterized by the low peak intensity, but at the expense of a decrease in the encapsulating properties.

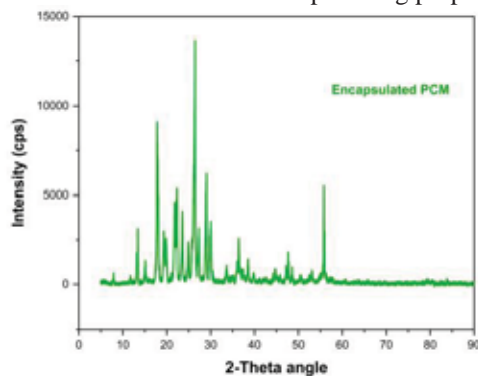


Figure 3. XRD pattern of the Encapsulated PCM.

### 3.4. FTIR Analysis of Encapsulated PCM

As can be seen in Fig. 4, the FTIR spectrum of Methyl palmitate PCM has a very strong and broad peak at  $3360\text{ cm}^{-1}$ , which corresponds to the O-H stretching frequency. It was the C-H stretching vibration of the aliphatic chain that contributed to the peaks at  $2925$  and  $2855\text{ cm}^{-1}$ .

Medium bending vibration of methylene/methyl (at  $1459\text{ cm}^{-1}$ ) and C-O stretching of alcohol (at  $1066\text{ cm}^{-1}$ ) are responsible for the peaks in the infrared spectrum. The in-plane rocking vibration of the methylene group was blamed for the  $728\text{ cm}^{-1}$  peak. The N-H bending vibration of the

amine group was observed as a peak at  $3145\text{ cm}^{-1}$  in the FTIR spectra of the melamine.

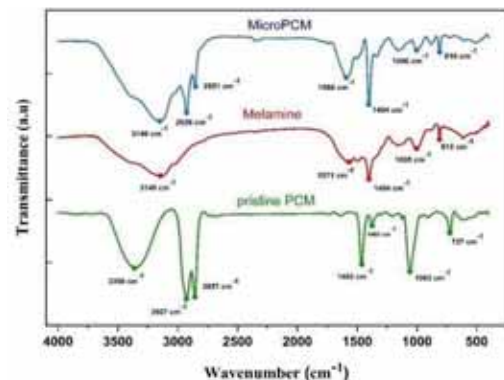


Figure 4. Chemical Compatibility test results

The N-H bond in-plane bending vibration was attributed to the prominent peak at  $1560\text{ cm}^{-1}$ . Similarities between the  $810\text{ cm}^{-1}$  peak and the stretching vibration of triazine rings were found. Table 1 provides a concise summary of the FTIR analysis results. The absorption peaks of PCM and the melamine-formaldehyde shell can be seen in the FTIR spectra of the Encapsulated PCM, with no evidence of a shift. This not only confirmed that the PCM was successfully encapsulated in the melamine-formaldehyde shell material, but also demonstrated there was no chemical contact between the core and the shell material [6].

### 3.5. Phase change and latent heat properties of the Encapsulated PCM

Results from a DSC study comparing the phase change behaviour of pure PCM and Encapsulated PCM are shown in Fig. 5. As can be seen from the data, the pure Methyl palmitate PCM showed a single-peak congruent phase change both during cooling and melting. It's interesting to see that the pure PCM goes straight from an isotropic liquid to a triclinic solid as it freezes [7].

Encapsulated PCM showed unusual behaviour during its phase transformation, with dual peaks appearing during the cooling process and a single peak appearing during melting, just like pure PCM. It was at an onset temperature of  $20^\circ\text{C}$  that the Encapsulated PCM's two peaks, the rotator and stable crystal phase, were activated.

The occurrence of the dual crystallization peak can be attributed to (a) the containment of the Methyl palmitate PCM within a closed shell, and the transition of the core PCM inside the shell from the heterogeneously nucleated liquid phase to the rotator phase and then to the Beta phase at temperatures of  $20^\circ\text{C}$  and  $14.21^\circ\text{C}$ , respectively, as the temperature of the Encapsulated PCM [8].

The DSC results (Fig. 5) indicated that peaks contributed 46.9% and 51.4%, respectively, to the latent heat of crystallization at approximately  $20^\circ\text{C}$  and  $14^\circ\text{C}$ .

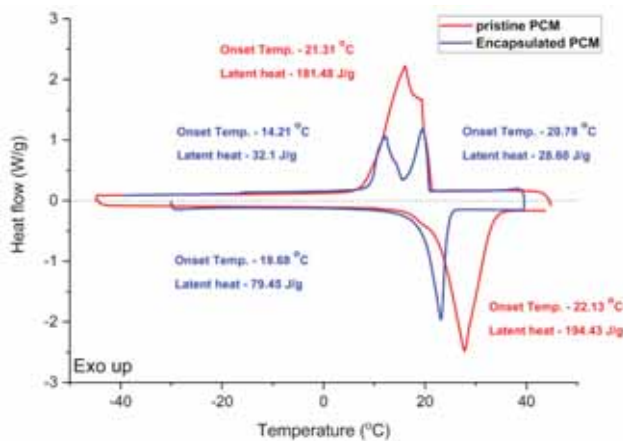


Figure 5. DSC graphs of PCM and Encapsulated PCM.

However, the nucleation kinetics of the PCM inside the shell material was also significantly affected by the degree of supercooling (the difference between the onset temperatures of melting and cooling processes).

Keeping in view the enthalpies associated with both the melting and freezing processes, we can calculate the Encapsulated PCM's encapsulation efficiency (EE) to be 36.9 %, as shown in Eq. (1). As a result, this is favored as a more appropriate parameter than the encapsulation ratio for gauging the PCM's performance in the MF shell. Latent heat enthalpies of encapsulated PCMs may be changed by adjusting the core-to-shell ratio.

To this end, we used Eq. (3) to calculate the thermal energy storage capacity ( $\epsilon$ ) of the Methyl palmitate PCM contained within the MF shell material, and discovered that the Encapsulated PCM displayed a very high i.e. 88.7%. This showed that almost complete the Methyl Palmitate PCM enclosed could efficiently store and release the energy via phase transformation [9].

$$R = \frac{\Delta H_{m, NP}}{\Delta H_{m, PCM}} \times 100 \quad \text{---- (1)}$$

$$EE = \frac{\Delta H_{m, NP} + \Delta H_{c, NP}}{\Delta H_{m, PCM} + \Delta H_{c, PCM}} \times 100\% \quad \text{---- (2)}$$

$$\epsilon = \frac{\Delta H_{m, NP} + \Delta H_{c, NP}}{R} \times 100\% \quad \text{---- (3)}$$

where,  $\Delta H_{m, PCM}$  and  $\Delta H_{c, PCM}$  are the latent heat of melting and freezing of pure PCM and  $\Delta H_{m, micro-PCM}$  and  $\Delta H_{c, micro-PCM}$  are the latent heat of melting and freezing of Encapsulated PCM, respectively.

The pure PCM's behavior during solid-to-liquid and solid-to-solid phase transitions within the shell material may be a factor in the Encapsulated PCM's lowered latent heat. Some researchers suggest that the lower PCM content of the shell material may also contribute to the reduced latent heat enthalpy [10].

As indicated in Table 4, significant thermal properties of the encapsulated PCM were in good agreement with those of other MPCMs of related types. The Encapsulated PCM

created in this study has demonstrated promising phase transition characteristics, significant latent heat potential, and excellent thermal energy storage capacity, according to the DSC results.

### 3.6. Thermal stability of the Encapsulated PCM

As can be seen in Fig. 6, TGA was used to test the thermal stability of both pure PCM and Encapsulated PCM. The findings of the tests, which were conducted using the tangential approach, showed that the single-step mass loss/degradation of the pure PCM began at 100 °C and was finished at 184 °C, with a mass loss of 99% leaving the residual contents behind in the crucible. The Encapsulated PCM was broken down into two stages: This mass loss was primarily attributable to the evaporation of water and other molecules from the microcapsules' outer surfaces during the first step of their decomposition [11].

When the temperature was raised even higher, cracks appeared in the MF shell material, exposing the PCM contained within to the high heat and leading to its decomposition at 130°C. The FESEM picture of the broken microcapsules following full disintegration of the PCM is shown in the Fig. 6 to support this finding.

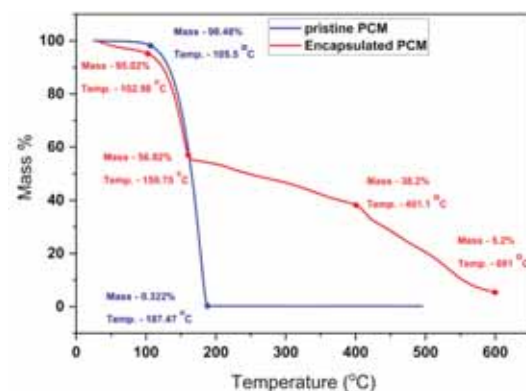


Figure.6. TGA graphs of PCM and the Encapsulated PCM.

At 160 °C, all the PCM in the shell dissolved, resulting in a 39% mass loss. A sudden mass loss was detected between 387.9 °C and 415.8 °C, and the decomposition proceeded up to 610 °C. The second step of decomposition began at about 188 °C and was associated with the loss of the shell material (MF) [12, 13].

The TGA results show that the Encapsulated PCM had great thermal stability, since the decomposition temperature was much higher than the PCM's operational temperature range (21°C to 26°C), making it ideal for low temperature TES applications [14].

### 3.7. Thermal conductivity of the Encapsulated PCM

Encapsulated PCM and pure PCM were found to have thermal conductivities of 0.1789 and 0.1587 W/m K, respectively. Based on the test results, it indicates that Encapsulated PCM had a somewhat lower thermal conductivity than pure PCM. Since the outer MF polymeric shell material has very low thermal conductivity, it is possible that its presence contributes to this marginal reduction [15].

One positive effect of Encapsulated PCM's low thermal conductivity was that it made for an effective heat insulator. However, the PCM's melting and freezing behaviour within the shell would have been slightly changed due to the shell material's insulating tendency towards heat transfer, as a result of the shell's poor thermal conductivity. This may also have contributed to the lowered latent heat enthalpy of the Encapsulated PCM. Nonetheless, the Encapsulated PCM has proven to be thermally stable and to be a good heat insulator [16].

#### IV. CONCLUSIONS

1. Microstructural analysis clearly showed that Encapsulated PCM formed into spherical capsules with a slightly rough exterior. The elemental composition of the Encapsulated PCM has been further verified by the presence of carbon, nitrogen and oxygen.
2. The particle size study found that the microcapsules had a Z-average size of 380 nm, which provided strong support to the development of the micro/nano sized Encapsulated PCM particles with desirable surface energy features.
3. The XRD findings highlighted the Encapsulated PCM's amorphous form. However, it was anticipated that the microcapsules would have a lot of energy on the inside, which would make their shells more resistant to cracking.
4. Minor peak changes seen in the FTIR spectra might be attributable to physical amalgamation between the PCM (core) and the MF (shell) during synthesis, confirming their chemical stability.
5. The Encapsulated PCM showed impressive phase change behaviour, with a huge latent heat potential. However, the Encapsulated PCM still showed good phase transition characteristics as applicable for thermal energy storage despite the presence and phases during the crystallization process and a relatively low supercooling degree.
6. A high thermal energy storage capability ( $\epsilon$ ) of 87 % was demonstrated by the Encapsulated PCM, characterizing its efficiency in storing and releasing the energy during phase change.
7. The operating temperature limits of pure PCM for the TES application were far lower than those of the Encapsulated PCM, which remained thermally stable up to 131°C. The Encapsulated PCM was able to insulate against heat because of its low thermal conductivity as well.
8. For low-temperature thermal energy storage applications, as-synthesized Encapsulated PCM with the aforementioned advantages can be a promising and practical option.

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