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Production and Characterization of Encapsulated Phase Change Materials for Thermal Energy Storage

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Termal Enerji Depolaması için Kapsüllenmiş Faz Değişim Malzemelerinin Üretimi ve Karakterizasyonu

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Abstract

Phase-change materials (PCM) have become commercially available as a result of advanced microencapsulation applications that have increased the thermal and mechanical properties of these materials. Microencapsulation can be achieved by using different techniques and various core and shell materials. In this study, the synthesis of microcapsules with a coconut oil core and melamine formaldehyde shell has been carried out using the in-situ polymerization method, and the main important properties of this microencapsulated PCM, such as physical, chemical, and thermal, have been analyzed. The microcapsules were characterized using scanning electron microscopy (SEM), Energy Dispersive X-Ray (EDX), Fourier Transform Infrared (FT-IR) spectroscopy, X-Ray diffraction, Differential Scanning Calorimetry (DSC) and Thermo-gravimetric (TG) analysis. The synthesized capsules were 1-5 µm in diameter, spherical with dents on their surfaces. Analyses showed that the core of the microcapsules melted at 21.1 °C and crystallized at 9.7 °C. Latent heats for melting and crystallization were obtained to be 105.1 and 76.3 J/g, respectively. TGA showed that encapsulated PCM decomposed in two steps and that its chemical stability was good. Encapsulation ratio was 78.59 percent, whereas the specific heat capacity and thermal conductivity of synthesized capsules were 2.726 J/gK and 0.296 W/mK, respectively. The capsules were found to have potential for slurry preparation for thermal applications.

Keywords: *Phase-change materials; Encapsulation; In-situ polymerization; Thermal energy storage.*

Öz

Faz değiştiren malzemeler (FDM), bu malzemelerin termal ve mekanik özelliklerini artıran gelişmiş mikrokapsülleme uygulamalarının bir sonucu olarak ticari olarak kullanılabilir hale gelmiştir. Mikrokapsülleme, farklı teknikler ve çeşitli çekirdek ve kabuk malzemeleri kullanılarak gerçekleştirilebilir. Bu çalışmada, in-situ polimerizasyon yöntemi kullanılarak hindistancevizi yağı çekirdekli ve melamin formaldehit kabuklu mikrokapsüllerin sentezi gerçekleştirilmiş ve bu mikrokapsüllenmiş FDM'nin fiziksel, kimyasal ve termal gibi temel önemli özellikleri analiz edilmiştir. Mikrokapsüller, taramalı elektron mikroskobu (SEM), Enerji Dağıtıcı X-Işını (EDX), Fourier Dönüşümlü Kızılötesi (FT-IR) spektroskopisi, X-Işını kırınımı, Diferansiyel Taramalı Kalorimetri (DSC) ve Termo-gravimetrik (TG) analiz kullanılarak karakterize edilmiştir. Sentezlenen kapsüller 1-5 µm çapında, küresel ve yüzeylerinde ezikler vardı. Analizler, mikrokapsüllerin çekirdeğinin 21,1 °C'de eridiğini ve 9,7 °C'de kristalleştiğini göstermiştir. Erime ve kristalleşme için gizli ısılar sırasıyla 105,1 ve 76,3 J/g olarak elde edildi. TGA, kapsüllenmiş FDM'nin iki adımda ayrıştığını ve kimyasal kararlılığının iyi olduğunu göstermiştir. Kapsüllenme oranı %78,59 iken, sentezlenen kapsüllerin özgül ısı kapasitesi ve termal iletkenliği sırasıyla 2,726 J/gK ve 0,296 W/mK olarak belirlenmiştir. Kapsüllerin termal uygulamalar için bulamaç hazırlama potansiyeline sahip olduğu sonucuna varılmıştır.

Anahtar Kelimeler: *Faz değişim malzemeleri; Kapsülleme; Yerinde polimerizasyon; Termal enerji depolama.*

1. Introduction

Today, in many applications in practice, various methods, such as dyeing or isolation, are employed for the targeted release of substances that are to be used as the main agent based on certain timing or environmental conditions, or for avoiding their interacting with the environment until they reach the target zone. These methods also diversify with the emergence of new technologies and new necessities. Encapsulation is one such method. Encapsulation is the process of coating solid or liquid particles with a thin film (Chen *et al.* 2014), generally of polymeric materials (Chen and Fang 2011). Capsules, which have been utilized in different fields for many years, are now produced at micro- and nanoscales with the advancement of technology. Over the last years they have received much attention and found a much wider application area (Song *et al.* 2018). The encapsulation technique has recently been used in the production of capsules, the core material of which is a phase-change material (Chen *et al.* 2014). Industrial applications of ePCM and its derivatives are increasing rapidly. Micro- and nano-capsules have been introduced into industries such as pharmaceuticals, textiles, paint, food, cosmetics, electronics, heating-cooling, and energy systems (Benita 2005; Chen and Fang 2011; Chen et al. 2014; Delgado et al. 2013). Composed of a wide range of

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shell and core materials, they have the potential to eliminate, or at least minimize, the challenges or to significantly contribute to increasing efficiency.

Since ePCMs can be synthesized using different shell and core materials, every unique capsule brings about unique features; hence, different uses. And the overall manufacturing process has certain effects on the physical, and sometimes chemical, properties of the resulting capsules. For this reason, PCM capsules can be structurally different, even if not methodologically. In this respect, even in the case of capsules obtained using the same shell and core materials, it will be possible to obtain unique materials since the final capsules would probably have different structural, dimensional, superficial, chemical, and physico-chemical properties, etc.

ePCMs are most commonly used in thermal energy storage, heat transfer, and temperature regulation and control applications; however, they are also very attractive for improving the thermal performance of woven fabrics (Chen and Fang 2011).

Studies on the use of ePCM in solutions, which are prepared by mixing capsules with carrier fluids and mostly referred to as ePCM slurry (ePCM-S), have also been conducted. In the last decade, the use of ePCM-S as the working fluid operating in heat transfer applications has also been in focus. Applications include secondary cooling cycles in electronic circuit cooling systems and heating ventilation systems (Tyagi et al. 2011; Zhang et al. 2010) as well as cooling photovoltaics (Gschwander and Schossig) and solar thermal panels (Baronetto et al. 2014). It was shown in a 4-month experiment in a ceiling system with encapsulated PCM that constant temperature was achieved and flow rate was significantly reduced (Griffiths and Eames 2007).

The shell of the capsules prevents the core material from interacting with the carrier liquid, which increases the stability of the material and prevents the agglomeration of the particles. In addition, when the capsules' core is in liquid form, they allow the material still to be used as solid particles and balance the volume change, which will occur during the phase change, within the shell. While, it is also among the findings that the total heat transfer coefficient decreases due to the decrease in effective thermal conductivity and the increase in fluid viscosity (Alvarado *et al.* 2007), encapsulation increases the surface-to-volume ratio of the phase-change material, (Farid *et al.* 2004) and by taking advantage of the latent heat of fusion of the phase change material, significantly increases the overall thermal capacity and heat transfer efficiency of the carrier fluid and thereby the thermal system.

Therefore, in addition to the material of the capsules to be synthesized, the method of encapsulation and the preparation of the obtained capsules and their slurries also arise as separate engineering problems that need to be tackled and solved. For this purpose, many studies have been carried out on the preparation and properties of ePCM-S, and important results have already been achieved (Chen and Fang 2011).

Generally, according to the mechanism of encapsulation, methods by which encapsulation of the target material is achieved are classified into three main groups: physical, chemical and physicochemical. Although the encapsulation methods are quite diverse, there are positive and negative aspects to each method that can lead to being successful or unsuccessful in ensuring a certain desired property or feature. In this study, microencapsulated PCM (mePCM) was synthesized via in-situ polymerization method using coconut oil (CO) as the core and melamine formaldehyde (MF) as the shell. The resulting capsules were characterized by SEM, FT-IR, EDX, DSC, and TG analyses.

2. Materials and Methods

In this section, information about the materials, including the shell and core materials, which are the direct components used in capsule production, and other chemicals used as auxiliary agents in the synthesis process, such as surfactants, as well as all laboratory equipment, test and measurement devices, and a detailed outline of methods used for synthesis and characterization during and after synthesis, is given.

2.1. Selection of Shell and Core Materials

The selection of shell materials and core materials should be made together (Chen and Fang 2011) depending on the purpose of use. It is a very demanding and meticulous process, since the properties expected from the resulting material are very different and diverse, and the core-shell material pair should be resistant to all conditions of the medium to be used in so that it serves the purpose.

Materials to be selected as shell material should have major general characteristics such as (1) good flexibility, (2) good sealing, (3) durability, and (4) low reactivity. Many organic and inorganic materials with the above properties are suitable for use as shell materials. While the most commonly used shell material is polymers, polystyrene, polymethylmethacrylate, Arabic gelatin-gum, gelatin, amino plastics, urea formaldehyde resin, melamine formaldehyde resin, gelatin formaldehyde resin, and other similar materials can be listed among shell materials reported in the literature (Su et al. 2017).

The selection of the core material to be used in the production of ePCMs is also very important, and it is a process that must be done meticulously, taking several parameters into consideration. As with shell materials, there are some main features that core materials must have, in addition to the intended purpose and ambient and operating conditions that will apply during use. These can be listed as:

(1) appropriate phase change temperature or temperature range,

(2) high phase change latent heat,

(3) good thermal conductivity,

(4) low density change between phases and

(5) low reactivity (Chen and Fang 2011, Yeşilyurt *et al.* 2019).

It can be seen in the literature that paraffins are suitable and practical substances widely preferred in the production of ePCM (Farid *et al.* 2004, Chen and Fang 2011, Pan 2013, Daou *et al.* 2016). As a matter of fact, paraffin wax is the most common core material preferred in encapsulation processes (Ghasemi *et al.* 2022). Non-paraffin organic PCMs are mostly in the form of acids, which can roughly be classified into six main groups: lauric, palmitic, stearic, myristic, capric, and caprylic (Sharma et al. 2015). The materials in the fatty acid groups are known for their excellent phase-change properties (Hasnain 1998). Despite the fact that inorganic PCMs have advantages such as high thermal conductivity and high energy storage density (Bruno et al. 2015), they are not preferred due to subcooling and phase separation during liquid to solid phase change (Atkin and Farid 2015). On the other hand, ideal core materials can also be obtained from mixtures of organic or inorganic PCM to take advantage of both groups of materials (Chen and Fang 2011).

When the core material of a capsule is considered thermodynamically, it is important that the latent heat of fusion required to melt the unit mass of material is as high as possible (Yeşilyurt and Çomaklı 2023), as this will ensure the maximum amount of heat is absorbed or stored. It is also preferable that the volumetric change of the core PCM between phases is very small and the vapor pressure is low since it will be contained in a closed volume. High thermal conductivity and density, as well as consistency and stability of phase changes over time, have been identified as critical factors for processes. Kinetic nucleation and crystal growth rate should be large. It should not be toxic, its economic life should be long, it should not be harmful to health, and it should be easily supplied and low-cost in comparison to its benefits (Hale et al. 1971; Akçay 2006; Garg et al. 1985; Pasupathy et al. 2008).

Despite the fact that, once the shell has been formed, residual formaldehyde can be found, which causes environmental and health problems during the polymerization process (Qiu *et al.* 2019), melamine-formaldehyde (MF), as well as urea-formaldehyde (UF), is widely used as shell material due to its good chemical stability and mechanical strength (Chandel and Agarwal 2017). In the present study, coconut oil and melamine formaldehyde were selected as the core and shell materials, respectively.

2.2. Synthesis Method of Coconut oil/Melamine Formaldehyde Microcapsules

Although the microencapsulation technique has been known for decades, the process is thought to be too expensive for practical application. However, thanks to the development of new synthesis methods and the improvement of existing methods with new technical and technological means, costs have been reduced, and mePCM has found a variety of application areas, to which new ones are being added day by day. Nowadays, the number of methods is around twenty-five and is classified into three main groups: chemical, physical, and physicochemical. However, mostly chemical methods are implemented, and in-situ polymerization, being highly developed and commercialized among other chemical methods (Duan 2015), is the most commonly used method (Zondag et al. 2016). With the in-situ polymerization method, it is possible to produce the best quality microcapsules in terms of diffusion tightness of their walls and capsule sizes ranging from 5 to 100 □m. Therefore, in-situ polymerization is widely used in the manufacture of ePCM (Qiu et al. 2019).

Micro- and nano-encapsulation is a technique widely used in various industries (such as food, pharmaceutical, textile, agriculture, biomedical, and coating technologies). This technique aims to protect active substances from environmental conditions, to provide controlled release, and to increase their stability. There are many methods in the production of micro- and nano-capsules, but the in-situ polymerization method is an effective and widely used method that allows the capsule core to be coated with a polymeric shell (Jia et al. 2016; Alehosseini and Jafari 2019).

The capsules obtained with the in-situ polymerization method, which is a highly efficient method in the production of micro- and nano-capsules, have a wide

range of applications in many fields such as food, pharmaceutical, energy, and material sciences thanks to their advantages such as controlled release, stability, and biocompatibility (Silva et al. 2017; Phan et al. 2022; Alehosseini and Jafari 2019; Srinivasaraonaik et al. 2020). The capsules produced with this method create a great innovation in the industry by ensuring the protection of the core material they contain.

In this method, the core material is in an emulsion or suspension while the polymerization reaction is carried out directly in the dispersion medium. As a result, the core material is surrounded by a polymeric shell and a stable capsule structure is formed (Li *et al.* 2018). In-situ polymerization is a method that combines two immiscible liquids, such as water and organic solvents, which involves complementary, direct-acting, organic intermediates that will react with each other to form pre-condensates. In this method all polymerization occurs in a continuous phase rather than in two phases, as is the case in interfacial polymerization (Nadaroğlu et al. 2022). The distinction is that there is no reactant in the core material during in-situ polymerization (Qiu et al. 2019). In in-situ polymerization, the monomer and catalyst dissolved in the continuous phase are separated from the core material, and the pre-polymer gradually forms on the surface of the core material during the advancement of the polymerization reaction (Erdoğan 2017). Numerous studies that have employed this method in encapsulation processes have been reported in the literature (Zhao and Zhang 2011).

2.3. Synthesis Procedure for the In-Situ Polymerization Method

In-situ polymerization occurs when monomers are polymerized directly in the encapsulation process in the presence of a specific reactant or catalyst. The procedure for applying this method includes the following basic steps:

- Dispersion of Core Material: The core material is dispersed in a water-based medium to form an emulsion. A homogeneous distribution of droplets is achieved by using emulsifiers (surfactants).
- Addition of Monomers and Crosslinkers: The monomers and crosslinkers (melamine-formaldehyde in the present study) required for polymerization are added. The melamine-formaldehyde monomers and crosslinkers were prepared as a pre-polymer solution as shown in Figure 1.(a), which was subsequently introduced into the emulsion system via a dropping funnel under continuous stirring to ensure controlled polymer shell formation as shown in Figure 1.(b).
- Initiation of Polymerization: The reaction is initiated by temperature or pH control.
- Shell Formation: At this stage, monomers polymerize around the core to form a thin polymeric shell around it.
- Solvent Removal: Once the polymer shell is fully formed, the system is continued to be mixed for a certain period of time. Once solvent removal and cross-linking are complete, the capsules are separated by filtration or centrifugation.
- Hardening and Stabilization: The capsules are dried and stabilized.



(a) (b)

Figure 1. Pre-polymer preparation (a) and addition of shell material through the dropping funnel to the medium on the mixer (b)

2.4. Testing Equipment and Characterization Methods

2.4.1. Scanning electron microscope (SEM) and Energy Dispersive X-Ray (EDX)

Scanning electron microscopy (SEM) involves a surface scan with an electron beam performed by devices with high discrimination capabilities in the examination of the microstructures of samples. In an SEM device, Energy Dispersive X-ray (EDX) analysis can also be performed, which helps determine the elemental composition of materials. The data obtained by EDX analysis consists of the spectra having peaks that correspond to the elements that make up the actual composition of the analyzed sample.

SEM and EDX analyses, for analysis of the structural morphology and elemental composition of the synthesized microcapsules, were performed by using the Zeiss Sigma 300 SEM device that exists within the inventory of Atatürk University Eastern Anatolia Advanced Technologies Application and Research Center (DAYTAM). In this way, information about the surface morphology, particle size, and physical structures of the sample being examined is obtained. In SEM analysis, as in other alternative analysis methods such as Transmission Electron Microscope (TEM) or Optical Microscope (OM),

it is possible to obtain information about the diameter, shell thickness, clustering, surface profile and roughness, cracking, breakage or similar deformations or pores in the structure, and the sphericality of the capsules as well as the regularity and continuity of the sphericality (Alva et al. 2017).

2.4.2. Fourier Transform Infrared (FT-IR) Spectroscopy Analysis

A Fourier Transform Infrared Spectrophotometer (FT-IR) is used to characterize organic or inorganic compounds (Ono *et al.* 2011). In infrared (IR) absorption spectroscopy, IR rays, which have a longer wavelength than visible light, are absorbed by the material, causing vibration or rotational movements on the bonds in the molecules of the sample's structure. Making use of these energy changes occurring at the molecular level, an IR spectrum that can be used to characterize the material is obtained. The absorption peaks that occur as a result of the frequencies formed by the vibration of the bonds formed between the atoms comprising the substance yield, in a sense, the fingerprint of the sample (Lin and Wang 2012). Every substance has a unique spectrum. In the organic matter spectra, the 2000 cm^{-1} part is more detailed and is referred to as the "fingerprint region." In this region, the spectrum is taken twice as wide so that more detailed information about the structure is obtained (Gündüz 2007; Ergin et al. 2013).

To investigate the chemical structure of mePCM, an FTIR analysis in the range of 4000-400 cm^{-1} was performed, through service procurement, by Ataturk University, DAYTAM, using a Bruker VERTEX 70V FT-IR spectroscopy.

2.4.3. Differential Scanning Calorimeter (DSC) Analysis

The DSC analysis technique involves measuring the sample's energy change as a function of time or temperature against a reference (alumina pan Al_2O_3) at a given heating rate. The DSC device basically measures the energy delivered to the sample to equalize the temperature difference between the reference and the sample. The values obtained as a result of these measurements are plotted as a distribution graph showing on the horizontal axis the temperature and on the vertical axis the energy transferred to the sample. While the intersection point of the tangents drawn at the point where the graph peaks gives the temperature value at which the sample begins to melt ($T_{melting}$), the amount of heat transferred to the sample during melting, that is the integration of the area under the peak, gives the latent heat of fusion ($\Delta H_{melting}$). Using the built-in software functions, the heat capacity (C_p) of the sample can be directly determined through the measurement data.

In the study, Differential Scanning Calorimeter (DSC) analysis was performed by using the NETZSCH STA 409PC/PG device that exists within the inventory of Atatürk University, Engineering Faculty, Chemical Engineering Department, in order to determine the heat absorption and diffusion properties of coconut oil and synthesized microcapsules. Melting enthalpy (ΔH_m) and crystallization enthalpy ($-\Delta H_c$) and relevant temperatures were determined by DSC analysis.

2.4.4. Thermo-Gravimetric (TG) analysis

Thermo-gravimetric analysis (TG) is a thermal analysis in which physical and chemical changes are measured at a constant heating rate as a function of time and temperature. The sample is placed in the heating chamber inside the appliance on a balance arm acting as a sensitive microbalance or in a crucible pot hanging therefrom. The position of the balance arm is kept constant by an electro-optic system by applying a torque that varies depending on the weight of the sample. The change in the torque values expresses the changing mass. On the other hand, the heating chamber is constantly controlled to provide the required temperature profile (i.e. heating rate) as a function of time (Pal et al. 2018).

In TG analysis, in response to the change in temperature, a change in mass (usually in the form of a decrease) occurs. These changes in the mass of the sample are attributed to decomposition of organic and inorganic content, absorption of gases, desorption of volatile contaminants (monomers, solvents, plasticizers, and other additives), et cetera, based on the point and percentage of the change. Detection and determination of a number of polymeric events, such as diffusion and permeability of gases and polymer degradations, can be done in an oxidative, inert, or vacuum environment (Pal et al. 2018; Shojaei and Azhari 2018; Mudalige et al. 2019). Polymers can be identified by pyrolysis of the samples, and thus their characterization can be made. TG tests are also implemented to estimate the long-term stability of polymer samples in various thermal environments (Pal et al. 2018).

That is, TG is the measurement of sample weight/mass against the temperature of the sample or time. The sample is generally heated up at a constant heating rate or maintained at a constant temperature. Meanwhile, the change in the mass of the sample (generally a loss) is recorded as a function of time and/or temperature. From these records, called thermograms, different polymer

types can be identified by examining certain threshold values of the weight and the temperature values corresponding to these values (Chandrasekaran and Sharma 2019). In addition, based on the experimental results reported in the literature and known properties of materials, the residual percentage of weight at temperatures above the temperature at which the sample is completely burnt can be attributed to other impurities and catalyst particles that are present in the sample (Shojaei and Azhari 2018).

Oxidation occurs when the sample is heated under an air/oxygen atmosphere; thus, the catalyst contained in the sample can be used to quantify impurities. When nitrogen gas is used, oxidation is prevented, and thus, it means that weight loss is directly related to defects and/or functional groups in the structure of the sample (Shojaei and Azhari 2018). TGA analysis was carried out using the NETZSCH STA 409PC/PG device that exists within the inventory of Atatürk University, Engineering Faculty, Chemical Engineering Department.

3. Results and Discussion

Using the in-situ polymerization method, CO/MF microcapsules, with sizes ranging from 1 to 4 µm in size, have been successfully synthesized and characterized. Dried microcapsules can be seen in Figure 2.



Figure 2. Dried CO/MF microcapsules

3.1. Results of SEM analysis

Coconut oil encapsulated with melamine formaldehyde (MF) was examined by SEM analysis, images of which are given in Figure 3. When the SEM images were examined, it was seen that there were crater-like pits on the surface of the capsules. SEM images clearly indicate that the pits on the capsule surfaces do not form holes in the shell material but rather appear to be dents. As a result of research on the existence, causes, and effects of such surface imperfections on the performance of the capsules in the thermal cycle, similar surface malformations were found to have been reported in some studies in the literature (Zhao and Zhang 2011, Delgado *et al.* 2012). Zhao and Zhang (2011) reported that the surface profile of malformed capsules improved with increased thermal cycles; hence, they did not pose a threat to capsule structure or functioning.

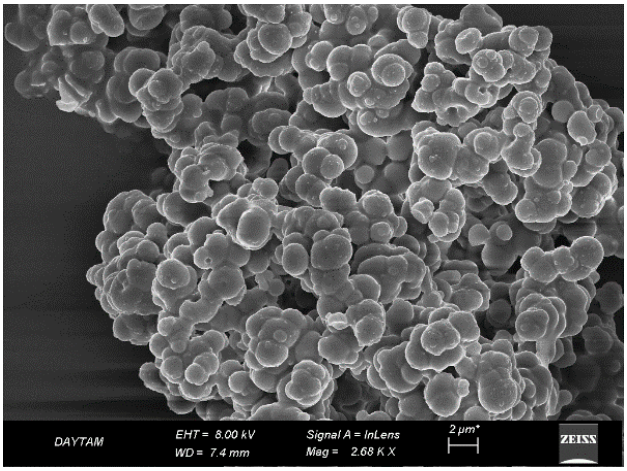


Figure 3. SEM image of the mePCM obtained in this study

Although it is understandable that the shell material, which undergoes thermal expansion and contraction during heat exchange, would have a more uniform structure over time, it is concluded that obtaining a uniform surface morphology in production is important since the improvement of an uneven shell structure may lead to cracks or breaks in certain areas due to excessive thinning and thermal fatigue. Indeed, capsules with insufficient shell thickness were reported to have failed and deteriorated after a certain number of thermal cycles (Kong *et al.* 2016).

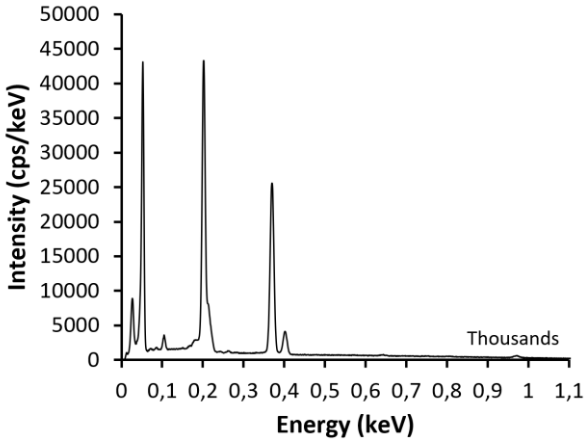


Figure 4. EDX spectrum of mePCM

The chemical compositions of PCM microcapsules were also investigated by performing EDX analysis. The EDX spectrum given in Figure 4 reflects the elemental composition of CO/MF microcapsules. According to the obtained spectrum, it is seen that the analyzed sample is mostly composed of light elements. The peak around 0.2 keV probably corresponds to the carbon (C) Kα peak. This

is due to fatty acids, which are the main components of coconut oil, and the carbon content of the MF polymer. The prominent peak in the range of 0.3-0.4 keV probably belongs to the oxygen (O) Kα peak. Oxygen may be due to formaldehyde components in the MF resin and possible surface oxidation. The peak around 0.4 keV in the spectrum indicates the nitrogen (N) Kα peak. Since the MF polymer contains a significant amount of nitrogen in its structure, the presence of this peak confirms the presence of the MF capsule shell. The presence of additional small cells with lower density can be attributed to the trace elements or impurities in the sample. Lack of high energy peaks of metal elements (Si, Al, Fe, etc.) in the spectrum indicates that the capsule shell does not contain inorganic fillers and is composed of organic compounds. Overall, the EDX graph confirms that the CO core was successfully encapsulated in the MF shell and that the sample contains mainly carbon, oxygen, and nitrogen.

3.2. FTIR Spectroscopy-Characterizations of Microcapsules and Verification of Core Content

In the FTIR spectrum given in Figure 5, a broad band is observed around 3200-3600 cm^{-1}. This band is due to the stretching vibrations of -OH and -NH groups in the melamine-formaldehyde shell. This broadening may be evident due to hydrogen bonds. The peaks observed in the 2800-3000 cm^{-1} range indicate aliphatic C-H stretching vibrations originating from long-chain fatty acids of coconut oil. This is an important indicator confirming the presence of core material. A peak observed in the 1700-1750 cm^{-1} region belongs to ester carbonyl (C=O) stretching vibrations and may originate from triglycerides in coconut oil or possible oxidation products.

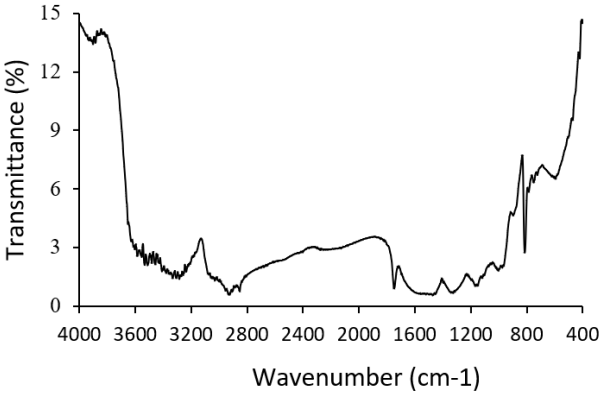


Figure 5. FTIR spectra of microcapsules

The melamine-formaldehyde shell itself does not have a significant carbonyl absorption, so signals in this region indicate the presence of encapsulated oil. Furthermore, the bands around 1500–1600 cm^{-1} are related to the C=N and C-N vibrations of the melamine ring and indicate the successful formation of the shell. The bands around 1000-1200 cm^{-1} are related to the existence of C-O-C and C-N bonds and are evidence supporting the formation of the polymer structure. The sharp peaks below 800 cm^{-1} reflect the characteristic fingerprint region of the MF polymer. The overall spectrum shows that the coconut oil core is successfully encapsulated in the melamine-formaldehyde shell and carries the spectral signatures of both components.

3.3. Results of DSC Analysis

Both pure coconut oil and melamine formaldehyde/coconut oil (MF/CO) microcapsules were examined with differential scanning calorimetry. Using nitrogen gas at a 20 ml/min flow rate, the MF/CO microcapsules test specimen was heated from 0 °C to 50°C at 5°C/min heating rate and then cooled from 50°C to 0°C again at a 5.00°C/min rate, whereas the heating-cooling cycle for the CO specimen was in the range of 0°C - 300°C, all other parameters intact. The results were, as expected, consistent with the literature.

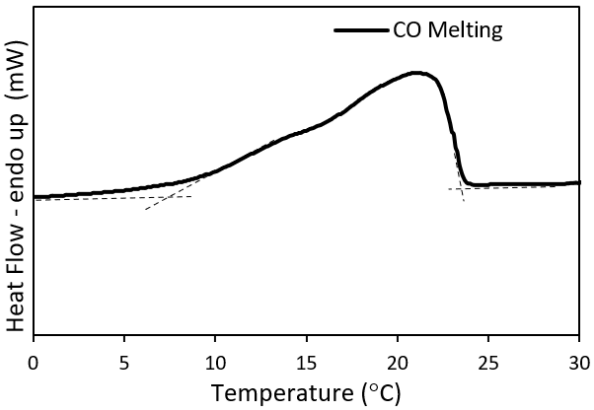


Figure 6. DSC curve for Coconut Oil

Evaluating the DSC curve of coconut oil, shown in Figure 6, the melting and crystallization temperatures were found to be 21.1, and 9.7, respectively. Integrating the area under the peaks or the curves, the latent heat of fusion (melting) (ΔHm) and latent heat of crystallization (-ΔHc) were calculated as 105.1 and 76.3 J/g, respectively.

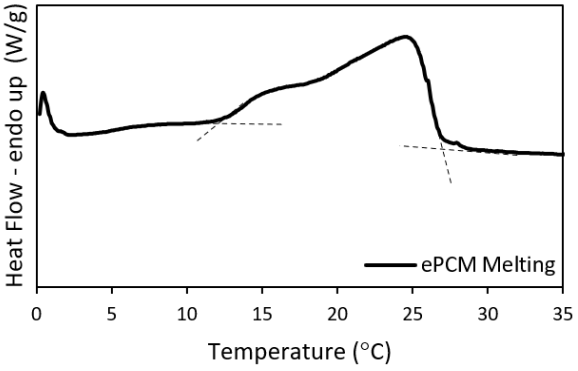


Figure 7. DSC curve for microencapsulated coconut oil

Evaluating the DSC curve of synthesized microcapsules, shown in Figure **7**, the melting temperature of microcapsules was found to be 24.6°C, whereas the crystallization temperature, being a little bit lower, was found to be 5.8°C. The latent heat of fusion (melting) (ΔH_m) and latent heat of crystallization ($-\Delta H_c$) were found to be 82.6 and 18.9 J/g, respectively.

3.4. Results of Thermo-Gravimetric (TG) Analysis

In order to determine the thermal stability of the microcapsules synthesized in this study, TG analysis was conducted from ambient temperature (~30°C) to 750°C at a heating rate of 5°C/min under a gas flow of 20 mL/min in a nitrogen environment to remove all released gases and to prevent thermo-oxidative degradation (Höhne *et al.* 2003).

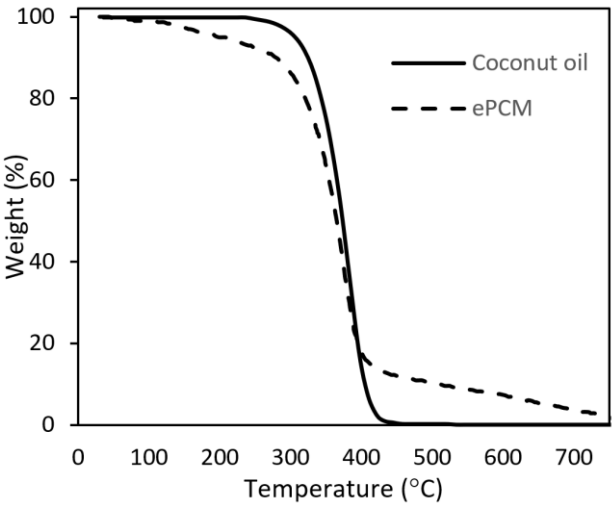


Figure 8. Thermograms of TG analyses for coconut oil and ePCM

The thermograms for CO and MF/CO mePCM given in Figure 8 show that the critical 5% mass loss for coconut oil occurred at 303°C and decomposition at 339°C. On the other hand, 5% mass loss of MF/CO ePCM occurred at 207°C and appears to have decomposed in two steps at 144°C and 334°C, respectively. Although organic coconut oil decomposed without leaving residue after 400 degrees, MF/CO ePCM has been found to have left around 2% due to its nitrogen content.

3.5. Encapsulation Ratio

The encapsulation ratio refers to the ratio of the core and shell materials that make up the capsule. It is the expression of the ratio of the latent heat of encapsulated PCM determined by DSC analysis ($H_{micro-PCM}$) to the latent heat obtained for the pure core material (H_{PCM}) in percentage (Alva *et al.* 2017) and is calculated as shown in Equation 1 (Liu *et al.* 2019).

$$R = \left(\frac{\Delta H_{micro-PCM}}{\Delta H_{PCM}}\right) \times 100\% \qquad (1)$$

As the latent heat of the core material, H_{PCM}, and the latent heat of the microcapsules, $H_{micro-PCM}$, were obtained as 105.1 (J/g) and 82.6 (J/g), respectively, the encapsulation rate of the microcapsules was calculated to be 78.59%, using the equation above.

3.6. Encapsulation Efficiency

For a completely reversible phase change, the latent heats for melting and crystallization must be the same. If there is a difference between these two values, it indicates that PCM has a sub-cooling problem or has a incongruent melting - solidification processes.

$$E = \frac{\Delta H_{melting,ePCM} + \Delta H_{crystallization,ePCM}}{\Delta H_{melting,PCM} + \Delta H_{crystallization,PCM}} \times 100 \qquad (2)$$

The encapsulation efficiency (E) is the percentage expression of the sum of melting and freezing latent heats of the ePCM to that of the PCM and is calculated as given in Equation 2 (Liu *et al.* 2019). Higher encapsulation efficiency means high mechanical strength and tightness. Phase change enthalpy of ePCM is a powerful function of encapsulation ratio and encapsulation efficiency, and encapsulation efficiency is a critical parameter in determining the amount of PCM to be encapsulated (Salunkhe and Shembekar 2012) in order to achieve the required performance in a specific application. In this study, the encapsulation efficiency (E) of synthesized capsules was calculated to be 55.95.

3.7. Density

The bulk density of microcapsules was measured to be 1.0253 g/cm^3. On the other hand, as the weight ratio of shell and core materials can be determined by the encapsulation ratio, the properties of the resulting capsules can also be calculated based on the properties of constituents. In this regard, the density of microcapsules calculated using the following formula (Goel *et al.* 1994) can be used as an confirmatory figure for the measurements.

$$\rho_{capsule} = \left(\frac{d_{core}}{d_{capsule}}\right)^3 \frac{\rho_{core}}{W_{core}}$$
$$\rho_{capsule} = \left(\frac{1,9}{2,0}\right)^3 \times \left(\frac{0,903}{0,7859}\right) \qquad (3)$$

The density value was also calculated using Equation 3 (Qiu *et al.* 2019), taking the average capsule diameter as 2.0 µm and the core diameter as 1.9 µm, whereas the density of the core (ρ_{core}) is taken as the mean of the solid and liquid phases. The calculated density was 0.9851 g/cm^3 and is very consistent with the measured value. This conformance also verifies the consiste0ncy of average capsule diameter.

3.8. Specific Heat Capacity and Thermal Conductivity

Specific heat capacity (C_p) is a physical quantity peculiar to the material and is the amount of heat required to raise the temperature of a gram of mass by 1 degree Celsius. It is also a function of temperature, which means that the amount of heat required for each unit temperature increase is not equal, or to put it in reverse, the amount of heat that any material can hold at each temperature value, that is, its heat capacity at that temperature, will be different. The specific heat capacity of water was taken to be 1 by definition, and the unit calorie was defined as the amount of energy that is required to increase the temperature of 1 gram of water from 13.5 degrees to 14.5 degrees. Thus, C_p values for each temperature value can be calculated differently at every other point of a continuous DSC curve. DSC data is given as shown in Equation 4.

$$DSC = m \times C_p \times sensitivity \times heating\ rate$$
$$[\mu V] = mg \times (\frac{J}{gk}) \times (\frac{\mu V}{mW}) \times (\frac{K}{s}) \quad (4)$$

Herefrom, C_p can then be calculated as shown in Equation 5 ($Netzsch$ 2019).

$$C_p = \frac{DSC\big(SigDif(HeatFlow - Baseline)\big)}{m \times heating\ rate \times sensitivity}$$
$$C_p = \frac{\mu V}{mg \times \left(\frac{K}{s}\right) \times \left(\frac{\mu V}{mW}\right)} = \left(\frac{J}{gK}\right) \quad (5)$$

The specific heat capacity of the microcapsules was measured as 2.7911 J/gK at 303K. The specific heat capacity of microcapsules can also be calculated using the energy balance, with the equation below (Goel *et al.* 1994, Qiu *et al.* 2019).

$$C_{p,capsule} = \frac{(W_{core} \times C_{core} + W_{shell} \times C_{shell})\rho_{core}\rho_{shell}}{(W_{core} \times \rho_{shell} + W_{shell} \times \rho_{core})\rho_{capsule}} \quad (6)$$

where W is the weight fraction. The C value calculated with this formula can be used to verify the C value obtained in the DSC analysis. The C value of ePCM is calculated as the area under the DSC curve between the onset and end points determined by the tangent method, that is, the ratio of the enthalpy of the sample obtained by integrating the curve function between these points to the sample mass and the temperature change of the sample between these points. The C value calculated using Equation 6, using catalog values for the C and values of core and shell materials, was 2.7262 J/gK.

The thermal conductivity of the microcapsules can be measured using a thermal properties analyzer, like a Decagon KD2 Pro, or be calculated using the composite sphere approach as given by Equation 7 (Qiu *et al.* 2016, Qiu *et al.* 2019).

$$\frac{1}{k_{capsule}d_{capsule}} = \frac{1}{k_{core}d_{core}} + \frac{d_{capsule} - d_{core}}{k_{shell}d_{capsule}d_{core}} \quad (7)$$

Using Equation 7, the thermal conductivity of CO/MF core/shell microcapsules was calculated to be 0.2955 W/mK. **Table 1** details the thermal properties of core PCM, coconut oil, and the microencapsulated MF/CO capsules, based on analyses and calculations made in this study.

Table 1. Thermal Properties of Coconut oil and MF/CO microcapsules

	Melting				Crystallization				R*	E**	C	k
	Enthalpy (J/g)	Temperature (°C)			Enthalpy (J/g)	Temperature (°C)					J/gK	W/mK
		Onset	Peak	End		Onset	Peak	End				
CO	105.1	7.3	21.1	23.6	76.3	13.7	9.7	3.0	-	-	3.135	0.321
ePCM	82.6	12.3	24.6	27	18.9	8.2	5.8	2.5	78.59	55.95	2.726	0.296

* Encapsulation Ratio, ** Encapsulation Efficiency

The thermal characteristics of the synthesized CO/MF microcapsules were found to be consistent with values reported in similar studies. For instance, Qiu *et al.* (2016) reported a melting enthalpy of 244 J/g for the paraffin core and 213.5 J/g for the mePCM, and thermal conductivities for the same were reported to decrease from 0.358 W/mK to 0.34 W/mK, which exhibits a similar trend as in the present study. The thermal conductivity value of 0.296 W/mK, obtained in this study, is typical for polymer-shelled MEPCMs and comparable to similar materials reported in the literature. It is also seen in Table 1 that, upon encapsulation, the melting enthalpy decreased by about 21% and the crystallization enthalpy by about 75%. A pronounced decrease in crystallization enthalpy has been frequently reported in MF-based CO microcapsules (Qiu *et al.* 2016, Saraç *et al.* 2019). The asymmetry between melting and freezing enthalpies arises from restricted molecular mobility and subcooling effects caused by the polymeric shell, which limit nucleation and crystal growth during solidification. Nevertheless, the high encapsulation ratio (78.6%) and stable melting behavior confirm the successful phase-change functionality of the synthesized MEPCM.

4. Conclusions

The PCM capsules synthesized within the scope of this study were prepared to establish the infrastructure and ensure the method and to improve and test the materials and instruments for future production of new and original micro/nano capsules for future research.

mePCMs were successfully synthesized using coconut oil as the core and melamine-formaldehyde as the shell through the in-situ polymerization method. CO/MF mePCM were successfully produced with an appropriately controlled narrow particle size distribution, with a stirring rate of 2000 rpm. Comprehensive characterization confirmed successful encapsulation and favorable thermal properties. SEM analysis revealed spherical capsules with diameters between 1 and 5 µm, and they were shown to have a relatively uniform and compact surface profile with smooth morphology, while FT-IR and EDX results verified the presence of both core and shell components. Melting and crystallization temperatures and latent heats of the PCM and mePCM were determined using the DSC analyses, which demonstrated that the melting and crystallization enthalpies decreased from 105.1 J/g and 76.3 J/g (pure CO) to 82.6 J/g and 18.9 J/g (MEPCM), respectively, corresponding to an encapsulation ratio of 78.6%. The slight decrease in latent heat is consistent with literature and attributed to the thermal resistance of the polymeric shell.

In addition, TG analysis showed that the microcapsules decomposed in two steps and confirmed good thermal stability, with decomposition temperatures above 330°C. The measured thermal conductivity (0.296 W/m·K) and specific heat capacity (2.726 J/gK) indicate effective heat transfer performance for low-temperature storage systems.

Compared with literature data, the prepared MEPCM exhibited comparable or superior encapsulation efficiency and thermal conductivity, confirming its potential for use in PCM slurries for cooling, solar, and HVAC applications. These findings lay a solid foundation for future research focusing on improving cycling stability and scaling up the synthesis process for practical thermal energy storage systems.

Based on all these findings, it can be concluded that the prepared ePCMs have good potential for use in the preparation of slurries to be used in low –temperature heat transfer fluids.

Declaration of Ethical Standards

The authors declare that they comply with all ethical standards.

Credit Authorship Contribution Statement

Author 1: Conceptualization, Methodology / Study design, Software, Validation, Formal analysis, Investigation, Resources, Data curation, Writing – original draft, Writing – review and editing, Visualization.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability Statement

The authors declare that the main data supporting the findings of this work are available within the article.

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