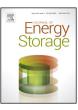
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Journal of Energy Storage

journal homepage: www.elsevier.com/locate/est



Preparation and thermal performance of methyl palmitate and lauric acid eutectic mixture as phase change material (PCM)



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ARTICLE INFO

Article history: Received 22 September 2016 Received in revised form 25 April 2017 Accepted 24 August 2017 Available online 9 October 2017

ABSTRACT

A series of binary mixtures of Methyl Palmitate (MP) and Lauric Acid (LA) were prepared and investigated, aiming for potential phase change material (PCM) for thermal energy storage systems. The thermal analysis of the PCM binary mixtures was investigated by means of Differential Scanning Calorimetry (DSC). A theoretical and experimental determination of the eutectic mixture was established. The results indicated that the eutectic binary mixture of 60%MP and 40%LA has desirable properties of phase transition temperatures within the comfort temperature range ($T_m = 25.6 \,^{\circ}\text{C}$, $T_f = 20.2 \,^{\circ}\text{C}$) and high latent heat capacity ($\Delta H_m = 205.4 \,\text{J/g}$, $\Delta H_f = 205.8 \,\text{J/g}$). The paper experimentally studied the other important thermo-physical properties required for modelling and stimulating the PCM in any storage systems such as thermal conductivity, enthalpy curve, phase diagram, specific heat, thermal diffusivity, and density. The thermal stability test indicated that the eutectic mixture had reliable thermal performance upon thermal cycling. Based on all these results, the MP-LA eutectic mixture is a promising material for thermal energy storage.

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1. Introduction

The limited reserves of fossil fuel, rapid growth of global energy consumption, and the increased environmental concerns of greenhouse gas emissions have shed the light on the importance of having an effective utilization of energy. According to the U.S. Energy Information Administration (EIA), The total energy consumption by the residential and commercial building sectors was estimated to be 41% of the total U.S energy consumption in 2015 [1]. Heating and cooling of buildings alone account for 58% of the total energy consumption in the residential and building sectors [2], and therefore account for over 20% of U.S. energy consumption. Reducing this energy consumption through gains in efficiency is a key method for reducing dependency on fossil fuels. Phase change materials (PCMs) have potential for reducing residential energy requirements and reducing energy demand [3,4]. PCMs are unique in that they store and release large amounts of energy at nearly constant temperature during phase transitions. They have been used in buildings for thermal regulation, reducing energy consumption, and reducing indoor temperature swing. To achieve these objectives, PCMs in buildings are usually incorporated into the building through wallboards, ceiling, and floor by enclosing the PCM in microscopic polymer films that form a sort of panel that can be installed on top of the ceiling, behind the wallboards, or under the floor [3]. Another common technique is incorporating the PCM through gypsum, cement paste, and mortar by impregnating the PCM directly through the construction material to form a matrix and therefore increasing the thermal mass of the building [5–7].

Developing a suitable eutectic PCM for room-temperature applications in buildings is very challenging. The American Society of Heating, Refrigerating and Air-Conditioning Engineers (ASHRAE) standard 55-2013 [8] reported that the average comfort temperature ranges from 19 °C to 27 °C. Paraffins with suitable melting temperature have been studied by many researchers [9-11]. However, their major drawback of high cost has limited their potential use for energy savings in buildings [12]. Salt hydrate PCMs are limited because of the supercooling and phase separation upon cycling [13,14]. In contrasts, fatty acids feature a long-term stability upon cycling, economic feasibilities, little or no supercooling, smaller volume change during phase transition, and nontoxicity [15–17]. Methyl esters have the benefits of high latent heat, lower cost, chemical stability, no corrosiveness to buildings materials, and less flammability [18,19]. However, the major limitations of fatty acids and methyl esters are the low thermal conductivity, and that their individual melting temperatures are

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higher than the comfort temperature for buildings. Therefore, eutectic mixtures can be prepared to adjust the melting temperature of the individual PCMs to the comfort range. Many studies have found that eutectic mixtures of fatty acids possess desirable characteristics for building applications. For example, Sari et al. [20] investigated a eutectic mixture of Capric acid and Stearic acid with suitable melting temperature of 26.04 °C and melting latent heat 176.6 l/g. Wen et al. [21] prepared a eutectic mixture of Capric acid and Lauric acid with melting temperature of 19.09 °C and melting latent heat 141.5 J/g. Khawaji et al. [22] studied a eutectic mixture of decanoic acid and tetradecanoic acid with melting temperature of 20.5 °C and melting latent heat of 153 J/g. Moreover, the study was one of a few studies to investigate the other thermophysical properties of the eutectic mixture such as specific heat capacity, thermal conductivity, and thermal diffusivity. An extensive review of suitable PCMs for buildings can be found in references [23,24].

It can be concluded that previous studies concerning eutectic mixtures usually lack experimental determination of the thermal stability upon cycling. Because of the nature of the eutectic mixture, the precise determination of the eutectic composition ratio is very crucial for consistent performance and long term stability. Therefore, the eutectic composition for the mixture must be determined in fine steps around the theoretical eutectic point, and experimentally tested over several thermal cycles, otherwise, an off-eutectic composition will lead to a phase transition separation of the individual components upon thermal cycling. Furthermore, the important thermo-physical properties such as the specific heat capacity, thermal conductivity, and thermal diffusivity, for both liquid and solid phase, were not reported for eutectic mixtures in most of previous studies. Only a few existing studies examined these additional thermo-physical properties in a detailed manner. Such information is required for modeling the optimum amount, effectiveness, and position of PCMs before being integrated into a thermal system. The current organic based eutectic mixtures with a suitable phase transition temperature for thermal comfort in buildings typically have lower latent heats than other PCMs, reducing their effectiveness. Moreover, most previous research has been focused on mixtures of fatty acids with fatty acids, paraffin with paraffin, methyl esters with methyl esters or salt hydrates with salt hydrates [24-28]. Reports on mixture of fatty acid with methyl ester to form PCM eutectic mixtures are very few so far. Because of the high latent heat of methyl esters, and the other advantages of fatty acids mentioned above, eutectic mixtures consisting of fatty acids and methyl esters are very promising and are expected to have excellent thermal performance - comparable to that of individual PCMs - but within the comfort temperature range. In this study a eutectic mixture of a methyl ester with a fatty acid, namely a methyl palmitate and lauric acid binary system, is studied as phase change energy storage material for thermal regulation in buildings. The aim was to develop a PCM with suitable phase transition temperature and with high energy storage capacity compared to current PCMs, then to provide detailed information on thermo-physical data such as thermal conductivity, enthalpy curves, phase diagrams, thermal cycling effects, specific heat, thermal diffusivity, and density. These data are very important to simulate the actual behavior of PCM in any thermal energy storage application.

2. Materials and methods

2.1. Materials

Methyl Palmitate (MP, 99% purity) and Lauric Acid (LA, 99% purity) were supplied by Sigma Aldrich and used as components in preparation of MP/LA binary eutectic mixture. The chemical data and theoretical thermal properties for the individual PCMs are listed in Table 1. The components of MP/LA mixture were heated up to 50 °C and mixed homogeneously by stirring for 30 min.

2.2. Theoretical prediction of MP/LA binary mixture

The phase change temperature, binary mixture, and the optimum mixing ratio for a binary mixture PCM can be theoretically predicted by thermodynamic calculations using the, Eq. (1) [29]. The latent heat of fusion for the binary eutectic mixture can be calculated using the model proposed by Zhang et al., Eq. (2) [25].

$$T_m = \frac{H_i}{\frac{H_i}{T_i} - R \ln X_i}, i = A, B \tag{1}$$

$$H_{m} = T_{m} \sum_{i=0}^{n} \left[\frac{X_{i} H_{i}}{T_{i}} + X_{i} \left(C_{P,i}^{Liq} - C_{P,i}^{Sol} \right) \ln \frac{T_{m}}{T_{i}} \right], i = A, B$$
 (2)

Where ΔH_i is the latent heat of fusion for components A and B in units of J/mole, and T_i is the melting temperature of the individual PCM components A and B in Kelvin. T_m is the predicted melting temperature of the eutectic mixture in Kelvin, X_i is the molar ratio of components A and B, R is the universal gas constant, H_m is the latent heat of fusion for the binary mixture in J/mole, $C_{P,i}^{Liq}$ and $C_{P,i}^{Sol}$ are the specific heats at constant pressure of the component "i" for both the solid and liquid phase in J/mol K.

The predicted phase diagram using formula (1) and (2) and the theoretical data is shown on Fig. 1. The theoretical eutectic point for the MP/LA binary mixture was found at 60/40 molar ratio, with melting temperature of $24.0\,^{\circ}\text{C}$ and latent heat capacity of $206.6\,\text{J/g}$ for melting.

2.3. Thermal characterization

The investigation of MP/LA eutectic mixture included several experiments around the theoretical eutectic point until an ideal PCM mixture at a certain mass percentage is found with a single desirable sharp melting peak and high latent heat. The melting onset temperature (T_{om}) , melting temperature (T_m) , The freezing onset temperature (T_{of}) , Freezing temperature (T_f) , and latent heat of fusion (ΔH) were measured using a Seiko DSC6200 colorimeter (Seiko Instruments) and DSC2010 (TA instruments). The DSC measurements were performed using aluminum sample pans and

Table 1Chemical data and theoretical thermal properties for the individual PCMs.Name.

	Scientific Name	Molecular Formula	CAS Number	Melting point [°C]	Latent heat capacity [J/g]	specific heat [J/g.K]	Density [g/ml]
Methyl Palmitate (MP)	Methyl hexadecanoate	C ₁₇ H ₃₄ O ₂	112-39-0	29.0	229.5	1.75	0.852
Lauic Acid (LA)	Dodecanoic acid	$C_{12}H_{24}O_2$	143-07-7	44.0	188.7	2.01	0.883

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