Thermal characteristic investigation of eutectic composite fatty acid as heat storage material for solar heating and cooling application

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Thermal characteristic investigation of eutectic composite fatty acid as heat storage material for solar heating and cooling application

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Abstract. A composite phase change material (CPCM) of myristic acid/palmitic acid/sodium myristate (MA/PA/SM) and of myristic acid/palmitic acid/sodium laurate (MA/PA/SL) were impregnated with purified *damar* gum as called *Shorea Javanica* (SJ) to improve the thermal conductivity of CPCM. The thermal properties, thermal conductivity, and thermal stability of both CPCM have investigated by using a Differential Scanning Calorimetry (DSC) thermal analysis, hot disc thermal conductivity analyzer, and Simultaneous Thermal Analyzer (STA), simultaneously. However, a chemical compatibility between both fatty acid eutectic mixtures and SJ in composite mixtures measured by Fourier Transform Infra-Red (FT-IR) spectrophotometer. The results were obtained that the thermal conductivity of MA/PA/SM/SJ and MA/PA/SL/SJ eutectic composite phase change material (CPCM) were improved by addition 3 wt.% and 2 wt.% of *Shorea javanica* (SJ), respectively, without occur a significant change on thermal properties of CPCM. Moreover, the absorbance spectrum of FT-IR shows the good compatibility of SJ with both MA/PA/SM and MA/PA/SL eutectic mixtures, the composite PCM also present good thermal performance and good thermal stability. Therefore, it can be noted that the purified Shorea Javanica proposed, the as high conductive material in this study was able to improve the thermal conductivity of eutectic PCM without any significant reduction on its thermo-physical and chemical properties and can be recommended as novelty composite phase change material for thermal energy storage application.

1. Introduction

Thermal energy storage (TES) plays an important role in generating the solar energy to use in a wide range application such as heating and cooling, solar power, and industrial waste heat recovery [1-4]. The latent heat thermal energy storage (LHTES) material known as phase change materials (PCMs)

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offer more advantage over sensible heat thermal energy storage (SHTES) materials, particularly the small temperature different between melting and solidifying point, small volume, and low weight per unit of storage capacity. PCMs absorb heat as storage energy during the heating process and release it during cooling [5]. However, phase change material (PCM) that used in thermal energy storage (TES) application have classified into two categories which are inorganic and organic phase change materials (PCMs). Although the inorganic PCMs group have great heat storage capacity with a wider range of phase transition temperature. But they have numbers of disadvantages such as subcooling, corrosion, phase separation, phase segregation, and lack thermal stability, compared to organic PCMs which only have a problem with low heat transfer rate due to low thermal conductivity [6-8].Organic phase change materials (PCMs) such as paraffin and fatty acids have been used in a wide application of thermal energy storage due to their high thermal storage capacity, a wide range of phase transition temperature, low subcooling, and have a good reliability [7]. However, the common problem of these organic PCMs is their low heat transfer rate due to low thermal conductivity, which leads to large temperature gradients. Therefore, enhancement the thermal conductivity of organic phase change materials (PCMs) became one of the main issues in a wide application of thermal energy storage (TES) [9].

Numbers of different methods have been proposed for enhancing the thermal conductivity of PCMs. Encapsulation was one of few methods proposed in order to improve the thermal conductivity of PCMs. On the other hand, this method was handled the leakage problem properly and bring a high cost of production [8]. The thermal conductivity of PCM can be improved as well by adding high conductive porous material using simple mixing method, solution casting method or impregnation method [10-13].

However, in this work, we propose a modification by combination two existed methods of simple mixing and impregnation in order to prepare the composite fatty acids eutectic mixture as composite phase change material (CPCM) by adding Shorea javanica, which is obtained from purified damar gum. The study was performed to analyze the improvement of thermal conductivity of prepared eutectic composite fatty acid with different composition of Shorea Javanica and to evaluate its thermal performance and chemical compatibility of prepared CPCM.

2. Materials and methods

2.1. Materials

Two kind of fatty acid eutectic mixture, myristic acid/palmitic acid with 5 wt.% sodium myristate (MA/PA/5 wt.%SM) and myristic acid/palmitic acid with 10 wt.% sodium laurate (MA/PA/10 wt.% SL) were propose as based phase change material (PCM) as conducted by Fauzi, Metselaar in previous study [14]. A natural damar gum or *Shorea javanica* which obtained from conventional plantation farm in Indonesia was proposed as high conductive porous material. However, additional purification processes were required in order to remove the impurity content from *Shorea Javanica* (SJ).

The purification process was conducted by using an organic solvent method which was introduced previously by Setianingsih, 1992 [15]. Toluene (C_6H_5-CH_3) with a molecular weight (MW) 92.14 $g.mol^{-1}$ (Fisher Scientific) and *Shorea Javanica* (SJ) in composition ratio 1:8 wt.% stirred at 2000 rpm for 20 minutes at ambient temperature in order to obtain a completed dissolves solution of *Shorea Javanica* (SJ). Moreover, 1 wt.% of activated charcoal (AC) manufactured by Acros Organic was added for decolorizing of SJ solutions which stirred for 15 minutes at 45 ^oC. The sedimentation and filtration process was conducted to separate the impurities from the solutions. Subsequently, the pure solution of Shorea Javanica (SJ) was placed in a rotary evaporator to vaporize the solvent toluene, and thus the button product of evaporation process was dried with oven with temperature 80 ^oC for 8 hours in order to obtain pure *Shorea Javanica* powder. A dried powder of *Shorea Javanica* needs to be grinded in a rotary ball mill for further separated by sieve shaker to obtain the same size particle of *Shorea Javanica* in 100 μm [16].

2.2. Preparation and analysis methods

A modification mixing-impregnation method was used to prepare a novel eutectic composite phase change material (CPCM) of MA/PA/SM/SJ and MA/PA/SL/SJ. The eutectic mixtures of MA/PA/5wt.%SM and MA/PA/10wt.%SL separately were placed into a jacketed flask reactor along with different mass composition of SJ in 1, 2, 3, 4, and 5 wt.% under vacuum pressure and hot heat transfer fluid (HTF) circulation was maintained the temperature stable at 70 ^{o}C. However, a stirrer mixing was applied to homogenous distribution of SJ in MA/PA/SM and MA/PA/SL eutectic mixtures. These preparation processes performed for 2 hours and ended by drying process at ambient temperature.

Each composition of prepared eutectic composite MA/PA/SM and MA/PA/SL with different mass fraction SJ simultaneously were evaluated their thermal conductivity, thermal properties, and thermal stability using thermal conductivity analyzer (Hot Disc, TPS 2500 S), thermal Differential Scanning Calorimetric (DSC) analyzer (Metler Toledo, DSC^{1} Stare system), and Simultaneously Thermal Analyzer (STA 6000, Perkin Elmer), respectively. Furthermore, the Fourier transform infrared spectroscopy (FT-IR, Bruker Tensor 27) used to identify the chemical compatibility between both fatty acid eutectic mixtures and *Shorea Javanica*. The analysis was performed using ATR sample compartment with MIR spectra in the wavenumber range of 4000 – 400 cm^{-1}.

Figure 1. Thermal performance test setup.

Moreover, the thermal performance of MA/PA/SM/SJ and MA/PA/SL/SJ eutectic composite were evaluated with customized test setup device as shown in Fig. 1. The setup was consisting of 2 fluid circulation bath for both hot and cold medium circulated into the chamber. The 2 cylindrical glass tubes were attached inside the chamber and direct contact with heat transfer fluid (HTF). Subsequently, 6 gr of two couple samples of MA/PA/SM; MA/PA/SL eutectic mixtures and MA/PA/SM/SJ; MA/PA/SL/SJ eutectic composites were placed separately in those glass tube in different turn. The alterations temperature of these samples against endothermic and exothermic time was recorder by thermocouples (J-type, Omega) connected to data acquisition (DAQ) system. The setting temperature of hot and cold heat transfer fluid (HTF) set above of melting temperature and below of solidification temperature of each prepared samples which are 65 ^{o}C and 30 ^{o}C, respectively.

3. Results and discussion

3.1. Thermal characteristic of Eutectic composites

In the previous study, Fauzi et al. was reported that the myristic acid/palmitic acid (MA/PA) eutectic mixture have a better improvement on their thermal properties by adding 5 wt.% sodium myristate (SM) and 10 wt.% sodium laurate (SL), respectively, in term of reducing the phase transition temperature and increasing the latent heat of fusion [14]. The studies were obtained the melting temperature of MA/PA reduced 11.5% and 10.12% by adding 5 wt.% SM and 10 wt.% SL, while the latent heat of fusion improved about 15.53 % and 12.24%, respectively. Nevertheless, these additions of acid-based surfactants do not show a significant improvement in thermal conductivity of eutectic PCM. Therefore, *Shorea Javanica* was proposed in this study as a high conductive material to enhance the thermal conductivity of prepared PCM with consideration no much effect on degrading the thermal properties.

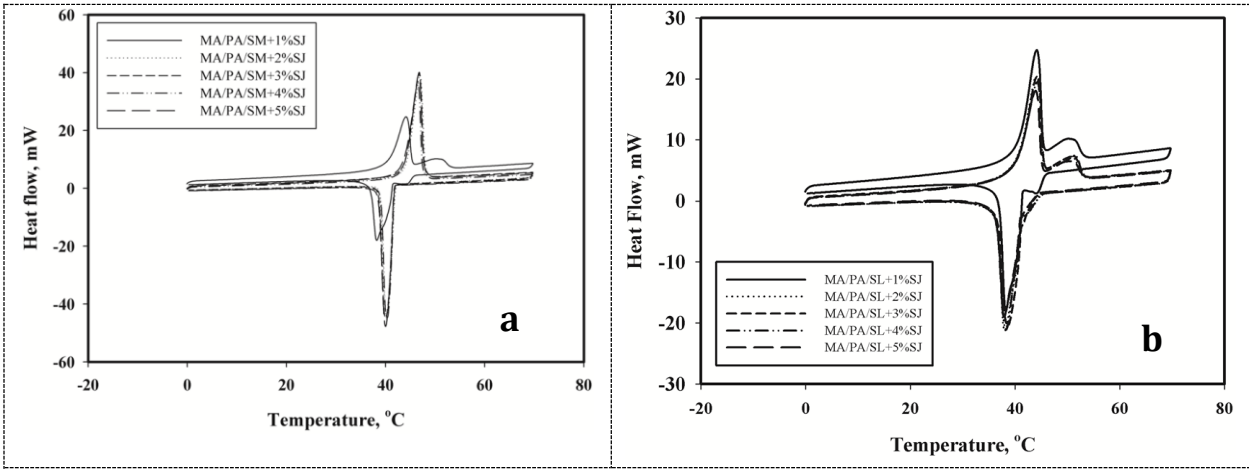

Figure 2. DSC curves of eutectic mixture composites with different mass fraction SJ: a). MA/PA/SM/SJ; b). MA/PA/SL/SJ.

The DSC curve in Fig. 2.a and Fig. 2.b presents the thermal properties of CPCM in a different composition of SJ. The melting temperature (T_m) and solidification temperature (T_s) of MA/PA/SM/SJ and MA/PA/SL/SJ interpreted from onset point, while the latent heat of fusion during melting ($\Delta H_{f,m}$) and solidification phase (ΔH_{fs}) were obtained from interpolating of charts peak area. The result as shown in Table 1 indicated that the phase transition temperature of MA/PA/SM/SJ and MA/PA/SL/SJ eutectic composite rises up against increasing percentage composition of SJ, and in the same time, its latent heat of fusion tend to decrease with increasing SJ composition.

The thermal conductivity of MA/PA/SM/SJ and MA/PA/SL/SJ eutectic composite in a different composition of SJ were listed in Table 2 which shows that the thermal conductivity of those composite mixture has increased simultaneously with increasing the percentage composition of SJ. These results noted that the highest amount of thermal conductivity was obtained at MA/PA/SM and MA/PA/SL with addition 5 wt. % SJ which are 0.492 and 0.393 $Wm^{-1}K^{-1}$, respectively. Nevertheless, the thermal properties of MA/PA/SM and MA/PA/SL with 5 wt.% SJ present an unexpected decrease on latent heat of fusion which has a significant drop to 167.38 and 153.91 Jg^{-1},compared to the latent heat of fusion of initial MA/PA/SM and MA/PA/SL eutectic mixture as 179.12 and 174.47 Jg^{-1}, respectively. However, an improvement of thermal conductivity without a significant drop of latent heat of fusion instead shows by MA/PA/SM+3%SJ and MA/PA/SL+2%SJ eutectic composite mixtures. Moreover, the thermal conductivity of MA/PA/SM/3%SJ and MA/PA/SL/2%SJ eutectic composite shows a great improvement by increased 101.65% and 62.55%, respectively, compared to thermal conductivity

value of their initial mixture [14]. Meanwhile, the latent heat of fusion only presents a slight decreased which is 0.93% and 7.50%.

Tabel 1. Thermal properties of MA/PA/SM and MA/PA/SL/SJ eutectic mixture with Shorea Javanica.

Composite phase change materials (CPCM)	T_m *(°C)*	$\Delta H_{f,m}$ *(J/g)*	T_s *(°C)*	$\Delta H_{f,s}$ *(J/g)*
MA/PA/SM + 1% SJ	40.54	179.92	41.46	183.55
MA/PA/SM + 2% SJ	43.20	176.39	41.95	179.95
MA/PA/SM + 3% SJ	43.96	177.45	41.73	180.85
MA/PA/SM + 4% SJ	43.89	169.35	41.75	172.29
MA/PA/SM + 5% SJ	43.75	167.38	41.67	175.63
MA/PA/SL + 1% SJ	40.64	169.26	41.56	169.70
MA/PA/SL + 2% SJ	40.20	161.27	41.36	159.26
MA/PA/SL + 3% SJ	40.48	158.69	41.50	156.68
MA/PA/SL + 4% SJ	40.26	157.45	41.40	155.68
MA/PA/SL + 5% SJ	40.08	153.91	41.30	153.14

Numbers of studies have been using a different kind of porous materials to improve the thermal conductivity of PCMs. Karaipekli, Sarı [17] proposed a different mass fraction of expanded graphite (EG) and carbon fiber (CF) to the enhanced thermal conductivity of stearic acid (SA). The result of these study reported that thermal conductivity of SA was increased with increasing the mass fraction of EG and CF and indicated an insignificant decrease in its latent heat of fusion at the same time. Sarı and Karaipekli [18], studied the effect of addition expanded graphite (EG) into paraffin in improvement the thermal conductivity of paraffin. The results were indicating that the thermal conductivity of paraffin/EG composite mixture was increased with increasing the mass fraction of EG. The decreasing of latent heat of fusion also occurred by increasing the composition of EG in paraffin in this study. In the extended work, these authors evaluated the thermal characteristic and thermal conductivity improvement of capric-myristic acid/expanded perlite and some fatty acid compounds with expanded graphite. They also obtained that these porous material were able to improve the thermal conductivity of PCMs and also reduced its latent heat of fusion insignificantly [19, 20].

Tabel 2. Thermal conductivity MA/PA/SM/SJ and MA/PA/SL/SJ composite PCM with different composition SJ.

Composite phase change materials (CPCMs)	**Thermal conductivity, $Wm^{-1}K^{-1}$**
MA/PA/SM + 1% SJ	0.463
MA/PA/SM + 2% SJ	0.475
MA/PA/SM + 3% SJ	0.488
MA/PA/SM + 4% SJ	0.489
MA/PA/SM + 5% SJ	0.492
MA/PA/SL + 1% SJ	0.373
MA/PA/SL + 2% SJ	0.382
MA/PA/SL + 3% SJ	0.387
MA/PA/SL + 4% SJ	0.387
MA/PA/SL + 5% SJ	0.393

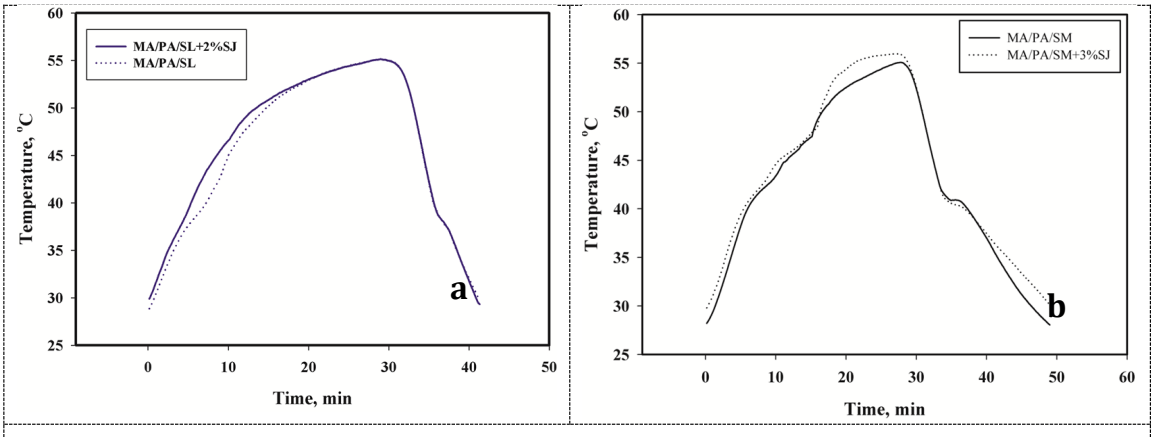

Figure 3. Curves of thermal performance heat storage and heat release: a). MA/PA/SM/SJ; b). MA/PA/SL/SJ.

Moreover, the improvement of thermal conductivity of MA/PA/SM+3%SJ and MA/PA/SL+2%SJ composite mixtures were proved by the improvement of their heat transfer rate as seen in Fig. 3.a and Fig. 3.b. The figures show the heat storage and heat release duration time needed for MA/PA/SM+3%SJ and MA/PA/SL+2%SJ eutectic composites comparison with their initial non-composite eutectic mixtures MA/PA/5wt.%SM and MA/PA/10wt.%SL to change the phase from solid to liquid and vice versa. Both curves show the phase transition time of MA/PA/SM+3%SJ and MA/PA/SL+2%SJ eutectic composites, and noticed that the duration time for both composite PCMs to reach the melting and solidification points were 8.4 and 6.7 minutes; 8 and 6 minutes, respectively. This phase transition time shows shortest than their initial mixtures of MA/PA/5wt.% SM and MA/PA/10wt.%SL which is 9.7 and 10.6 minute; 11.3 and 7.16 minute, respectively. However, the same identic result also reported by Zang and Fang [10] in other work proposed paraffin and paraffin/expandable graphite (P/EG) and reported that the heat storage time of paraffin was longer 27.4% than P/EG and heat release time paraffin was longer 56.4% than P/EG. It is noted that the heat transfer rate of CPCM was higher than PCMs. In addition, the heat transfer process during the heat storage was controlled by natural convection, whereas in the heat release, the heat transfer controlled by thermal conduction. Hence, the increase of conductivity coefficient of eutectic CPCM had brought a significant effect on enhancement the heat transfer rate during release the heat than heat storage process. Therefore, the CPCM need shorter time once heat release process than storage process [10].

3.2. Compatibility SJ with MA/PA/SM

The FT-IR spectra as seen in Fig. 4.a and Fig. 4.b shows the absorbance peak for each functional group of chemical structures of MA/PA/SM, MA/PA/SM+3%SJ, and MA/PA/SL, MA/PA/SL+2%SJ compared to pure SJ spectrum, respectively. The spectra indicated have the same frequency in every peak range of wavenumber between MA/PA/SM eutectic mixture and MA/PA/SM+3%SJ eutectic composite; MA/PA/SL eutectic mixture and MA/PA/SL+2%SJ eutectic composite, respectively. It is mean that the addition of 3 wt. % and 2% of *Shorea Javanica* (SJ) in MA/PA/SM and MA/PA/SL eutectic mixtures did not occur any chemical reaction in the composite mixtures. Therefore, it can be noted that the changing of thermal properties of MA/PA/SM/SJ and MA/PA/SL/SJ eutectic composites was not caused by the chemical interaction between SJ and initial eutectic PCM. Subsequently, it was due to physical properties of *Shorea Javanica* (SJ) which has a high melting point and low latent heat of fusion, and thus contributed to changing the thermal properties of eutectic CPCM.

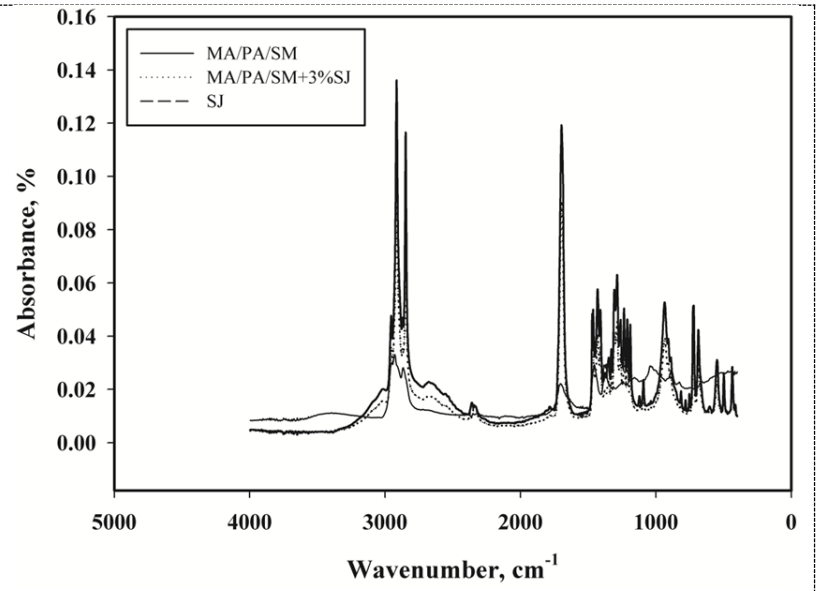

Figure 4. FT-IR curves of MA/PA/SM and MA/PA/SJ.

3.3. Thermal stability of MA/PA/SM/SJ

The thermal stability curve of MA/PA/SM+3%SJ and MA/PA/SL+2%SJ eutectic composites was demonstrated in Fig. 5.a and Fig. 5.b which is shows that the prepared eutectic composites do not show any mass degradation within work temperature 30 C to 160 ºC. The weight degradation of MA/PA/SM+3%SJ and MA/PA/SL+2%SJ have appeared once the working temperature at 168.7 ºC and 164.28 ºC, and thus reaches an optimum weight degradation at work temperature 289.39 ºC and 272.64 ºC, respectively. Thus, these results indicate that the MA/PA/SM+3%SJ and MA/PA/SL+2%SJ eutectic composites presented a good stability to apply as a CPCM in thermal energy storage application (TES) with operation temperature below of 160 ºC.

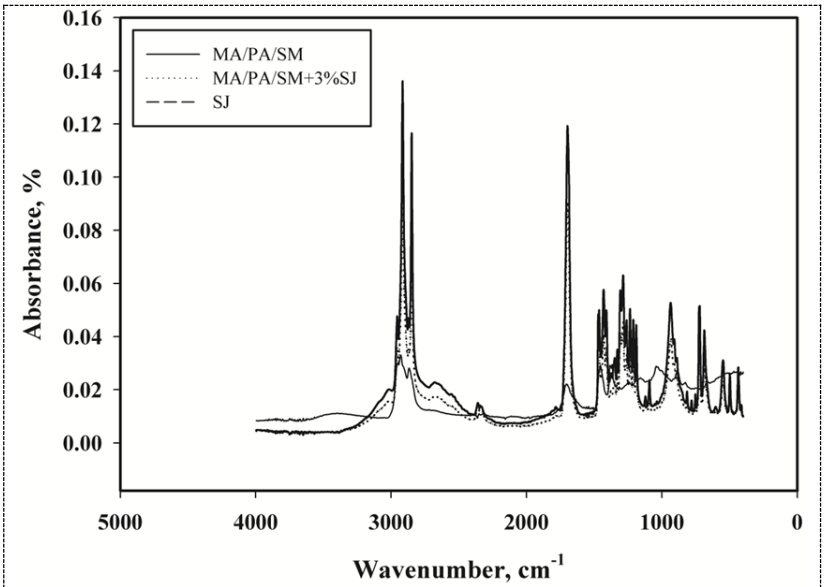

Figure 5. Curve thermal stability of MA/PA/SM+3%SJ.

Thermal stability of composite phase change materials (CPCMs) also been studied by other researchers. Kim and Drza [12], measured thermal stability of paraffin/xGnP composite PCM at range temperature 30 to 600 ^{o}C. The mass loss of CPCMs started to decomposition at 200 ^{o}C and reach to total decomposition at 280 ^{o}C. In the other work, Jeong, Jeon [21] analyzed thermal decomposition of n-octadecane/diatomite CPCMs at range temperature of 30 to 400 ^{o}C. In this study obtained that the weight decomposition of n-hexadecane/diatomite composite was 50% lower than pure n-hexadecane PCM at operation temperature 200 ^{o}C. According to these studies can be noted that CPCMs have a higher thermal stability compared to the pure PCMs.

4. Conclusions

The preparation and thermal characteristic analysis of two novel eutectic composites phase change material (CPCM) which involves the myristic acid/palmitic acid/sodium myristate (MA/PA/SM) and myristic acid/palmitic acid/sodium laurate (MA/PA/SL) with Shorea Javanica (SJ) have been evaluated in this current study. The thermal conductivity of these CPCMs was simultaneously increased with increasing the composition of SJ as 1, 2, 3, 4, and 5 wt. %, respectively. But, CPCM with composition SJ 3 wt. % and 2% with MA/PA/SM and MA/PA/SL respectively that shows a good improvement on thermal conductivity without significant impact in decreasing the latent heat of fusion of the CPCM mixtures. The eutectic composite of MA/PA/SM+3%SJ and MA/PA/SL+2%SJ also indicate a good thermal performance, no chemical reaction between each component in the mixture and has a good thermal stability without occurs weight degradation within temperature work 30 ^{o}C to 160 ^{o}C. Therefore, those may conclude that the Shorea Javanica (SJ) is acceptable to use as a porous material to improve the thermal conductivity of composite phase change material (CPCM).

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