



The role of vegetable oil in water based phase change materials for medium temperature refrigeration

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ABSTRACT

This research aims to investigate development of water based phase change materials (PCM) for cold thermal energy storage (CTES). Mixtures of water with small amount vegetable oil addition were chosen as candidates of the PCM that were considered to be suitable for medium temperature refrigeration application with temperature range of products between $-1\text{ }^{\circ}\text{C}$ and $+5\text{ }^{\circ}\text{C}$. The PCM candidates were tested experimentally through DSC and T-history method. The results showed that esters of vegetable oils played very important role on the solubility of the vegetable oil in water. The esters made the investigated vegetable oil (soya oil or corn oil) mix well in water solutions and they worked as nucleating agents that could lower freezing point and reduce super-cooling of the water. It was found that addition of vegetable oils by 5% to 10% in water solution could decrease the freezing temperature from $0\text{ }^{\circ}\text{C}$ down to respectively $-3.5\text{ }^{\circ}\text{C}$ to $-6.5\text{ }^{\circ}\text{C}$ and could also minimize degree of super-cooling of the PCM candidates.

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1. Introduction

Thermal energy storage is considered as one of the most perspective technologies for increasing the efficiency of energy conversion processes and effective utilization of available sources of heat [1,2]. In the last few years, much attention has been paid to the latent heat thermal energy storage systems (LHTES) due to their various applications [3–6]. The technology is also effective to reduce gap of unbalances between energy supply and demand [7–10]. Among various applications of thermal energy storage, heat or cold accumulation of temperature ranging from $-50\text{ }^{\circ}\text{C}$ to $120\text{ }^{\circ}\text{C}$ has a greater market potential that can be carried out using wide range of phase change materials (PCMs) [11]. Application of PCMs as latent heat thermal energy storage (known as LHTES technique) can provide larger capacity of energy storage per unit mass and transfer heat at relatively constant temperature [12–16]. The technique, therefore, can use a smaller size system with narrow temperature range during phase change process compared with sensible heat storage (SHS) [17–19].

For LHTES applications, many materials have been investigated. They included organic, inorganic and mixture of both organic and inorganic materials [20]. Materials with high thermal capacity and

constant range of phase change temperature are considered as good PCMs [21]. PCM made of organic solid-liquid has attracted much attention to various applications for their excellence properties [22–25]. Organic PCMs such as paraffin and fatty acid are reported in [26,27]. Paraffin is the most popular organic PCM, which has very small degree of super-cooling and wide range of phase change temperature. Paraffin is also chemically stable during phase change process [28]. However, paraffin waxes have disadvantages due to their low thermal conductivities, low latent heat, flammability and high change in volume [29].

Fatty acid ($\text{CH}_3(\text{CH}_2)_{2n}-\text{COOH}$) has advantages compared to paraffin. Fatty acid has more accurate melting point, high latent heat, congruent melting and solidification temperatures, small degree of super-cooling, low vapor pressure, little volume change during phase change process and low cost for energy storage. Paraffin is also non-toxic, non-corrosive to metals, thermally and chemically stable, and non-flammable [30–34]. However, fatty acid has unpleasant odor compared with paraffin [35]. In order to eliminate the unpleasant odor, some researchers recommended replacing them with their derivative fatty acid esters, which could be obtained via the esterification of fatty acids with alcohols. The effect of alcohols on the thermal properties of fatty acid esters was relatively excessive [36].

The main drawback of TES technology is how to develop effective PCMs for storing energy [37]. Phase change temperature and latent heat of fusion are the two basic parameters of PCMs [38].

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Therefore, the selection of PCMs with suitable phase change temperature is critical [39]. Other parameters include thermal stability and energy storage characteristics [40]. In addition, high degree of super-cooling could make evaporation temperature of refrigerator system lower and certainly reduce cooling efficiency [41,42,43].

Water is often employed as PCMs because of its reliability, stability, low cost, high specific heat, high density, and high latent heat capacity of 335 kJ/kg. Water also has good safety issue [44–46]. Furthermore, it has been implemented in many applications especially in air conditioning systems for shifting peak loads. Water, however, has a high degree of super-cooling [47,48]. Water cannot be applied as PCM for CTES of operating temperatures below 0 °C, due to its freezing and melting point at 0 °C [49,50]. One way to make water applicable as PCMs at below 0 °C is by adding nucleation agent to trigger heterogeneous nucleation and to eliminate its super-cooling [51].

A PCM for medium temperature refrigeration application should be able to maintain product temperatures between –1 °C and +5 °C. In this application, evaporating temperature of refrigeration system is usually lower than –8 °C [52]. PCMs utilized for this application, therefore, should be melting and freezing at temperature range between –6 °C and –4 °C with assumption that the PCM would be placed in the inlet-air after evaporator. Such PCMs could be organic based materials (paraffin), salt solutions, or water based materials [53]. For salt-based PCMs, their freezing and melting temperatures can be lowered down by increasing salt concentration of the PCM solution. However, salt solutions are corrosive and have lower latent heat than water [54,55,56]. Another way to squeeze down freezing and melting temperatures of water is by adding antifreeze liquid [57].

To date, there are few studies on utilization of organic PCMs made of water with nucleating agents of vegetable oils. Vegetable oil contains various types of fatty acids. Fatty acids and their fatty acid ester or eutectic mixtures also have many superior properties as organic PCM materials [58–61]. Fatty acid esters are new material for organic PCMs. Unfortunately very limited thermal data is available in literatures [62]. Fatty acids are derivatives of materials readily found in nature and labeled as bio-based materials [63]. Another advantage of water based PCMs with vegetable oil nucleating agent is that vegetable oils offer a continuous supply [64–66].

This paper reports an investigation on very small vegetable oil (soya and corn oil ester) solution in tap water as PCM material alternatives for medium temperature refrigeration applications. The little amount of vegetable oil ester in the investigated water solution would make properties of the solution similar to the physical and thermal properties of water. This could make the

solution become strong PCM candidates for below 0 °C temperature applications. Other results such as super-cooling of water and the influence of vegetable oil addition in reducing super-cooling of the PCM candidates are also discussed. This paper also explains how water and vegetable oil ester can properly be mixed to become applicable PCM solutions.

2. Experimental

2.1. Materials

Materials tested in this study were natural ester oils commonly called “vegetable oils” extracted from soya bean and corn. These vegetable oils were used without further purification. Soya and corn oil esters were chosen because they contain poly-unsaturated fatty acids (PUFA) which make its freezing and melting temperatures relatively low. Chemical composition of soya and corn oil ester was tested with Gas Chromatography Mass Spectrometry (GCMS). The test results are presented in Tables 1 and 2. The tables show that commercial soya and corn oil esters are composed mainly by methyl esters of 53.89% and 38.54%, respectively. The soya and corn oil also contains benzene (16.4%) and (17.45%), 1,3-cyclohexadiene (6.85%) and (8.29%), beta-sesquiphellandrene (11.55%) and (23.83%), and others of about 11.68% and 11.89%.

The main composition of soya and corn oil esters is methyl ester. Methyl ester is a small ester with single carbon chain. Small esters are soluble in water. Esters are derived from carboxylic acids in which one hydroxyl (–OH) group replaced by one alkyl (–O) group [61]. In water solutions, certain acid molecules of ester having –OH cluster would be ionized by releasing hydrogen (H) atom to generate ion H⁺. Although esters can't hydrogen bond with themselves but esters can hydrogen bond with water molecules. One of the slightly positive hydrogen atoms in a water molecule can be sufficiently attracted to one of the single pairs on one of the oxygen atoms in an ester for a hydrogen bond to be formed. There is also, of course, dispersion forces and dipole–dipole attractions between the ester and the water molecules. Forming these attractions releases energy. This helps to supply the energy needed to separate water molecule from water molecule and ester molecule from ester molecule before they can mix together [67]. This explains why small esters (soya and corn oil esters) dissolve in water. In higher chain esters such as ethyl and propyl esters, as chain lengths increases, the hydrocarbon parts of the ester molecules start to get in the way. By forcing themselves between water molecules, they break the relatively strong hydrogen bonds between water molecules without replacing them. This makes the process energetically less profitable, and so solubility of esters decreases.

Table 1
Chemical composition of commercial soya-oil ester

Component name	Formula	Area (%)
Benzene, 1-(1,5-dimethyl-4-hexenyl)	C ₁₅ H ₂₂	16.04
Zingiberene, 1,3-Cyclohexadiene, 5-(1,5-dimethyl-4-hexenyl)	C ₁₅ H ₂₄	6.85
Cyclohexene, 1-methyl-4-(5-methyl-1-methylene-4-hexenyl)	C ₁₅ H ₂₄	11.32
Dodecanoic acid, methyl ester (CAS) Methyl laurate	C ₁₃ H ₂₆ O ₂	5.30
4-Octenoic acid, methyl ester	C ₉ H ₁₆ O ₂	0.59
Beta-sesquiphellandrene	C ₁₅ H ₂₄	11.55
6,7-Dihydroxy-5,8,13,14-pentaphenotetrone	C ₂₂ H ₁₀ O ₆	0.36
Hexadecanoic acid, methyl ester	C ₁₇ H ₃₄ O ₂	12.64
Dodecanoic acid, (2,2-dimethyl-1,3-dioxolan-4-yl) methyl ester	C ₁₈ H ₃₄ O ₄	
Hexadecanoic acid, (2,2-dimethyl-1,3-dioxolan-4-yl) methyl ester	C ₂₂ H ₄₂ O ₄	24.14
2-Heptadecanone, 1- (2,2-dimethyl-1,3-dioxolan-4-yl) methoxy	C ₂₃ H ₄₄ O ₄	
10-Octadecenoic acid, methyl ester	C ₁₉ H ₃₆ O ₂	5.79
Nonadecanoic acid, methyl ester	C ₂₀ H ₄₀ O ₂	5.43

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