Fatty acids/1-dodecanol binary eutectic phase change materials for low temperature solar thermal applications: Design, development and thermal analysis

Rohitash Kumar a,b,⇑, Sumita Vyasa, Ambesh Dixit b

a Defence Laboratory Jodhpur, Rajasthan 342011, India
b Department of Physics & Center for Solar Energy, Indian Institute of Technology Jodhpur, Rajasthan 342011, India

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A B S T R A C T

Thermal phase diagrams are computed for binary mixtures of different fatty acids with 1-dodecanol to design new eutectic phase change materials with melting temperatures and latent heat of fusion ranging from 15.5 to 19 °C and 183 to 188 kJ kg⁻¹, respectively. The lauric (LA), myristic (MA) and palmitic (PA) fatty acids with 1-dodecanol eutectic compositions are identified and experimentally validated using differential scanning calorimetric measurements. The measured eutectic compositions are 29:71, 17:83, and 10:90 wt% for LA-DE, MA-DE, and PA-DE binary systems respectively. The melting temperatures and latent heat of fusion for these eutectic compositions are 17, 18.43, 20.08 °C and 175.3, 180.8, 191 kJ kg⁻¹ respectively. The computed and measured composition weight fractions, melting temperature and latent heat of fusion values are in good agreement. The suitable melting temperature and considerably large latent heat of fusion for these designed and developed new eutectic phase change materials make them suitable for latent heat thermal energy storage systems for low temperature solar thermal applications such as building heating/cooling and body cooling clothing applications.

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1. Introduction

The energy requirement/demand is increasing day by day due to the continuously population increasing across the world. The associated developments such as industrialization, enhanced living comforts also demand the everyday increasing energy consumption simultaneously. These developments have led to the over exploitation of fossil fuels to fulfill the energy requirements and use of refrigeration gases in air conditioning systems, which are causing the global warming and ozone layer depletion (Memon, 2014). Approximately 80% of the annual energy requirement of the world is fulfilled by the fossil fuels, which releases ~30 Gt CO₂ to the environment (Agency, 2012). The global temperature will increase by ~2.6 °C and 3.6 °C by 21st and after 22nd century respectively with current energy generation trends (IEA, 2015). Thus, the preventive measures are essential to minimize the dependency on fossil fuels and refrigeration gases, to reduce such adverse effects. This compels to increase the use of renewable energy alternatives to meet the continuously increasing energy demands.

The major fraction of energy production is consumed for building heating/cooling applications to improve the thermal comfort by minimizing the temperature fluctuations (Henze, 2005; Wyse, 2011). The utilization of air conditioners and space heating systems are major sources for electricity load variation in hot and cold climatic regions. Hence, renewable energy based energy efficient heating and cooling systems are required to alleviate the energy and climatic problems. Renewable energy sources, such as solar, wind, sea, and geo thermal energies are intermittent and leading to the energy demand and supply gap (Khudhair and Farid, 2004). The intermittency problem associated with such renewable energy sources can be reduced by thermal energy storage systems integration with renewable energy production systems. The use of thermal energy storage systems in conjunction with renewable energy sources will also assist in the enhancement of energy efficiencies of building heating/cooling systems (Mehling et al., 2002; Haussmann et al., 2002).

The use of latent heat thermal energy storage system (LHTESS) is more useful over sensible heat thermal energy storage system (SHTESS) because of their higher energy storage density (Abhat, 1983; Lane, 1983, 1986; Dincer and Rosen, 2002). LHTESS offers
additional advantages such as small thermal energy storage vessel requirement, release/absorption of thermal energy in a small temperature range (around phase change temperature of phase change material) compared to SHTESS (Zalba et al., 2003; Kumar et al., 2016a). The phase change materials (PCMs) based thermal energy storage devices have attracted attention for their use in buildings’ floor, ceiling, and walls to enhance their thermal mass for thermal comfort applications. Inorganic, organic and eutectic PCMs have been explored for these applications (Sharma et al., 2009). Inorganic PCMs such as hydrated salts offer high latent heat of fusion and high energy storage density as compared to the organic PCMs (Al-Abidi et al., 2012). However, these PCMs suffer from low thermal stability during charge/discharge cycles, large supercooling requirement, incongruent melting, etc. (Lane, 1986; Dincer and Rosen, 2002; Zalba et al., 2003). The thermophysical properties, such as long term thermal stability in conjunction with relatively good latent heat of fusion of organic fatty acids make them suitable candidates for above mentioned applications (Farid et al., 2004; Tyagi and Buddh, 2007; Rozanna et al., 2004; Feldman et al., 1989; Hasan and Sayigh, 1994; Sari and Kaygusuz, 2003; Cedeno et al., 2001; Inoue et al., 2004; Murat, 2014). However, the melting temperature range of these fatty acids PCMs is relatively higher than the desired temperature range for practical solar thermal applications in building comforts. In addition, these fatty acids also suffer from relatively low latent heat of fusion.

The eutectic compositions, which are specific compositions of two or more materials with different melting temperatures, at which material has sharp melting and solidification without any phase segregation, may provide the desired thermophysical properties for required applications (Mehling and Cabeza, 2008). The melting temperature of eutectic mixture is the lowest in the entire composition range of mixtures of individual phase change materials. Eutectic compositions of fatty acids may provide low melting temperature range PCMs and thus, making them suitable for building heating/cooling applications. Usually, the mass fraction ratio, latent heat of fusion, and melting/solidification temperatures of eutectic mixtures are characterized using differential scanning calorimeter (DSC) measurements. Zhang et al. determined a eutectic composition for binary system of capric acid (CA) and Lauric acid (LA) consisting 65 wt% CA and 35 wt% LA with 18 °C melting temperature and 141 kJ/kg latent heat of fusion (Zhang et al., 2001). Kauranen et al. and Peippo et al. investigated several eutectic mixtures of binary fatty acids with melting temperature ranging from 10.2 to 18.5 °C and latent of heat ranging from 138 to 185 kJ/kg for thermal energy storage applications (Kauranen et al., 1991; Peippo et al., 1991).

There are other developments on different kinds of fatty acid esters via esterification of fatty acids using alcohols for enhanced phase change temperature and to reduce the bad odor problem of fatty acids (Sari and Kariapekili, 2009; Sari et al., 2011a,b; Costa et al., 2012; Agafonova et al., 2011; Feldman et al., 1986; Aydin and Aydin, 2012; Aydin, 2013). Sari et al. synthesized butyl and isopropyl stearate for thermal energy storage applications in buildings (Sari et al., 2009). The phase change temperature and latent heat of fusion of butyl stearate and isopropyl stearate are 23.67, 22.12 °C and 121, 113.1 kJ/kg respectively.

The latent heat of fusion of eutectic compositions of fatty acids is relatively low, which leads to the poor thermal energy storage density. Thermal energy storage system with such materials will be quite bulky to compensate the low energy storage density. Thus, there is a need to design and develop PCMs with considerably higher latent heat of fusion, desired melting temperature range 15–20 °C, stability against thermal cycling, low supercooling, congruent melting, non-toxic and lower cost with ease of availability. We designed new eutectic compositions of LA, MA and PA fatty acids with 1-deodecanol for low temperature solar thermal applications. In this paper, computation of such eutectic compositions with their melting point and latent heat of fusion are discussed and their experimental validation has been carried out using DSC measurements. The designed eutectic systems exhibit reasonably higher latent heat of fusion and melting temperatures in the desired temperature range.

2. Computational and experimental methods

2.1. Computational details

New phase change materials (PCMs) with desired melting point and latent heat of fusion properties can be designed and developed by mixing two or more materials in specific ratios. The solid-liquid behavior of mixtures of different materials can be modeled using the Gibbs excess energy approach (Diarcce et al., 2015). If a binary homogeneous liquid system containing two chemical substances A and B at constant pressure P does not form solid solution and the liquid is cooled, the relationship between solidified mole fraction of substance i (A or B) and respective change in enthalpy with their melting temperature for ideal solution may be represented by

\[ \ln x_i = \frac{\Delta H_i(T,P)}{R} \left( \frac{1}{T_{m,i}} - \frac{1}{T_m} \right) \]  \tag{1} 

Eq. (1) is the Schroder-Van Laar equation. Here, \( x_i \) is the mole fraction of substance i in liquid phase; \( \Delta H_i(T_P) \) is change in enthalpy at temperature \( T \) and pressure \( P \); \( T_{m,i} \) denotes the melting temperature of pure substance; \( T_m \) represents the temperature of the system and \( R \) represents the ideal gas constant, 8.315 J mol\(^{-1}\) K\(^{-1}\).

For liquid-solid phase transformation, \( \Delta H(T_P) \) is the molar enthalpy of fusion for substance i at temperature T and pressure P. The Liquidus compositions for substances A and B at various temperatures can be calculated using Eq. (1) and used for computing eutectic temperature and compositions at the intersecting point of their liquids lines (Diarcce et al., 2015).

The enthalpy of fusion for a system consisting multiple substances can be computed using following Eq. (2)

\[ H_m = T_{m} \sum_{i=1}^{n} \left[ x_i \Delta H_{m,i} + x_i (C_{PSA} - C_{PSB}) \ln \frac{T_{m,i}}{T_m} \right] \]  \tag{2} 

Here, \( C_{PSA} \) and \( C_{PSB} \) are specific heats at constant pressure in liquid and solid state for \( i \)th substance respectively. \( T_{m,i} \) and \( T_m \) are the melting temperatures of eutectic composition and pure i\textsuperscript{th} substance respectively (Kumar et al., 2016b). The above calculated enthalpy of fusion for eutectic composition comprises the solid and liquid specific heat contribution of pure chemical substances. As the organic phase change materials’ molecules are large and the difference between solid and liquid phase specific heats is very small compared to the molar enthalpy, hence the contribution of liquid and solid specific heats part in total enthalpy of eutectic composition becomes insignificant (<4%) as compared to molar enthalpy of fusion part (Kumar et al., 2016b). Hence, we ignored the second term of Eq. (2) and it becomes

\[ H_m = T_{m} \sum_{i=1}^{n} \left[ x_i \Delta H_{m,i} \right] \]  \tag{3} 

We used Schroder-Van Laar model to compute solid-liquid phase diagrams for lauric acid (LA) – 1-dodecanol (DE), myristic acid (M A) – 1-dodecanol (DE), and Palmitic acid (PA) – 1-dodecanol (DE) binary systems and determined eutectic point. The latent heat of fusion for these binary eutectic mixtures is calculated using Eq. (3).
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