



Preparation of phase change material emulsions with good stability and little supercooling by using a mixed polymeric emulsifier for thermal energy storage

Fangxian Wang, Xiaoming Fang*, Zhengguo Zhang*

Key Laboratory of Enhanced Heat Transfer and Energy Conservation, The Ministry of Education, School of Chemistry and Chemical Engineering, South China University of Technology, Guangzhou 510640, China

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ABSTRACT

A mixed polymeric emulsifier consisting of polyvinyl alcohol (PVA) and polyethylene glycol-600 (PEG-600) was explored for preparing high-performance phase change material emulsions (PCMEs) containing the paraffin with a melting point of 62–64 °C. After the effects of the mass ratio of PVA to PEG-600, the mass ratio of the mixed emulsifier to the paraffin and the homogenization rate on the size distribution, viscosity and dispersion stability of the PCMEs containing 20 wt% paraffin were systematically investigated, the optimal emulsifying process parameters have been determined to be the mass ratio of PVA to PEG-600 of 50:50, the mass ratio of the mixed emulsifier to paraffin of 1:5 and the homogenization rate of 10,000 rpm. Accordingly, the PCMEs with the paraffin mass fractions varying from 10 wt% to 30 wt% were prepared. It is found that the paraffin has been well dispersed in water in the form of sphere-like droplets with average diameters ranging from 3 μm to 11 μm in the obtained PCMEs. Significantly, the PCMEs with different mass fractions of the paraffin exhibit no supercooling, owing to the function of the mixed polymeric emulsifier as a nucleating agent. The apparent specific heat of PCMEs are 1.51–2.18 times as high as that of water at the phase transition temperature region, due to the existent of the paraffin in them. Their apparent thermal conductivity gradually decreases with an increase in the mass fraction of the paraffin, while the viscosity increases significantly with mass fraction of the paraffin at the same temperatures. Furthermore, the pumping power consumption of the PCMEs shows a drastic reduction as compared to that of water at the same heat storage capacity. It is revealed that the PCMEs show great potential for use as a novel heat transfer fluid (HTF) in thermal energy storage (TES) systems.

1. Introduction

Latent functional thermal fluids are dispersions of some amount of phase change materials (PCMs) into conventional heat transfer fluids (HTFs), which have been considered as an effective route for reaching heat transfer enhancement, due to their enhanced specific heat and energy storage capacity along with acceptable viscosity for pumping [1]. Generally, there are four categories of the latent functional thermal fluids, which include ice slurry, clathrate hydrate slurry, microencapsulated PCM slurry and PCM emulsion (PCME). Although the ice slurry, clathrate hydrate slurry and microencapsulated PCM slurry possess large energy storage density, they suffer from high cost [2,3]. Moreover, it is difficult for microencapsulated PCM slurries to maintain a stable and consistent flow state if the PCM capsules are large; and their shells increase heat transfer resistance [4]. By contrast, the PCMEs, manufactured by incorporating PCMs into water with the aid of

emulsifiers, possess several advantages, including simple preparation process, negligible thermal resistance, good operation stability and low cost [5,6]. Consequently, PCMEs show great potentials for use as novel HTFs with improved specific heat in various fields, such as solar thermal utilization, air-conditioning systems, electronic systems, and so on [7–11].

In order to accelerate the practical applications of PCMEs, extensive researches have long been conducted on them, which mainly focus on preparing stable PCMEs by investigating the suitable processing conditions including emulsifier type and amount along with stirring speed [6,12–15], studying their thermal properties [9,16] and rheological behavior [17–21], evaluating their heat transfer performance in different systems [11,22,23], together with exploring strategies to reduce their supercooling [24]. Note that stability and supercooling are the two important issues hampering the development and application of PCMEs. Specifically, for reducing supercooling of PCMEs, adding

* Corresponding authors.

E-mail addresses: cexmfang@scut.edu.cn (X. Fang), cezhang@scut.edu.cn (Z. Zhang).

Nomenclature			
A	Sectional area of the pipe (m^2)	v	Velocity (m s^{-1})
C_p	Apparent specific heat ($\text{kJ kg}^{-1} \text{K}^{-1}$)	V	Volume
d	Inside diameter of the pipe (m)	<i>Greeks</i>	
f	Fanning friction factor (–)	γ	Shear rate (s^{-1})
K	Fluid consistency coefficient (–)	η_p	Efficiency of the pump (–)
L	Length of pipe (m)	μ	Viscosity (Pa s)
m	Mass flow rate (kg s^{-1})	ρ	Density (kg m^{-3})
n	Dimensionless flow behavior index (–)	τ	Shear stress (Pa)
ΔP	Pressure drop (Pa)	<i>Subscript</i>	
P_p	Pumping power (W)	<i>PCME</i>	Phase change material emulsion
q	Volume flow rate ($\text{m}^3 \text{s}^{-1}$)	<i>w</i>	Water
Q	Energy storage capacity (kJ)		
Re_{MR}	Reynolds number (–)		
T	Temperature ($^{\circ}\text{C}$)		

nanomaterials [25–27] and solid paraffin wax with higher phase change temperature [28,29] and employing appropriate emulsifiers [6,30] as nucleating agents have been proved to be effective methods [4]. However, although the addition of nanomaterials can effectively reduce the supercooling degree of PCMEs, the nanomaterials dispersed in PCMEs are prone to aggregate and settle; the addition of solid paraffin wax with higher phase change temperature gives rise to an increase in viscosity of PCMEs, especially at the temperatures lower than its phase transition point [28]. Comparatively, utilizing appropriate emulsifiers should be an advisable approach, because the obtained PCMEs not only do not contain other additives but also can be expected to possess small supercooling as well as good stability through the tight wrapping of the PCM colloid particles with the emulsifiers. Günther et al. [30] studied the supercooling phenomenon for the hexadecane/water emulsions containing various surfactants; they found that the supercooling degree was about 12 $^{\circ}\text{C}$ for the PCME emulsified by sodium dodecyl sulfate (SDS) and 7.6 $^{\circ}\text{C}$ for the one fabricated with the help of polyoxyethylene sorbitan monopalmitate (Tween 40). Golemanov et al. [6] prepared a series of PCMEs by employing various composite surfactants, including Tween surfactants and Brij surfactants with different numbers of carbon atoms in their hydrophobic tail, α -olefin sulfonate, betaine, and so on; it was found that although the stability and supercooling of the PCMEs were improved in the presence of long-chain surfactants, their supercooling degrees were still greater than 4 $^{\circ}\text{C}$. All these researches suggest that the type of the surfactants and the composition of the mixed emulsifiers have distinct impact on the stability and phase change behavior of the obtained PCMEs. Apparently, it is still highly necessary to explore novel composite emulsifiers for preparing the PCMEs with good stability as well as little supercooling.

Polymeric emulsifiers, including polyvinyl alcohol (PVA), lignosulfonate, carboxymethyl cellulose, ethylene oxide/propylene epoxide copolymer, etc., possess relatively large molecular mass as compared with common surfactants and have been widely used in many fields, such as oil recovery, drug delivery systems and plastics products [31–33]. It has been reported that polymeric emulsifiers can play a dual role of improving the mechanical properties of the interface film and increasing the affinity between the dispersed phase and continuous phase, by adsorbing at the oil/water interface [34,35]. Moreover, it has been found that polymer can be used as a highly active nucleating agent to accelerate the crystal growth of isotactic polypropylene [36]. Based on the above reports, it can be inferred that polymeric emulsifiers are suitable for preparing PCMEs with good stability and small supercooling. However, little work has been done on the preparation of PCMEs by using polymeric emulsifiers by now.

In this work, a kind of mixed polymeric emulsifiers consisting of PVA and polyethylene glycol-600 (PEG-600) was explored for

preparing the PCMEs containing a paraffin with a melting peak temperature of 62–64 $^{\circ}\text{C}$. Firstly, the effects of the mass ratio of PVA to PEG-600, the mass ratio of the mixed emulsifier to the paraffin, and the homogenization rate on the size distribution, stability and viscosity of the obtained PCMEs were investigated systematically, with the purpose of optimizing the mass ratio and concentration of the mixed emulsifier. Secondly, the PCMEs with different mass fractions of paraffin were prepared using the optimized mixed emulsifier under a homogenization rate of 10,000 rpm, and the morphology and size distribution of the obtained PCMEs were characterized. Thirdly, the thermophysical properties, rheological behavior and viscosities of the PCMEs with different mass fractions of paraffin were measured. Finally, the pumping power consumptions for transporting the PCMEs containing different concentrations of paraffin were calculated based on their thermophysical properties and viscosities.

2. Experimental section

2.1. Materials

Paraffin (melting point was 62–64 $^{\circ}\text{C}$) was purchased from Huayong Paraffin Co, Ltd. Polyvinyl alcohol (PVA) AH-26, whose degree of alcoholysis was 97.0–98.8% and molecular weight was about 110,000, was supplied by Sinopharm Chemical Reagent Co., Ltd. Polyethylene glycol-600 (PEG-600) were purchased from Shanghai Chemical Reagent Co., Ltd.

2.2. Preparation of PCMEs

The PCMEs were prepared by emulsifying the paraffin into water on a rotor-stator homogenizer (model FJ200-SH, Shanghai Specimen Model Factory) at 80 $^{\circ}\text{C}$ for 5 min using a mixture of PVA and PEG-600 as the emulsifier. In order to obtain the PCMEs with small droplets as well as good fluidity and dispersion stability, the effects of the mass ratio of PVA to PEG-600, the mass ratio of the mixed emulsifier to the paraffin and the homogenization rate on the size distribution, viscosity and dispersion stability of the PCMEs containing 20 wt% the paraffin were systematically investigated. Specifically, for investigating the effect of the mass ratios of PVA to PEG-600, a series of the PCMEs were prepared at the mass ratios of 30:70, 40:60, 50:50, 60:40, and 70:30, respectively, under the condition of the mass ratio of the mixed surfactant to paraffin fixed at 1:5 and the homogenization rate fixed at 10,000 rpm. To explore the effect of the mass ratio of the mixed emulsifier to the paraffin, another series of the PCMEs were prepared at the mass ratios of 1:10, 3:20, 1:5, 1:4 and 3:10, respectively, under the condition of the mass ratio of PVA to PEG-600 fixed at 50:50 and the homogenization rate fixed at 10,000 rpm. Moreover, several PCMEs

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