Synthesis and Heat Transfer Performance of Phase Change Microcapsule Enhanced Thermal Fluids

Polyalphaolefins (PAOs) are widely implemented for electronics cooling, but suffer from a low thermal conductivity of about 0.14 W/mK. However, adding thermally conductive, phase-change-material (PCM) particles to a PAO can significantly improve the fluid thermal properties. In this paper, PCM microcapsules and silver-coated PCM microcapsules were synthesized using the emulsion polymerization method and the thermal performance of PCM fluids was studied in a microchannel heat sink and compared with that of the pure PAO. A test loop was designed and fabricated to evaluate the synthesized PCM fluids and it was found that fluid with uncoated PCM microcapsules has a 36% higher heat transfer coefficient than that of the pure PAO. Additionally, the heat transfer coefficient of PCM fluids with silver-coated PCM microcapsules was also 27% higher than that of pure PAO, but lower than that of fluids with uncoated PCM microcapsules. The thermal resistance of the uncoated PCM fluid was about 20% lower than that of the pure PAO fluid at the same pumping power, despite the PCM fluid’s higher viscosity. Pumping tests were run for several hours and showed no evidence of particle accumulation or settling within the heat transfer loop. [DOI: 10.1115/1.4030234]

Keywords: phase change materials (PCM), thermal conductivity, heat transfer fluids

1 Introduction

Interest in engineered suspensions of micro-/nanoparticles in liquids has increased in recent years, particularly due to the potential for higher fluid thermal conductivities, resulting in smaller and more compact heat exchangers [1–8]. Several researchers [9–13] have experimentally investigated the convective heat transfer of aqueous metal oxide or nitride nanoparticles in circular tubes and results show enhancement of the heat transfer when nanoparticles are added to the cooling fluid. For example, Chein and Chuang [14] studied copper oxide/water mixtures in a microchannel heat sink and their experimental results indicated higher energy absorption of thermal fluids compared with pure water at low flow rates. However, no heat transfer enhancement was observed at higher flow rates.

Microencapsulated PCMs, i.e., PCMs encapsulated in a micron-sized shell, can mitigate PCM leakage and potential undesirable interactions between the PCM and the base fluid.[3,4,15–22] Rao et al. [23] and Dammel and Stephan [24] conducted experimental studies on the convective heat transfer characteristics of microencapsulated PCM particles suspended in water in minichannels. At low particle concentrations (below 5%), higher heat transfer was found at all flow rates. Higher heat transfer coefficients were achievable with higher particle concentration, however only at low flow rates. The results also showed that the maximum enhancement occurred if the inlet temperature of the fluids was slightly below the theoretical melting temperature of the PCM. The main enhancement provided by PCM is a significant increase in thermal storage density due to the latent heat of melting [17]. However, thermal conductivity of PCMs is relatively low. For example, the PCM wax and PAO fluid have comparable thermal conductivities of around 0.14 W/mK. Low thermal conductivity restricts heat transfer rate as well as accessibility to thermal energy stored away from heat transfer interfaces. Therefore, there is a need for microencapsulated PCMs with high thermal conductivity in addition to high heat capacity.

A combination of these two advantages to enhance the heat transfer properties of a heat transfer fluid is rarely reported. In our previous research, we developed a comprehensive process to microencapsulate paraffin wax PCMs in melamine-formaldehyde resin shell for high heat capacity and suppressed supercooling [22]. In this study, thermal conductivity of the PCM microcapsules is further enhanced with silver coating on the surface of melamine-formaldehyde resin shell. The as-produced PCM microcapsules, with and without a silver coating, are dispersed in PAO to make phase changeable, thermal conductive fluids, and their convective heat transfer characteristics are investigated in a heat transfer loop.

2 Experiment

2.1 Synthesis and Property Characterization. The microcapsules, consisting of paraffin wax in resin shell, were synthesized using the well-developed in situ polymerization method, as detailed in previous publications [15,22]. Typically, to produce prepolymers, melamine and formaldehyde are added into distilled water with adjusted pH at around 8.5 and kept at 70°C. Meanwhile, paraffin wax is added into water with surfactant and adjusted pH at around 4.0, then mechanically stirred at 60°C to make an oil-in-water emulsion. The as-produced prepolymer solution is then added to the emulsion, in which melamine-formaldehyde shell is formed and condensed on the surface of the paraffin-wax droplets. The final fluid is filtered and the solid white product is washed with distilled water and acetone, and then dried at 60°C overnight.

To enhance the thermal conductivity of the microcapsules, a thin layer of silver was coated on the surface of the PCM microcapsule particles. Coating of a silver layer on surface of microPCMs was performed in an aqueous solution based on so-called silver mirror reaction [25–27]. Typically, Tollens’
Table 1  Properties of PCM samples dispersed in PAO with the PCM weight percentage of 20%

<table>
<thead>
<tr>
<th>Sample</th>
<th>Shell material</th>
<th>Latent heat (J/g)</th>
<th>Ag (Vol. %)</th>
<th>Ag shell thickness (nm)</th>
<th>Thermal conductivity (W/mK)</th>
<th>Viscosity (cP)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pure PAO</td>
<td>N/A</td>
<td>0</td>
<td>0</td>
<td>NA</td>
<td>0.140</td>
<td>7.3</td>
</tr>
<tr>
<td>microPCM</td>
<td>Polymer</td>
<td>42.6</td>
<td>0</td>
<td>NA</td>
<td>0.152</td>
<td>11.8</td>
</tr>
<tr>
<td>PCM_Ag1</td>
<td>Polymer rough</td>
<td>33.0</td>
<td>2.2%</td>
<td>73</td>
<td>0.153</td>
<td>13.1</td>
</tr>
<tr>
<td>PCM_Ag2</td>
<td>Polymer rough</td>
<td>29.0</td>
<td>3.4%</td>
<td>117</td>
<td>0.239</td>
<td>13.6</td>
</tr>
<tr>
<td>PCM_Ag3</td>
<td>Polymer smooth</td>
<td>32.0</td>
<td>2.4%</td>
<td>82</td>
<td>0.251</td>
<td>11.2</td>
</tr>
</tbody>
</table>

reagent is produced by adding ammonia aqueous solution dropwise into a silver nitrate aqueous solution while stirring, until the fluid turns back to a clear and transparent solution. PCM microcapsules are then added into this solution while stirring. Sufficient amount of reducer such as glucose or formaldehyde solution is added into the suspension dropwise to produce a silver layer on the surface of the microcapsules. The mixture is filtered after reaction, and the black-to-gray powder is then washed with water and acetone and dried for future use.

By controlling the surface properties of the uncoated microcapsules, silver can be coated uniformly (in sample PCM_Ag1 and PCM_Ag3) or roughly (in sample PCM_Ag2), which affects their heat transfer properties. Silver layers are smooth coating on the original microcapsules. To create a rough silver layer on the microcapsules, the melamine–formaldehyde resin surface needs to be activated by stirring in diluted tin chloride solution for 10 min and then washed twice with distilled water [25,26]. After that, the same process as described above was performed, using surface-activated microcapsules instead of the original ones. The experimental results are summarized in Table 1.

The structure of the silver-coated PCM microcapsules was examined with energy-dispersive spectra (EDS) using a scanning electronic microscope (SEM, Hitachi SU-70), as shown in Fig. 1. The phase transition processes and latent heat capacity of the PCM microcapsules and their dispersions in PAO fluids were measured using the differential scanning calorimetry (DSC, TA-Q100). The weight and volume percentage of silver in the core-multi-shell particles could be determined by comparing the latent heat of microcapsules before and after silver-coating.

The thermal conductivity of the pure PAO and the dispersions of PCM microcapsules in PAO were measured at room temperature using a 3-o-wire method [4,28,29] and the dynamic viscosity of the pure PAO and PCM microcapsules fluids was measured using a commercial viscometer (Brookfield DV-I Prime) at room temperature. It is interesting that the PCM-Ag microcapsules with smooth silver coating are significantly more thermal conductive than the rough ones, even though the silver fraction of the later (3.4 vol. %, in PCM-Ag2) is apparently larger than that of the smooth coated PCM-Ag3 microcapsules (2.4 vol. %). Meanwhile, PCM-Ag1 and PCM-Ag2 with rough silver coating elevate viscosity of heat transfer fluids more significantly than PCM-Ag3. This indicates that the rough coating of silver does not form a continuous Ag layer but discrete silver clusters on the surface of microPCMs, which is less effective in enhanced thermal conductivity. As a result, it can be expected that the PCMAg3 fluid with smooth silver coating layer performs better as a heat transfer fluid than that of the other two PCM-Ag samples with rough silver-coating.

2.2  Heat Transfer Measurements. A heat transfer loop was designed and built specifically for evaluating the heat transfer performance of the PCM fluids in a liquid-cooled cold plate, and a schematic of the loop is shown in Fig. 2. The hold-up volume of the entire system is about 80 ml and the individual components in the heat transfer loop include: a reservoir tank, a centrifugal pump, a Coriolis flow meter, two heat exchangers, and a micro-channel cold plate (the test section). The reservoir tank is full of liquid and allows the fluid level to be monitored. A stirrer is used to make sure the stable and uniform distribution of the microcapsules in fluids. A centrifugal pump circulates the fluid through the entire system and the flow rate is measured using a flow and density meter and a transmitter-display. The coolant is preheated before entering the test section and cooled upon leaving the test section using two heat exchangers, which consist of two copper cold plates with internal criss-crossed fins brazed together. The temperatures of the working fluids (50/50 aqueous ethylene glycol) in the secondary loops of these two heat exchangers are controlled by two thermal baths. The test section consists of a Microcool CP-101 microchannel cold plate and a 10 × 10 mm ceramic heater soldered to the bottom. The temperature of the heater was monitored using two T-type thermocouples attached to the bottom of the cold plate. Two digital multimeters were used to measure the voltage and current being supplied to the heater wires and the power input is adjusted using a variable transformer. The

Fig. 1 (a) TEM images of the polymer shell of PCM microcapsules and SEM image of (b) PCM-microcapsules before silver coating, (c) with smooth silver coating, and (d) with rough silver coating. EDS analysis of (e) silver and (g) carbon in the smooth-coated microcapsule and (f) (h) that in rough-coated microcapsules are also shown.
cold plate is incorporated into a milled Teflon block to reduce heat losses. Four K-type thermocouples were placed in the system to measure the bulk temperature of the coolant after each heat exchanger and the test section. All thermocouples were calibrated in a thermal bath and the maximum deviation was $0.2^\circ C$.

3 Results and Discussion

3.1 Pressure Drop Results. The microchannel cold plate was tested with pure PAO, PAO suspensions containing uncoated PCM microcapsules, and PAO suspensions containing PCM microcapsules with a smooth silver coating (A3). Flow rates from

Fig. 2 Schematic diagram (left) of the experimental setup for heat transfer measurement and a photograph of the microchannel (right)

Fig. 3 (a) Pressure drop in the cold plate at different inlet temperatures and (b) friction factor comparison: PAO, PCM, and PCM-Ag3

Fig. 4 (a) Heat transfer coefficient and (b) Nusselt number at different inlet temperatures
60 ml/min to 220 ml/min, heater power input from 30 W to 70 W, and inlet temperatures from 20 °C to 35 °C were tested. Additionally, pressure drop tests were conducted without heat input at four different fluid inlet temperatures. The results are depicted in Fig. 3(a) and as expected for this range of Reynolds number, the pressure drop increases linearly with flow rate. It was also found that the fluids with PCM particles have a higher viscosity than pure PAO.

Figure 3(b) compares the friction factor, \( f = \frac{\Delta P \cdot D_b \cdot A_f^2}{\rho \cdot L \cdot V^2} \), obtained for each of the three different fluids (PAO, PCM, and PCM-Ag3) in the experiments. All results were also compared with the Darcy friction factor for a rectangular duct based on the channel width/height ratio \([30]\), \( f = \frac{84.9}{Re} \). It can be seen that the results for the three fluids are in good agreement with the numerical model, given the uncertainties in the viscosities of the fluids, the dimensions of the microchannel, and the increased pressure drop in the entrance and exit of the cold plate and the developing regions of the microchannel.

### 3.2 Heat Transfer Results

The trials with heat input were conducted over a range of flow rates and heater powers and the average heat transfer coefficients were calculated for each power input using the equation \( h = \frac{Q}{A_{suf} \cdot \Delta T} \). The experimental results are plotted in Figs. 4(a) and 4(b). For the pure PAO tests, the heat transfer coefficient increases as the temperature is increased, due to a slight increase in the fluid’s heat capacity with temperature. However, the absorption of heat by the melting of the PCM results is governed by a much more complex relationship between the heat transfer coefficient, the flow rate, the inlet temperature, and the heat input. The heat transfer coefficients for the PCM fluids increase as the inlet temperature is increased from 20 °C to 30 °C, but then decreases at higher temperatures. The amount of PCM that melts in the cold plate depends on the inlet temperature, flow rate, and heat input and the greater the amount of PCM that melts in the cold plate, the higher the heat transfer coefficient becomes. However, at inlet temperatures above 30 °C, a portion of the PCM has melted before entering the test section, decreasing the amount of PCM available for phase change in the cold plate, thus resulting in decreased heat transfer coefficients.

The fluids containing the silver-coated PCM microcapsules had lower heat transfer coefficients than the fluid containing PCM particles with no metallic coating, which is surprising given the higher thermal conductivity of the silver-coated PCM fluid. The silver-coated PCM microcapsules do have a lower latent heat than the pure PCM particles, thus it is suspected that this reduction in latent heat is dominating the improvement in thermal conductivity. As described above, the latent heat is responsible for altering the temperature gradients near the heat transfer surfaces, which increases the flow of heat into the fluid.

As shown in Fig. 4, the Nusselt numbers (Nu) of the PCM fluids are higher than those of pure PAO and PCM-Ag3, due to the higher heat transfer coefficients. The Nusselt numbers of PCM-Ag3 are lower than those of the pure PAO because of the higher thermal conductivity of the PCM-Ag3 fluid.

Using the equation below, the pumping power and the thermal resistance were also calculated in order to compare the thermal performance of the pure PAO, PCM, and PCM-Ag3 fluids.

\[
P_p = \frac{V \cdot \Delta P}{Q} \quad (1)
\]

\[
R_{th} = \frac{T_{max} - T_{in}}{Q} \quad (2)
\]

The results are plotted in Fig. 5. It can be found that, as expected, the pumping power of the pure PAO decreases with increasing
inlet temperature, due to the fluid’s viscosity decreasing with temperature. At 30°C, the thermal resistance of the PCM fluid is about 20% lower than that of the PAO over the range of flow rates tested and the heat transfer coefficient of the PCM fluid is about 36% higher than that of the PAO. However, as discussed, the advantage of this large increase in heat transfer rate is reduced by the increase in pumping power that is required to pump the PCM fluid. As can be seen in the figure, a similar result is found at other temperatures.

Based on the above data, significant improvement on heat transfer performance is achieved with the use of PCM microcapsules with large latent heat capacity, in comparison to the pure PAO fluids. It appears that the addition of silver coating can potentially lead to two competing effects: (1) increase fluid thermal conductivity and (2) decrease the latent heat of the PCM microcapsules. Future research needs to be performed to find an optimum to balance the thermal conductivity and latent heat capacity of PCM-Ag microcapsules for heat transfer applications.

3.3 Extended Duration, Constant Flow Rate Experiments. To investigate the stability of the PCM-enhanced fluids, experiments were run for several hours without heat input. The tests were conducted with the PAO fluids containing silver coated PCM microcapsules for 3 hr and the pressure drop versus time is plotted in Fig. 6. The pressure drops were relatively constant over the duration of these tests, which indicates that no accumulation of PCM particles was occurring in the cold plate. The small pressure drop variation can be attributed to fluctuations in the room temperature and flow rate.

4 Conclusions

PCM fluids with uncoated and silver-coated microcapsules were synthesized and characterized, and their heat transfer performance was compared with that of pure PAO, which was the base fluid for the PCM fluids. The PCMs fluid with uncoated PCM microcapsules had a 36% higher heat transfer coefficient relative to pure PAO over a range of flow rates and temperatures, when compared at the same flow rate. The thermal resistance of this PCM fluid at a given pumping power was about 20% lower than that of pure PAO. A silver-coated PCM-Ag3 fluid also showed an increased heat transfer coefficient, which was greater than that of pure PAO, but less than that of the uncoated PCM fluid. The decrease of heat transfer coefficient with silver coating is due to the lower latent heat capacity of PCM-Ag3, even though its thermal conductivity is enhanced. Pumping tests conducted over several hours showed no effect of either particle accumulation or settling within the heat transfer loop. From the results, it is clear that this PCM fluid would provide improved cooling relative to pure PAO in heat transfer applications.

Acknowledgment

The author Yang acknowledges the financial support from National Science Foundation (NSF) under Grant No. 1336778. The author Lawler acknowledges the financial support from Department of Energy (DE-FG02-07ER86295).

Nomenclature

\[ Re = \text{Reynolds’s number} \]
\[ T_{in} = \text{fluid inlet} \]
\[ T_{max} = \text{maximum heater temperature} \]
\[ T_{avg} = \text{average heater surface temperature} \]
\[ V = \text{volumetric flow rates} \]
\[ \Delta P = \text{pressure drop} \]
\[ \rho = \text{fluid density} \]
\[ \nu = \text{kinematic viscosity} \]
\[ \Theta = \text{degree} \]

References


Journal of Heat Transfer

SEPTEMBER 2015, Vol. 137 / 091018-5


