Characterization of Physical, Thermal and Hydrodynamic Properties of Microencapsulated Phase Change Material Slurry as a Heat Transfer Fluid

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ABSTRACT

There has been a growing interest in developing new fluids aiming to increase the efficiency of heat transfer systems. However, specifically-designed fluids were often too expensive. A good approach was incorporation of phase change materials (PCM) into the fluids. Yet, it brought about particular problems often associated with the miscibility of PCM with carrier fluids and with meeting the increased pumping power needs. With rapidly developing nanotechnology and introduction of new methods, micro-/nano-encapsulated PCM (ePCM) now seems to offer a solution. Thanks to encapsulation, the phases are always separated from each other during the heat transfer process, by which the advantages associated with two-phase flow at the micro level can be achieved and the advantages of single phase fluid at the macro level can be exploited. Volumetric slurry concentrations below 25% can be assumed to be Newtonian and under ideal conditions, the Nusselt number of the ePCM slurry is 1.5 to 4 times higher than that of the single phase flow. In this study, the use of ePCM slurries as heat transfer fluids and characterization of ePCM slurry as a working fluid in a heat transfer system is addressed.

Keywords—ePCM slurry; slurry characterization; hydrodynamic characteristics; heat transfer fuid; encapsulation

1.0 INTRODUCTION

Heat Transfer Fluids (HTF) are many. As the cheapest and readily available natural fluids, air and water are the most common fluids that have been used as HTF. Depending on the particular needs, water-based solutions as well as some chemicals compounds and solution thereof have also been used as HTF, and in some special cases CFC coolants and the likes are employed. In short the need for HTF with better properties or for one possessing the right properties that are required by particular applications is allways in consideration.

Today, it is probably no big deal to obtain specifically designed HTF that would meet any requirement, however, cost, related to both research & development and characterization, is a major concern.

One specifically designed HTF is micro/nano encapsulated phase change material slurries (ePCM-S), which is trending since the last decade as it became a no-longer-

expensive option thanks to the rapid development in the nanotechnology and highly developed methods for synthesizing micro or nanoscale capsules entrapping phase change materials (PCM) inside. Besides, theare are readily available, commercial, micro-/nano-encapsulated PCM which can be used in preparing ePCM-S. This article presents a brief of methods and mathematical equations and correlations that can be used in the characterization of ePCM-S.

2.0 CHARACTERIZATION OF ePCM-S

2.1 Thermo-Physical Characteristics

mePCM slurry with water as carrier fluid was prepared using microcapsules comprised of coconut oil as core and melamine formaldehyde as shell. The thermal and physical properties of microcpapsules used in the slurry preparation is given in Table 1 below.

Table 1. Thermal and physical properties of mePCM used in slurry, as obtained by DSC analysis.

		CO	**ePCM**
Melting	Enthalpy	105.1	82.6
	Onset Temperature	7.3	12.3
	Peak Temperature	21.1	24.6
	End Temperature	23.6	27
Crystallization	Enthalpy	76.3	18.9
	Onset Temperature	13.7	8.2
	Peak Temperature	9.7	5.8
	End Temperature	3.0	2.5
Encapsulation Ratio		-	78.59
Encapsulation Efficiency		-	55.95
Specific Heat		3.135	2.726
Thermal Conductivity		0.321	0.296

The flow and heat transfer properties of the fluid operating in a thermal system depend on independent parameters such as density, viscosity, thermal conductivity, melting and freezing temperatures and melting latent heat. Therefore, taking into account the concentration of PCM by weight, the values of these parameters for the fluid are calculated on the principles of conservation of mass and energy [1].

2.1.1 Density

In order to calculate the bulk density and specific heat capacity of the fluid, the density of the core and shell materials, the capsule and the carrier fluid, as well as the specific heat capacities must be known separately. Using the values in Table 1, the densities of the capsule and ePCM-S HTF are calculated as:

$$\rho_{cap} = \rho_{c,ave} * (1 - R) + \rho_{sh} * R \quad (1)$$

$$\rho_{sl} = \rho_{fl} * (1 - \phi) + \rho_{cap} * \phi \quad (2)$$

where, $\rho_{c,ave}$ is the average of the densities of core PCM in solid and liquid states.

The density value can also be calculated with the following equation

$$\rho_{cap} = \left(\frac{d_c}{d_{cap}}\right)^3 \frac{\rho_c}{W_c} \quad (3)$$

using composite sphere approach [2–4] or be directly measured.

2.1.2 Specific Heat Capacity

Specific heat capacities for capsule and slurry, using the values from Table 1, are calculated by the below equation [2, 3]:

$$C_{p,cap} = \frac{(R \times C_c + (1 - R) \times C_{sh})\rho_c \rho_{sh}}{(R \times \rho_{sh} + (1 - R) \times \rho_c)\rho_{cap}} \quad (4)$$

or by equation 5 [5]:

$$C_{p,cap} = R * C_{p,c} + (1 - R) * C_{p,sh} \quad (5)$$

However, essentially, the specific heat capacity of the capsule, as a function of temperature, also changes depending on whether the temperature is in the phase change temperature range or not (Figure 1). When the temperature is not within the phase change temperature range, the specific heat capacity of

the capsule is considered equal to the specific heat capacity of the core, while the specific heat capacity in this range is expressed as [6]:

$$C_{p,cap} = C_{p,c} + \left\{\frac{\pi}{2}\left(\frac{h_{s,fl}}{T_{m,e} - T_{m,o}} - C_{p,c}\right) sin\pi\left[\frac{T - T_{m,o}}{T_{m,e} - T_{m,o}}\right]\right\} \quad (6)$$

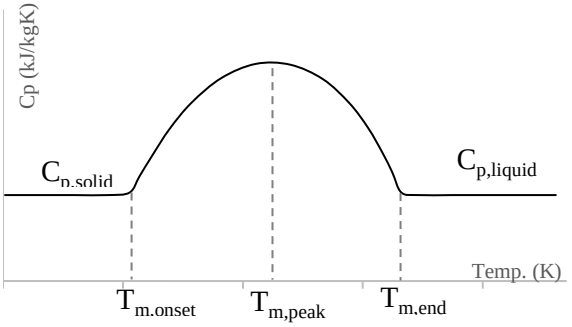

Figure 1. Change of ePCM's specific heat capacity with temperature (assuming equal Cp values of PCM in solid and liquid state) [6]

And the bulk specific heat capacity of the slurry can be calculated as [4, 5]:

$$C_{p,sl} = \xi * \mathrm{C_{p,cap}} + (1 - \xi) * C_{p,fl} \quad (7)$$

where ξ is the mass fraction of the capsule in the fluid.

2.1.3 Thermal Conductivity

Next, the thermal conductivity of the capsules can be obtained using equation 8 [5, 7]

$$\frac{1}{k_{cap} d_{cap}} = \frac{1}{k_c d_c} + \frac{d_{cap} - d_c}{k_{sh} d_{cap} d_c} \quad (8)$$

which can then be plugged into the following equation in order to obtain the thermal conductivity of the ePCM-S by Maxwell's relation.

$$\frac{k_{sl}}{k_{fl}} = \frac{2 + \frac{k_{cap}}{k_{fl}} + 2\phi\left(\frac{k_{cap}}{k_{fl}} - 1\right)}{2 + \frac{k_{cap}}{k_{fl}} - \phi\left(\frac{k_{cap}}{k_{fl}} - 1\right)} \quad (9)$$

Rearranging equation 9, thermal conductivity of the slurry can be expressed and calculated as [4]:

$$k_{sl} = k_{fl}\left[2 + \frac{k_{cap}}{k_{fl}} + \frac{2\phi\left(\frac{k_{cap}}{k_{fl}} - 1\right)}{2} + \frac{k_{cap}}{k_{fl}} + \phi\left(\frac{k_{cap}}{k_{fl}} - 1\right)\right] \quad (10)$$

However, the interaction between the fluid particles as well as the movement thereof also has an improving effect on thermal conductivity. Therefore, the effective thermal conductivity of the fluid should be expressed as the product of thermal conductivity of the fluid calculated in Equation 10 and a coefficient f reflecting the above-mentioned effects:

$$k_{eff} = f * k_{sl} \quad (11)$$

Here f is chosen according to the value of the slurry density and Peclet number, which is an expression of the transport rate by diffusion to the transport rate by advection in the solution. So f can be calculated as below, picking up the values of B and m depending on the value of Peclet number:

$$f = 1 + B + \phi Pe_{cap}^{m}$$

$$\begin{Bmatrix} B = 0, m = 1.5 & Pe < 0.67 \\ B = 1.8, m = 0.18 & 0.67 \le Pe \le 250 \\ B = 3.0, m = 1/11 & Pe > 250 \end{Bmatrix} \quad (12)$$

The Peclet number for spherical micro/nano particles can be obtained as:

$$Pe_{cap} = \frac{\gamma d_{cap}^2}{\alpha_{fl}} \quad (13)$$

where α is the thermal diffussivity of the base fluid, water in this case, γ is the shear rate, and d is the capsule diameter [4].

2.1.4 Viscosity

Finally, the viscosity of the slurry needs to be determined. The following theoretical

formula, developed by Einstein, can be used to calculate the viscosity of dilute suspensions of hard shell spheres.

$$\mu = \mu_0(1 + k\phi) \tag{14}$$

where μ is the viscosity of the suspension, μ_0 is the viscosity of the pure solvent, ϕ is the concentration by volume and k is a constant dependent on the shape, rigidity and Brownian motion of the particles, and determined to be 2.5 for rigid particles in spherical form (glass spheres) [8].

Considering that the suspension reacts the particle additions as a hydrodynamically homogeneous medium; by adding a particle with a volume dV to a solution, with the viscosity of μ, consisting of pure liquid in volume of A, which already contains similar particles of volume V, a theoretical viscosity formula applicable to higher concentrations can be obtained [8]. Thus, the increase in viscosity of the solution can be expressed in general as:

$$\begin{aligned} \mu + d\mu &= \mu\left(1 + k\frac{\mathrm{dV}}{\mathrm{A+V}}\right) \\ d\mu/\mu &= 1 + k\frac{\mathrm{dV}}{\mathrm{A+V}} \end{aligned} \tag{15}$$

When this differential change is integrated over the total change interval; the general formula:

$$\mu = \mu_0\left(1 + \frac{V}{A}\right)^k = \mu_0(1 - \phi)^{-k} \tag{16}$$

is obtained. Here, ϕ, as mentioned above, is the concentration by volume.

The assumption that the suspension is hydrodynamically homogeneous is valid only when the particles are sufficiently separate so that their interactions, except for causing an overall increase in viscosity, can be neglected. Particles moving relative to each other under the shearing motion of fluid collide and roll over each other until they separate again under further shearing. It has been shown that the duration the spherical particles are in contact with each other is directly proportional to the concentration. During this contact, a certain volume of liquid remains motionless around the point of contact. Therefore, it is clear that the effect of two spheres in contact with each other on viscosity will be greater than the two spheres that are separated from each other.

The volume of liquid that gets immobilized around the spheres is proportional to the volume of the spheres. Therefore, this stationary volume is expressed as $qV\phi$, q being an experimentally-determined proportionality factor. Therefore, the dead volume *A* must be subtracted from the liquid volume and added to particle volume *V*. Considering that the two colliding spheres will behave like a non-spherical particle during the time of contact, the shape factor k should also vary slightly with the concentration; however, this effect is negligible as it is very minor compared to the effect of immobilized liquid [8]. Consequently, the Equation 16 should be altered to:

$$\begin{aligned} \mu &= \mu_0\left(1 + \frac{V + qV\phi}{A - qV\phi}\right)^k \\ \mu &= \mu_0(1 - \phi - q\phi^2)^{-k} \end{aligned} \tag{17}$$

It was reported that the most suitable curve (given in Figure 2) for the experimental data obtained with Ostwald viscometers is yielded by Equation 17 having the value of k = 2.5 and q = 1.16.

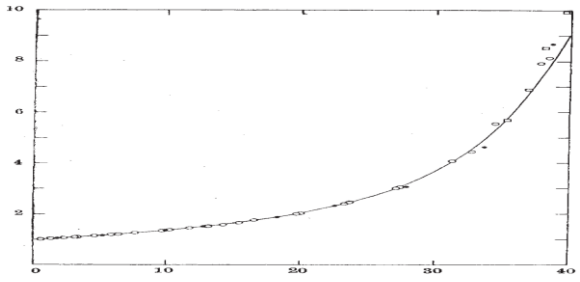

Figure 2. Relative viscosity of suspensions containing glass spheres as a function of volume fraction [Theoretical curve: values of Equation (17) for k = 2.5, q = 1.16] [8].

When this graph is examined, it is seen that the relative viscosity is quite small for values below 5%, and it is very close to 1 for values such as 1% and 2%. For this reason, we also decided to prepare the slurry with the ratios of 1%, 1.5% and 2% in order to prevent a significant increase in viscosity of HTF.

$$\mu_{sl} = \mu_{fl}\left(1 - \phi - A_{\phi}\phi^{2}\right)^{2,5} \quad (18)$$

where ϕ is the volumetric concentration of ePCM in the solution. Aϕ, which depends on the size, shape and type of the particle, is the proportionality constant that can be determined experimentally (indicated by q in the Vand formulation).

Wang et al. [9] compared their experimentally obtained viscosity values to the values calculated by Equation 18 (seen in Figure 2 above). The graph in Figure 3 shows that there is an acceptable agreement between the calculations and the experimental results.

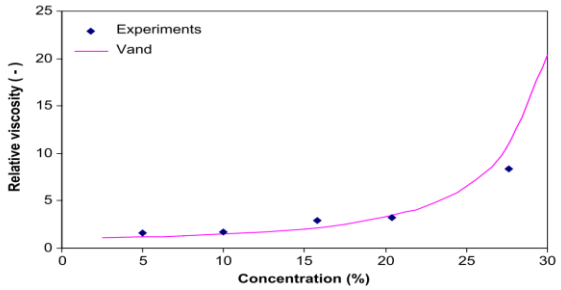

Figure 3. Change of relative viscosity with capsule concentration [9]

This correlation given by Vand [8], was used, in the general form below, in the calculation of solution viscosities and was reported to be valid by Goel et al. [2] and Qiu et al. [3]. Qiu et al. [3], in their study on the properties of ePCM-S and their use in solar systems, stated that $A\phi$ ratio constant to be 3.7 commercial microcapsules.

$$\frac{\mu_{sl}}{\mu_{fl}} = (1 - \phi - 3{,}7\phi^{2})^{-2,5} \quad (19)$$

As can be seen in Figure 2 , there is a very high agreement between the relative viscosity values obtained by the formula and the measurement results, especially when the concentration by volume is below 25%. Thomas [10] reported that this ratio was 97.5%, but with increasing concentration value, the agreement gradually disappeared, for example, it decreased to 60% for 40% and 8.7% for 60% concentration, so it is not reliable for high concentrations. Thomas [10], also created a better fitting variant of Equation 19, which he expressed as:

$$\mu_{eff} = \mu_{fl} + 2.5\phi + 10.05\phi^{2} + 0.00273\exp(16.6\phi) \quad (20)$$

However, since the solution concentration is low in our study (1%, 1.5% and 2%), Equation 19 offers an acceptable level of agreement, and this equation is used for concentrations below 25% in the literature.

3.0 Results and Discussion

Compared to water, although the specific heat capacity of the slurry is lower, within the melting temperature range, ePCM slurry has a significantly higher effective specific heat capacity, boosted by the latent heat of fusion.

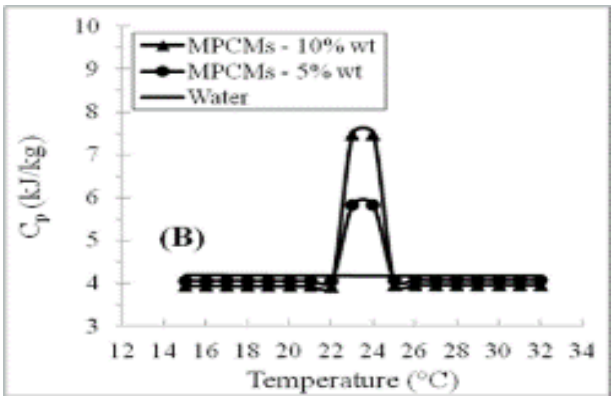

Figure 3. Specific heat capacity of ePCM-S and water [4]

On the other hand the increase in the viscosity of the slurry was almost negligible in

comparison. The pumping power required to draw same amount of heat is much less in ePCM-S than in pure water.

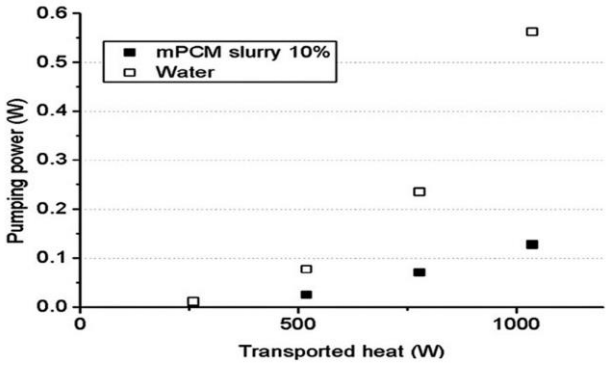

Figure 8 Pumping power versus heat drawn [11, 12]

4.0 Conclusions

Despite the fact that it is more viscous than water and causes more pressure drop, has an improved heat transfer coefficient the, the results show a significantly higher overall heat transfer coefficient for slurryi thus indicate that ePCM-S can be an advanced HTF.

Athough heat transfer properties is directly proportional to the ePCM concentration, there is a critical point under which the concentration should be kept due to increased pumping requirements and the fact that it decreases the thermal conductivity of the fluid outside the phase change range: 10%.

5.0 References

[1] Yamagishi Y, Takeuchi H, Pyatenko AT, Kayukawa N. Characteristics of microencapsulated PCM slurry as a heat-transfer fluid. *AIChE Journal* (1999) **45**(4):696–707. doi:10.1002/aic.690450405.

[2] Goel M, Roy SK, Sengupta S. Laminar forced convection heat transfer in microcapsulated phase change material suspensions. *International Journal of Heat and Mass Transfer* (1994) **37**(4):593–604.

[3] Qiu Z, Li P, Wang Z, Zhao H, Zhao X. PCM and PCM Slurries and Their Application in Solar Systems. In: Zhao X, Ma X, editors. *Advanced Energy Efficiency Technologies for Solar Heating, Cooling and Power Generation*. Cham: Springer International Publishing (2019). p. 101–141.

[4] Languri EM, Shaaban AH, Kong, Minsuk, Alvarado JL. Numerical Simulation of a Microencapsulated Phase Change Material Slurry Flowing in a Helical Coil Heat Exchanger. In: *Proceedings of the ASME International Mechanical Engineering Congress and Exposition, November 15-21, 2013, San Diego, California, USA*. New York, N.Y.: American Society of Mechanical Engineers (2013).

[5] Guo Y, Zhang X, Yang L, Xu C, Du X. The Heat Transfer of Microencapsulated Phase Change Material Slurry and Its Thermal Energy Storage Performance of Combined Heat and Power Generating Units. *Energies* (2017) **10**(10):1662. doi:10.3390/en10101662.

[6] Kuravi S. *Numerical Study Of Encapsulated Phase Change Material (epcm) Slurry Flow In Microchannels*. Doktora Tezi. University of Central Florida. Orlando, Florida (2009).

[7] Qiu Z, Ma X, Zhao X, Li P, Ali S. Experimental investigation of the energy performance of a novel Micro-encapsulated Phase Change Material (MPCM) slurry based PV/T system. *Applied Energy* (2016) **165**:260–271. doi:10.1016/j.apenergy.2015.11.053.

[8] Vand V. Theory of Viscosity of Concentrated Suspensions. *Nature* (1945) **155**(3934):364–365. doi:10.1038/155364b0.

[9] Wang X, Niu J, Li Y, Wang X, Chen B, Zeng R, et al. Flow and heat transfer behaviors of phase change material slurries in a horizontal circular tube. *International Journal of Heat and Mass Transfer* (2007) **50**(13-14):2480–2491. doi:10.1016/j.ijheatmasstransfer.2006.12.024.

[10] Thomas DG. Transport characteristics of suspension: VIII. A note on the viscosity of Newtonian suspensions of uniform spherical particles. Journal of Colloid Science, 20(3), 267-277. *Journal of Colloid Science* (1965) **20**(3):267–277. doi:10.1016/0095-8522(65)90016-4.

[11] Jurkowska M, Szczygieł I. Review on properties of microencapsulated phase change materials slurries (mPCMS). *Applied Thermal Engineering* (2016) **98**:365–373. doi:10.1016/j.applthermaleng.2015.12.051.

[12] Delgado M, Lázaro A, Mazo J, Marín JM, Zalba B. Experimental analysis of a microencapsulated PCM slurry as thermal storage system and as heat transfer fluid in laminar flow. *Applied Thermal Engineering* (2012) **36**:370–377. doi:10.1016/j.applthermaleng.2011.10.050