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# Thermal characteristics of paraffin in a spherical capsule during freezing and melting processes

# Keumnam Cho\*, S.H. Choi

School of Mechanical Engineering, Sungkyunkwan University, 300 Chunchun-dong, Changan-ku, Suwon 440-746, South Korea

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#### Abstract

The objective of the present study was to investigate the thermal characteristics of paraffin in a spherical capsule during freezing and melting processes. Experiments were performed with paraffin, i.e., *n*-tetradecane, and a mixture of *n*-tetradecane (40%) and *n*-hexadecane (60%) and water. The parameters were the Reynolds number and the inlet temperature during the freezing process and initial temperature during the melting process for a PCM-thermal-storage system. The phase-change period for the capsule at the edge of a storage tank was shorter than that at the center of the storage tank due to smaller porosity at the center than the edge of the storage tank. Water showed a bigger of subcooling than paraffin. It took longer for water to reach the dimensionless thermal-storage capacity of 1 during the freezing process, and shorter during the melting process than that for paraffin. The average heat-transfer coefficients were affected by the inlet or initial temperature and Reynolds number more during the melting process than during the freezing process due to a natural-convention effect during the melting process. The average heat-transfer coefficients for paraffin were larger by a maximum of 40% than those for water during the freezing and melting processes.  $\mathbb{C}$  2000 Elsevier Science Ltd. All rights reserved.

Keywords: Spherical capsule; Freezing; Melting; PCM-thermal-storage system; Heat transfer coefficient

### 1. Introduction

A thermal-storage system has the advantage of an efficient use of energy by reducing the imbalance of an electric demand between daytime and nighttime in summer. It is classified as sensible and latent heat-storage systems. The latent heat-storage system is superior to the sensible heat-storage system since the former reduces the installation area and the expense due to a large thermal-storage capacity and constant

phase-change-temperature during freezing and melting processes. The latent heat-storage system is classified as static and dynamic systems. The ice-in-coil, ice-oncoil, ice-ball, and ice-lens systems are the static systems, whereas the ice-harvest, ice slurry, and phasechange-material slurry systems are the dynamic systems.

The ice-ball system has the advantage of relatively large thermal area and easy installation. Saitoh [1] who used *n*-heptadecane ( $C_{17}H_{36}$ ) as a phase-change-material reported that a spherical capsule had the best thermal-storage performance among spherical, plate, cylindrical, and coil shapes. Kamiya [2] and Choi et al. [3] investigated the overall thermal-storage characteristics of a spherical-capsule system with respect to the

<sup>\*</sup> Corresponding author. Tel.: +82-331-290-7445; fax: +82-331-290-5849.

E-mail address: keumnam@yurim.skku.ac.kr (K. Cho).

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#### Nomenclature

- A Surface area of spherical capsule  $(m^2)$
- c Specific heat of PCM (kJ kg<sup>-1</sup> K)
- d Diameter of spherical capsule (m)
- *H* Heat of fusion (kJ kg<sup>-1</sup>)
- *h* Average heat-transfer coefficient (W m<sup>-2</sup> K<sup>-1</sup>)
- *n* Total number of spherical capsules
- *Q* Thermal-storage capacity (kJ)
- q Heat transfer rate (W)
- *Re* (Porosity) Reynolds number,  $\rho Ud/((1 \varepsilon)\mu)$
- Ste Stefan number,  $c(T_{\rm fi} T_{\rm m})/H_{\rm m}$
- T Temperature ( $^{\circ}$ C)
- t Time (s)

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U Average velocity of circulating fluid (m s<sup>-1</sup>)
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V Volume (m<sup>3</sup>)

#### Greek symbols

- $\varepsilon$  Porosity of capsule in storage tank,  $V_{\rm ci}/(V_{\rm ci} + V_{\rm c})$
- $\mu$  Viscosity of circulating fluid (kg m<sup>-1</sup> s<sup>-1</sup>)
- $\rho$  Density of circulating fluid (kg m<sup>-3</sup>)

#### Subscripts

- c Capsule
- ci Circulating fluid
- fi Inlet condition of circulating fluid
- i Initial
- m Melting
- o Outside of capsule
- tot Total

#### Superscript

+ Dimensionless

temperature difference between the inlet and outlet of a storage tank.

Chen and Yue [4] and Saitoh and Hirose [5] investigated thermal characteristics in a thermal-storage tank by adopting one-dimensional axial model. Benenati and Brosilow [6] reported that the porosity distribution in the radial direction of a rockbeds system varied within the distance of 4.5-5 rock diameters from the wall of a storage tank, and the fluctuation in the porosity distribution decreased near the center of the storage tank. Lerou and Froment [7] experimentally investigated velocity and temperature distributions in the radial direction of a rockbeds storage tank and reported that the velocity and temperature fluctuations near the tank wall were bigger than those at the center due to the different porosity distribution in the radial direction of the tank. Thus, the thermal characteristics in the spherical capsules of a thermal-storage system may be different in the radial direction of the storage tank due to the different porosity distribution in the radial direction of the tank, a phenomenon requiring further investigation of the local thermal characteristics in the spherical capsule in not only axial but also radial directions of the storage tank.

A commercial spherical-capsule system utilizes water with an inorganic material. The inorganic material which is a nucleating agent is used both for reducing a subcooling effect in a spherical capsule and making solidification temperature near 0°C. The subcooling phenomenon usually deteriorates the thermal performance of the spherical-capsule system. Arnold [8] investigated the effect of the degree of subcooling on the thermal characteristics of the spherical-capsule with water by varying the capsule diameter.

Thermal-storage materials which had higher melting temperature than water but similar latent-heat capacity with water are required for the spherical-capsule system. The candidates are clathrate and paraffin. Clathrate has the problem of restriction of CFC and HCFC refrigerants and shows a relatively high degree of subcooling. The clathrate with a melting temperature from 5 to 15°C was investigated by Chung et al. [9] and Kim et al. [10]. Paraffin has the advantages of a wide range of melting temperature, almost negligible degree of subcooling and phase separation. Lim et al. [11] and Kim [12] investigated the thermal characteristics of paraffin for heating. Choi et al. [13] measured and reported melting temperatures and fusion energies of both *n*-Tetradecane and *n*-Hexadecane. The paraffin which has a higher melting temperature than water is required for use in the thermal-storage system.

The objective of the present study was to investigate the thermal characteristics of paraffin in a spherical capsule during freezing and melting processes.

#### 2. Experimental apparatus and procedure

The present experiments were performed with two kinds of paraffins: *n*-tetradecane ( $C_{14}H_{30}$ ) and a mixture of *n*-tetradecane 40% and *n*-hexadecane ( $C_{16}H_{34}$ ) 60%. The purity of both paraffins was 94%. Paraffin is non-poisonous, chemically stable, has almost negligible



Fig. 1. Measured temperature and fusion energy of paraffins and distilled water by differential scanning calorimeter (DSC).

degree of subcooling during nucleation and a small volume change during a phase-change process.

Fig. 1 shows melting temperature and fusion energy of paraffins and distilled water measured by a differential scanning calorimeter (DSC). The measured melting temperature of distilled water was  $0.81^{\circ}$ C. Since the melting temperature of distilled water is  $0^{\circ}$ C, the value of  $0.81^{\circ}$ C was applied for calibrating the measured melting temperature of paraffins. The calibrated melting temperature ( $T_{\rm m}$ ) and fusion energy ( $H_{\rm m}$ ) were 3.64°C and 172.1 kJ/kg for *n*-tetradecane and 6.81°C and 120.9 kJ/kg for the mixture. Water with an inorganic material was used for comparison with paraffin.

A schematic diagram of the present experimental apparatus is shown in Fig. 2. A detailed diagram of the test section is shown in Fig. 3. An acryl storage tank had a diameter ratio (ratio of inner diameter of the storage tank to the outer diameter of a spherical cap-



- 1. Test section
- 2~3. Constant temperature baths
- $4 \sim 8$ . Flow control valves
- 11. PC 10. Data acquisition system
  - Fig. 2. Schematic diagram of the present experimental apparatus.

9. Flow meter



- 1. Test section
- 4. Inner insulator
  - 5. Outer insulator
- 7. Metal screen
- 8. Steel scrubber

2. Spherical capsule

- 3. Honeycomb
- 6. Thermocouple

- Fig. 3. Detailed diagram of the test section.



Fig. 4. Temperature variation inside and on the surface of a capsule filled with *n*-tetradecane along the centerline of the tank during freezing process ( $T_{fi} = -4^{\circ}C$  (*Ste* = 0.0983), Re = 16).

sule) of 8.4, an inner diameter of 220 mm, a wall thickness of 10 mm, and a height of 173 mm. The spherical capsules were piled up in seven layers in the storage tank. The average porosity  $(\varepsilon)$  of the capsules in the storage tank was 0.47. The porosity was defined as the circulating fluid volume  $(V_{ci})$  divided by the addition of circulating fluid volume to capsule volume  $(V_c)$ . The outside of the storage tank was thermally insulated by glasswool. Stainless steel funnels were used at the inlet and outlet of the storage tank. Metal scrubber was installed inside each funnel and a honeycomb section was used behind the funnel to make uniform flow. Hollow spherical capsules, made of high-density polyethylene, had an inner diameter of 24.5 mm and a thickness of 0.8 mm. Paraffins and water with an inorganic material were used to fill the capsules. The initial temperature  $(T_i)$  of the capsules before starting the experiment and the inlet temperature  $(T_{\rm fi})$  of the storage tank were controlled by a constant-temperature bath which circulated a 40% aqueous solution of ethylene glycol. The flowrate was measured by a rotameter which was calibrated within  $\pm 1\%$  accuracy. The temperatures inside and at the surface of capsules and the temperature of circulating fluid were measured by copper-constantan thermocouples which were calibrated within  $\pm 0.15^{\circ}$ C by using a standard RTD. The measured temperatures were recorded by a 60 channel data-acquisition system.

The key experimental parameters were the (porosity) Reynolds number and inlet temperature. The Reynolds numbers were 8, 12 and 16, which were calculated by using the outside diameter of a capsule and the average



Fig. 5. Temperature variation inside and on the surface of a capsule filled with *n*-tetradecane along the centerline of the tank during melting process ( $T_{\rm fi} = 10^{\circ}$ C (*Ste* = 0.0818),  $T_{\rm i} = -4^{\circ}$ C, *Re* = 16).

porosity in the storage tank. The inlet temperatures were -7, -4, and  $-1^{\circ}$ C, and the initial temperature of the experiment was  $10^{\circ}$ C during the freezing process. The inlet temperature was  $10^{\circ}$ C, and the initial temperatures were -7, -4,  $-1^{\circ}$ C during the melting process. Stefan number (*Ste*) was calculated by using inlet temperature during the freezing and melting processes.

The initial temperature inside the storage tank was set to  $10^{\circ}$ C by a constant temperature bath (part 3 in Fig. 2), and another bath (part 2 in Fig. 2) was set to one of the inlet temperatures before the freezing-process experiment. The freezing-process experiment was started by connecting the bath (part 2 in Fig. 2) to the test section and turning off the bath (part 3 in Fig. 2). The melting-process experiment was performed in the reverse order of the freezing-process experiment.

#### 3. Results and discussion

#### 3.1. Temperature inside a spherical capsule

The temperature variations inside and on the surface of a spherical capsule filled with *n*-tetradecane located along the centerline of the storage tank are shown in Fig. 4 during the freezing process and in Fig. 5 during the melting process when the Reynolds number was 16. The inlet temperature was  $-4^{\circ}C$  (*Ste* = 0.0983) during the freezing process and  $10^{\circ}C$  (*Ste* = 0.0818) during the melting process. In both the figures, the three interior temperatures indicated as 1, 2, and 3 are shown. The temperature at location 2 for the seventh layer is higher than that at location 3 during the freezing process and lower than that at location 3 during the melting process while the opposite is true for other



Fig. 6. Temperature variation on the center of a capsule filled with *n*-tetradecane and of working fluid around the capsule (freezing:

 $T_{\rm fi} = -4^{\circ} C$  (*Ste* = 0.0983),  $T_{\rm i} = 10^{\circ} C$ , *Re* = 16; melting:  $T_{\rm fi} = 10^{\circ} C$  (*Ste* = 0.0818),  $T_{\rm i} = -4^{\circ} C$ , *Re* = 16).

layers. The reason is that the capsule at the center of the seventh layer was located near the outlet of the storage tank and thus affected by the circulating fluid at the outlet of the tank. In particular, the temperature difference between 2 and 3 at the first and the fourth layers during the melting process causes the naturalconvection inside a capsule, a phenomenon which will be mentioned later. The temperature at the surface of the capsule was always lower than the temperature inside the capsule during the freezing process. During the phase-change process, the temperature at the center of the capsule was almost constant, but the temperatures at the other locations than the center decreased since the heat removal was larger than the solidification energy. Grodzka [14] reported that the temperature during a phase-change process decreased since the

heat removal was larger than the solidification energy when a freezing process was slow and the temperature gradient inside a capsule increased due to the impurity of paraffin.

The time for the capsules at the first layer along the centerline of the storage tank to reach the inlet temperature of  $-4^{\circ}$ C was less by 20 min than that at the seventh layer during the freezing process. The time for the capsules at the first and seventh layers along the centerline of the storage tank to reach the inlet temperature of  $10^{\circ}$ C was less by 25 min than that at the fourth during the melting process as shown in Fig. 5.

The temperature variation of the capsule filled with n-Tetradecane located at the center and edge of the first and seventh layers of the storage tank were measured to investigate local temperature variations in



Fig. 7. Effect of the Reynolds number and inlet temperature on the phase-change period at the center of a capsule filled with *n*-tetradecane located at the center of the fourth layer of storage tank (freezing:  $T_i = 10^{\circ}$ C; melting:  $T_{fi} = 10^{\circ}$ C (*Ste* = 0.0818)).

the axial and radial directions of the storage tank. Fig. 6 shows the temperature variation of the circulating fluid around the capsule when the inlet temperature was  $-4^{\circ}C$  (*Ste* = 0.0983) during the freezing process and was 10°C (*Ste* = 0.0818) during the melting process with the Reynolds number at 16. The temperature gradient in the liquid-sensible-heat region was larger than that in the solid-sensible-heat region. The reason being that the thermal diffusivity of paraffin in the liquid phase is smaller by 26% than that in the solid phase.

#### 3.2. Phase-change period in storage tank

The phase-change period is defined as the period from start to end of phase change, as indicated by temperature deviation of  $\pm 0.2^{\circ}$ C from the slope of the phase-change temperature. The phase-change period for the capsule at the first layer was shorter than that for the capsule at the seventh layer, and the phasechange period for the capsule at the edge of the storage tank was shorter than that at the center of the storage tank as shown in Fig. 6. The reason is that the porosity at the center of the tank is smaller than that at the edge of the tank at the same layer of the storage tank as reported by Benenati and Brosilow [6]. The differences of phase-change period between the first and seventh layers and between the center and edge of the storage tank were larger during the freezing process than during the melting process.

Fig. 7 shows the effect of the Reynolds number and inlet or initial temperature on the phase-change period



Fig. 8. Effect of phase-storage material on phase-change period at the center of a capsule located at the center of the fourth layer of storage tank (freezing:  $T_{\rm m} - T_{\rm fi} = 8^{\circ}$ C,  $T_{\rm i} = 10^{\circ}$ C; melting:  $T_{\rm m} - T_{\rm i} = 8^{\circ}$ C,  $T_{\rm fi} = 10^{\circ}$ C).

at the center of a capsule filled with *n*-tetradecane located at the center of the fourth layer of the tank. The phase-change periods during the freezing and melting processes decreased as the Reynolds number increased and the inlet or initial temperature decreased. As the inlet or initial temperature increased from -7 to  $-1^{\circ}$ C, the phase-change periods during the freezing and melting processes increased by 88% and 7%, respectively, when the Reynolds number was 8 and by 53% and 11%, respectively, when the Reynolds number was 16. The dotted line in Fig. 7 showed the phase-change period subtracted by the subcooling period for the inlet temperature of  $-1^{\circ}$ C. The subcooling period was 11 min for the Reynolds number of 8 and 4 min for the Reynolds number of 16. The effect

of the inlet or initial temperature on the phase-change period during the freezing process was severer than that during the melting process.

Fig. 8 shows the effect of the thermal-storage material on the phase-change period at the center of a capsule and at the center of the fourth layer of the storage tank when the difference between the inlet or initial temperature and phase-change temperature was approximately 8°C. The subcooling period for the capsule filled with water was 76 min when the Reynolds number was 8. The dotted line in Fig. 8 shows the phase-change period for water subtracted by the subcooling period. The phase-change period during the freezing process for the capsule filled with paraffin was shorter by  $16 \sim 72\%$  than that with water. The phase-



Fig. 9. Effect of the inlet temperature and Reynolds number on the dimensionless thermal-storage capacity for a capsule filled with *n*-tetradecane in storage tank (freezing:  $T_i = 10^{\circ}$ C; melting:  $T_{fi} = 10^{\circ}$ C (*Ste* = 0.0818)).

change period during the melting process for whole phase-change materials was almost equal to that during the freezing process.

#### 3.3. Dimensionless thermal storage capacity $(Q^+)$

The total thermal-storage capacity in a storage tank can be calculated by the following equation:

$$Q_{\text{tot}} = \int_0^{t_1} q \, \mathrm{d}t + \int_1^{t_2} q \, \mathrm{d}t + \dots + \int_{n-1}^{t_n} q \, \mathrm{d}t \tag{1}$$

The total thermal-storage capacity varies with the phase-change material used and experimental parameters. Thus, the dimensionless thermal-storage capacity  $(Q^+)$  is defined as the relative contribution of the storage material and each experimental parameter as shown below:

$$Q^{+} = \frac{\int_{0}^{t} q \, \mathrm{d}t}{Q_{\mathrm{tot}}} \tag{2}$$

Fig. 9 shows the effect of the inlet or initial temperature of circulating fluid and Reynolds number on the dimensionless thermal-storage capacity for the capsule filled with *n*-Tetradecane. The time to reach the dimensionless thermal-storage capacity of 1 decreased during the freezing process as the inlet temperature decreased and was almost the same during the melting process as the initial temperature decreased. For the Reynolds number of 8, it took longer by 30% to reach the



Fig. 10. Effect of the storage material on the dimensionless thermal-storage capacity for a capsule in storage tank (freezing:  $T_{\rm m} - T_{\rm fi} = 8^{\circ}$ C,  $T_{\rm i} = 10^{\circ}$ C, Re = 16; melting:  $T_{\rm m} - T_{\rm i} = 8^{\circ}$ C,  $T_{\rm fi} = 10^{\circ}$ C, Re = 16).

dimensionless thermal-storage capacity of 1 than for the Reynolds number of 16 during phase-change processes. The dimensionless thermal-storage capacity was affected by the inlet temperature and Reynolds number during the freezing process, whereas it was not much affected by the initial temperature and Reynolds number during the melting process.

Fig. 10 shows the effect of the storage material on the dimensionless thermal-storage capacity when the difference between the inlet or initial temperature and phase-change temperature was approximately  $8^{\circ}$ C and the Reynolds number was 16. It took 27 min longer for water to reach the dimensionless thermal-storage capacity of 1 than for paraffin during the freezing process. However, it took 10–17 min less for water to reach the dimensionless thermal-storage capacity of 1 than for paraffin during the melting process.

## 3.4. Average heat transfer coefficient, $h_0$

The local heat-transfer coefficient on the surface of a capsule could not be estimated accurately since the temperature difference between the capsule and working fluid around the capsule was as small as  $\pm 0.15^{\circ}$ C. Thus, the average heat-transfer coefficient instead of the local heat-transfer coefficient in the storage tank was obtained by the following equation:

$$h_0 = \frac{q}{nA.\Delta T} \tag{3}$$

where the temperature difference  $(\Delta T)$  was the difference between the average temperature at the surface of the capsule and that of the circulating fluid at the first and seventh layers of the storage tank. The heat-transfer rate (q) was calculated by multiplying the tempera-



Fig. 11. Effect of the inlet temperature and Reynolds number on the average heat transfer coefficient for a capsule filled with *n*-tetradecane in storage tank (freezing:  $T_i = 10^{\circ}$ C; melting:  $T_{fi} = 10^{\circ}$ C (*Ste* = 0.0818)).

ture difference between the inlet and outlet of the storage tank by the mass flowrate and specific heat of the circulating fluid.

Fig. 11 shows the effect of the inlet or initial temperature and Reynolds number on the average heattransfer coefficient for the capsule filled with *n*-Tetradecane in the storage tank. The average heat-transfer coefficients increased by 10-25% during the freezing process and by 60-220% during the melting process as the inlet or initial temperature decreased from -1 to  $-7^{\circ}$ C. The average heat-transfer coefficients increased by approximately 30% during the freezing process and by 70-250% during the melting process as the Reynolds number increased from 8 to 16. The average heat-transfer coefficients were affected by the inlet or initial temperature and Reynolds number more during the melting process than during the freezing process. The reason is that the natural convection occurred inside the capsule during the melting process since the surface temperature of the capsule was higher than the inside temperature of the capsule. When the heat transfer coefficient data were compared with the predicted values in the literature by Whitaker [15] for flow of gases in a packed bed, the heat transfer coefficient data during freezing and melting processes agreed with approximately 18% with the predicted values.

Fig. 12 shows the effect of storage material on the average heat-transfer coefficient for the capsule in the storage tank when the difference between the inlet or initial temperature and phase-change temperature was approximately  $8^{\circ}$ C. The average heat-transfer coefficient was the largest for the *n*-Tetradecane, then, the



Fig. 12. Effect of the thermal-storage material on the average heat-transfer coefficient for a capsule in storage tank  $(T_m - T_{fi} = 8^{\circ}C)$  tank (freezing:  $T_m - T_{fi} = 8^{\circ}C$ ,  $T_i = 10^{\circ}C$ ; melting:  $T_m - T_i = 8^{\circ}C$ ,  $T_{fi} = 10^{\circ}C$ ).

mixture, and the least for water during the freezing process, whereas it was the largest for the mixture, then, the *n*-Tetradecane and the least for water during the melting process. In general, the average heat-transfer coefficients for paraffin were larger by up to 40% than those for water during freezing and melting processes.

#### 4. Conclusions

A summary of the present study is given below.

- The local phase-change temperatures of paraffin in a capsule were different during freezing and melting processes due to the difference between heat removal and solidification energy.
- 2. The phase-change period for the capsule at the edge

of a storage tank was shorter than that at the center of the storage tank because the porosity at the center was smaller than the edge of the storage tank.

- 3. The phase-change period during the freezing process for the capsule filled with paraffin was shorter by 16–72% than that with water. The difference in the phase-change period during the melting process for paraffin and water was less than during the freezing process. Water with an inorganic material showed a bigger degree of subcooling than paraffin.
- 4. It took longer for water to reach the dimensionless thermal-storage capacity of 1 during the freezing process, and shorter during the melting process than that for paraffin.
- 5. The average heart-transfer coefficients were affected by the inlet or initial temperature and Reynolds number more during the melting process than

during the freezing process due to the natural-convection effect during the melting process.

6. The average heat-transfer coefficients for paraffin were larger by up to 40% than those for water during freezing and melting processes.

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