Fundamental Study of Latent Cold Heat Energy Storage by Means of Oil Droplets at Low Freezing Point*

(Numerical Calculation of Motion and Solidification Characteristics of Oil Droplets Ascending in a Cold Water Solution due to Buoyancy)

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We have proposed a numerical calculation method for predicting the solidification characteristics of paraffin oil droplets (tetradecane, $\text{CH}_3(\text{CH}_2)_{12}$, melting point 5.8°C , diameter from 4 mm to 6 mm) ascending in a cold water solution layer (a mixture of ethyleneglycol ($\text{CH}_2\text{OH} \cdot \text{CH}_2\text{OH}$) and water, temperature from -2°C to -10°C) due to buoyancy. The oil droplets started to ascend with several initial velocities and initial temperatures ($10^\circ\text{C} - 60^\circ\text{C}$) in the water solution layer. The oil droplets were decelerated by hydraulic resistance of the water solution flow. The water solution flowed vertically downward at a low velocity of 0.1 m/s. The temperature of the oil droplet decreased due to direct-contact heat transfer with the cold water solution, and the oil droplet started to solidify when its surface temperature reached its freezing point. The solidified fraction of the oil droplet was calculated by taking into account a combination of the flow behavior and solidification characteristics of the oil droplet. As a result, the motion of the oil droplet, the completion height and time taken for solidification were determined. The parameters used were the diameter, initial velocity and initial temperature of the oil droplet, and temperature and velocity of the water solution.

Key Words: Heat Energy Storage, Solidification, Numerical Analysis, Oil Droplet, Direct-Contact Heat Exchange, Buoyancy, Ascending Velocity

1. Introduction

The authors⁽¹⁾⁻⁽²⁾ previously reported a new latent cold heat energy storage method via solidification of oil droplets injected into a layer of cold water solution. Those papers provided the experimental results of the solidified fraction of oil droplets ascending in the layer of cold water solution, in terms of oil droplet diameter, oil flow rate from the injection nozzle, oil temperature and water solution temperature. However, they were insufficient to elucidate the effects of those parameters on the solidification rate of oil droplets due to the limitation of experimental condi-

tion ranges. Therefore, it is necessary to predict the solidified fraction of oil droplets in wide ranges of these parameters, by numerical calculation. The previous results⁽²⁾, obtained using a simple physical model which dealt with a constant velocity of ascending oil droplets and homogeneous temperature profile in the oil droplet under the solidification process, indicated that the solidified fraction of oil droplets increased with increasing temperature of oil injected from the nozzle and the water solution, while it decreased with increasing ascending velocity of oil droplets in the layer of cold water solution.

However, we must take into account the fact that the ascending droplets are decelerated by the flow resistance of water solution, which flows downward, and there arises a temperature distribution in the oil droplet during the solidification process. Therefore, it is necessary to clarify the solidification characteristics of oil droplets in terms of flow behavior and

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temperature distribution of ascending oil droplets in the layer of flowing cold water solution, with the help of numerical calculation.

The purpose of this paper is to propose a suitable physical model with which to predict the solidified fraction of ascending oil droplets, taking account of flow behavior and physical properties of both oil droplets and water solution. The present numerical calculation has been carried out to determine the effects of oil droplet diameter, flow rate and temperature of oil as a solid-liquid phase change material, which is injected from the nozzle, and the flow rate and temperature of the water solution (30% mass fraction of ethyleneglycol) as a cold heat energy transfer medium on the solidified fraction and completion time of the oil droplets.

Nomenclature

a : thermal diffusivity
 C_D : flow drag coefficient
 c : specific heat
 d : diameter of oil droplet
 g : acceleration due to gravity
 K : assumed mass coefficient
 N : the number of meshes in numerical calculation
 Nu_{li} : internal Nusselt number
 Nu_{lo} : external Nusselt number
 Nu_{so} : external Nusselt number of solid sphere (solidified oil droplet)
 n : time step
 Pr : Prandtl number
 R : solidification fraction of frozen oil droplet
 Re : Reynolds number
 r : radius of oil droplet
 r_i : intersurface radius of solidified oil droplet
 r_o : outer radius of oil droplet
 T : temperature
 T_c : water solution temperature
 T_m : melting point of tetradeane (5.8°C)
 T_n : initial temperature of oil droplet or injection temperature of oil droplet as latent heat energy storage material
 ΔT : temperature range of apparent specific heat of tetradeane, which is assumed to be equivalent to the amount of solidification heat
 t : time
 Δt : time interval
 t_{cf} : completion time of solidifying oil droplets
 U_c : flow velocity of water solution
 U_n : initial velocity of oil droplet or injection flow velocity of oil from the nozzle
 U_p : absolute velocity of oil droplet
 We : Weber number

Y : vertical distance from the nozzle outlet
 Y_o : vertical distance from the nozzle outlet to the top surface of the water solution layer (1.6 m)
 Y_{cf} : ascending height of freezing oil droplets from start of solidification of oil droplet to completion of oil droplet solidification
 Y_m : vertical distance where the oil droplet starts to solidify in calculation
 α : heat transfer coefficient of oil droplet intersurface
 λ : thermal conductivity
 μ : viscosity
 ν : kinetic viscosity
 ρ : density
 σ : intersurface tension between oil droplet and water solution
 τ : time from start of moving oil droplet

subscripts :

c : water solution
 dl : tetradeane in a liquid state
 ds : tetradeane in a solid state

2. Physical Model for Numerical Calculation

2.1 Numerical calculation model for solidification of single oil droplet

The previous experimental results revealed that the packing factor of oil droplets in water solution layer was very small, and hence it was possible to deal with the flow behavior of each oil droplet as a single oil droplet. Therefore, the present numerical calculation was performed using the single oil droplet model, in which the single oil droplet ascended in the water solution layer and the freezing oil droplet was dealt

Fig. 1 Solidification proceeds uniformly on entire surface of rolling droplet

with as a solid sphere when it started to solidify from its surface. For a low Reynolds number below 200 in the present research, there appeared a maximum local heat transfer coefficient of the oil droplet at the top of the ascending oil droplet surface, which was at the front stagnation point, as shown in Fig. 1⁽³⁾. As a result, the freezing oil droplet rotated due to the appearance of the higher density part of the ascending oil droplet at the top of oil droplet surface; rotation of the freezing oil droplet occurred in every direction of the oil droplet surface, which allows the growth of the solidifying layer of the ascending oil droplet to be almost uniform under the solidification process. Therefore, the one-dimensional solidification model indicated in Fig. 2 was used for the present numerical analysis of the solidification characteristics of ascending oil droplets in a cold water solution layer. In Fig. 2, it is seen that the oil droplet is a sphere having a radius of $r_o(d_o/2)$ and the intersurface of the freezing oil droplet is located at a radius of r_i where the inside of r_i is in a liquid state, while outside of r_i is in a solid state. The diagrams on the right side in Fig. 2 show the tendencies of some physical properties of oil in both liquid and solid states in the oil droplet. The physical properties of tetradecane⁽⁴⁾, that is, specific heat c_d , density ρ_d and thermal conductivity λ_d , are expressed as a function of temperature in both liquid and solid phases for numerical calculation, as follows.

For specific heat c_d (J/(kg · °C)) ;

• liquid : $c_{dl} = 4.18T + 1.98 \times 10^3$ (1)

• solid : $c_{ds} = 1.80 \times 10^3$ (2)

For density ρ_d (kg/m³) ;

• liquid : $\rho_{dl} = 770$ (3)

• solid : $\rho_{ds} = 810$ (4)

For thermal conductivity λ_d (W/(m · °C)) ;

• liquid : $\lambda_{dl} = 4.59 \times 10^{-7}T^2 - 1.93 \times 10^{-4}T + 0.173$ (5)

• solid : $\lambda_{ds} = -0.03 \times 10^{-6}T^2 - 7.13 \times 10^{-3}T + 0.375$ (6)

The unit of temperature T is degrees Celsius (°C) and the apparent specific heat includes the melting heat of tetradecane (229.1 kJ/kg) in the temperature range ΔT around its melting point of 5.8°C. The value of ΔT was assumed to be 0.14°C in consideration of the stability of numerical computation.

The numerical calculation for the solidification of the oil droplet was carried out to solve the following one-dimensional thermal conduction equation with spherical coordinates.

$$c\rho\partial T/\partial t = (1/r^2)\partial(r^2\lambda\partial T/\partial r)\partial r. \quad (7)$$

The boundary conditions at the center and outside diameter of the spherical oil droplet in Eq. (7) were expressed as follows.

For $r=0$ at the center,

$$\partial T/\partial r = 0. \quad (8)$$

For $r=r_o$ on the outside diameter,

$$-\lambda\partial T/\partial r = \alpha(T - T_r). \quad (9)$$

The value of the overall heat transfer coefficient α between the intersurface of the solidification front of the oil droplet and the water solution can be given by

$$\alpha = (\lambda_c/d_p)(1/Nu_{io} + (\lambda_c/\lambda_{di})(1/Nu_{ii})). \quad (10)$$

where the external Nusselt number Nu_{io} ⁽⁵⁾ of the outer surface of the oil droplet and internal Nusselt number Nu_{ii} ⁽⁶⁾ of the intersurface between the solidification layer and liquid oil core in the oil droplet shown in Fig. 2 are

$$Nu_{io} = 2 + 0.90Re^{0.5}Pr^{0.5} \quad (11)$$

$$Nu_{ii} = 3.75 \times 10^{-3}(U_p d_p / V_{di})[V_{di}(c_{di}\rho_{di})/\lambda_{di}] / [1 + (\mu_{di}/\mu_c)]. \quad (12)$$

The Prandtl number Pr of the water solution is

$$Pr = V_c/(\lambda_c/(c_c\rho_c)). \quad (13)$$

The value of the heat transfer coefficient α for the solidified oil droplet can be given by

$$\alpha = (\lambda_c/d_p)Nu_{so} = (\lambda_c/d_p)(2 + 0.57Re^{0.5}Pr^{1/3}). \quad (14)$$

where Nu_{so} is the average Nusselt number of the solid sphere proposed in the previous paper⁽⁷⁾, assuming that the natural convection in the oil droplet can be neglected. The entire surface of the ascending oil

Fig. 2 Calculation model to determine solidification characteristics of oil droplet

droplet was assumed to solidify instantaneously in the calculation model, since the experimental results revealed that the entire surface of the oil droplet solidified spreads over the entire surface of the oil droplet within 0.1 second. The number of meshes in the radial direction for the present numerical calculation was selected to be 80 and the time step was fixed at 0.1 second in consideration of the convergence and accuracy of the numerical computation.

2.2 Calculation model of solidification and flow behavior of a single oil droplet

Figure 3 shows a combination of two calculation models for solidification and flow characteristics of a single oil droplet ascending in a water solution layer. The diagrams on the right and left sides of Fig. 3 show the liquid-solid phase-change behavior and flow velocity profile of the ascending oil droplet, respectively.

First, an oil droplet starts to move upward from the nozzle in the water solution layer at the initial velocity U_n and temperature T_n . The oil droplet which ascends with decreasing temperature due to the direct heat exchange between it and the water solution, is decelerated by the flow resistance of the water solution flow, which flows downward at a velocity of U_c ($U_c \leq 0$ m/s). The oil droplet starts to solidify at the position Y_m , as shown on the left diagram in Fig. 3. The ascending velocity of the solidified oil droplet decreases markedly, due to the larger flow drag coefficient brought about by the solid surface of the solidified oil droplet as compared with that of the liquid surface of the oil droplet under the liquid state. Eventually, the solidification of the oil droplet is completed at a height of Y_{CF} , after the freezing front

in the oil droplet reaches the center of the oil droplet.

2.3 Calculation model of oil droplet movement

The governing equation of momentum balance for an ascending or descending single particle in a liquid layer was given by Ref. (8) as

$$\begin{aligned} (\pi d_p^3 \rho_d / 6) dU_p / dt = & [\pi d_p^3 (\rho_c - \rho_d) g / 6] \\ & - \pi d_p^2 C_D \rho_c (U_p - U_c)^2 - (K \pi d_p^3 \rho_c / 6) dU_p / dt \\ & - 1.5 (\pi \rho \mu_c)^{1/2} \int_0^\tau (dU_d r) [1 / (t - \tau)^{1/2}] d\tau, \end{aligned} \quad (15)$$

where U_p and U_c are the absolute velocities of the particle and fluid, respectively, C_D is the flow drag coefficient, K is the assumed mass coefficient, τ is the time between the starting time of the particle movement and the time t defined in Eq. (15). With regard to the right-hand-side terms in Eq. (15), the first term corresponds to the gravitational force, the second corresponds to the flow drag, the third to the force based on the assumed mass of fluid and the fourth to the viscous force based on the Basset term. The assumed mass coefficient was selected to be 0.5 for a sphere under constant acceleration motion⁽⁹⁾.

The flow drag coefficient C_D for a single oil droplet in the present calculation is given by⁽¹⁰⁾

$$\text{for } Re = 1 \sim 80; C_D = 2.28 Re^{-0.193} We^{-0.0326} \quad (16)$$

$$\text{for } Re > 80; C_D = 0.195 Re^{0.344} We^{0.622}, \quad (17)$$

where

$$\text{Reynolds number } Re = (U_p - U_c) d_p / \nu \quad (18)$$

$$\text{Weber number } We = \rho_c (U_p - U_c) d_p / \sigma, \quad (19)$$

C_D for a single solidified oil droplet is given by the following Eqs. (20) and (21) proposed by Refs. (11) and (12):

$$\text{for } Re = 1 \sim 700; C_D = (24 / Re) (1 + 0.15 Re^{0.687}) \quad (20)$$

Fig. 3 Combination of calculation models for solidification characteristics and velocity variation of oil droplet

for $Re=700\sim 1.5\times 10^5$;

$$C_D = \left[\sqrt{\frac{24}{Re}} + 0.34 \left(Re^{0.06} + \frac{1}{1.72 + 0.018 Re} \right) \right]^2, \quad (21)$$

Regarding the present calculation procedure, first the flow velocity U_n of the ascending oil droplet was calculated using the flow drag coefficient C_D in Eqs. (16) and (17) with the initial flow velocity of U_n at time $t=0$ s, while the flow velocity of the solidified oil droplet at a height of Y_m , where the oil droplet started to solidify, was calculated using the values of C_D in Eqs. (20) and (21) instead of Eqs. (16) and (17), respectively.

As for numerical computation of the momentum equation (15) using the differencing scheme, the calculation model indicated in Fig. 4 was used for determining the flow velocities of a single oil droplet and a solidified oil droplet. The ascending velocity of the oil droplet injected from the nozzle into the water solution layer was assumed to be constant after taking a sufficient amount of time for the oil droplet to reach terminal velocity. The time taken to the terminal velocity of the oil droplet was divided by $(N-1)$, as shown in Fig. 4, and the oil droplet velocity U_p^n at each time step n was determined by repeating numerical computations. Boundary conditions and initial conditions are as follows.

- Initial velocity

For oil droplet calculation, the initial velocity was defined by the injection velocity of the oil droplet from the nozzle.

For a solidified oil droplet, the initial velocity was defined by the oil droplet velocity when the oil droplet started to solidify.

- Velocity condition at time step $n=N$ ($t=\infty$), $dU_p/dt=0$.

The number of grid meshes was $N=400$ and time step was $\Delta t=0.05$ s, which were determined by taking into account associated computational cost and accuracy. The numerical computations were repeated until the error of two iterative values of flow velocity U_p^n was within 10^{-6} m/s. The diameter of oil droplets for numerical calculation was determined by taking into consideration of the experimental results.

3. Computational Results and Discussion

3.1 Numerical calculation results of a single oil droplet under the solidification process

Figure 5 shows the calculated temperature variation of an oil droplet (5.4 mm in diameter), which remained stationary a water solution layer, in the radial direction with time at an initial temperature of 20°C and when its surface temperature was maintained at 0°C . It is seen that the oil droplet

temperature decreases with time and the solidification front appears at the position where its temperature drops suddenly from 5.8°C . Here, the solidification fraction, R , of the oil droplet under the freezing process was defined as the mass ratio of the amount of solidified oil droplet to total mass of the oil droplet,

$$R = \frac{(4\rho_{ds}\pi/3)(r_o^3 - r_i^3)/[(4\rho_{dl}\pi r_i^3/3) + (4\pi\rho_{ds}/3)(r_o^3 - r_i^3)]}{\rho_{ds}(r_o^3 - r_i^3)/[\rho_{dl}r_i^3 + \rho_{ds}(r_o^3 - r_i^3)]}, \quad (22)$$

where the volumetric change of the oil droplet during the solidification process was neglected and r_i corresponded to the distance from the center of the oil droplet to the solidification front.

3.2 Numerical calculation results of the movement of flowing oil droplet

Numerically calculated and experimental ascending velocities U_p of the oil droplet and the solidified oil droplet in the vertical direction (Y -direction) are shown in Figs. 6(a)–(c) at a superficial water solution velocity of $U_c = -0.007$ m/s. The position $Y=0$ m corresponds to the exit of the nozzle,

Fig. 4 Calculation model to determine ascending velocity of oil droplet

Fig. 5 Example of calculation of temperature distribution in an oil droplet

Fig. 6 Ascending velocity of oil droplet determined by calculation and experiments

$Y=1.6$ m corresponds to the upper surface of the water solution layer, and U_p is the injection velocity of the oil droplet from the nozzle. In Fig. 6, the solid lines indicate the calculated velocity profile of the oil droplet, dotted lines indicate the calculated velocity profile of the solidified oil droplet, and open circles indicate the measured velocities for both the oil droplet and the solidified oil droplet. The position of the oil droplet in the Y direction where it started to solidify in the present calculation was taken from the experimental results, and the initial velocity of the solidified oil droplet in the calculation was assumed to be the velocity of the ascending oil droplet at the same position as where it started to solidify. It is seen in Fig. 6 that the calculated results for the oil droplet and the solidified droplet are in good agreement with experimental ones, except for the data at around the beginning of solidification. At the beginning of solidification, the measured oil droplet velocity decelerated gradually with the height of the water layer since the velocity data were obtained by averaging the velocities of some oil droplets of different diameters. In contrast, the calculated velocity dropped suddenly because the calculation model for the oil droplet was shifted to that for the solidified oil droplet at the position where the oil droplet solidified. However, the calculation results agree well, on the whole, with those of experiments.

3.3 Numerical calculation results of solidification characteristics of oil droplet in consideration of oil droplet velocity

3.3.1 A comparison between numerical and experimental results Figure 7 shows the variation of flow velocity U_p (fine solid lines in Fig. 7) and the solidified fraction R (solid-lines in Fig. 7) of the oil droplet with the position Y of the ascending oil droplet in the vertical direction for various initial oil

Fig. 7 Variations of ascending velocity U_p and solidification rate R of oil droplets with vertical location Y

droplet temperatures T_n from 20°C to 60°C. In Fig. 7, it is found that the velocity of every oil droplet decreases at a constant rate after the oil droplet is injected at an initial injection velocity of $U_n=0.307$ m/s, and then the velocity of each oil droplet with different initial temperatures drops suddenly in the order of the initial temperature of the oil droplet in the range of $Y=0.2-0.9$ m because the oil droplet starts to solidify. Finally, the velocity of each solidified oil droplet reaches a constant terminal velocity. It is noted that the solidification fraction increases markedly with an increase in the position Y of the solidified oil droplet at the beginning of the solidified and then the rate of increase of the solidification fraction decreases with progress of the solidification process since the thermal resistance of the oil droplet increases with increasing the thickness of the solid layer formed in the oil droplet. Based on

the results in Fig. 7, it can be said that the numerical results of the solidification fraction are in good agreement with the experimental ones (open circles in Fig. 7) at a position of $Y_o=1.6$ m.

3.3.2 Relationship between time periods as well as ascending distance of the oil droplet until the completion of solidification, and various other parameters

Figure 8 indicates the variation in the completion time t_{CF} of the solidification of the oil droplet with the initial temperature T_n of the oil droplet injected from the nozzle, for various water solution temperatures T_c . From the results in Fig. 8, it is noted that t_{CF} increases with increasing initial temperature of the oil droplet, the relation of which shows the convex curve shown in Fig. 8, and the tendency for T_{CF} to increase with an increase in T_n becomes weaker in the range of low water solution temperature.

The relationship between the position Y_{CF} of the

Fig. 8 Completion time of solidification t_{CF} vs initial droplet temperature T_n

Fig. 9 Ascending height of complete solidification Y_{CF} vs initial droplet temperature T_n

oil droplet in the Y direction at which the oil droplet solidifies completely and the initial temperature T_n of the oil droplet injected from the nozzle is presented in Fig. 9. As shown in Fig. 9, Y_{CF} increases with increasing T_n , the relation of which follows the convex curve, and the rate of increase of Y_{CF} relative to T_n decreases gradually with increasing T_n . Moreover, Y_{CF} decreases with decreasing water solution temperature T_c because the completion time of oil droplet solidification is shortened with increasing heat flow into the oil droplet, which is caused by the large temperature difference between the oil droplet and the water solution, and the ascending velocity of the oil droplet decreases with an increase in the viscosity of the water solution, according to the decrease of temperature with an increase of staying time of the oil droplet in the water solution layer.

Figure 10 shows the relationship between the completion time t_{CF} of solidification of the oil droplet and oil droplet diameter d_p for various water solution temperatures T_c . It is clear that t_{CF} increases exponentially with increasing oil droplet diameter. The variation of the oil droplet position Y_{CF} with the oil droplet diameter d_p is shown in Fig. 11. Y_{CF} also increases exponentially with increasing oil droplet diameter. This increase of Y_{CF} can be explained by the fact that the increase of the oil droplet volume with an increase in its diameter allows an increase of the time period until the solidification front reaches the center of the oil droplet and a decrease of the staying time of the oil droplet in the water solution layer due to an increase in the ascending velocity of the oil droplet.

Figure 12 indicates the variation of the completion time t_{CF} of oil droplet solidification with velocity U_c of the water solution for various water solution temperatures T_c . The minus sign with water solution velocity U_c corresponds to the flow direction of

Fig. 10 Completion time of solidification t_{CF} vs droplet diameter d_p

Fig. 11 Ascending height for complete solidification Y_{CF} vs droplet diameter d_p

Fig. 12 Completion time of solidification t_{CF} vs velocity of water solution U_c

Fig. 13 Ascending height for complete solidification Y_{CF} vs velocity of water solution U_c

gravitational acceleration. The results in Fig. 12 reveal that t_{CF} varies to a lesser extent with increasing water solution velocity. This phenomenon can be

explained by the fact that the heat transfer coefficient of the oil droplet surface increases to a lesser extent with increasing water solution velocity, since the relative velocity between the ascending oil droplet and descending water solution is reduced by decreasing the ascending oil droplet velocity due to an increase in flow drag. The relationship between the position Y_{CF} of the oil droplet when it starts to solidify and the water solution temperature T_c is presented in Fig. 13, in which, it is seen that Y_{CF} decreases considerably with increasing water solution velocity because decrease in the ascending velocity of the oil droplet results in the start of its solidification at a shorter distance from the nozzle. Finally, from the present calculation results, it is concluded that the oil droplet solidifies completely below the oil droplet position of $Y_{CF}=4$ m in the vertical direction.

4. Concluding Remarks

The following are the conclusions reached after conducting a numerical calculation study of the motion and solidification characteristics of an oil droplet ascending in a cold water solution by buoyancy.

(1) The ascending velocities of the oil droplet and the solidified oil droplet could be calculated using the momentum equation related to a single droplet model which dealt with each flow drag as a liquid sphere or solid sphere depending on the phase state.

(2) It was found that the solidified fraction of the oil droplet ascending in the water solution layer could be predicted by combining the velocity solution of a single droplet ascending in a viscous fluid layer with the calculation model for the solidification of a single droplet.

(3) The numerical results revealed that the time period and the position of the oil droplet from the nozzle in the vertical direction, when the oil droplet solidified completely, increased with increasing initial temperature of the oil droplet injected from the nozzle and the diameter of the oil droplet. It was also found that the completion time of solidification of the oil droplet remained almost constant with increasing water solution velocity, while the position of the oil droplet which solidified completely was lowered with increasing water solution velocity.

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