

MELTING CHARACTERISTICS OF CNT-DISPERSED LATENT HEAT STORAGE MATERIAL IN RECTANGULAR TANK WITH VERTICAL HEAT TRANSFER SURFACE

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An important issue in heat storage technology is the increase in heat storage density and heat storage rate. To increase the density, a method that employs latent heat storage and uses latent heat in addition to sensible heat is examined. For the latter problem, it is considered effective to increase the thermal conductivity of the heat storage material. However, the latent heat storage of melting requires that the melt convection be not impaired in addition to the increase in thermal conductivity. Dispersive fluids can convect even if the dispersoid is solid. It is presumed that it is possible to improve the heat exchange performance while maintaining the fluidity by using a material with high thermal conductivity as the dispersoid. This study shows the melting performance evaluation results of the latent heat storage material having a carbon nanotube (CNT) dispersion system with high thermal conductivity, which enhances the thermal conductivity of the latent heat storage material and does not hinder the free convection. A rectangular vessel (width 50 mm, depth 50 mm, height 150 mm) with a heated vertical surface was used for the latent heat storage experiment. The evaluation of melting speed was carried out by comparing the latent heat storage amount between a 0.100mass% CNT-dispersed latent heat storage material and a latent heat storage material single phase sample. The effect of increasing thermal conductivity by CNT dispersion was observed in the first half of the melting latent heat storage process. However, since the increase in viscosity due to CNT dispersion causes convection inhibition, the melting rate in the latter half of the latent heat process of melting is reduced. This report shows that the melting acceleration due to CNT dispersion was in the first half range up to a melting rate of 25% regardless of the heating surface temperature.

Key Words : CNT, free convection, latent heat storage, phase change

1. INTRODUCTION

To speed up the latent heat storage of melting, it is required to increase the thermal conductivity of the latent heat storage material and promote convective heat transfer. It is possible to achieve both improvement of thermal conductivity and promotion of convection by dispersing fine high thermal conductivity materials in the latent heat storage material. Carbon nanotubes (CNT) are very small substances and have very high thermal conductivity (MWCNT: Multi-Wall Carbon Nanotube, 1400 to

3000 W/(m·K))¹⁾. Yulong et al.²⁾ reported that the heat transfer coefficient of 0.5 mass% MWCNT dispersed water was more than 3.5 times that of water at $Re = 800$. Dispersion of CNTs in a latent heat storage material can be expected to promote melting heat transfer. The authors confirmed that the thermal conductivity increased by up to 10%³⁾ with respect to the paraffin single phase by measuring the MWCNT dispersed latent heat storage material of 1.0 mass% or less by the hot wire method. It was feared that the increase in viscosity would lead to convection inhibition because the shear stress of the latent heat

storage material containing 0.5 mass% of MWCNT doubled⁴⁾ at a shear rate of 500 /s. The authors also reported⁵⁾ that the increase in viscosity decreased the melting rate in experiments in a rectangular heat storage tank with a horizontal heating bottom.

This study shows the experimental evaluation results of the melting characteristics of the MWCNT dispersed latent heat storage material in a rectangular (width 50 mm, depth 50 mm, height 150 mm) vessel with vertical heating surface. This paper describes the results of comparative evaluation between the CNT-dispersed latent heat storage material and single-phase sample using the heating surface temperature as a parameter.

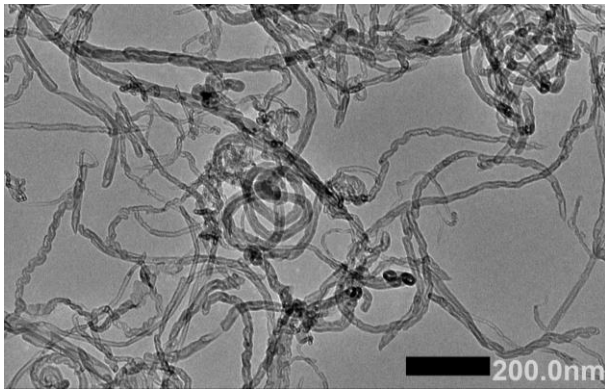


Fig.1 Electron micrograph of MWCNT.

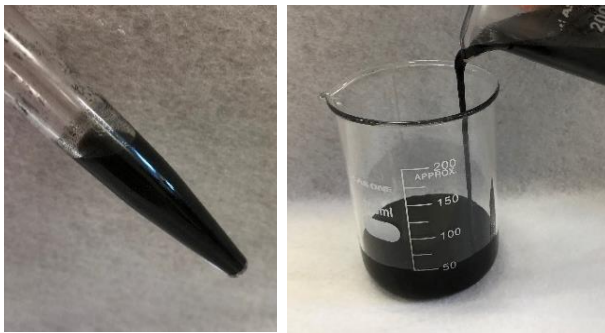


Fig.2 External appearance of MWCNT dispersed Tetracosane at liquid phase: $T >$ Melting point 60.5°C .

Table 1 Mass composition ratio of test samples [mass%].

Dispersoid	Surfactant		Dispersion medium Tetracosane
	Anion	Nonion	
MWCNT			
φ_{CNT}	φ_{sa}	φ_{sn}	φ_{tet}
-	-	-	100
0.100	0.010	0.010	99.880

2. TEST SAMPLES AS LATENT HEAT STORAGE MATERIAL

Fig.1 shows the transmission electron micrograph of the multi-walled carbon nanotube (MWCNT) as dispersoid used in this study. MWCNT has a

structure in which CNTs are stacked in a nested manner. The average size of the test MWCNT is 17 nm in diameter and 5 μm in length. The thermal properties are thermal conductivity $3000 \text{ W}/(\text{m}\cdot\text{K})$, true density $1350 \text{ kg}/\text{m}^3$, and specific heat $0.6691 \text{ kJ}/(\text{kg}\cdot\text{K})$. These values were provided by the manufacturer.

Fig.2 shows the external appearance of multi-walled carbon nanotubes dispersed latent heat storage material as a test sample. The test sample has a black color in both the solidified state and the melted state. The MWCNT dispersed latent heat storage material has fluidity when the continuous phase is melted.

Table 1 shows the mass composition ratio of the test samples. Tetracosane (Tokyo Chemical Industry Co., Ltd., $\text{CH}_3(\text{CH}_2)_{22}\text{CH}_3 = 338.6538 \text{ g/mol}$, purity $> 99\%$, melting point 50.6°C) was used as the dispersion medium. Two types of surfactants were used to disperse MWCNT in the latent heat storage material. The first one is nonionic surfactant (polyoxyethylene alkyl ether NIKKOL BT-7) as a wetting agent, and the second one is anionic surfactant (linear alkylbenzene sulfonate, Kao Co., Ltd. Neoperex G-15) for dispersion in continuous phase. The mass composition ratio of the two types of surfactants is as small as 10 mass% of MWCNT, respectively.

Carbon nanotubes were dispersed using an ultrasonic homogenizer (Sonics VCX-500, 20kHz, 500W) according to the following procedure.

1. Mix the nonionic surfactant with the carbon nanotubes.
2. Set a beaker containing tetracosane in a constant temperature device at 95°C to melt the continuous phase substance.
3. Put the mixture of 1 and the anionic surfactant in melting-tetracosane and add ultrasonic waves (20kHz, $500\text{W}\times 70\%$) for 360 seconds to disperse.

A 50g sample was produced by this process at one time. It was confirmed by observation that MWCNT separation and aggregation in the sample did not occur at 70°C for 50 days or more.

3. EXPERIMENTAL APPARATUS AND METHOD

Fig.3 shows the configuration diagram of the experimental equipment consisting of the test unit, heat transfer surface temperature control system, and temperature data collection system. The heat storage tank as the test section is made of an acrylic plate with 8mm in thickness. The inner dimensions are height of H 150 mm, a width of W 50 mm, and a depth of D 50

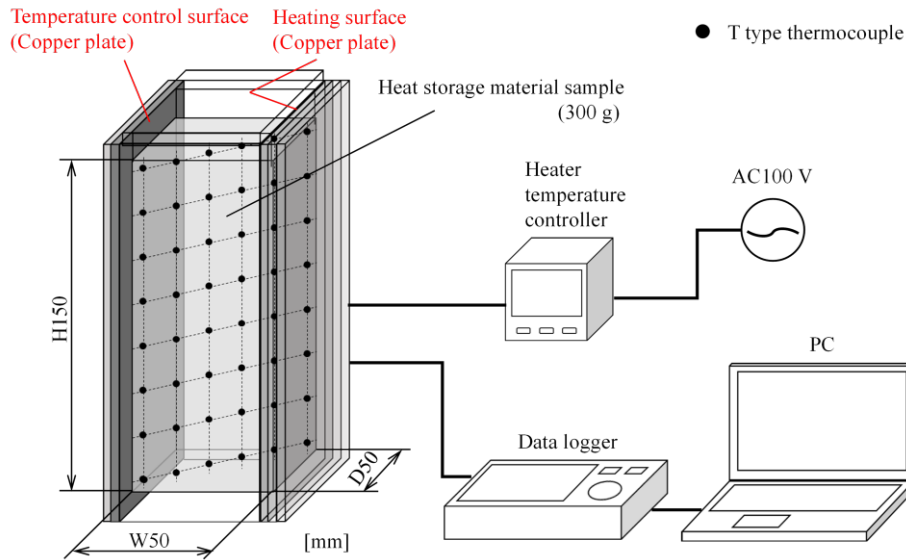


Fig.3 Configuration diagram of the experimental equipment.

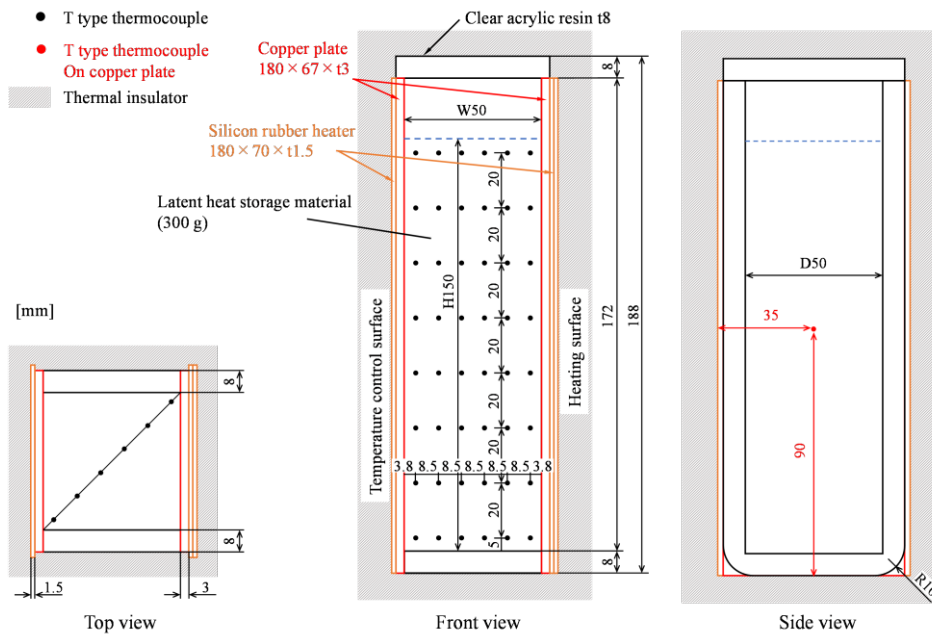


Fig.4 Test section of latent heat storage vessel

mm. It is possible to observe the latent heat storage material sample because the heat storage tank is transparent. The amount of the experimental sample placed in the heat storage tank is 300 g.

Fig.4 shows the dimensions of the test section and the thermocouple arrangement. A copper heat transfer surface with a thickness of 3 mm and an electric silicon rubber heater are placed on the left and right sides of the test section. The temperature of the heat transfer surface can be controlled to the desired temperature by a temperature controller. Right side heat transfer surface is a heating surface that has double 64W electric silicon rubber heaters. The left side heat transfer surface is a heat retaining surface that has a single 64W electric silicon rubber heater. The temperature of the heating surface and the heat retaining surface is controlled by ON / OFF of

heater input. The ON / OFF of heater input is controlled by the temperature controller based on the temperature information from the thermocouple attached to the surface of the heat transfer surface. The heat transfer surface temperature can be controlled within ± 0.5 °C during the stable period of temperature. The heat loss of the heat storage tank is reduced by a 50 mm thick thermal insulation cover. The test using the heat flow sensor showed that the heat loss was 15 W/m² or less in the experimental range of this study, and the heat loss on the side wall surface with respect to the amount of heat input was 1.3% or less. Forty-eight sets of thermocouple networks ($\phi 0.1$ mm, T-type thermocouples) are used to measure the temperature distribution in the heat storage tank.

The sample of tetracosane single phase is white in

the solidified solid phase and becomes transparent when it melts into a liquid phase. Therefore, it is possible to visualize and observe the melting process. On the other hand, MWCNT-dispersed tetracosane is impossible to measure the amount of melting using visualization data, because it is always black opaque. Therefore, the melting interface is estimated from the measured temperature data, and the amount of melting is evaluated.

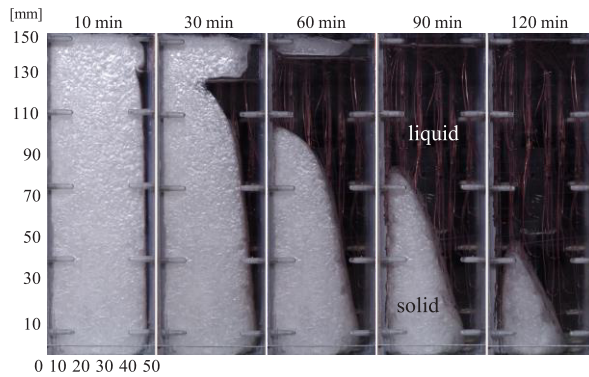


Fig.5 Photo of melting process: tetracosane, 75 °C.

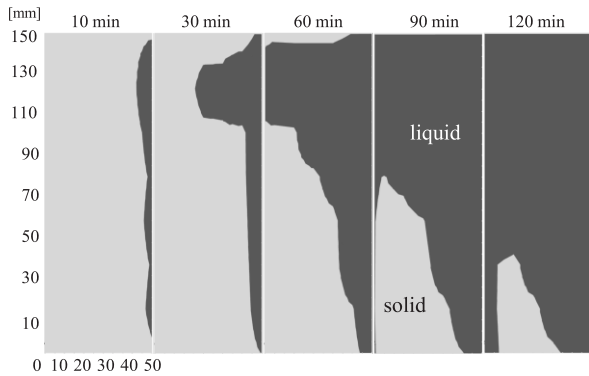


Fig.6 Estimated melting process by MP50.6 °C, 75 °C.

Fig.5 shows a photograph of the melting process of tetracosane visualized and observed with the heating surface temperature set to 75°C. It is observed that the molten liquid phase (black) spreads from the vertical heat transfer surface on the right side, and the solid phase (white) on the upper right side of the tank decreases rapidly due to melting convection.

Fig.6 shows the melting process in the heat storage tank two-dimensionally, with the melting point of Tetracosane at 50.6°C as the melting interface. The liquid phase shown in gray spreads from the upper right side first. It is the same tendency as the result shown in the previous figure.

Fig.7 shows the time variation of the melting amount ratio. The lines in this figure indicates the calculated value from the visualized image, and the dots show the melting amount ratio data estimated

from melting point temperature. The melting amount ratios estimated from the temperature agree with the data calculated by visualization within 4.7% regardless of the heating surface temperature ($T_h = 60$ to 80°C). The method by temperature estimation is judged to fully satisfy the role of calculating the melting ratio of this study

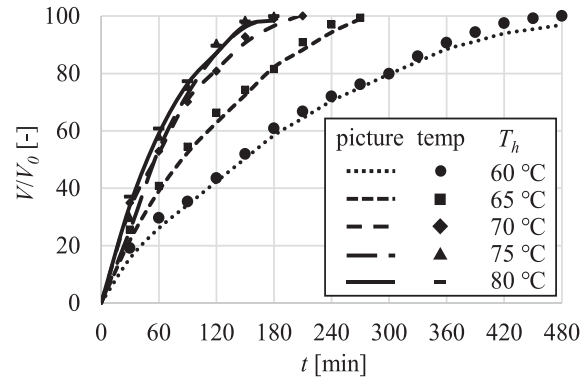


Fig.7 Variation of melting rate with time.

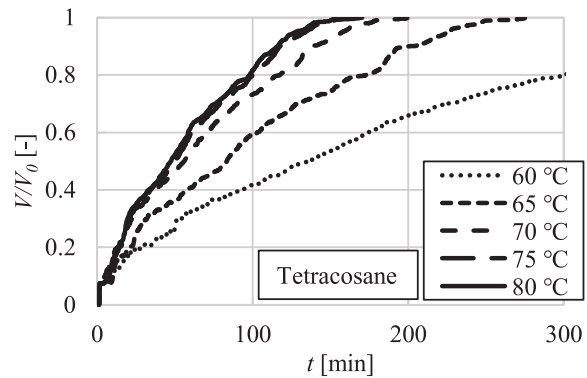


Fig.8 Ave. temp. of latent heat storage material with time.

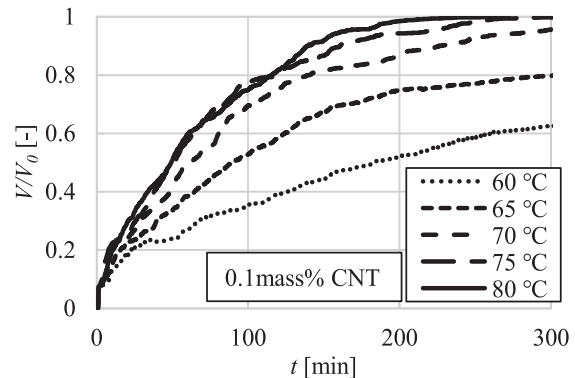


Fig.9 Ave. temp. of latent heat storage material with time.

The temperature of the latent heat storage material sample is kept uniform at the start of the experiment by keeping both heat transfer surfaces temperature at $40^\circ\text{C} \pm 1.0\text{K}$. The experiment is started by raising the heating surface temperature to the temperature of each experiment condition. The amount of melting heat per time Q_{lat} is calculated by differentiating the

amount heat of melting.

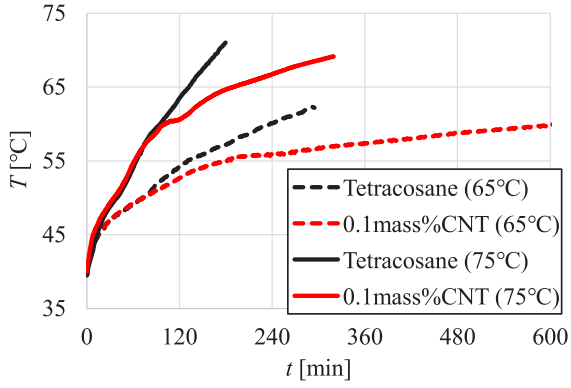


Fig.10 Ave. temp. of latent heat storage material with time.

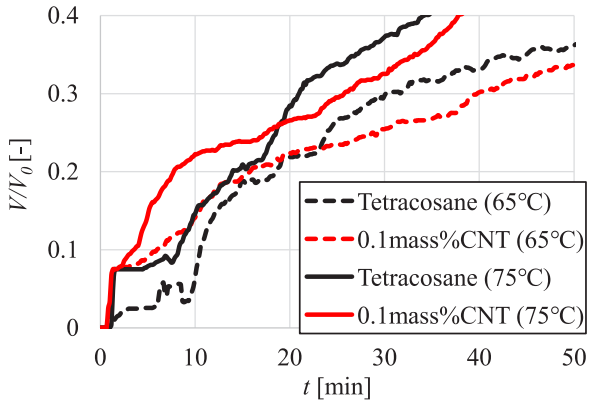


Fig.11 Melting rate with time

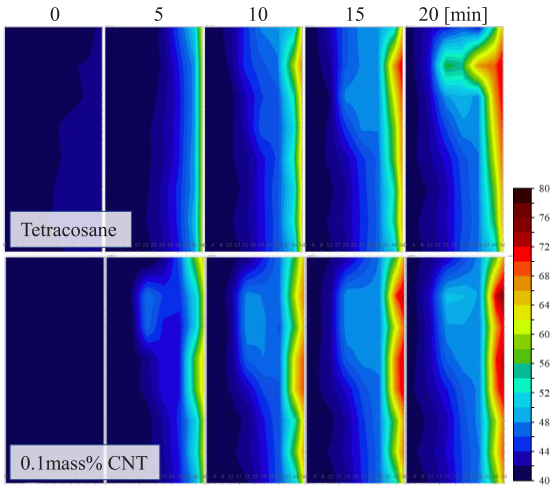


Fig.12 Temperature distribution

4. RESULTS AND DISCUSSION

Fig.8 and 9 indicate the time history of the melting ratios of the Tetracosane single-phase and 0.1 mass% MWCNT-dispersed samples, respectively. The gradient of both melting rate increases with the temperature of the heating surface.

Figure 10 shows the variation over time of the total average temperature in the thermal storage tank at heating surface temperatures $T_h = 65$ and 75°C , respectively. The black line in the figure shows the

data for the Tetracosane single phase and the red line shows the data for the 0.1 mass% MWCNT dispersion sample, where the dotted line is at $T_h = 65^\circ\text{C}$ and the solid line is at $T_h = 75^\circ\text{C}$. It can be observed that the temperature reverses between the Tetracosane single phase and the MWCNT dispersion sample at 51°C ($T_h = 65^\circ\text{C}$) and 57°C ($T_h = 75^\circ\text{C}$).

Figure 11 compares the time evolution of the melting rate in Tetracosane single phase and 0.1 mass% MWCNTs at the initial stage below 40% melting fraction for the heated surface temperatures of $T_h = 65^\circ\text{C}$ and 75°C . The black line in the figure shows the data of Tetracosane single phase and the red line shows the data of 0.1 mass% MWCNT dispersed sample, where the dotted line is $T_h = 65^\circ\text{C}$ and the solid line is $T_h = 75^\circ\text{C}$. The melting rate of the MWCNT-dispersed sample is higher than that of the MWCNT-free sample by less than about 25%.

Figure 12 shows the change in temperature distribution at the heating surface temperature $T_h = 75^\circ\text{C}$ at 5-minute intervals from the start of heating until 20 minutes later. The upper figure is the Tetracosane single phase and the lower figure is the MWCNT dispersion sample of 0.1 mass%. From the figure, the MWCNT dispersion sample has a wider temperature distribution at high temperature, and the heat transfer between 10mm from the heating surface is much better.

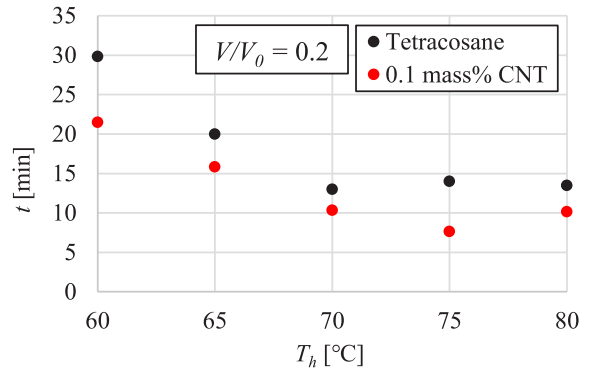


Fig.13 $V/V_0 = 0.2$ reached time with temp of heat transfer

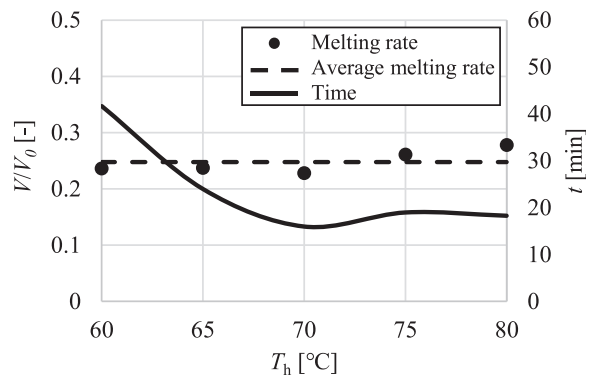


Fig.14 Time to reverse the melting rate with and without MWCNT.

Fig.13 indicates the variation of the reach time to the melting ratio $V/V_0=0.2$ with the heating surface temperature T_h . The time required to reach the melting ratio $V/V_0=0.2$ tends to be shorter for the MWCNT-dispersed sample regardless of the heating surface temperature conditions.

Fig.14 shows the variation of the melting ratio at which reverse occurs and its time with the heating surface temperature. The first vertical axis in the figure indicates the melting rate at the time of reversal, and the second vertical axis is the time.

The broken line in the figure is the average value of the melting rate, and the solid line shows the time. The shortening of the heat storage time by dispersing MWCNT is a melting ratio of 25% regardless of the heating surface temperature condition. There is no change in the required time in the heating surface temperature range of over $70\text{ }^\circ\text{C}$ in the experiments of this study.

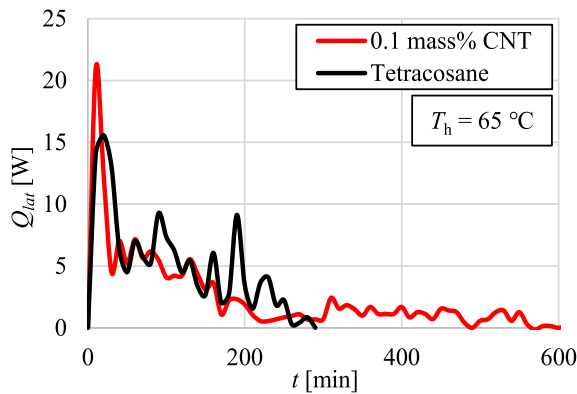


Fig.15 Melting speed with time: $T_h=65\text{ }^\circ\text{C}$.

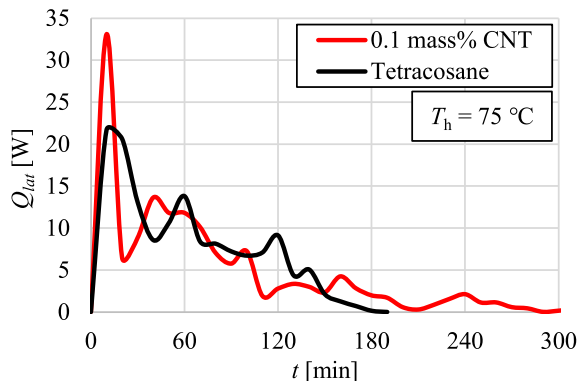


Fig.16 Melting speed with time: $T_h=75\text{ }^\circ\text{C}$.

Fig.15 and 16 indicate the variation of melting rate with time on each the heating surface temperatures $T_h = 65$ and $75\text{ }^\circ\text{C}$, respectively. In the first half of the experiment, the melting rate of the MWCNT dispersed sample shows a large value at any heating surface temperature. The higher thermal conductivity of the MWCNT dispersed sample than without one causes this phenomenon. However, in the latter half of the experiment, it can be observed that the value of the without MWCNT sample takes a larger value.

The effect of melting convection heat transfer increases with increasing of volume ratio of liquid phase in the latent heat storage experiment. The viscosity of the MWCNT dispersed sample is higher than that of the without one. It is presumed that suppression of melting convection due to high viscosity caused a decrease in melting rate.

5. CONCLUSIONS

A melting experiment was conducted using a rectangular tank with a vertical heating surface using a MWCNT dispersed latent heat storage material. The following results were obtained by comparison with the experimental results using the undispersed sample.

1. Dispersion of MWCNT in the latent heat storage material increased the melting rate early in the latent heat storage process. The increasing of melting rate was observed in $V/V_0 \leq 25\%$ range regardless of the heating surface temperature in this experiment.
2. The time required to complete the melting in the entire heat storage tank was significantly increased by the MWCNT dispersion.
3. It was suggested that the improvement of thermal conductivity by MWCNT dispersion brought about an increase in the melting rate in the first half, and that the increase in viscosity by MWCNT dispersion works to suppress convective heat transfer in the second half.

ACKNOWLEDGMENT: This publication was subsidized by JKA through its promotion funds from KEIRIN RACE.

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