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On the heat removal characteristics and the analytical model of a thermal energy storage capsule using gelled Glauber's salt as the PCM

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Abstract

An analytical and experimental investigation was performed on a heat removal process of the thermal energy storage (TES) capsule, using gelled Glauber's salt. Transient heat flux at the capsule wall was measured for various cooling conditions. In cases where the initial temperature was lower than the saturation temperature of the phase change material (PCM), numerical analyses were performed, approximating the process by a relaxation phenomenon where crystals grew around crystal nuclei. The results of the numerical solution agreed very well with the experimental results. © 2001 Elsevier Science Ltd. All rights reserved.

1. Introduction

Usage of the thermal energy storage (TES), utilizing waste energy, natural energy, cheaper midnight electricity and so forth, is very important in considering energy saving to preserve the environment. The latent heat thermal energy storage, having higher storage density, has been remarked upon especially [1]. At the same time, the defect of the latent thermal energy storage has also been pointed out. Since an adequate phase change material (PCM) with its melting point around the desired temperature range is difficult to find, inorganic materials such as sodium sulfate hydrate or calcium chloride hydrate were considered as the PCM recently. The storing phenomenon of the heat reaction accompanied with the crystallization of the solution has been discussed [2,3].

In this paper, the characteristics of a thermal energy storage capsule, using PCM composed of a mixture of a sodium sulfate water solution and a gelled material to prevent the separation of the produced crystal, are discussed analytically and experimentally. Decahydrate of a monoclinic system, called Glauber's salt, appears in the sodium sulfate water solution in a temperature range below 32.4°C. The crystal dissolves in its own crystal water at a temperature range above 32.4°C. This paper treats the chemical process where Glauber's salt separates from the water solution below 32.4°C. In the TES system, using the sodium sulfate water solution as the PCM, heat produced or absorbed by the following chemical reaction between the decahydrate crystal and the water solution is utilized.

$$Na_2SO_4 + 10H_2O \Longleftrightarrow Na_2SO_4 \cdot 10H_2O$$

The PCM used in this paper includes more than 10 water molecules for one sodium sulfate molecule. Therefore, the concentration of the solute (sodium sulfate) and the saturation temperature increase by the chemical reaction process from the right to the left of the above equation. As a result, the heat storage process occurs over a wide temperature range. The heat storage process, where the hydrate crystal dissolves in water solution, progresses through smoothly due to the heat supply. However, as for the heat removal process, where the crystal grows in the water solution, the delay of the reaction usually occurs due to supersaturation [4]. Therefore, it is es-

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Nomenclature

a	thermal diffusivity
C	crystallized fraction of sodium sulfate molecules
c	specific heat
E	heat of reaction stored in PCM
H	rate of heat generation per unit volume
M	volume of PCM in a capsule
Q	heat of reaction stored in a capsule per unit increment of temperature rise
q	heat flux at a capsule wall
R	radius of a capsule wall
r	a coordinate in radial direction (Fig. 9)
T	temperature
t	time
λ	thermal conductivity
ρ	density
α	a constant in Eq. (6) [s^{-1}]

Subscripts

H	saturated situation at temperature T
I	initial value of a heat removal process
M	PCM
P	capsule container
R	accumulated value from 14°C to T°C
RT	total heat of reaction
W	value for circulating water
1	inner surface of a capsule wall
2	outer surface of a capsule wall

sential to clarify the characteristics of the heat transfer in a heat removal process, relating to the crystallization and the crystal growth in a supersaturated solution.

Telkes [5,6] has remarked on the properties of the sodium sulfate decahydrate for a long time, and has achieved a variety of investigations mainly from practical viewpoints. She tried to develop materials promoting a crystal nucleation in a supersaturated situation, and discussed the cost and the safety of the material. Moreover, Telkes [7] also measured a cooling curve, i.e., a time-dependency of the temperature, of sodium sulfate solutions for various concentrations. The apparatus was enclosed in a container and was cooled from the surroundings by air. However, there was discussion neither about a physical model of the crystal nucleation nor a heat transfer phenomenon in the PCM. Biswas [8] showed the fraction of sodium sulfate decahydrate in a solution under various solute concentrations and temperatures. Marks [9] carried out research to prevent a retreat of the material under a repeated usage of heat storage and release. Thus, most of the previous works have been focused on properties of the material, and the researches from a heat transfer viewpoint seem to be insufficient.

Some of the authors [10,11] investigated a heat transfer phenomenon in the gelled sodium sulfate decahydrate as a heat conduction problem with heat generation and absorption, experimentally and analytically. From the results, it was clarified that the phenomena in a heat storage process can be well approximated by a heat conduction model with heat absorption. The local equilibrium of a solute concentration was assumed to be at a saturated concentration corresponding to the temperature of the place. On the other hand, it was also found that a relaxation phenomenon of crystal growth should be considered in an analysis of the heat removal process.

In this paper, the heat removal characteristics of a cylindrical TES capsule, which contains a sodium sulfate decahydrate as the PCM, are measured precisely and its analytical model is proposed.

2. Experiment

2.1. Experimental apparatus

The experimental apparatus is composed of a test section, two constant temperature baths set at different temperatures and pumps supplying water from the constant temperature baths to the test section.

Details of the test section are shown in Fig. 1. In the test section, a heat storage section of a cylindrical shape is fixed concentrically, inside a container of a hollow cylinder made of vinyl chloride, which has a diameter of

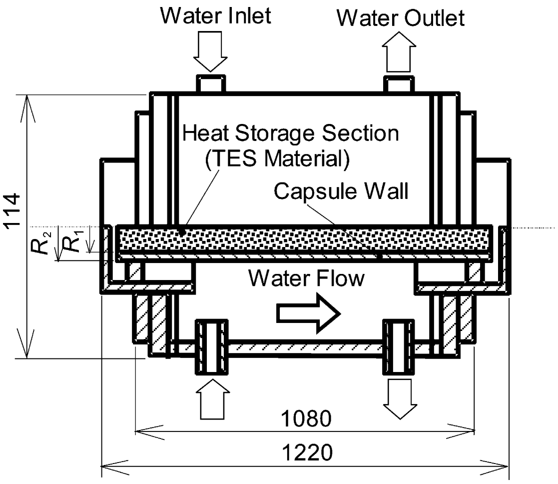

Fig. 1. Details of the test section.

114 mm and a length of 1080 mm. Four inlets and outlets for water are prepared around each end of the hollow cylinder at intervals of 90°. Water at a constant temperature is supplied from the constant temperature bath into the test section, flowing through a gap between the heat storage section and the container along the axial direction. On the inner wall of the container, thermocouples are installed to measure the water temperature at three locations, i.e., at the inlet, the middle section and the outlet.

A structure of the heat storage section is shown in Fig. 2. The above-mentioned gelled sodium sulfate decahydrate is filled in a polypropylene capsule, which has a length of 1220 mm, an inner diameter of 24.3 mm and a thickness of 1 mm. On the outer surface of the capsule, thermocouples are placed at the locations 300, 600 and 900 mm from the water inlet. A foil of nichrome heater of a 0.03 mm thickness to calibrate a heat flux is wrapped around the capsule. Both surfaces of the heater are electrically insulated by a Teflon tape thinner than 0.1 mm. On top of the outer Teflon tape, a heat flux meter is placed. It is composed of three layers of silicone sheets, which have ten sets of series thermocouples in the middle sheet. The heat flux meters are set at places of 400 and 800 mm from the water inlet. The uncertainty of the heat flux meter is 0.06 W for the flow rate of the whole capsule. The heat flux meter responds within an accuracy rate of 0.1% within 5 s.

2.2. Calibration of the heat flux meter

The heat flux meter was calibrated using the foil heater. The measurement was carried out for every 5°C from 15°C to 30°C. The linear relation was obtained and it did not depend on the water temperature. In the following experiments, the heat flux was determined using this relation. The foil heater was used only to calibrate the heat flux meter and never operated in the following experiments.

2.3. Measurement of sensible heat and the heat of reaction

The sodium sulfate in the TES capsule exists as a compound of the decahydrate crystal and the water solution at a temperature range below 32°C. When its temperature rises, a solute concentration at the saturated state increases, since the crystal melts into the solution. During the process, heat absorption occurs. On the other hand, when the temperature drops, an inverse process occurs. However, the solution usually falls into a supersaturated state and the reaction does not progress quickly.

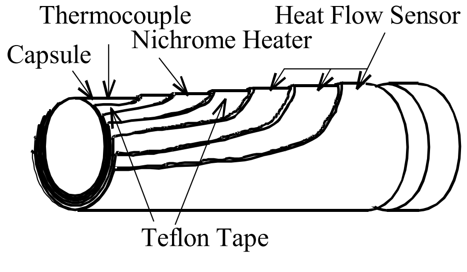

Fig. 2. A structure of the heat storage section.

2.3.1. Measuring procedure

In advance of a measurement of heat removal, sensible heat and the heat of reaction stored in the capsule at each temperature were measured using the apparatus described in Section 2.1. The temperature increment was set to be 2°C. The procedure is shown as follows:

(1) Two constant temperature baths were set at 14°C and 16°C, respectively. Water from the lower temperature bath was circulated through the test section for a long time until the whole capsule reached a steady state.

(2) By operating the selector valve, the test section was immediately connected to the bath, set at 16°C. Then, a time-dependency of heat flux flowing into the capsule was measured using the heat flux meter.

(3) Beside process (2), temperature of the bath, set at 14°C, was reset to 18°C.

(4) The measurement of process (2) was performed for a long time until the indication of the heat flux meter converged to almost 0 and the whole capsule reached a steady state at 16°C. Then, the selector valve was operated, again, to connect the test section to the bath set at 18°C prepared in process (3). Then, a time-dependency of heat flux was measured again.

In the same way, the above processes (3) and (4) were repeated step by step until the temperature reached 38°C. Hence, heat stored in the capsule for every 2°C was measured.

2.3.2. Results of measurement

The results of the measurement are shown in Fig. 3. In the temperature ranges lower than 16°C and higher than 34°C, the values are almost the same, as expressed

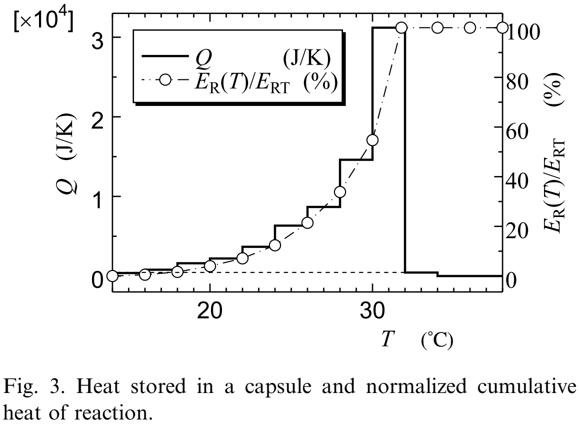

Fig. 3. Heat stored in a capsule and normalized cumulative heat of reaction.

in the figure using a dotted line. At these temperature ranges, heat is stored as sensible heat. Hence, the ordinate of Fig. 3 above the dotted line corresponds to the heat of reaction. The amount of heat stored in the capsule for each temperature increment of 2°C was measured for more than 3 h. The time duration was needed to confirm that the indication of the heat flux meter reached 0. According to the sensitivity of the heat flux meter, uncertainty of the heat flux measurement through the experiments was found to be small. During 3 h of measurement, instantaneous values of the measured heat flux varied at most ±2.5%. This was because the temperature of the circulating water showed a periodic variation of 0.1°C in maximum. However, uncertainty of Fig. 3 never exceeded 1%, since the instantaneous values were averaged for a long time.

A ratio of cumulative heat of reaction $E_{\mathrm{R}}(T)$ for a temperature range from 14°C to T°C against the total heat of reaction E_{RT} stored in the capsule, which corresponds to the area above the dotted line of Fig. 3, is also shown in the same figure. $E_{\mathrm{R}}(T)$ coincides with E_{RT} at about 32°C. It is because the sodium sulfate decahydrate in the PCM dissolves completely in the water solution above the temperature. According to the thermal properties of sodium sulfate, decahydrate should disappear and melt in its own crystal water at the critical temperature of 32.4°C. It is slightly above the measured value. It indicates that a slightly larger amount of water is contained in the PCM, and the solution falls under saturated condition at the temperature above 32°C.

2.4. Measurement of heat removal

2.4.1. Measuring procedure

Heat removal was measured by the following procedure:

(1) Hot water, kept at a constant temperature of T_{F} in the constant temperature bath, is circulated around the capsule for a long time, until the whole capsule has a uniform temperature equal to the water temperature. Under the initial condition, a part or all of the heat of reaction is stored in the capsule.

(2) Water in the constant temperature bath is cooled slowly and continuously, and heat stored in the capsule is delivered to the circulating water, until the water temperature drops to around 14°C. Time variations of the circulating water temperature, the capsule wall temperature and heat flux at the capsule wall are measured.

2.4.2. Results of measurement

Experiments were performed for the case where a cooling rate of the bath was 1.6×10^{-3} K/s. Results are shown in Figs. 4(a) and (b) and in Fig. 5(a) and (b) for initial temperatures T_{I} of 30°C and 34°C, respectively. In Figs. 4(a) and 5(a), a solid line and a dotted line show the water temperature around the capsule and the temperature at the outer surface of the capsule, respectively. Figs. 4(b) and 5(b) show heat fluxes at the capsule wall. Uncertainty of the heat flux measurement was within 1 W/m^2. As water temperatures near the inlet and the outlet of the test section showed almost the same value throughout the experiments, heat transfer phenomena in the capsule were considered to be a one-dimensional problem in radial direction. Experiments were performed under various combinations of the cooling rate and the initial temperature.

2.5. Discussions on typical patterns of heat flux variation in heat removal process

Patterns of heat flux variations in two cases shown in Figs. 4(b) and 5(b) are considerably different. In the case

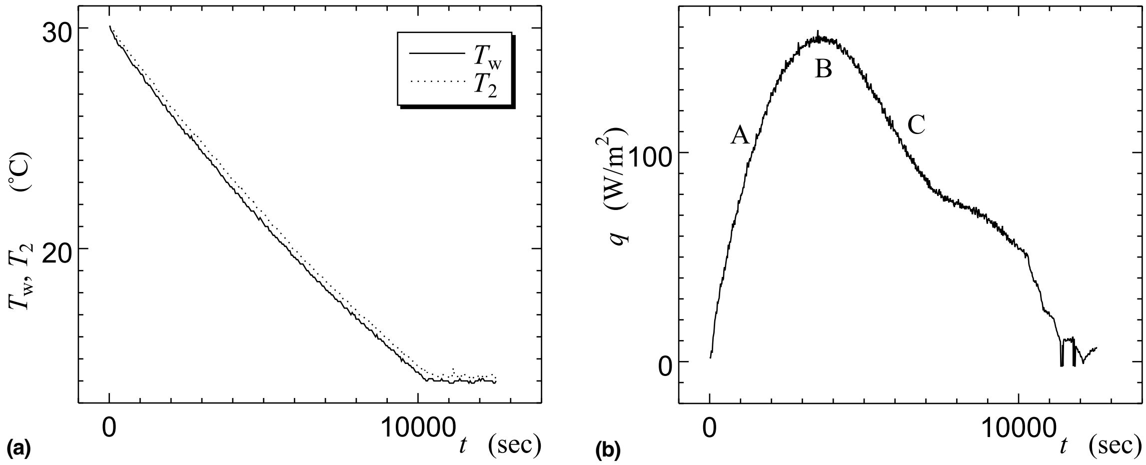

Fig. 4. Heat removal characteristics at $T_{\mathrm{I}} = 30$°C: (a) temperature variations of water and capsule outer wall; (b) heat flux variation at the capsule wall.

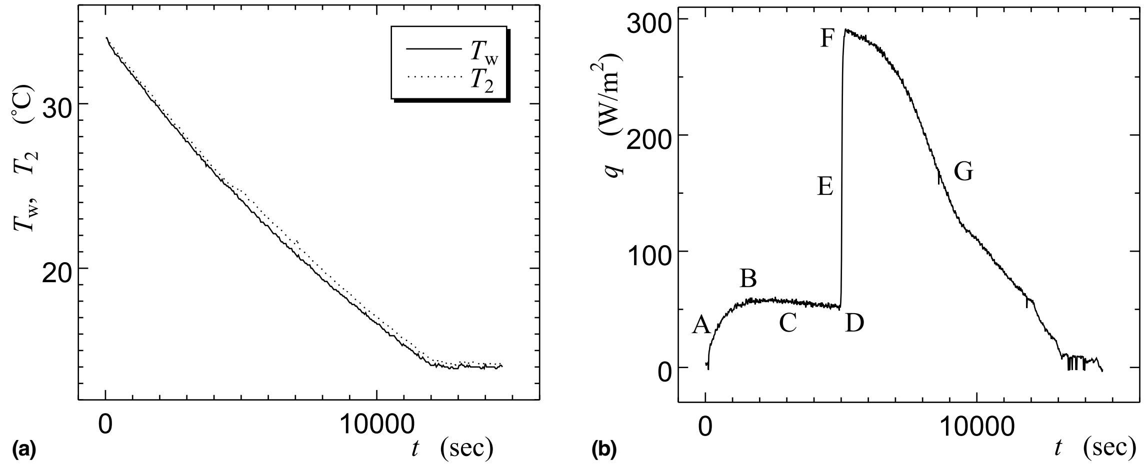

Fig. 5. Heat removal characteristics at $T_{\rm I} = 34$°C: (a) temperature variations of water and capsule outer wall; (b) heat flux variation at the capsule wall.

of $T_{\rm I} = 30$°C as shown in Fig. 4(b), heat flux increases initially (A), reaches to a maximum value (B) and decreases gradually thereafter (C). On the other hand, in the case of $T_{\rm I} = 34$°C as shown in Fig. 5(b), heat flux has a small maximum value (B), decreases slightly (C), then, increases abruptly (E), reaches to a maximum value (F) and finally decreases gradually (G).

The above-mentioned typical difference in the heat flux variation arises from different processes of crystal formations of sodium sulfate water solution to the decahydrate. The difference in process depends on the initial temperature $T_{\rm I}$ exceeding the maximum saturation temperature 32°C or not, as explained in Section 2.3. In the case of Fig. 4(b) where the initial temperature $T_{\rm I}$ is lower than 32°C, a lot of small decahydrate crystals already exist in the PCM at the beginning, and the heat of reaction is delivered mainly along with a crystal growth phenomenon by cooling. In the case of Fig. 5(b) where the initial temperature $T_{\rm I}$ is higher than 32°C, however, no decahydrate crystal exists in the PCM initially. The water solution loses sensible heat by cooling and falls in a supersaturated condition. Then, a lot of crystal nuclei appear in the supersaturated water solution when the amount of the nuclei exceeds a certain limit (point D in Fig. 5(b)). Crystals begin to grow rapidly around the nuclei with a release of the heat of reaction (E–F).

Figs. 4(b) and 5(b) show the heat delivered from the whole capsule. However, the above results should be the accumulation of phenomena in each small volume of element within the PCM. On the basis of the relation shown in Fig. 3, Fig. 6 represents qualitatively a relation between a temperature T of the small element and a fraction of crystal molecules to solution molecules of sodium sulfate C included in the element. On a segment P–Q, no crystal exists, and the PCM is in a state of solution. On a curve Q–R, a saturated liquid and the crystal coexist in an equilibrium state. In the area surrounded by the curve Q–R and coordinate axis, the PCM is in a supersaturated state. The heat removal process occurs in this area and on the line P–Q–R. For the case of Fig. 4, the initial situation is located on a point X in Fig. 6, where a saturated liquid and the crystal coexist in an equilibrium state. As the PCM is cooled down, the situation moves into a supersaturated state along a path X–Y, making the crystal grow in a relaxation process. For the case of Fig. 5, on the other hand, the initial situation is located on point U in Fig. 6, where no crystal exists. Then, the situation moves along a path U–Q–V–W by cooling. In a process moving from Q to V, crystal nuclei appear in the solution. When an amount of the crystal exceeds a certain limit (V), the process shifts to a relaxation process (V–W), where crystals grow rapidly.

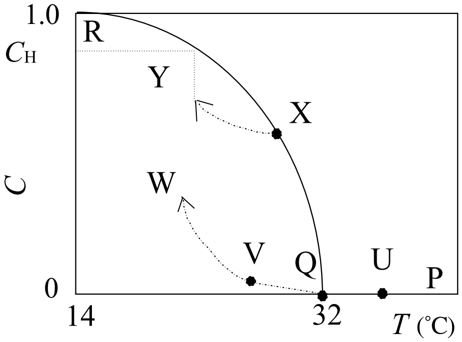

Fig. 6. Paths for heat removal processes.

In this paper, as the first report, an analytical model for the case of Fig. 4(b), where crystals already exist in the PCM in the initial condition, is discussed. The case of Fig. 5(b), which includes the crystal nucleation, will be discussed in the following report.

3. Analysis

3.1. Basic equations

Transient temperature distributions and heat flux variations of the TES capsule (the PCM and the container) for the processes where the PCM was cooled and delivered the heat of the reaction with the existence of crystal nuclei were analyzed.

Heat transfer phenomena in the capsule follow approximately one-dimensional heat conduction in the radial direction with an inner heat generation. Hence, a cylindrical coordinate system was used. Thermal properties were assumed to be constant, not being affected by temperature change. The inner heat generation of the PCM corresponds to the heat of reaction due to the crystal growth. A method to estimate a transient inner heat generation is described later. Basic equations, initial conditions and boundary conditions are shown below.

$$\frac{\partial^2 T_{\rm i}}{\partial r^2}+\frac{1}{r}\frac{\partial T_{\rm i}}{\partial r}+\frac{H_{\rm i}}{\lambda_i}=\frac{1}{a_{\rm i}}\frac{\partial T_{\rm i}}{\partial t} \quad (0\leqslant r\leqslant R_2, t\geqslant 0, {\rm i}={\rm P},{\rm M}), \tag{1}$$

$$T_{\rm i}=T_{\rm I} \quad (0\leqslant r\leqslant R_2, t=0, {\rm i}={\rm P},{\rm M}), \tag{2}$$

$$T_{\rm P}=T_2 \quad (r=R_2, t\geqslant 0), \tag{3}$$

$$T_{\rm P}=T_{\rm M} \quad (r=R_1, t\geqslant 0), \tag{4}$$

$$\lambda_{\rm P}\frac{\partial T_{\rm P}}{\partial r}=\lambda_{\rm M}\frac{\partial T_{\rm M}}{\partial r} \quad (r=R_1, t\geqslant 0), \tag{5}$$

where subscripts $\rm i = P,\ M$ denote the PCM $(0 < r < R_1)$ and the capsule container $(R_1 < r < R_2)$, respectively, and $H_{\rm P} = 0$.

3.2. Rate of heat generation in the process of the crystal growth

The rate of heat generation H in the basic equation (1) depends on the temperature and the cooling process. As mentioned in Section 2.5, the initial temperature $T_{\rm I}$ is lower than 32°C in this report. Therefore, the process treated here is basically a crystal growth where crystal nuclei or small crystals of decahydrate already exist in the sodium sulfate solution at the initial condition and increase their sizes in a supersaturated solution.

Such a process is considered as a relaxation process, and it can be approximated by the following relation.

$$\frac{{\rm d}C}{{\rm d}t}\approx\alpha(C_{\rm H}-C), \tag{6}$$

where $C_{\rm H}$ denotes a fraction of crystallized sodium sulfate molecules in the saturated state at the temperature T (Fig. 6), and satisfies a formula $C_{\rm H} = 1 - E_{\rm R}(T)/E_{\rm RT}$ in relation to Fig. 3. α is a constant and the reciprocal of a relaxation time.

The following relation can be obtained.

$$H=\frac{E_{\rm RT}}{M}\frac{{\rm d}C}{{\rm d}t}. \tag{7}$$

Using Eqs. (6) and (7), the following relation can be derived:

$$H\approx\alpha\left(\frac{E_{\rm RT}}{M}\right)(C_{\rm H}-C). \tag{8}$$

3.3. Numerical analysis

A numerical analysis by the control volume method was performed on the basis of Sections 3.1 and 3.2. As for the PCM $(0 < r < R_1)$, sizes of control volumes in the radial direction were decided unevenly to obtain the same volume for each element. The number of elements was selected to be either 61 or 121 to keep a good rate of accuracy. The container wall $(R_1 < r < R_2)$ was also finely divided into five elements.

In the numerical analysis, the temperature T and the fraction of crystal C were solved simultaneously at each element and time step. A rate of heat generation was calculated using Eq. (8). The constant α in Eq. (8) should be a characteristic value of the PCM used in the experiment. As for the boundary condition at the outer surface of the capsule wall, a measured temperature variation was used as a function of time. The fraction of crystal $C_{\rm H}(= 1 - E_{\rm R}(T)/E_{\rm RT})$ for each temperature was estimated from the measured relation shown in Fig. 3.

As the PCM used in this research was a gelled mixture, thermal properties were unknown and had to be decided in advance. The heat capacity $c\rho$ was determined from the sensible heat in Fig. 3, subtracting the effect of a heat capacity of the container. The thermal conductivity was measured and determined as $\lambda = 0.60$ W/(m K). For thermal properties of the container (polypropylene), reference values [12] were used.

4. Comparison of experimental and analytical results and discussions

4.1. Discussion on constants of the PCM

The constant α, which is the characteristic value of the PCM, was determined.

On the basis of the analytical model mentioned in Section 3, numerical calculations were performed for various values of α under a condition of an initial tem-

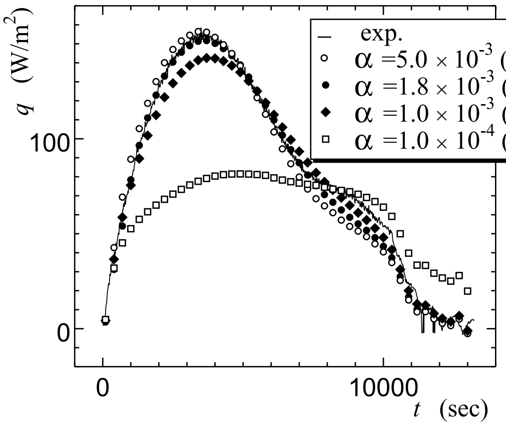

Fig. 7. Dependency of heat removal process on a value of α ($T_\mathrm{I} = 30$°C, cooling rate 1.6×10^{-3} K/s).

perature of 30°C and a cooling rate of 1.6×10^{-3} K/s. Results of the numerical calculations are shown in Fig. 7. The experimental results for the same condition as in Figs. 4(a) and (b) are also shown in the figure for a comparison. The solid line denotes the experimental result. It is found in Fig. 7 that the numerical result for $\alpha = 1.8 \times 10^{-3}$ s^{-1} coincides quite well with the experimental result. Although it is difficult to decide the value for α exactly, the numerical result by changing the constant to $\alpha = 5.0 \times 10^{-3}$ s^{-1} still agrees approximately well. It means that if the value for α is approximately equal to 1.8×10^{-3} s^{-1}, quite reasonable numerical results can be obtained.

Hence, in the following numerical calculations, the value $\alpha = 1.8 \times 10^{-3}$ s^{-1}, which was obtained from a single experimental condition as mentioned above, will be used as the characteristic value of the material.

4.2. Comparison of experimental and analytical results under various cooling rates

Experiments were performed for initial temperatures of 30°C and 26°C, respectively, varying the cooling rate in the range from 1.6×10^{-3} to 5.0×10^{-4} K/s. Variations of heat flux at the capsule wall were measured and compared with the numerical results. Figs. 8(a) and (b) and Figs. 9(a)–(c) show the results for initial temperatures of 30°C and 26°C, respectively. The result for the initial temperature of 30°C and the cooling rate of 1.6×10^{-3} K/s has already been shown in Fig. 7.

It is clear from Figs. 8 and 9 that transient heat removal processes for various initial temperatures and cooling rates can be analyzed accurately by the method proposed in this paper. It means that a constant α depends upon the type of the material but is independent of the temperature and the cooling rate. It is also found from Figs. 8 and 9 that the maximum value for the heat flux increases and the time required to reach the maximum heat flux is shortened by increasing the cooling rate. The reason can be explained as follows. By increasing the cooling rate, the degree of non-equilibrium increases in a shorter time and the right-hand side of Eq. (8) also increases. This causes an increase in the rate of heat generation H in Eq. (8) and the heat stored in the capsule is delivered in a shorter time. The total amount of heat delivered from the capsule is set by the initial temperature and the final temperature, and does not depend on the cooling rate.

The value for a constant α depends on PCM and the gelled material. It is a very important value, but very complicated and difficult to find, especially because the composition of the gelled material is unknown. Hence, obtaining a physical meaning of the value will be necessary in the future.

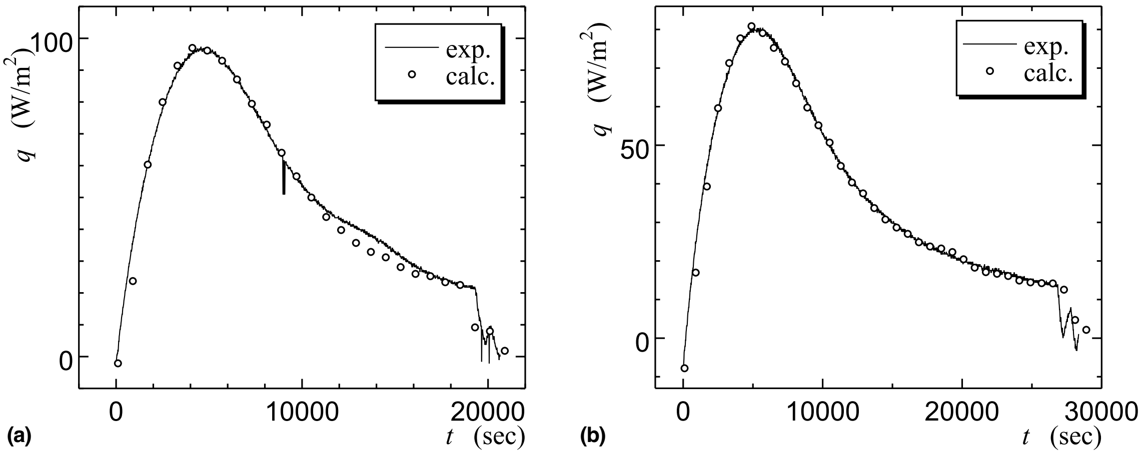

Fig. 8. Comparison of experimental and analytical results for $T_\mathrm{I} = 30$°C: (a) cooling rate 8.0×10^{-4} K/s; (b) cooling rate 5.6×10^{-4} K/s.

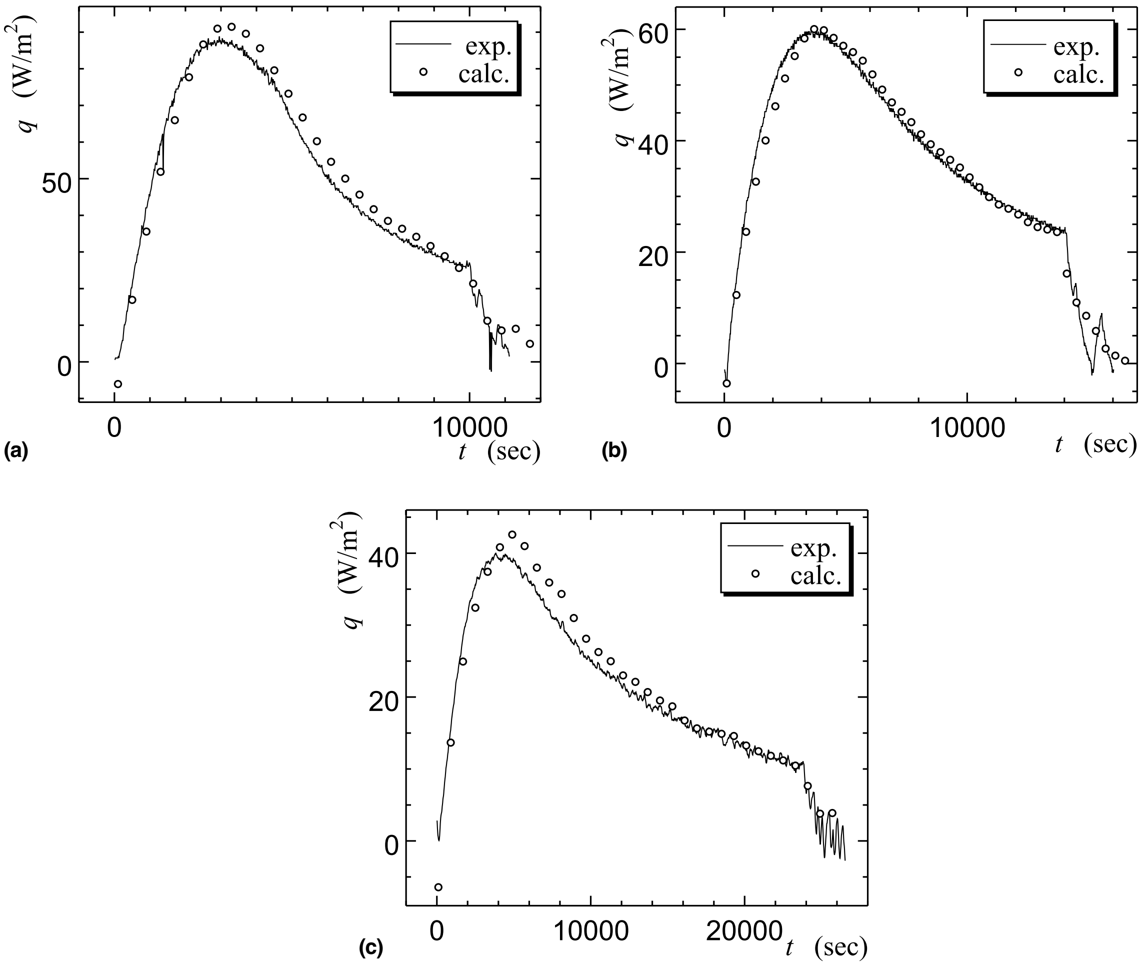

Fig. 9. Comparison of experimental and analytical results for $T_I = 26°C$: (a) cooling rate 1.2×10^{-3} K/s; (b) cooling rate 1.0×10^{-3} K/s; (c) cooling rate 5.0×10^{-4} K/s.

5. Conclusions

The heat generated in a cylindrical TES capsule with the gelled sodium sulfate decahydrate inside as the PCM was considered as a sensible heat and the heat of the reaction arose by the relaxation process of the crystallization. The characteristics were discussed on the basis of experimental and analytical results. The conclusions were made as follows:

(1) An apparatus for accurate measurement of the heat flow through TES capsule was designed, and the amount of heat in the reaction being stored for each increment of temperature was clarified for the temperature range from 14°C to 38°C. It was also shown that the TES used in this experiment was in a water solution state above 32°C.

(2) The mode of a crystallizing process, which occurs by cooling the TES capsule continuously, differs significantly depending on the initial temperatures being higher or lower than 32°C. When the capsule is cooled down from a temperature higher than 32°C to a temperature lower than 32°C, a process of crystal nucleation in a supersaturated solution appears. When the initial temperature is lower than 32°C, the heat removal process is explained by a phenomenon of a crystal growth where crystal nuclei or small crystals already existing in the solution increase in size.

(3) On the basis of the above conclusion (2), an analytical model of a cooling process was proposed for the initial temperature lower than 32°C.

(4) Experiments were performed under two initial temperature conditions, 30°C and 26°C, respectively, and the cooling rates were also varied in the range from 1.6×10^{-3} to 5.0×10^{-4} K/s. The experimental results on heat flux were compared with the numerical calcu-

lations using the analytical model mentioned above. They agreed very well with each other.

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