Heat and Fluid Flow of Two Immiscible Liquid Layers in Vertical Cylindrical Container\textsuperscript{*}

(Application to Device for Sequential Production of Solid Spherical Shells in Liquid-Liquid-Gas Systems)

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Solid spherical shells of mm-order diameter can be applied in lightweight structural materials, buoyant catalytic agents, high-performance solid fuels and artificial organs. A device for sequential production of solid spherical shells using liquid-liquid-gas systems is comprised of a cylindrical container containing two immiscible liquids and a gas injection nozzle at the center of the bottom. By controlling the gas flow and the temperature field of the two liquid layers, liquid spherical shells are formed at the interface between the two immiscible liquids and they solidify during the upward motion. For stable production, it is necessary to correctly estimate the thermo-fluid flow and to control the temperature field in the two liquid layers with high accuracy. To develop a device for sequential production of solid spherical shells, natural convection heat transfer in the two immiscible liquid layers in the cylindrical container was studied. By solving the incompressible Navier Stokes equations and the energy equations for the upper and lower liquids, the flow patterns and thermal field of the two liquid layers were quantitatively investigated. Suitable conditions for the sequential production of solid spherical shells were determined.

Key Words: Multiphase Flow, Numerical Analysis, Computational Fluid Dynamics, Heat Transfer, Natural Convection, Solid Spherical Shells, Encapsulated Liquid Drops, Compound Fluids

1. Introduction

Solid spherical shells, which are produced by solidification of liquid spherical shells, have highly functional applications. They can be used in lightweight structural materials for space craft, high performance solid fuels, buoyant catalytic agents, artificial organs, energy storage systems and other applications. Generally, it is difficult to uniformly and sequentially produce mm-sized or larger spherical shells in a gravitational environment. To date, the sequential production techniques of mm-sized solid spherical shells have mainly been limited to a method utilizing solidification of liquid spherical shells formed through annular nozzles during free fall in the atmosphere\textsuperscript{1}. In this method, however, a very large-scale and high-cost drop tower (more than 10 m in height) is required to solidify molten spherical shells. The authors have developed a new simplified method for sequential production of mm-sized solid spherical shells utilizing a simple device with liquid-liquid gas systems, and succeeded in the production of HTS (Heat Transfer Salt: KNO\textsubscript{3} + NaNO\textsubscript{3} + NaNO\textsubscript{2}, 44 + 49 + 7 mol\%) solid spherical shells\textsuperscript{2}. The device consists of a cylindrical container holding two immiscible liquids and a gas injection nozzle installed at the center of the container bottom. Liquid spherical shells are formed sequentially at the horizontal interface between the two immiscible liquids by effectively controlling the gas jet flow rate and the properties of the liquids\textsuperscript{3}. The liquid spherical shells solidify while
they move upward to the surface of the upper liquid, which has a suitable temperature distribution, due to the buoyancy force. Compared with the previous method using annular nozzles, our method has the advantages that the device size is minimized (180 mm in height) and that high cooling efficiency can be expected due to the liquid-cooling system. However, some problems must be solved in order to develop a higher-performance device. The production frequency of this method is much smaller than that of the method using annular nozzles. Moreover, to stably produce solid spherical shells of valuable materials for industrial applications, the thermal and flow fields of liquids in the device must be quantitatively predicted and precisely controlled.

In this numerical analysis, the gas jet flow, the formation of liquid spherical shells and the buoyancy-driven motion are disregarded. Therefore, the thermal and flow fields of liquids in the liquid–liquid gas systems can be treated as the natural convection heat transfer of two immiscible liquid layers in a vertical cylindrical container with a horizontal liquid liquid interface and a free surface. Although there have been some experimental and numerical studies on such natural convection heat transfer problems, systematic and practical data concerning suitable temperature fields for solid spherical shell production seem to be lacking.

In the present paper, the thermal and flow fields of two liquid layers in a container are studied in detail in order to stably produce solid spherical shells using liquid–liquid gas systems. That is, the natural convection heat transfer of two liquid layers in a vertical cylindrical container, which is heated at the bottom and cooled at the upper part of the side wall, is studied experimentally and numerically. The effects of the liquid properties, the container geometry, the liquid depth and the cooling condition on the thermal and flow fields are clarified quantitatively. Furthermore, the validity of the numerical data for the temperature field is confirmed by comparison with the experimental data. Useful suggestions are also presented for the sequential production of mm-sized solid spherical shells.

2. Nomenclature

\[ A : \text{aspect ratio (ratio of radius to height of container)} = \frac{Z}{R} \]
\[ C_{sv} : \text{specific heat [J/(kg·K)]} \]
\[ g : \text{gravitational acceleration [m/s²]} \]
\[ Gr : \text{Grashof number} = \frac{R^3 g \rho \alpha (T_b - T_i) \beta}{\mu^2} \]
\[ k : \text{thermal conductivity [W/(m·K)]} \]
\[ L_z : \text{dimensionless z-direction length of cool section} \]
\[ N_i : \text{Prandtl number} = \frac{C_{p_i} \mu_i}{k_i} \]
\[ r, z : \text{dimensionless axisymmetric cylindrical coordinates} \]
\[ R : \text{radius of cylindrical container [m]} \]
\[ Re_s : \text{Reynolds number} = \frac{\rho_i R U}{\mu_i} \]
\[ t : \text{dimensionless time defined by Eq. (5)} \]
\[ T_i : \text{dimensionless temperature defined by Eq. (5)} \]
\[ T_b : \text{temperature at cool section [K]} \]
\[ T_i : \text{temperature at heated section [K]} \]
\[ T_i : \text{dimensionless temperature at center of liquid–liquid interface (at } r = 0 \text{ and } z = L_z) \]
\[ U : \text{characteristic velocity} = \sqrt{g \beta (T_b - T_i) R} \text{ [m/s]} \]
\[ Z : \text{height of cylindrical container [m]} \]
\[ \beta_i : \text{coefficient of thermal expansion [1/K]} \]
\[ \gamma : \text{density ratio} = \frac{\rho_1}{\rho_1} \]
\[ \kappa : \text{viscosity ratio} = \frac{\mu_1}{\mu_2} \]
\[ \lambda : \text{thermal conductivity ratio} = k_i/k_b \]
\[ \mu : \text{viscosity [Pa·s]} \]
\[ \rho_i : \text{density [kg/m³]} \]
\[ \psi_i : \text{dimensionless stream function defined by Eq. (5)} \]

Subscripts
\[ i : 1 \text{ (liquid 1) or 2 (liquid 2)} \]
\[ 1 : \text{liquid 1 (lower liquid)} \]
\[ 2 : \text{liquid 2 (upper liquid)} \]

3. Numerical Analysis

3.1 Basic equations

To clarify the heat and fluid flow of two liquid layers in the device, the natural convection heat transfer of two immiscible liquid layers in a vertical cylindrical container, which is heated at the bottom and cooled at the upper part of the side wall, is analyzed numerically. The following assumptions based on preliminary and experimental observations are used in the analysis.

(1) The fluids are Newtonian. The viscous dissipation term in the energy equation is neglected.

(2) All physical properties are constant except for the densities of liquids in the buoyancy term. The Boussinesq assumption is used.

(3) The viscosity and the density of gas are neglected.

(4) The fields of heat and fluid flow are axisymmetric.

(5) The wave on the interface between two immiscible liquids and the wave on the free surface are neglected. The Marangoni effect is also neglected.

(6) The heat flux at the free surface is zero.
Fig. 1 Schematic of theoretical model and coordinate system

A schematic view of the coordinate system and the theoretical model based on the above assumptions is shown in Fig. 1. The Navier-Stokes equations and the energy equations without viscous dissipation for viscous, incompressible and axisymmetric flow in terms of the stream function $\psi$ and vorticity $\omega$ are written in dimensionless form and in axisymmetric cylindrical coordinates $(r, z)$ as follows:

$$
\omega_r = \frac{1}{r} \frac{\partial \psi_z}{\partial r} - \frac{1}{r^2} \frac{\partial \psi_z}{\partial z} - \frac{1}{r} \frac{\partial \psi_r}{\partial r},
$$

$$
\frac{\partial \omega_r}{\partial t} + \frac{1}{r} \frac{\partial \psi_z}{\partial r} \frac{\partial \omega_r}{\partial r} + \frac{1}{r} \frac{\partial \psi_r}{\partial z} \frac{\partial \omega_r}{\partial z} - \omega_r \frac{\partial \psi_z}{\partial r} = \frac{1}{Re} \left( \frac{\partial^2 \omega_r}{\partial r^2} + \frac{1}{r} \frac{\partial \omega_r}{\partial r} + \frac{r^2}{\partial z^2} \right) - \frac{Gr_i}{Re} \frac{\partial T_i}{\partial r},
$$

and

$$
\frac{\partial T_i}{\partial t} + \frac{1}{r} \frac{\partial \psi_z}{\partial r} \frac{\partial T_i}{\partial r} + \frac{1}{r} \frac{\partial \psi_r}{\partial z} \frac{\partial T_i}{\partial z} = \frac{1}{Re_P} \left( \frac{\partial^2 T_i}{\partial r^2} + \frac{1}{r} \frac{\partial T_i}{\partial r} + \frac{r^2}{\partial z^2} \right),
$$

where $t$ indicates the dimensionless time, $T_i$ the dimensionless temperature, $Re$ the Reynolds number, $Pr_i$ the Prandtl number and $Gr_i$ the Grashof number, which are defined by Eq. (5). Subscript $i$ is equal to 1 or 2 and indicates liquid 1 (lower liquid) or 2 (upper liquid), respectively. Continuity equations are automatically satisfied by introducing $\psi_i$ which are related to the velocity components $v_i$ as shown in the following equations:

$$
\nu_r = \frac{1}{r} \frac{\partial \psi_z}{\partial z}, \nu_z = -\frac{1}{r} \frac{\partial \psi_r}{\partial r},
$$

where subscripts $r$ and $z$ indicate the radial and axial direction, respectively. All quantities are expressed in the following dimensionless forms:

$$
r = \frac{r}{R}, z = \frac{z}{L_c}, \psi = \frac{\psi}{U R}, \omega = \frac{\omega}{R \omega}, t = \frac{t}{U / R},
$$

$$
T_i = \frac{T_i - T_{ic}}{T_{ic} - T_c}, Re_i = \frac{\rho \mu U}{k_i}, Pr_i = \frac{c_p \mu}{k_i},
$$

and

$$
Gr_i = \frac{R^2 \rho g \beta \rho (T_i - T_c)}{\mu^2},
$$

where a prime denotes the dimensional value, $R$ indicates the container radius, $U$ the characteristic velocity ($U = \sqrt{g \beta(P_c - P_0) R}$), $T_i$ the temperature at the cool section, $T_c$ the temperature at the heated section, $\rho_i$ the density, $\mu_i$ the viscosity, $C_p_i$ the specific heat, $k_i$ the thermal conductivity, $g$ the gravitational acceleration and $\beta_i$ the coefficient of thermal expansion. Here, $Gr_i = Re_i^2, Gr_2 = Re_i^2 \beta_2 / \beta_1$ are introduced.

The boundary conditions are expressed as follows:

(a) at the bottom of the container $(z = 0)$.
No slip:

$$
\nu_r = \nu_z = 0,
$$

Uniform temperature:

$$
T_i = T_c;
$$

(b) at the liquid-liquid interface $(z = L_i)$.
No flow across the interface:

$$
\frac{\partial \psi_i}{\partial r} = 0,
$$

Continuity of tangential velocity:

$$
\frac{\partial \psi_i}{\partial z} = \frac{\partial \psi_i}{\partial z},
$$

Continuity of tangential stress:

$$
\frac{\partial}{\partial z} \left( \frac{1}{r} \frac{\partial \psi_i}{\partial r} \right) = - \frac{\partial}{\partial z} \left( \frac{1}{r} \frac{\partial \psi_i}{\partial r} \right),
$$

Continuity of temperature:

$$
T_i = T_c,
$$

Continuity of heat flow:

$$
\frac{\partial T_i}{\partial z} = \frac{\partial T_i}{\partial z},
$$

(c) at the free surface $(z = A)$.
No flow across the surface:

$$
\frac{\partial \psi_i}{\partial r} = 0,
$$

Zero tangential stress:

$$
\frac{\partial}{\partial z} \left( \frac{1}{r} \frac{\partial \psi_i}{\partial r} \right) = 0,
$$

Zero heat flux:

$$
\frac{\partial T_i}{\partial z} = 0;
$$

(d) at the side wall of the container $(r = 1)$.
No slip:

$$
\nu_r = \nu_z = 0,
$$

State of heat conduction $(r = 1, 0 \leq z \leq A - L_c)$:

$$
T_i = \frac{z}{A - L_c} + 1;
$$

Unifom temperature:

$$
T_i = 0;
$$

(e) at the cool section $(r = 1, A - L_c \leq z \leq A)$.
Axis of symmetry:

$$
T_i = 0;
$$

(f) along the central axis $(r = 0)$.
Axis of symmetry:
\[ \phi_t = \omega_t = \frac{\partial T_i}{\partial y} = 0, \quad (19) \]

where \( L_i \) indicates the dimensionless depth of liquid 1, \( \kappa = \mu_1/\mu_2 \) the viscosity ratio, \( \lambda = k_i/k_2 \) the thermal conductivity ratio, \( L_z \) the dimensionless \( z \)-direction length of the cool section around the upper part of the container side wall and \( A \) the aspect ratio, which is the ratio of \( R \) to the height \( Z \) of the cylindrical container. \( L_i, L_z \) and \( A \) are made dimensionless using \( R \).

The boundary conditions in Eqs. (7), (17) and (18) are introduced based on the experimental results shown in Section 4. By numerically solving the above boundary-value problem, the fields of heat and fluid flow of two immiscible liquid layers can be obtained. In this paper, \( G_R, P_r, \) the density ratio \( \gamma = \rho_1/\rho_2 \), \( \kappa, \lambda, A, L_i \) and \( L_z \) are the fundamental dimensionless parameters.

3.2 Numerical method of solution

The following method of numerical solution was used. Equations (1) to (3) were rewritten as finite difference equations using the centered spatial differences of second-order accuracy and backward time differences of second-order accuracy. The finite difference equations were solved using the implicit and SOR (successive over-relaxation) method. In the calculation mesh system, the radial step size was 0.025 and the axial step size was 0.025. The time step was 0.1. The SOR procedure was repeated until \( \phi_t, \omega_t \) and \( T_i \) changed by less than a specified tolerance per iteration. The tolerance was chosen to be \( 1 \times 10^{-4} \) for \( \phi_t, \omega_t \) and \( T_i \). When the values of \( \phi_t, \omega_t \) and \( T_i \) at a certain time were obtained, their values after \( DT \) were calculated by the same procedure. The initial conditions of computations were set as \( \phi_t = 0, \omega_t = 0, T_i = 1 \) and \( T_0 = 0 \) except for the values at the boundaries.

4. Results and Discussion

Generally, in the case of a high Rayleigh number \( Ra(=G_R \cdot P_r; \leq 10^6) \) for the natural convection heat transfer in the previous device \(^{19} \), a three dimensional analysis or an analysis based on a turbulent model must be used. However, considering that the ultimate goal of this study was to minimize the device size, and that we could qualitatively predict the heat and fluid flow in the device even from the analysis at relatively low \( Ra \), the numerical analysis and the experiments were performed only for a small device.

In the present experiment, the cylindrical container was made of brass with \( R \) of 15 mm, \( Z \) of 54 mm and thickness 2 mm. The coolant pipe, which was made of copper, was 8 mm in outer diameter and 5 mm in inner diameter, and was set around the upper part of the cylindrical container side wall. An electric heater was set beneath the cylindrical container bottom. The cylindrical container was coated with both an insulating material and a lagging material. The time-averaged mean temperature of the two liquid layers in the device was measured after a time long enough to ignore the effect of the initial state. Three thermocouples 1.0 mm in diameter set at intervals of 0.5 in the \( r \)-direction were used in temperature measurements by shifting them in the \( r \) and \( z \) directions. The lower liquid was 66 wt% glycerin-water or distilled water, and the upper liquid was silicone oil or kerosene. The atmospheric temperature was 280 to 283 K. The coolant was water, which was kept at 289 K in a constant-temperature bath and was circulated by a centrifugal pump. The physical properties of the liquids used here are shown in Table 1. The physical properties are the measured, reported or estimated values\(^{19,20} \) at 298 K, which corresponded to \( T_i = 0.5 \) because the temperature at the heated section was 305 K and the temperature at the cool section was 292 K (see Section 4.1). The \( \mu_i \) and \( \rho_i \) in Table 1 are the measured values. The experiments were performed under the following five conditions and the results were compared with the numerical results.

<table>
<thead>
<tr>
<th>Property</th>
<th>Liquid</th>
<th>Water (at 298 K)</th>
<th>Silicone Oil (at 298 K)</th>
<th>Water (at 298 K)</th>
<th>Kerosene (at 298 K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Specific heat</td>
<td>( c_p ) (J/kg K)</td>
<td>3.68 \times 10^3</td>
<td>1.70 \times 10^3</td>
<td>1.81 \times 10^3</td>
<td>2.03 \times 10^3</td>
</tr>
<tr>
<td>Thermal conductivity</td>
<td>( k_i ) (W/m K)</td>
<td>0.386</td>
<td>0.130</td>
<td>0.667</td>
<td>0.115</td>
</tr>
<tr>
<td>Prandtl number</td>
<td>Pr</td>
<td>117</td>
<td>127</td>
<td>6.29</td>
<td>23.7</td>
</tr>
<tr>
<td>Coefficient of thermal expansion</td>
<td>( \beta_1 ) (1/K)</td>
<td>4.27 \times 10^{-4}</td>
<td>1.08 \times 10^{-3}</td>
<td>2.73 \times 10^{-4}</td>
<td>5.50 \times 10^{-4}</td>
</tr>
<tr>
<td>Viscosity</td>
<td>( \mu_i ) (Pa s)</td>
<td>1.26 \times 10^{-2}</td>
<td>9.35 \times 10^{-3}</td>
<td>9.00 \times 10^{-4}</td>
<td>1.34 \times 10^{-3}</td>
</tr>
<tr>
<td>Density</td>
<td>( \rho_i ) (kg/m^3)</td>
<td>1170</td>
<td>935</td>
<td>997</td>
<td>815</td>
</tr>
</tbody>
</table>
water and the upper liquid was kerosene ($Gr_1 = 1.49 \times 10^9$, $Gr_2 = 9.02 \times 10^4$, $Pr_1 = 6.20$, $Pr_2 = 23.7$, $\gamma = 1.22$, $\kappa = 0.672$, $\lambda = 3.28$, $A = 3.6$, $L_1 = 0.5$, $L_2 = 0.65$).

Because the numerical and experimental results revealed that the thermal and flow fields of two immiscible liquid layers in Cases 1～V were unstable at the initial stage and fluctuated periodically at the later stage, the results were expressed as time-averaged values.

In the condition that the bottom of the cylindrical container was at a uniform high temperature, the side wall of the container was adiabatic, the free surface was at a uniform low temperature and $Gr_1$, $Gr_2 \leq 0$, it was confirmed that the numerical result of heat flux at the bottom of the container was equal to the analytical result within ±0.05 % deviation. The comparison of thermal and flow fields between numerical and experimental results is shown below.

### 4.1 Results in Case I

Figures 2 (a) and (b) show the time-averaged temperature distributions along the container side wall and the container bottom in Case I, respectively. It is found that the temperature along the container side wall varies almost linearly in the z-direction, and that the temperatures at the container bottom and the cool section along the upper part of the container are almost constant values. These results correspond to the fact that the heat transfer at the container side wall and the bottom is mainly governed by heat conduction. The reason for this seems to be that the materials of the cylindrical container and the coolant pipe have high thermal conductivity. It is also recognized even in Cases II～V that the heat transfer at the container side wall and the bottom is governed by heat conduction, and that the temperatures at the heated section and cool section are almost constant values. Therefore, as the boundary conditions in the numerical analysis, the temperature distribution along the container side wall is assumed as the heat conduction state, and the temperatures at the container bottom and the cool section are assumed as the constant temperatures. Figure 3 shows the comparison of time averaged temperature distribution in the z-direction at $r=0$ and $r=0.5$ between the numerical and the experimental results. Because the liquid spherical shells are accelerated and move upward along the central axis of the cylindrical container, a large temperature difference between liquid 1 and liquid 2 along the central axis is required for rapid solidification which increases the shell hardness in the earlier stage and prevents the molten shells from breakdown. That is, rapid solidification realizes a small-sized device and the stable production of solid spherical shells. Because the temperature in liquid 2 along the central axis was almost constant except for small regions near the liquid-liquid interface and near the free surface in the previous experiment, the value $\Delta T$ is defined as the temperature difference along the central axis between the temperature at the liquid-liquid interface and the temperature at the middle point of liquid 2 ($z=L_1+|A-L_1|/2$). These two points roughly correspond to the liquid shell production point and the solidification point, respec-

![Fig. 2 Experimental results of time averaged temperature distribution of cylindrical container side wall and bottom](image-url)

![Fig. 3 Comparison of time averaged temperature distribution in z-direction at r=0 and r=0.5 between numerical and experimental results in Case I (Experimental uncertainty in $T_i = \pm 22.6\%$ and in $z = \pm 5.0\%$ at 20:1 odds)](image-url)
tively. From Fig. 3, it is found that the numerical results agree quantitatively with the experimental results, and that $\Delta T$ is about 0.293. The dash dotted line in Fig. 3 indicates the numerical result of the $z$-direction temperature distribution at $r=0$ under the adiabatic condition at the container side wall. This condition means the boundary condition $\partial T_s/\partial r = 0$, not the boundary condition in Eq. (17) at $r=1$ and $0 \leq z \leq A - L$. In this case, the temperature throughout liquid 2 except in the small region near the liquid-liquid interface becomes high and $\Delta T$, which is almost 0.169, is smaller than that obtained using the boundary condition in Eq. (17). This result shows that the adiabatic condition at the container side wall is not suitable for the production of solid spherical shells. Similar tendencies were confirmed even in Cases II to V. Figure 4 shows the numerical results of the time dependent temperature $T_s$ at the liquid-liquid interface and at $r=0$. Large fluctuation of $T_s$ is not desirable for producing solid spherical shells because the change of physical properties of liquids depending on the temperature at the liquid-liquid interface and at $r=0$ may cause unstable formation of molten spherical shells and may result in solidification of the liquid-liquid interface. Therefore, it is important to check the relations of various parameters to the value $T_s$. From Fig. 4, it is found that the change of $T_s$ is periodic (the period is $t \approx 80$) and is relatively large. Figures 5 (a) and (b) show the numerical results of the time dependent isothermal lines and stream lines in two liquid layers. In Fig. 5, the left end dash dotted line indicates the central axis of the cylindrical container and the right end indicates the container side wall. In this section, the values of isothermal lines correspond to $T_s=0.1n$, where $n=1, 2, \cdots, 10$, and the values of stream lines correspond to $\psi_s=0, \pm 0.0005, \pm 0.001, \pm 0.003, \pm 0.005$. From Fig. 5, it is found that there are two vortices in liquid 2 and two or three vortices in liquid 1, and that the vortices alternate repeat the contraction and the expansion motions. The time periods of these motions in liquids 1 and 2 are different. The period of liquid 1 is $t \approx 80$ and that of liquid 2 is $t \approx 40$. The period of liquid 1 is about twice as long as that of liquid 2, and is almost the same as that of $T_s$. This result seems to suggest that the thermal and flow fields depend on the motion of liquid 1 rather than that of liquid 2, and that resonance vibration may occur in the thermo fluid flow in the two liquid layers system.

4.2 Results in Cases II to V

Figure 6 shows the comparison of time averaged temperature distribution in the $z$ direction at $r=0$.
and $r=0.5$ between the numerical and the experimental results in Case II, i.e., the case of different $L_1$ from Case I. From Fig. 6, it is found that the numerical results agree quantitatively with the experimental results, and that the value $\Delta T$ is relatively large (it is about 0.440). Figure 7 shows the numerical results of the time-dependent $T_i$ at the liquid-liquid interface and at $r=0$. The change of $T_i$ is periodic and similar to that in Case I, but the amplitude of $T_i$ is smaller than that in Case I. The period is $t=24$. Figure 8 shows the numerical results of the time-dependent streamlines in two liquid layers. From Fig. 8, it is found that there are two vortices in liquid 2 and one vortex in liquid 1. It is also found that the vortex motion in liquid 1 and liquid 2 alternately repeats contraction and expansion, and that the two vortices in liquid 2 are induced by the upward flow near the side wall from the liquid-liquid interface and downward flow near the side wall from the cool section, respectively. These two periods of liquid 1 and liquid 2 are almost the same, about 24. Consequently, small $L_1$ is suitable for the production of solid spherical shells compared with Case I because it results in the large $\Delta T$ and small change of $T_i$. Furthermore, it was reported that small $L_1$ resulted in high production frequency of liquid spherical shells. Therefore, $L_1=0.5$ is used as a fixed condition in the following numerical and experimental analyses.

Figure 9 shows the comparison of time-averaged temperature distribution in the $z$ direction at $r=0$ and $r=0.5$ between the numerical and the experimental results in Case III, i.e., the case of different $L_2$ from Case II. From Fig. 9, it is found that the numerical results agree quantitatively with the experimental results. Compared with Case II, the temperature throughout liquid 2 except near the liquid-liquid interface becomes high and $\Delta T$ becomes small (=0.408) owing to the small cool section. It is also found that the amplitude of $T_i$ is as small as that in Case II and the schematic of behavior of vortices is similar to that in Case II. The figures for $T_i$ and streamlines are omitted. The time period of $T_i$ is almost equal to that in Case II and is about 24.

Figure 10 shows the comparison of time-averaged temperature distribution in the $z$ direction at $r=0$ and $r=0.5$ between the numerical and the experimental results in Case IV (Experimental uncertainty in $T_i=\pm21.4\%$ and in $\varepsilon=\pm5.0\%$ at 20:1 odds).
results in Case IV, i.e., the case of different $A$ from Case II. From Fig. 10, it is found that the numerical results agree quantitatively with the experimental results. Compared with Case II, the temperature throughout liquid 2 becomes low owing to the small distance between the cool section and the liquid-liquid interface, and $\Delta T$ becomes large ($=0.472$). It is also found that the amplitude of $T_i$ is as small as that in Case II and the schematic behavior of vortices is similar to that in Case II. The figures for $T_i$ and stream lines are omitted. The time period of $T_i$ is smaller than that in Case II and is about 20 owing to the smaller system.

The thermal and flow fields of two liquid layers are considered in the case that liquid 1 is distilled water and liquid 2 is kerosene. Figure 11 shows the comparison of time-averaged temperature distribution in the $z$ direction at $r = 0$ and $r = 0.5$ between the numerical and the experimental results in Case V. From Fig. 11, it is found that the numerical results agree quantitatively with the experimental results, and that a relatively large $\Delta T$ can be obtained, which is similar to that in Case II and is about 0.401. Compared with Cases I ~ IV, where liquid 1 is 66 wt% glycerin water and liquid 2 is silicone oil, the thermal and flow fields in Case V are not periodic and are irregular. Figure 12 shows the numerical results of the time-dependent $T_i$ at the liquid-liquid interface and at $r = 0$. The change of $T_i$ is also irregular and the amplitude of $T_i$ is larger than that in Cases I ~ IV. Figure 13 shows the numerical results of the time-dependent stream lines in two liquid layers. From Fig. 13, it is found that the vortex in liquid 1 just below the liquid-liquid interface moves toward the side wall, then moves toward the central axis of the cylindrical container, and repeats these motions. It is also found that the two or three vortices in liquid 2 induced by upward flow from near the liquid-liquid interface and by downward flow from near the cool section appear. However, the motions of vortices in the two liquid layers are not periodic owing to high $Ra$.

In Cases I ~ V, a small difference between the numerical and experimental results of temperature at $r = 0$ and $r = 0.5$ near the container bottom and near the liquid-liquid interface occurs due to the thin thermal boundary layers and the difficulty of precise temperature measurements. The tendency that the numerical value of temperature at the free surface is larger than the experimental value, is also recognized in Cases I ~ V owing to the boundary condition in Eq. (15). In some cases, it is found that the qualitative tendencies of the liquid 2 temperature at $r = 0$ and $r = 0.5$ do not agree with the experimental results. However, it can be said in general that the numerical results agree well with the experimental results despite rough assumptions and complex phenomena. Therefore, the thermal and flow fields of two liquid layers can be accurately predicted using the numerical code developed here.

5. Desirable Running Condition of Device for Sequential Production of Solid Spherical Shells

In the sequential production of solid spherical shells in liquid-liquid gas systems, the liquids are often selected so that the stable liquid spherical shells

![Fig. 11](image1)
![Fig. 12](image2)
![Fig. 13](image3)
are produced at the liquid liquid interface. Then, systematic numerical analysis is carried out to clarify the effects of the dimensionless parameters $A$, $L_1$ and $L_2$ on the thermal and flow fields in detail which yields useful data for constructing a high-performance device. All the numerical results in this section are time-independent. The thermal and flow fields of liquids can be obtained for a steady state, where the temperature at the side wall of the cylindrical container is given as the heat conduction state based on the experimental results in Section 4. In the numerical analysis, the method used to obtain the solution at a certain time is the same as that described in Section 3.2. To obtain the numerical solution in a steady state, the calculation is repeated until $|\phi^{N+1} - \phi^N|/\Delta t$, $|\omega^{N+1} - \omega^N|/\Delta t$ and $|T^{N+1} - T^N|/\Delta t$ become less than $1 \times 10^{-6}$, where the superscript $N$ indicates the value at the $N$th time step. In this section, the parameters $Gr_1$ = $Gr_2$ = $1.0 \times 10^4$, $Pr_1$ = $Pr_2$ = 1, $\gamma$ = 2, $\kappa$ = 1, $\lambda$ = 5, $A$ = 2, $L_1$ = 1.0 and $L_2$ = 0.5 are set as the referential data for systematic analysis. The thermal and flow fields are analyzed only in the case of small $Gr_i$ in order to realize low-cost computation and obtain basic qualitative data for practical industrial applications.

Figure 14 shows the $x$-direction temperature distribution along the central axis of the cylindrical container for various values of $A$, and Fig. 15 shows the relationship of $\Delta T$ to $A$ for the same calculation conditions as those in Fig. 14. From Figs. 14 and 15, the value of $\Delta T$ has a maximum at $A$ = 3. Therefore, it is suggested that $A$ = 3 is suitable for sequential production of solid spherical shells because it yields large $\Delta T$. Figure 16 shows the stream lines in two liquid layers for various values of $A$ for the same calculation conditions as those in Fig. 14. In this section the values of $\psi$ correspond to $\psi$ = 0, ±0.0005, ±0.005, ±0.01, ±0.05. From Fig. 16, it is found that the effect of $A$ on the flow pattern or the behavior of vortices is not very strong. Figure 17 shows the $z$-direction temperature distribution along the central axis of the cylindrical container for various values of $L_1$, and Fig. 18 shows the relationship of $\Delta T$ to $L_1$ for the same calculation conditions as those in Fig. 17. From Figs. 17 and 18, it is found that $T_z$ and $\Delta T$ become small for $L_1$ within 0.4 $\leq L_1 \leq$ 0.8 and within 0.9 $\leq L_1 \leq$ 1.4, but that $T_z$ and $\Delta T$ become large for $L_1$

Fig. 14 Numerical result of effect of aspect ratio $A$ on temperature distribution along central axis in two liquid layers

Fig. 15 Numerical result of effect of aspect ratio $A$ on temperature difference $\Delta T$ along central axis between two liquid layers for same calculation conditions as in Fig. 14

Fig. 16 Numerical result of effect of aspect ratio $A$ on stream lines in two liquid layers for same calculation conditions as in Fig. 14

Fig. 17 Numerical result of effect of lower liquid 1 depth $L_1$ on temperature distribution along central axis in two liquid layers
within $0.8 \leq L_1 \leq 0.9$. This result corresponds to the large vortex near the central axis in liquid 1 reversing its direction of motion within $0.8 \leq L_1 \leq 0.9$, which is indicated by the stream lines in Fig. 19 obtained for the same calculation conditions as those in Fig. 17. The largest $\Delta T$ is recognized at the smallest $L_1=0.4$ in the analysis, and this result indicates that small $L_1$ is desirable for sequential production of solid spherical shells. This is the same result as that in Section 4.

Figure 20 shows the z-direction temperature distribution along the central axis of the cylindrical container for various values of $L_c$, and Fig. 21 shows the relationship of $\Delta T$ to $L_c$. Figure 22 shows the stream lines in two liquid layers for various values of $L_c$. Figures 21 and 22 are obtained for the same calculation conditions as those in Fig. 20. From Figs. 20～22, it is found that the effect of $L_c$ on $\Delta T$ and the flow pattern is weak in the range of $0.2 \leq L_c \leq 1.0$. However, because the relations of $T_i$ and $\Delta T$ to $L_c$ are almost linear, precise control of $T_i$ and $\Delta T$ may be possible by changing $L_c$. The effects of other parameters $Gr_1(300 \leq Gr_1 \leq 30 000)$, $Gr_2(100 \leq Gr_2 \leq 300 000)$, $Pr_1(0.1 \leq Pr_1 \leq 30)$, $Pr_2(0.01 \leq Pr_2 \leq 10)$, $\gamma(2 \leq \gamma \leq 60)$, $\kappa(0.1 \leq \kappa \leq 30)$ and $\lambda(0.01 \leq \lambda \leq 100)$ on the thermal and flow fields of two liquid layers were also investigated. The figures for these results are omitted here; however, it is found that large $Gr_1$, $\gamma$ and $\lambda$ yields large $\Delta T$ and that small $Gr_2$, $Pr_2$ and $\gamma$ also yield large $\Delta T$. It is also found that $\Delta T$ has a maximum value for various values of $Pr_1$.

6. Conclusions

The heat and fluid flow of two immiscible liquid layers in a vertical cylindrical container were studied both experimentally and numerically with the aim of constructing a high-performance device for production of solid spherical shells using liquid-liquid gas systems. The main features of this study were as follows.

(1) The validity of numerical results obtained here was confirmed from the fact that the numerical data of temperature in the two liquid layers agreed well with the experimental data. Therefore, the heat and fluid flow of two liquid layers, which were closely related to the performance of the device for production of solid spherical shells, could be accurately predicted using the numerical code developed here.

(2) The effects of various parameters on the thermal and flow fields of two liquid layers were clarified quantitatively, and the basic data for a
high performance simple device for producing solid spherical shells were presented. In particular, high thermal conductivity of the container material, small $L_s$, large $L_r$, and suitable $A$ resulted in a large temperature difference between the liquid-liquid interface and the upper liquid along the central axis of the cylindrical container. These results provide useful data to minimize the device size, and realize a short cooling distance and the stable production of solid spherical shells, because rapid solidification prevents breakdown of the molten spherical shells after formation.

References