Numerical and Experimental Study of Solidification in a Spherical Shell

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The present study explores numerically and experimentally the process of a phase-change material (PCM) solidification in a spherical shell. At the initial state, the PCM liquid occupies 98.5% of the shell. The upper segment of 1.5% contains air, which flows in as the solidification progresses. In the experiments, a commercially available paraffin wax is used. Its properties are engaged in the numerical simulations. The investigation is performed for solidification in spherical shells of 20 mm, 40 mm, 60 mm, and 80 mm in diameter at the wall uniform temperature, which varied from 10°C to 40°C below the mean solidification temperature of the phase-change material. Transient numerical simulations are performed using the FLUENT 6.2 software and incorporate such phenomena as flow in the liquid phase, volumetric shrinkage due to solidification, and irregular boundary between the PCM and air. The numerical model is validated versus the experimental results. Shrinkage patterns and void formation are demonstrated. Dimensional analysis of the results is performed and presented as the PCM melt fractions versus the product of the Fourier and Stefan numbers. This analysis leads to a generalization that encompasses the cases considered herein.

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1 Introduction

Phase change in spherical geometry is of great interest from the theoretical point of view and is of importance for the development of processes based on the use of latent heat. A considerable amount of literature on this subject exists, which reflects experimental, theoretical, and numerical investigations of melting and solidification. Theoretical studies on melting and solidification within spherical enclosures were performed by several researchers [1–8]. Advances in the field are summarized in Refs. [9,10] concerning its theoretical and practical aspects.

In the literature on solidification, a spherical shell containing a phase-change material (PCM) is usually treated as being completely filled by the molten material. This means that the PCM touches the entire internal surface of the shell. Then, if a uniform temperature is applied to the shell, solidification would be concentric, as demonstrated recently by Chan and Tan [11]. Actually, this physical situation is commonly treated in the literature. For instance, Ismail [12] summarized the available results and compared the position of the interface reported in various investigations as a function of time. The reported deviations from concentricity are related to a nonuniform distribution of the heat-transfer coefficient on the outside of the shell, as presented in Ref. [13]. The density difference between the liquid and solid is commonly neglected in those models. This may suit water freezing in some cases. For other materials and, in particular, paraffins, the solid density is higher than that of the liquid. This means that the solid will occupy only a part of the volume initially occupied by the liquid, while the remaining fraction of the enclosed volume will be a void. In casting [14], formation of voids is recognized as a major problem, as it is also in the design of latent heat thermal energy storage (LHETS) systems based on the melting-solidification cycles [15]. For instance, the solid shape and structure will affect the rate of consequent melting [16], thus affecting the entire cycle. Also, the design of a storage system must address such questions as shell shape, strength, orientation, packaging, etc. For instance, a packed bed of spheres [10] could collapse under its own weight due to the voids in the shells.

As mentioned above, the studies reported in the literature concern an initially completely filled spherical shell. However, it is practically impossible to have a rigid spherical shell filled 100% with liquid due to, e.g., thermal expansion. For example, if just a 0.5 mm high segment is filled with air in the upper part of a shell that is 80 mm in diameter, its volume is negligible being 0.012% of the shell volume, but its diameter is as large as 12.6 mm. As it is obvious that no solidification can occur inside the segment, its presence has a significant effect on the resulting solid shape. The profound effect of this feature has been recently demonstrated by Revankar and Croy [15].

Studies of solid-liquid phase-change are characterized by considerable difficulties encountered in both experimentation and modeling. In particular, temperature measurement inside the enclosure can affect the process itself. For this reason, visualization is widely used in melting studies. In solidification, however, visualization can be rather difficult if the solid is not transparent, hence it is rarely done [15]. Thus, it is common in the literature to report the overall results of measurements [18]. However, the overall measurements may not reflect voids or other structural features. As for the modeling, a significant effort is required to describe such features as solid-liquid volume change, natural convection in the melt, possible motion of the solid in the liquid, and, specifically in solidification, the curvilinear solid-liquid interface and formation of voids. Thus, while a pioneering work in this direction has been done back in the 1970s by Shamsundar and Sparrow [19], it is not surprising that the works that attempt at solving these problems of curvilinear geometries have started to appear only very recently [20–22].

In this study, both experiments and numerical modeling are reported. The experimental approach is similar to that employed by Glaich et al. [23] and involves thin plastic shells that are cut in halves during the process of solidification. The experimental findings serve to validate the numerical model and illustrate the essential features obtained numerically. The modeling is based on the method successfully used for melting by Assis et al. [24].

2 Modeling

2.1 Numerical Model. The numerical method has been extensively described in Ref. [24], and only the essential features are
presented here. The physical model is shown in Fig. 1. In the initial state, the liquid PCM fills 98.5% of the enclosed space. From above, the PCM is exposed to air, which is allowed to enter the shell gradually, as the process of solidification advances.

The properties of the PCM are based on a commercially available material—RT27 (Rubitherm GmbH, Berlin, Germany). As the material does not have a “sharp” melting temperature, a melting interval is used, with the solidus and liquidus temperatures set at 28°C and 30°C, respectively. The other properties are: latent heat $L=170$ kJ/kg, sensible heat capacity in liquid/solid state $c_p = 2.4 / 1.8$ kJ/kg-K, thermal conductivity in solid/liquid state $k = 0.24 / 0.15$ W/m-K, constant density of $\rho_l = 760$ kg/m$^3$ in the solid state, and dynamic viscosity of $\mu_l = 3.42 \times 10^{-3}$ kg/m-s in the liquid state. Variable density was defined in the liquid state with $\rho_l = 760$ kg/m$^3$ at 30°C and the volumetric expansion coefficient $\beta_l = 0.5 \times 10^{-3}$ K$^{-1}$. In the “mushy” state, where the solid and liquid phases coexist between 28°C and 30°C, the density varies linearly from 870 kg/m$^3$ at 28°C to 760 kg/m$^3$ at 30°C. It is assumed that both solid and liquid phases are homogeneous and isotropic, and the solidification process is axisymmetric. The molten PCM and the air are incompressible Newtonian fluids, and laminar flow was assumed in both. A density-temperature relation is used for air. In the simulations, the initial temperature of the bath was kept typically at 10°C or 7°C, which maintains a desired temperature within 0.1°C. A typical run took from a few days to several weeks.

2.2 Experimental. In the present study, 0.1 mm thick plastic shells are used, which are cut with the PCM after the latter had solidified completely or at the intermediate stages of the process [25]. Solidification of RT27 has been conducted in a Neslab RTE 7 circulating thermostatic bath, which maintains a desired temperature within 0.1°C. The bath was kept typically at 10°C or 20°C below the mean solidification temperature of the PCM. In order to be consistent with the simulation, the shell was inserted in the bath when the PCM temperature was about 32°C, i.e., few degrees above its mean melting temperature.

Two series of the experiments were performed. The first series was conducted in shells of 30 mm and 70 mm in diameter up to complete solidification. The shells were cut in halves, and the solid shape recorded. The second series was conducted in shells of 40 mm in diameter, where each shell was withdrawn from the bath at a prescribed time. The shell was cut in halves, the remaining liquid was removed, and the solid shape was recorded.

3 Results and Analysis

3.1 Solidification Patterns. An example of the series of complete solidification of paraffin RT27 in a shell 70 mm in diameter is shown in Fig. 2. One can see in the figure that a large void is formed in the upper part of the shell. This shrinkage resembles the cavity reported in Ref. [19] and can also be compared with similar voids reported recently for spherical [15] and vertical cylindrical enclosures [20–22]. We note that its location adjacent to the initially unfilled space in the upper part of the shell is consistent with the conclusion of Sulfredge et al. [17] that “with a degassed liquid, the distribution of solidification voids is largely determined by the location and pattern of the initial shrinkage voids.”

The shrinkage observed in Fig. 2 is further illustrated in Fig. 3 where the entire simulated process of solidification is presented...
for a shell 80 mm in diameter at $\Delta T = 20^\circ$C. The hues are shown in terms of the thermal conductivity, thus clearly separating between the solid, liquid, and air. The initial and final volumes occupied by air are denoted in the figure for the sake of clarity.

One can see from Fig. 3 that an upper shrinkage is formed, reflecting the experimental findings of Fig. 2 for a similar case. Its boundaries are defined by the initial level of the liquid PCM inside the shell. It appears that the numerical model succeeds to reflect formation of a characteristic solid phase shape up to the final state, which includes only solid and air, as shown by different colors.

The findings of Revankar and Croy [15] indicate that in some cases an additional central void can be formed when a spherical shell is not completely filled with liquid. That result was obtained for cyclohexane in a relatively small shell 2.54 cm in diameter. According to Revankar and Croy [15], “a large bubble” is formed at the top of the shell, while “several void dendrites” appear at its center. We note that the liquid-solid shrinkage of cyclohexane at 7.8% is much smaller than that of RT27, which is similar to $n$-octadecane.

Figure 4 shows our intermediate experimental and numerical results for the case of $D = 40$ mm and $\Delta T = 20^\circ$C. The numerical

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**Fig. 3** Simulated solidification with upper void ($D=80$ mm and $\Delta T=20^\circ$C)

**Fig. 4** Solidification with central void ($D=40$ mm and $\Delta T=20^\circ$C): (a) numerical, and (b) experimental [25]
results are shown in terms of the melt fraction (Fig. 4(a)) defined as the melted mass divided by the total mass of the PCM. Thus, the melt fraction is zero when solidification is complete. The experimental images are recorded immediately after the shells have been cut, and the remaining liquid has been poured out (Fig. 4(b)). Accordingly, the simulation results are also shown with the liquid phase “removed.” It appears that a remarkable agreement is obtained between the simulation and the experiment, providing validation for the model. Actually, Fig. 4 reflects, for a relatively small shell, the process leading to the formation of a central void, similar to the experimental findings of Revankar and Croy [15]. We note that the last picture of Fig. 4(a) does not show complete solidification, as the numerical approach is unable at this stage to model vacuum or formation of “void dendrites” at the center. However, this picture corresponds to the situation in which about 95% of the PCM has already solidified. Thus, the process is presented almost entirely. To the best of the authors’ knowledge, no simulation similar to that leading to Fig. 4 has been reported in the literature.

3.2 Melt Fraction. The melt fraction of the PCM, defined above, is shown for various cases in Fig. 5(a). We note here that the melt fraction reflects rather accurately the amount of heat to be removed from the system for full solidification. One can see that the full solidification time, which depends on the shell size and temperature difference, varies from 5 min to 150 min. On the other hand, the curves for various cases are similar in their shape.

3.3 Dimensional Analysis. The dimensionless parameters that define the process can be obtained from the dimensionless form of momentum and energy equations. The dimensionless energy conservation equation contains the following dimensionless groups:

(a) The Fourier number

$$Fo = (k/pc_p)(t/R^2)$$  \hspace{1cm} (1)

(b) The Stefan number

$$Ste = cpT/T_w$$  \hspace{1cm} (2)

where $\Delta T$ is the difference between the PCM mean melting temperature $T_m$ and the wall temperature $T_w$. In the present study, the Stefan number varies from about 0.1 ($\Delta T = 10^\circ C$) to 0.4 ($\Delta T = 40^\circ C$).

In order to account for the free convection contribution, the Grashof number was used in the analysis of melting [24]. However, the convective motion has been found insignificant in the solidification processes. For instance, in the process reflected in Fig. 3, i.e., for $D = 80$ mm and $\Delta T = 20^\circ C$, the velocities in the liquid phase did not exceed 1 mm/s at the beginning of the process, and 10 min later the velocities already decreased to the values below 0.1 mm/s, practically disappearing later. This result is consistent with the analysis of Viskanta and Gau [26], who established that the natural convection contribution is negligible for $Ste < 1$, and with the work of Yao [27] who showed that this contribution rapidly decreases with time. It is obvious that for smaller shells, e.g., that correspond to Fig. 4, the motion is even less significant. Thus, the Grashof number has been excluded from the present analysis. We note that the Prandtl number is about 35 for the material used in the present study, i.e., $Pr \gg 1$.

An application of the dimensionless groups introduced above leads to the generalized results presented in Fig. 5(b), where the melt fraction is shown versus the product of the Fourier and Stefan numbers $FoSte$ for all cases considered in the present study. Thus, the analysis in the present study is consistent with that introduced by Shamsundar and Sparrow [19]. One can see that all curves practically merge into a single curve, notwithstanding the differences in void formation illustrated in Figs. 3 and 4. Thus, we conclude that the generalization is valid for $Ste \leq 0.4$.

Also, this result provides an additional indication concerning the insignificant role of convection. On the other hand, one should bear in mind that volume shrinkage due to solidification [19] does not vary uniformly in the shell, but it is somewhat higher at the center and decreases from the center toward the shell surface. This contributable to the phase-change process expresses itself in the irregular shapes of the resulting solid phase.

An analysis of the results of Fig. 5(b) yields the following expression for the melt fraction:

$$MF = [1 - 4.5(FoSte)^{1/2}]^2$$  \hspace{1cm} (3)

This correlation, also shown in Fig. 5(b), is valid for the range of parameters explored in the present study and, in particular, for $Ste \leq 0.4$ and $Pr \gg 1$.

Summarizing the findings of the present study, we note that the conditions that determine the shape and structure of the voids in different cases have not been established yet. There are indications that the cooling rate and shell size are the major factors.

The results of the present investigation have been obtained for the cases in which air was allowed to flow into the shell to fill the
space formed by PCM shrinkage. In cases where the shell is sealed, the air is compressed on melting and expands on solidification, thus closing a full cycle. We assume that the trends discovered in the present study are quite general, and the results of the analysis performed herein will be valid also when additional features are taken into account.

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Nomenclature

- \( c_p \): specific heat at constant pressure (J/kg °C)
- \( D \): diameter (m)
- \( F_o \): Fourier number, \( (k/\rho c_p)l/R^2 \)
- \( k \): thermal conductivity (W/m °C)
- \( L \): latent heat (J/kg)
- \( P_r \): Prandtl number
- \( R \): radius (m)
- \( S_e \): Stefan number, \( c_pA/\rho k \)
- \( t \): time (s)
- \( T \): temperature (°C or K)

Greek Letters

- \( \beta \): volumetric expansion coefficient (1/K)
- \( \Delta \): difference
- \( \rho \): density (kg/m³)

References