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Abstract

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Lattice Boltzmann simulation of flow and heat transfer evolution inside encapsulated phase change materials due to natural convection melting

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Abstract: A comprehensive study of the melting process inside a capsule can potentially take full advantages of latent heat of phase change materials (PCMs). The present study was devoted to the problem of complex interaction of natural convection and melting of PCMs inside a spherical capsule under different sizes. The numerical results, simulated by lattice Boltzmann method (LBM), were compared with experimental data and published simulations. The results showed that LBM presented desirable accuracy compared to traditional computational fluid dynamics (CFD) methods. Then, the effects of non-uniform PCM properties, expressed by the solid/liquid thermal diffusivity ratio, on the melting rate were found to be nonlinear in different melting stages. The non-dimensional fully melting time reduced with the increase of the surface temperature and the capsule size, and the former compared to the latter could have a greater influence on the melting rate. Moreover, the non-dimensional fully melting time reduced when increasing the capsule diameter at the macro-scale; while there was a near-invariable non-dimensional fully melting time when the capsule size was changed at the micro-scale. The good understanding of the phase change process inside the capsule would provide essential information to develop a multi-scale model of microencapsulated PCM slurries.

Keywords: Melting; Phase change materials; Natural convection; Spherical capsule; Multi-scales; Lattice Boltzmann method

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Abbreviations:

Phase change material – PCM;

Computational fluid dynamics – CFD;

Lattice Boltzmann method – LBM;

Lattice Boltzmann – LB;

Multiple-relaxation-time – MRT.

24 Nomenclature

C	mushy zone constant	$\mathbf{S}$	relaxation matrix for flow field
C_p	specific heat of PCM [$\text{J}\cdot\text{kg}^{-1}\text{K}^{-1}$]	T	temperature [$^{\circ}\text{C}$]
C	lattice speed	Δt	unit lattice time
$\mathbf{F}$	total body force [$\text{N}\cdot\text{m}^{-3}$]	$\mathbf{u}$	velocity [$\text{m}\cdot\text{s}^{-1}$]
f_l	volume fraction of liquid	$\mathbf{u}_s$	velocity for solid phase [$\text{m}\cdot\text{s}^{-1}$]
f_i	density distribution function	Greek symbols	
$\mathbf{G}$	acceleration due to gravity [$\text{m}\cdot\text{s}^{-2}$]	α	thermal diffusivity [$\text{m}^2\cdot\text{s}^{-1}$]
g_i	enthalpy distribution function	β	thermal expansion coefficient [K^{-1}]
g^{eq}	equilibrium state of g_i	ρ	density [$\text{kg}\cdot\text{m}^{-3}$]
H	total enthalpy [$\text{kJ}\cdot\text{kg}^{-1}$]	λ	thermal conductivity [$\text{W}\cdot\text{m}^{-1}\text{K}^{-1}$]
L	latent heat of fusion [$\text{kJ}\cdot\text{kg}^{-1}$]	ν	kinematic viscosity [$\text{m}^2\cdot\text{s}^{-1}$]
$\mathbf{M}$	transformation matrix	Subscripts	
$\mathbf{M}$	moment vector in flow field	i	specified lattice direction
$\mathbf{N}$	moment vector in temperature field	l	liquid phase of PCM
$\mathbf{R}$	relaxation matrix for temperature field	s	solid phase of PCM

25 1. Introduction

26 Utilization of phase change materials (PCMs) for heat storage systems allows to extract a large
27 amount of energy within a tiny temperature variation, and thus has been applied to various fields,
28 such as building products [1-4], electronics cooling [5-7], and solar energy storage [8-10]. In
29 recent years, the encapsulated PCM (EPCM) and its slurry serve as a way to enhance

discharging/charging rate of the PCM by increasing the specific surface area. So far, encapsulated PCM and its slurry have been successfully applied in waste heat recovery [11], cooling systems [12, 13], and ground source heat pump systems [14].

In terms of the scale, the encapsulated PCM can be divided into macro-capsule, micro-capsule and nano-capsule with diameters of 1 mm to greater than 1 cm, from 1 μm to 1 mm, and less than 1 μm , respectively [15, 16]. In terms of the geometry, the encapsulated PCM can be classified into sphere, cylinder, and irregular shapes. In terms of the morphology, the encapsulated PCM can be categorized into mononuclear, polinuclear, matrix, and multi-wall [17]. In addition, there are two kinds of the melting mechanisms called unconstrained melting and constrained melting [18, 19]. The solid PCM will sink down due to gravity with direct contact melting at the bottom of the sphere during the unconstrained melting, while, in the constrained melting, the solid PCM is prevented from sinking to the bottom of the sphere. Different experimental and numerical studies have been performed to investigate PCM behaviors. The difference between the unconstrained melting process and constrained melting process was investigated via experiment methods under different surface temperatures and initial sub-cooling conditions [19]. Some scholars [18, 20, 21] used the conventional computational fluid dynamics (CFD) method to reproduce the constrained melting process inside the spherical capsule, and further analyze the effects of surface temperature, initial condition, and buoyancy-driven on the melting rate. However, the conventional CFD method would suffer from an empirical mushy zone constant, which was recommended between 10^4 and 10^7 [21, 22]. In contrast, the lattice Boltzmann (LB) model with the volumetric LB scheme can avoid the empirical mushy zone constant with no free parameters when it is used for simulating the melting process [23, 24]. The

evolution of temperature and mushy zone by using the lattice Boltzmann method (LBM) was investigated in various cavities such as square cavity [25-27], semicircle enclosure [28], an annulus between two coaxial vertical cylinders [29], and more complex structures [30, 31].

By mixing the encapsulated PCM and heat transfer medium like water, a functional thermal fluid called microencapsulated PCM slurry can be obtained. This slurry offers attractive opportunities for thermal energy transportation and heat transfer enhancement of heat exchangers [32]. The complex thermophysical properties and phase change behavior of the slurry made it difficult to predict the heat transfer of the slurry precisely. So far, many groups [33-38] presented various numerical models with a large amount of assumptions to analyze the heat and mass transfer with phase change in the microencapsulated PCM slurry. These assumptions included a uniform temperature inside the capsule, a single-phase non-Newtonian fluid instead of two-phase slurry, etc. However, the uniform temperature assumption made the melting rate present a linear decline inside of a non-linear trend when the thermal conductivity of the PCM was low. It was because this assumption ignored the drawdown of the melting rate caused by the inhibition of the low thermal conductivity of the PCM, resulting in a deviation between numerical results and experimental data [39]. To the best of our knowledge, based on the correlative algorithm [40, 41], it is reasonable to construct a multi-scale model without the above assumptions, which can provide a result that is more close to the reality. The main idea of the multi-scale model is an improved traditional slurry model by coupling the phase change information of the encapsulated capsules with heat transfer model of the slurry. Thus, it is critical to predict the behavior of the phase change inside capsules.

In our previous work [42], the constrained melting process inside a spherical capsule was

numerically investigated, and the simulations were then compared with the experimental data. The results showed that the effect of natural convection on the melting process decreased as the capsule size was decreased, and even could be ignored if the capsule diameter was less than 3 mm. In this study, the flow and heat transfer evolution of the PCM inside a spherical capsule was further simulated by using an enthalpy-based LBM. The numerical results were compared with the published simulation and experimental data. The effects of various parameters, such as surface temperature, capsule size, and non-uniform properties of the PCM, on the phase change process were analyzed. The results and their dimensionless forms were discussed in order to investigate the above influences of various parameters under different scales. These results would be useful to study microencapsulated PCM slurries on the heat transfer with phase change processes..

2. Methodologies and Models

2.1 Geometrical model

The geometrical model considered here is a spherical capsule filled with a solid PCM at an initial temperature T_{ini} , which could be a temperature lower than the PCM melting temperature T_{m} . For time $t > 0$, the surface temperature of the capsule T_{surf} was abruptly raised to a high value and the melting process commenced.

Note that the size of microcapsule is so small that the visualization of the melting patterns and the temporal evolution of the local temperature are hard to be measured through experiments. In this study, a spherical capsule with an internal diameter of 101.66 μm was presented to compare the results between LBM simulations and published experimental data [19], as shown in Fig.

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~~Fig. 1~~. In the experimental setup, eight points indicated as A-H were placed vertically along the symmetry axis that passed through the center of the sphere (point E). The distance between each point was 12.5 mm, except for point A which was only 10.3 mm away from point B and was used to monitor the chaotic temperature caused by the convection at the bottom of the spherical capsule (see Section 3.1). The point I, which had the same elevation as point E, was used to monitor the temperature next to the shell. The distance between points E and I was 50 mm.

Fig. 1 Spherical capsule and its section with different measurement points [19]. “+” and “-” values mean above or below the center of the capsule (point E), respectively.

For the experimental data used [19, 20], the initial temperature was 27.2°C, which was 1°C lower than the PCM melting temperature. The surface temperature was 40°C. Other properties of the PCM (n-octadecane) used are listed in ~~Table 1~~ ~~Table 1~~.

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Table 1 Thermophysical properties of n-octadecane [19, 20]

Parameter	Value
Melting temperature	28.2 °C
Density	772 kg·m ⁻³
Kinematic viscosity	5×10 ⁻⁶ m ² ·s ⁻¹
Specific heat	2330 J·kg ⁻¹ ·K ⁻¹
Thermal conductivity	0.1505 W·m ⁻¹ ·K ⁻¹
Latent heat of fusion	243.5 kJ·kg ⁻¹
Thermal expansion coefficient	0.00091 K ⁻¹

2.2 Total enthalpy-based lattice Boltzmann model

2.2.1 Assumptions

In this study, a total enthalpy-based lattice Boltzmann (LB) model [23, 24] was used to simulate the solid-liquid phase change heat transfer in a spherical capsule. It consists of an incompressible LB model for the velocity field and an enthalpy-based LB model for the temperature field. The moving solid-liquid phase interface was implicitly tracked through the liquid fraction, which can be obtained when the energy equation of PCM was solved. To simplify the physical problem, the following assumptions were made for the solid-liquid phase change heat transfer coupled with natural convection inside the spherical capsule.

a) Both the liquid and solid phases are homogeneous and isotropic, and thus their thermophysical properties are taken to be constants;

b) The density differences between solid and liquid phases are ignored;

c) Effective thermophysical properties are used to describe the PCM properties during the melting process, which are depended on the liquid fraction, specific heat and thermal conductivity;

d) Thermophysical properties of the PCM are assumed to be constants over the temperature range considered;

e) The density variation of the liquid PCM that drives natural convection is only considered in the body force terms (i.e. Boussinesq approximation) and;

f) The surrounding temperature around the spherical capsule is maintained constant.

Based on the above assumptions, the phase change process inside the spherical capsule could be considered as a symmetrical distribution around the perpendicular line through the center of

the spherical capsule. Hence, the flow of the liquid PCM could be assumed to be axisymmetric two-dimensional unsteady and incompressible [21].

2.2.2 Axisymmetric flow LB model

In the axisymmetric LB model for the velocity field, the multiple-relaxation-time (MRT) LB equation with an explicit treatment of the forcing term can be written as in Eq. (1) [43].

$$\mathbf{m}^*(r + \Delta r, t + \Delta t) = \mathbf{m}(r, t) - \mathbf{S}[\mathbf{m}(r, t) - \mathbf{m}^{\text{eq}}(r, t)] + \Delta t(\mathbf{I} - \mathbf{S}/2)\mathbf{F}_m(r, t) \quad (1)$$

where the superscript * means the effect of the solid phase that is not considered, $\mathbf{I}$ is the unit matrix, $\mathbf{M}$ is the transformation matrix for mapping velocity and moment spaces [44], $\mathbf{S}$ is the relaxation matrix in the moment space, $\mathbf{m} = \mathbf{M}f$ is the rescaled moment vector, and f is the density distribution function, $\mathbf{m}^{\text{eq}}$ is the corresponding equilibrium moment and can be expressed as follows [45].

$$\mathbf{m}^{\text{eq}} = \rho \left(1, -2 + 3\frac{\mathbf{u}^2}{c}, 1 - 3\frac{\mathbf{u}^2}{c}, \frac{u_r}{c}, -\frac{u_r}{c}, \frac{u_z}{c}, -\frac{u_z}{c}, \frac{u_r^2 - u_z^2}{c}, \frac{u_r u_z}{c^2} \right)^T \quad (2)$$

$$\mathbf{S} = \text{diag} \left(1.0, 1.0, 1.1, 1.0, 1.2, 1.0, 1.2, \left(0.5 + \frac{3v}{c_s^2 \Delta t} \right)^{-1}, \left(0.5 + \frac{3v}{c_s^2 \Delta t} \right)^{-1} \right) \quad (3)$$

where ρ , $\mathbf{u}$ and v mean the density, velocity and kinematic viscosity of the PCM, respectively, c_s means the sound speed in lattice space and is equal to $c/\sqrt{3}$. Base on the Boussinesq approximation, the forcing term in the moment space $\mathbf{F}_m$ is given by Eq. (4) [29].

$$\mathbf{F}_m = \left(0, 6\frac{\mathbf{F} \cdot \mathbf{u}}{c^2}, -6\frac{\mathbf{F} \cdot \mathbf{u}}{c^2}, \frac{F_r}{c}, -\frac{F_r}{c}, \frac{F_z}{c}, -\frac{F_z}{c}, 2\frac{F_r u_r - F_z u_z}{c^2}, \frac{F_r u_z + F_z u_r}{c^2} \right)^T \quad (4)$$

$$\mathbf{F} = \left(\frac{-2\rho v u_r}{r^2}, \rho |g| \beta (T - T_{\text{ref}}) \right) \quad (5)$$

where F_r and F_z are r- and z-components of the total body force $\mathbf{F}$, respectively, $|g|$ is the gravity acceleration. The thermal expansion coefficient β reflects the tendency of the PCM to change in

volume in response to a change in temperature, which was used to carry out the Boussinesq approximation.

In order to recover the axisymmetric momentum equations, the collision process of the fourth and sixth components of the moment vector $\mathbf{m}$ in Eq. ~~(1)(4)~~ should be modified [43].

$$m_3^*|_{\text{new}} = m_3^* - \frac{1}{r} \left[1 - \left(1 + \frac{6v}{c_s^2 \Delta t} \right)^{-1} \right] \Delta t^2 \left[\left(\frac{1}{12} + \frac{v}{2c_s^2 \Delta t} \right) m_1^{(1)} + 0.5m_7^{(1)} \right] \quad (6)$$

$$m_5^*|_{\text{new}} = m_5^* - \frac{1}{r} \left[1 - \left(1 + \frac{6v}{c_s^2 \Delta t} \right)^{-1} \right] \Delta t^2 m_8^{(1)} \quad (7)$$

where $m_1^{(1)}$, $m_7^{(1)}$ and $m_8^{(1)}$ can be calculated as follows.

$$m_i^{(1)} = (m_i - m^{\text{eq}}) / \Delta t + 0.5F_i \quad i = 1, 7, 8 \quad (8)$$

Based on the volumetric LB scheme [24], the velocity field should be calculated over the entire space that includes fluid and solid phases. Then, the effect of the solid phase can be considered by recalculating the density distribution function in Eq. ~~(1)(4)~~ through a linear interpolation as shown in Eq. (9).

$$f_i = f_i f_i^* + (1 - f_i) f_i^{\text{eq}}(\rho, \mathbf{u}_s) \quad (9)$$

where $\mathbf{u}_s$ is the velocity of the solid phase, whose value is set as 0 for the constrained melting.

Note that the density in Eq. ~~(9)(9)~~ should be first calculated using Eq. ~~(10)(40)~~ to ensure the local mass conservation. Here, for liquid phase ($f_i=1$), Eqs. ~~(1)(4)~~, ~~(9)(9)~~ and ~~(10)(40)~~ become a standard LB model, while for the solid phase ($f_i=0$), Eq. ~~(9)(9)~~ become $f_i=f_i^{\text{eq}}(\rho, \mathbf{u}_s)$, which means that the nonslip velocity $u=u_s$ can be strictly satisfied. In other words, there is no nonphysical flow in the solid phase.

The macroscopic quantities are calculated as follows.

$$\rho = \sum_i f_i^* - \frac{\Delta t}{2} \frac{\rho u_r}{r}, \quad \mathbf{u} = \left(\sum_i \mathbf{e}_i f_i + \frac{\Delta t}{2} \mathbf{F} \right) / \left(\sum_i f_i^* \right) \quad (10)$$

161 where $\mathbf{e}_i$ is the discrete velocity and given in Eq. (11).

$$\mathbf{e}_i = \begin{cases} c(0,0)^T & i = 0 \\ c(\cos[(i-1)\pi/2], \sin[(i-1)\pi/2])^T & i = 1-4 \\ \sqrt{2}c(\cos[(2i-1)\pi/4], \sin[(2i-1)\pi/4])^T & i = 5-8 \end{cases} \quad (11)$$

162 2.2.3 Axisymmetric thermal LB model

163 The total enthalpy distribution function $g(x,t)$ of the axisymmetric LB model, which used for
164 the temperature field, can be expressed as Eq. (12).

$$g_i(r + \Delta r, t + \Delta t) = g_i(r, t) - \omega_g [g_i(r, t) - g_i^{\text{eq}}(r, t)] \quad (12)$$

165 where the factor ω_g and the equilibrium distribution function g^{eq} are shown as follows [23, 29].

$$\omega_g = \left(1 + e_{ir} \frac{3\alpha}{rc^2} \right) / \left(0.5 + \frac{3\alpha}{\Delta t c^2} \right) \quad (13)$$

$$g^{\text{eq}} = \begin{cases} H - C_{p,\text{ref}} T + w_i C_{p,\text{ref}} T - w_i C_p T \frac{u^2}{2c_s^2} & i = 0 \\ w_i C_{p,\text{ref}} T + w_i C_p T \left[\frac{(\mathbf{e}_i \cdot \mathbf{u})}{c_s^2} + \frac{(\mathbf{e}_i \cdot \mathbf{u})^2}{2c_s^4} - \frac{u^2}{2c_s^2} \right] & i = 1-8 \end{cases} \quad (14)$$

166 where $C_{p,\text{ref}}$ is a reference specific heat, which was used to make the specific heat C_p and thermal
167 conductivity λ of the PCM decoupled [46], w_i is the weight coefficient, which equals to 4/9 when
168 $i=0$, and 1/9 for $i=1-4$, as well as 1/36 for $i=5-8$, C_p and α are different in the liquid domain and
169 solid one, as defined as follows.

$$\alpha = f_l \alpha_l + (1 - f_l) \alpha_s \quad (15)$$

$$C_p = f_l C_{p,l} + (1 - f_l) C_{p,s} \quad (16)$$

$$C_{p,\text{ref}} = 2C_{p,l} C_{p,s} / (C_{p,l} + C_{p,s}) \quad (17)$$

170 Here, the thermal diffusivity α and the thermal conductivity λ have the following relationship.

$$\alpha = \frac{\lambda}{\rho c} \quad (18)$$

171 The macroscopic variable, total enthalpy H , is calculated by Eq. (19).

$$H = \sum_i g_i \quad (19)$$

172 The volume fraction of the liquid f_l and temperature T can be determined by the
 173 thermodynamical relations as described in Eq. (20)(20) and Eq. (21)(21). Here, f_l is a local
 174 variable, representing the liquid fraction in the control cell of a lattice node, $f_l=0$ and $f_l=1$ mean
 175 that the lattice node is the solid part and liquid part, respectively. The value of f_l between 0 and 1
 176 represents the part that phase change happens.

$$f_l = \begin{cases} 0 & H \leq H_s \\ (H - H_s)/(H_l - H_s) & H_s < H < H_l \\ 1 & H \geq H_l \end{cases} \quad (20)$$

$$T = \begin{cases} H/C_{p,s} & H \leq H_s \\ H_s + \frac{H - H_s}{H_l - H_s}(T_l - T_s) & H_s < H < H_l \\ T_l + (H - H_l)/C_{p,l} & H \geq H_l \end{cases} \quad (21)$$

177 where H_s and H_l are the total enthalpy corresponding to the solidus temperature T_s and liquidus
 178 temperature T_l ($T \geq T_s$), respectively. The equal symbol means that the phase change process
 179 happened at a constant temperature, i.e. the melting temperature T_m in [Table 1](#)[Table 1](#).

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180 2.2.4 Model validation

181 In order to validate the enthalpy-based axisymmetric LB model, a baseline case of the melting
 182 process in an annulus between two coaxial vertical cylinders was tested [29], see [Fig. 2](#)[Fig. 2](#).

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183 The radii of the inner and outer cylinders are r_0 and $2r_0$, while the height l_z equals to $2r_0$. Initially,
 184 the annulus was filled with a solid PCM at the melting temperature T_m . Then, the wall
 185 temperature of the inner cylinder was raised to a high value T_b , while the outer cylinder wall was
 186 maintained at the melting temperature. The horizontal walls were supposed adiabatic. The PCM

considered was Paraffin RT35 and its thermophysical properties were given by Li et al. [29] in detail. The transient time was expressed by Fo number. The expressions of Nu and Fo were defined by Eq. (22).

$$Nu = \frac{\int_0^{l_z} q_r dy}{\lambda_l (T_b - T_m)}, \quad Fo = \frac{\alpha t}{l_z^2} \quad (22)$$

where T_m and T_b were the melting temperature and the temperature of the left vertical wall, respectively, l_z was the character height of the cavity, q_r was the local heat flux along the left wall, and λ_l and α were the thermal conductivity and thermal diffusivity of the liquid PCM, respectively.

Fig. 2 Schematic diagram of the melting process in an annulus at different time $t_1 < t_2$.

Here, the average Nu number on the left vertical isothermal wall, the evolution of the global liquid fraction, and the steady temperature field and flow field results were compared with the results of a published literature [29], see Fig. 3. It can be seen that those simulation results were very close to the published results. The maximum errors of Nu number and global liquid fraction was 3.50% and 3.06%, respectively. This indicated that the enthalpy-based axisymmetric LB model can predict the solid-liquid phase change heat transfer accurately.

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Fig. 3 Comparisons of the simulation results and results reported in [29].

2.2.5 Mesh resolutions test

In this work, the enthalpy-based axisymmetric LB model was carried out with FORTRAN programs on a personal computer with Intel(R) Core(TM) i5-6500 CPU and 8G RAM. Both the lattice length Δx and the lattice time Δt were set as 1.0, and thus the lattice speed $c=\Delta x/\Delta t$ was 1.0. The detailed thermophysical properties of the PCM used were listed in [Table 1](#). The viscosity in the lattice space was chosen as 2.0×10^{-2} , and the thermal diffusivity was 3.35×10^{-4} in order to keep the same Pr number that defined by Eq. (23). For both the flow field and the temperature field, the same transformation matrix $\mathbf{M}$ was used, and it was given in Eq. (24).

$$Pr = \frac{\nu}{\alpha} \quad (23)$$

$$\mathbf{M} = \begin{pmatrix} +1 & +1 & +1 & +1 & +1 & +1 & +1 & +1 & +1 \\ -4 & -1 & -1 & -1 & -1 & +2 & +2 & +2 & +2 \\ 0 & -2 & -2 & -2 & -2 & +1 & +1 & +1 & +1 \\ 0 & +1 & 0 & -1 & 0 & +1 & -1 & -1 & +1 \\ 0 & -2 & 0 & +2 & 0 & +1 & -1 & -1 & +1 \\ 0 & 0 & +1 & 0 & -1 & +1 & +1 & -1 & -1 \\ 0 & 0 & -2 & 0 & +2 & +1 & +1 & -1 & -1 \\ 0 & +1 & -1 & +1 & -1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & +1 & -1 & +1 & -1 \end{pmatrix} \quad (24)$$

Then, three different grid densities (i.e. 4096 nodes, 10206 nodes and 15197 nodes) were tested to make sure that the solution was independent from the adopted grid density, see Fig.4.

215 As a result, 10206 grids were found to be sufficient for the present work, which represented that
 216 there were 102 grids on the diameter.

Fig. 4 Variations of the global liquid fraction vs. time at different grid sizes.

3. Results and discussions

3.1 Comparison of numerical and experimental results

In this section, the constrained melting of the PCM inside a spherical capsule was simulated by the enthalpy-based axisymmetric LB model. The numerical results were compared with the experimental data concerned with a spherical capsule with the diameter of 101.66 mm. Firstly, a qualitative observation of the PCM melting was discussed. The experimental photographs of the constrained melting were shown in [Fig. 5](#), while the simulation results were presented in [Fig. 6](#). For the simulation results, the colorized temperature contours were shown on the left half of each circle, whereas the streamlines were drawn on the right half. Then, these observations were linked to the quantitative results of LBM simulations and detailed thermocouple readings, as shown in [Fig. 7](#).

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Fig. 5 Instantaneous photographs of the spherical capsule during the constrained melting [19, 20].

Fig. 6 Detailed temperature contours and streamlines for a wall temperature of 40°C at different time instants (the

red dashed line means the melting front).

By comparing each subpicture between ~~Fig. 5~~Fig-5 and ~~Fig. 6~~Fig-6, It can be seen that the

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243 LBM simulation results agreed well with the experimental data obtained from Tan [19, 20]. At
 244 the beginning, a thin layer of the liquid between the solid PCM and the spherical wall was
 245 formed, while the solid PCM remained a perfect sphere, as shown in ~~Fig. 5~~Fig. 5(b) and ~~Fig.~~
 246 ~~6~~Fig. 6(b). This is because the dominated factor of the PCM melting was the conduction in this
 247 stage. As time elapsed, a liquid layer was developed and thus the heat conduction was
 248 significantly reduced. An oval shape phase front was formed at the top of the solid PCM, while a
 249 waviness of the surface was observed at the bottom of the solid PCM, as illustrated in ~~Fig. 5~~Fig.
 250 ~~5~~(c)-(f) and ~~Fig. 6~~Fig. 6(c)-(f). This could be attributed to the natural convection, which occurred
 251 as a result of the warm liquid PCM rose along the hot spherical wall while the cool liquid PCM
 252 flowed down around the solid PCM. At the upper part of the spherical capsule, the “hot”
 253 spherical wall was higher than the “cold” solid PCM, which formed a steady thermal convection
 254 cell. On the contrary, the “hot” wall was lower than the “cold” solid PCM at the bottom part of
 255 the capsule. It would cause an intense natural convection and formed several chaotic convection
 256 cells until the solid PCM was melted completely, which further made the bottom of the solid
 257 PCM to be a waviness shape. Thus, a very interesting phenomenon can be observed in this stage
 258 that the thermal stable and unstable layers were co-existed in the spherical capsule. The
 259 horizontal stratified fluid layers can be observed in thermal stable layer, while there were several
 260 recirculating cells in the unstable layer. At the 120th minute, the position of the solid PCM was
 261 lower than the center of the sphere, thus clearly pointing to the preferential melting in the top
 262 half of the sphere, see ~~Fig. 6~~Fig. 6(g). In this stage, the solid PCM was a dome-shaped solid with
 263 top and bottom concave-down surfaces.

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Fig. 7 Comparison of the simulated and experimental results [19, 21] at different typical points (A, B, G).

Table 2 Comparison of the results between the use of conventional CFD method and LBM

Model	Software	Computed times (Parallel number)	Errors (%)		
			Location	Max	Average
Conventional CFD	FLUENT	348 min (1)	A	23.92	7.92
		225 min (2)	B	29.32	7.93
		178 min (4)	G	21.61	3.18
LBM	Visual studio (Fortran)	356 min (1)	A	11.09	4.23
		212 min (2)	B	17.89	8.26
		148 min (4)	G	12.79	2.81

In Fig. 7 and Table 2, the LBM simulations were compared with the conventional CFD simulations [21] and experimental data [19] at three measure points (A, B and G). The results showed that LBM had much higher parallelism than CFD, resulting in a higher efficiency of using LBM than that using CFD when the parallel number was more than two. Besides, the LBM results were much closer to the experimental data than the results obtained using the conventional CFD method [21]. For example, compared with the conventional CFD results at point A, the maximum and average errors of the LBM results can be reduced from 23.92% and 7.92% to 11.09% and 4.23%, respectively. From Fig. 7 (a) and (b), the disordered temperature data at the points A and B reflected the chaotic convection motion in the unstable layer at the bottom of the sphere. The chaotic temperature readings would be observed until there was no more solid PCM left in the capsule. From Fig. 7(c), the computed temperatures at

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point G were in a good agreement with the experimental data. The temperature curves were quite smooth, which was due to the stable nature of the liquid layer at the upper part of the sphere. It reflected that there was no strong convective motion existed, and confirmed that these points were located in the thermal stable layer. Similar figures were obtained at the points D, E, F and H, and thus they were not provided in ~~Fig. 7~~ Fig. 7.

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3.2 Effects of non-uniform properties of the PCM

To date, most studies on the phase change inside the spherical capsule considered the effects of different parameters such as the capsule size, the initial/surface temperature and the non-dimensional numbers (Ra , Pr , Ste) on the PCM melting process. The Pr number was previously defined in Eq. (23)(23). Ra and Ste numbers were defined in the following equation.

$$Ste = \frac{C_p (T_{surf} - T_m)}{L}, \quad Ra = \frac{|g| \beta (T_{surf} - T_m) l_z^3}{\nu \alpha} \quad (25)$$

where T_{surf} and T_m were the surface temperature of the spherical capsule and the melting temperature of the PCM, respectively, l_z was the character length, which can be set as the diameter, g was the acceleration of gravity, L was the latent heat of fusion, and β was the volume expansivity.

So far, there is insufficient studies focusing on the differences of the thermal diffusivity between the liquid and solid PCMs in existing non-dimensional parameters. In general, the thermal diffusivity of the solid PCM is larger than of the liquid state. It is a key parameter to the heat transfer process, and thus further affects the phase change process inside the capsule. In this study, a non-dimensional parameter (α_s/α_l), which represented the thermal diffusivity ratio of the solid PCM to the liquid PCM, was used to investigate the effect of non-uniform properties of the

PCM on the melting process. In order to observe an obvious regularity, the initial melting temperature in this section was set as 18.2°C (i.e. 10°C below the melting temperature). Here, the melting state was described by the global liquid fraction. Fig. 8 shows the variation in the global liquid fraction as a function of the time at different thermal diffusivity ratios (α_s/α_l). Based on the experiments [47], the thermal diffusivity ratio of paraffin varied from 1.1 to 1.7, and thus the present simulation was performed at six different ratios, which varied from 0.75 to 2.00 regularly to examine its effects on the PCM melting process. By increasing the ratio from 1.0 to 2.0, the time required for completing the PCM melting significantly decreased from 143 min to 85 min. However, the effect of the thermal diffusivity ratio on the dissolved time became was decreased with the increase of thermal diffusivity ratio.

Fig. 8 Variations in the global liquid fraction versus time at different thermal diffusivity ratios.

In order to further investigate the effect of the thermal diffusivity ratio on the melting rate, the variation in the melting rate as a function of the time and global liquid fraction at different ratios are shown in Fig. 9 (a) and (b) respectively. Fig. 9 (a) could be used to analyze the PCM melting behavior, while Fig. 9 (b) could be helpful when examining the magnitude of the melting rate under the same melting state (approximately expressed by the global liquid

fraction). It can be easily seen that there existed a rapid decrease in the melting rate at the beginning, which was due to the fact that the effect of the heat conduction was significantly reduced as the liquid layer progressed, as mentioned in Section 3.2. Then, the effect of the natural convection gradually became the dominated factor of the PCM melting, and thus the melting rate would fluctuate apparently because of the chaotic convection motion (as mentioned in Section 3.3). The transformation of the dominant factor from conduction to convection happened in a small range at different thermal diffusivity ratios (see the grey shadow area in Fig. 9Fig-9).

In addition, there was an interesting phenomenon in Fig. 9Fig-9(b) that the melting rate was stiffened to double as compared to its original one, when the thermal diffusivity ratio doubled at the conduction dominated stage. During the transformation stage (see the grey shadow area in Fig. 9Fig-9(b)), the effect of the thermal diffusivity ratio on the melting rate was rapidly weakened. The double increase in the thermal diffusivity ratio resulted in the melting rate increased by 64% when the global liquid fraction was about 0.13. As the global liquid fraction increases, this influence was somewhat enhanced: the melting rate was improved from 1.64 to 1.79 times when the thermal diffusivity ratio was expected to be doubled.

Fig. 9 Variations in the melting rate versus time and global liquid fraction at different diffusivity ratios.

3.3 Effects of the surface temperature and capsule size

In this section, the effects of the capsule size and the surface temperature on the melting process were discussed. Note that LBM was a mesoscale numerical method, and it was suitable for the simulations in both macroscale [24, 45] and microscale [48, 49]. In order to compare the results at different sizes and surface temperatures, a set of non-dimensional parameters was introduced in this section. The melting time was expressed by the Fo number. It was the ratio of the conduction transport rate to the quantity storage rate and was defined by Eq. (22). The different surface temperatures were expressed by the Ste number. It was the ratio of the sensible heat to the latent heat of the PCM and was defined by Eq. (25).

The simulation was performed at three different Ste numbers and six different capsule diameters, namely $Ste=0.08059$ ($T_{\text{surf}}=35^{\circ}\text{C}$), $Ste=0.13225$ ($T_{\text{surf}}=40^{\circ}\text{C}$), $Ste=0.18392$ ($T_{\text{surf}}=45^{\circ}\text{C}$) and $D_1=101.66\text{mm}$, $D_2=50.83\text{mm}$, $D_3=25.42\text{mm}$, $D_4=1.017\text{mm}$, $D_5=508.3\mu\text{m}$, $D_6=254.2\mu\text{m}$, respectively. The first three spherical capsules (D_1 - D_3) were in the macro-scale, while the others were in the micro-scale [15, 16]. Fig. 10 shows the variations in the global liquid fraction versus the non-dimensional time (Fo) at different Ste numbers and capsule sizes. The slopes of these graphs represented the melting rate. The highest melting rate occurred at the beginning of the melting process because of the heat conduction. For each capsule size, the global liquid fraction increased more rapidly with a larger surface temperature (or Ste), whereas the time required for completing the melting reduced with the increase of Ste . The effects of these parameters on the melting time and melting rate were different at macro- and micro- scales. In order to clearly illustrate the melting times under different conditions, the detailed information was provided in Table 3.

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Fig.10 Variations in the global liquid fraction versus Fo at different Ste numbers and capsule sizes.

Table 3 The end time of the melting process at different Ste numbers and capsule sizes

Diameter (mm)	$T_{\text{Surf}} (^{\circ}\text{C})$ Ste	Time (min) Fo	Diameter (μm)	$T_{\text{Surf}} (^{\circ}\text{C})$ Ste	Time (s) Fo
101.66	35 (0.08059)	274 (0.1331)	1017.0	35 (0.08059)	7.92 (0.6403)
	40 (0.13225)	129 (0.0625)		40 (0.13225)	4.56 (0.3689)
	45 (0.18392)	76 (0.0371)		45 (0.18392)	3.20 (0.2589)
50.83	35 (0.08059)	97 (0.1878)	508.3	35 (0.08059)	1.98 (0.6404)
	40 (0.13225)	51 (0.0987)		40 (0.13225)	1.13 (0.3690)
	45 (0.18392)	33 (0.0634)		45 (0.18392)	0.80 (0.2590)
25.42	35 (0.08059)	39 (0.3002)	254.2	35 (0.08059)	0.49 (0.6404)
	40 (0.13225)	20 (0.1557)		40 (0.13225)	0.29 (0.3690)
	45 (0.18392)	13 (0.1009)		45 (0.18392)	0.20 (0.2592)

It was interesting to note that for each surface temperature (or Ste), the actual melting time of a micro-capsule (D_4 - D_6) was always less than that of a macro-capsule (D_1 - D_3), while the non-dimensional form of the fully melting time showed an opposite trend. The former was mainly because the heat inputs to the capsule and PCM content were not the same when the surface temperature of the capsule maintained as constant, and thus the fully melting time was

always decreased with the decrease of the capsule size. The latter was because the convection effect was decreased with the decrease of the capsule size. During the melting process, the liquid PCM increased and thus the heat conduction was significantly reduced, which caused a long fully melting time. In the following discussion, the fully melting time was expressed by Fo instead of the actual time.

At the macro-scale, based on the data presented in ~~Fig. 10~~ Fig. 10(a)-(c) and ~~Table 3~~ Table 3, it can be extracted that 28.6% increase in the surface temperature was from 35°C to 45°C resulted in about 66.3-72.1% decrease in the melting time for each constant diameter, respectively. The effect of the surface temperature on the melting time slightly increased with the increase of the diameter of the capsule. For example, when the surface temperature was 45°C, the increase of the capsule diameter varied from 25.42 mm to 50.83 mm and from 50.83 mm to 101.66 mm resulted in 37.2-41.4% decrease in the melting time, respectively. The surface temperature had a more prominent effect on the melting time than the capsule size.

At the micro-scale, as shown in ~~Fig. 10~~ Fig. 10(d)-(f) and ~~Table 3~~ Table 3, the increase of the surface temperature varied from 35°C to 45°C, resulted in about 59.6% decrease in the fully melting time for each constant diameter, respectively. However, when the size of the capsule increased from 254.2 μm to 508.3 μm , or from 508.3 μm to 1.017 mm, there was a near-invariable fully melting time. In addition, there was an interesting phenomenon in ~~Fig. 10~~ Fig. 10 that the variation in the global liquid fraction was nearly linear at the macro-scale while a parabolic trend at the micro-scale was observed. It may be caused by the fact that the convection played a more important role than the conduction during the most of the melting process at the macro-scale, while the conduction was dominated at the micro-scale.

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4. Conclusions

Flow and heat transfer evolution of PCMs inside an encapsulated capsule due to natural convection melting was simulated using an enthalpy-based axisymmetric lattice Boltzmann model (LBM). The simulation results matched well with the experimental data, which presented that the thermal stable and unstable layers were coexisted in the spherical capsule during the melting process. Moreover, the results of LBM presented a desirable accuracy and efficiency compared to that of the traditional CFD methods in this situation.

In order to study the effect of the difference between the liquid thermal diffusivity and solid thermal diffusivity on the melting process, a dimensionless parameter called solid/liquid thermal diffusivity ratio was introduced. The fully melting time decreased by 40.6% (e.g. from 143 min to 85 min) when increasing the thermal diffusivity ratio from 1.0 to 2.0. During the melting process, however, the effect of the thermal diffusivity ratio on the melting rate was different. With continuous increase of the thermal diffusivity ratio, the melting rate enhancement was decreased rapidly with the progression of the melting process when the global liquid fraction was less than 0.13, and then the melting rate enhancement increased slightly from 1.64 to 1.79 times until the melting process was completed.

Finally, the effects of the surface temperature and capsule sizes on the melting time and melting rate were investigated at different scales by a set of dimensionless numbers, such as Ste and Fo . The surface temperature had a more significant effect than the spherical capsule size on the fully melting time. In addition, the effect of the capsule sizes on the melting process was different. At the macro-scale (i.e. diameters from 1 mm or greater than 1 cm), the double increase in the diameter of a capsule resulted in the fully melting time reduced by 35.8-37.3%; while at

the micro-scale (i.e. diameters from 1 μm to 1 mm), there was a near-invariable fully melting time when the capsule diameter was doubled.

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