Review on Numerical Simulations for Solidification & Melting of Nano-Enhanced Phase Change Materials (NEPCM)

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Abstract. A review on numerical simulations performed for solidification and melting process of Nano-Enhanced Phase Change Materials (NEPCM) is reported. The studies were conducted to understand the factors influencing the outcome such as nanoparticle fraction in the mixture, nanoparticle size, boundary conditions imposed and container geometry. It was found that while most studies investigated particle fraction effect, very few was conducted on the effect of nanoparticle size and shape. The numerical models applied to simulate the numerical works were compared. Most researchers applied enthalpy-porosity formulation coupled with finite volume method to perform the simulations. Most models solved a single macroscale domain at each iteration by assuming NEPCM as a single-phase substance which resulted in no attempt to model it as a two-phase substance which requires multidomain approach. Such dilemma is avoided when mesoscale method (Lattice Boltzmann Method - LBM) is applied.

1. Introduction

Over the recent years, researchers have dedicated their works to enhance the effectiveness of Phase Change Materials (PCM) as thermal energy storage system for various purposes. PCM relies on its ability to absorb and remove latent heat during its phase changing process in order to store and release heat energy as required. Such latent heat storage system is attractive as it offers high energy storage capacity during the process within acceptable range of temperature change [1, 2]. Although phase change process may occur in other different phase combinations, the most practical phase changes for PCM applications are liquid-solid (Solidification) and solid-liquid (Melting). In other words, PCM solidification allows for latent heat 'discharging' while PCM melting enables latent heat 'charging'.

Nanotechnology is manipulation of nano-sized particles to achieve new technological innovations. Choi et. al [3] proved that thermal conductivity of liquids can be enhanced by dispersing nanosized particles. In the following years, many studies have been performed on nanofluids which confirms its ability to enhance liquid thermophysical properties [4, 5]. Parallel with this advancement, nanoparticles have also been applied to improve the performance of PCM thus the term Nano-Enhanced Phase Change Materials (NEPCM) is introduced. In the year 2007, Khodadadi & Hosseinizadeh [6] added nanoparticles in PCM to investigate these fine particles influence on its performance during phase change process and the result was encouraging. The introduction of NEPCM calls for the need to further analyse the balance in between the benefits offered and the cost of developing the material. For such detail analysis and study, numerical simulation is the most

convenient and economical approach that have been widely used over the years. It allows various different conditions to be simulated by substituting the variables in the models used.

With the advancement of numerical methods and computational capability in the mid-20th century, solution to Navier-Stokes equations which were earlier limited and deemed to be tedious received more attention. Newtonian thermal-fluid flow problems were able to be predicted quantitatively by solving the set of mass, momentum and energy conservation equations. This scenario enabled for solidification / melting simulations to be conducted by considering both conductive and convective heat transfer mechanisms. As a result, enthalpy-porosity method (EPM) was developed [7, 8]. This approach is a single-domain formulation which adds porosity source term to the momentum equations to differentiate liquid and solid regions during the phase change process. A source term is also added to the energy equation via enthalpy terms to account for latent heat evolution during the same process.

In the 21st century, progress have been made to develop other approach which are able to solve solidification and melting phenomena. Mesoscale methods such as Lattice Boltzmann Method (LBM) have been proven to be able to yield acceptable results by modelling fluid flow at mesoscopic level in terms of local interactions between particles. LBM was first implemented for phase change material simulations in 2001 by Miller et.al [9].

Up to this moment, numerical simulations has been widely used to predict solidification and melting of NEPCM. To gain valuable results for further discussion and interpretation, it is vital that the model used is capable of producing acceptable results relative to the real physical phenomena. This review intends to summarize the progress made in the subject above by focusing on numerical simulations made for NEPCM phase change process which occur purely due to temperature difference. Any works with the objective to study the implications of having additional structures or external force were not included.

2. Factors Influencing NEPCM Solidification & Melting in Numerical Simulations

Since the early studies by numerical simulations on NEPCM solidification & melting, it has been found that the process relies on several important parameters. These factors have been investigated and the details are summarized in Table 1. All these presented works were validated by either previous experimental or numerical works and showed acceptable agreement.

2.1. Nanoparticle fraction in mixture

The main reason that NEPCM is widely used nowadays to replace conventional PCM as latent heat storage is the presence of dispersed nanoparticle such as copper (Cu), copper-oxide (CuO), aluminium-oxide (Al_2O_3) and Carbon Nanotube (CNT). These nanoparticles have high thermal conductivity which influences the overall NEPCM phase change behavior. Khodadadi & Hosseinizadeh [6] and Sebti et.al [10] found that increasing Cu nanoparticle volume fraction from 0 to 20% in a square and from 0 to 5% in an annulus NEPCM respectively will increase the thermal conductivity and heat transfer rate therefore reducing the total solidification time. Later, Elbahjaoui et.al [11] investigated solidification of rectangular NEPCM slabs containing Cu volume fractions of 0 to 8%. They found that the solidification rate also improved. Arasu et.al [12] simulated solidification and melting of annular NEPCM containing 0 to 10% volume fraction of Al_2O_3 and reported that the theoretical energy storage capacity of NEPCM becomes lower while the overall thermal conductivity and heat transfer rate increases. Kashani et.al [13] found that the heat transfer rate increases, and larger solid region exist with increment 0 to 8% of Cu volume fraction for solidification of square NEPCM. This larger solid region exists when the volume fraction increment is coupled with low wall temperature. Sharma et.al [14] concluded that the solidification time of NEPCM containing Cu decreases as the volume fraction increases from 0 to 20%.

Fan et.al [15] performed simulation for melting of rectangular NEPCM containing CNT nanoparticle. The CNT volume fraction was increased from 0 to 10% and as a result, the thermal conductivity increased, the melting rate improved while the latent heat decreases. At same time, they noticed an increase of mixture viscosity as well. Sushobhan & Kar [16] found the same observation by simulating melting of square NEPCM containing copper-oxide. Hosseini et.al [17] recorded decrease of 14.6% melting time, increase of 146% melting front penetration and increase of 44.2% liquid

fraction after the volume fraction of copper in annulus NEPCM was increased from 0 to 5%. Sebti et.al [18] simulated melting of square NEPCM dispersed with copper about the same percentage causing both the melting rate and thermal conductivity to increase. Bechiri et.al [19] found the same observation after simulating melting of semicircle NEPCM dispersed with Al_2O_3. Parsazadeh & Duan [20] found that the heat transfer, melting rate and viscosity increases when CuO nanoparticle is added from 0 to 7% to cylindrical NEPCM. Feng et.al [21] simulated melting of water NEPCM containing copper and increased the volume fraction from 0 to 10%. They found that the melting process expedited, and thermal conductivity increased. Next, Darzi et.al [22] and Jourabian et.al [23, 24] also simulated melting of the same NEPCM and nanoparticle but with increment of particle fraction from 0 to 3% and 0 to 4% respectively. The preceding researchers concluded that melting rate and thermal conductivity increases while the latent heat of fusion decreases. The presence of nanoparticles in conventional PCM greatly improves the overall NEPCM solidification and melting process

2.2. Nanoparticle Size

Nanoparticle size is another factor which influences NEPCM phase changing process. El Hasadi & Khodadadi [25] did the only research concerning this factor by applying numerical simulation. They reported thinning of frozen colloid layer over a given period despite of increased thermal conductivity after reducing Cu nanoparticle size from 5 nm to 2 nm in square water cavity. In addition to that, there was a change of solid-liquid interface shape from stable planar to unstable dendritic structure.

2.3. Boundary conditions

Boundary conditions set for the numerical model may also influence solidification/melting process. Kashani et.al [13] observed that by lowering the temperature of the wall in a square NEPCM coupled with increment of particle fraction, the solid region fraction increases during solidification. Sebti et.al [10] found that by increasing the temperature difference, ΔT from 5 to 20 between the outer and inner cylinder which constitutes an annulus, the total solidification time is reduced. Sharma et.al [14] witnessed in Figure 4(a) that as ΔT increases which is temperature difference between the inclined walls in a trapezium, the solidification process is expedited. Sebti et.al [18] simulated melting of paraffin-copper NEPCM and increased the temperature difference between hot wall and melting temperature. The result is melting process becomes faster. Mostafavinia et.al [26] studied melting of a square NEPCM by changing the position of heat source-sink pairs on top and bottom wall. They found out that the position of heat-source must be at separately dedicated walls to produce highest temperature difference, ΔT. This position coupled with placing the higher temperature wall on top will promote convection thus achieving the best melting performance. Ebrahimi & Dadvand [27] also performed simulation for the same condition but the position of heat source-sink were on the left and right walls. They found that the source-sink must be placed with alternate locations and order on the walls to achieve highest melting rate. Arici [28] studied on effect of placing the hot wall on vertical right and horizontal bottom walls. They found that bottom heating increases the stored energy by 26% and increases the melting rate. As conclusion, high temperature difference for container walls, appropriate positioning of source-sink or combination of both may provide desirable performance for the NEPCM; accelerated phase change process in solidification or melting.

2.4. Container Geometry

Sharma et.al [14] found that trapezoidal NEPCM cavity takes lesser time to completely solidify as compared to square cavity with the same internal area. In addition to that, increment of the inclination angle of inclined wall also contribute to higher solidification rate which is shown in Figure 4(b). Kashani et.al [29] simulated solidification of wavy cavity containing NEPCM and found that by increasing the waviness, solidification rate is enhanced. Surface waviness can be used to control solidification time. Elbahjaoui & Qarnia [30] studied the melting of rectangular NEPCM where the aspect ratio is changed. They observed that by increasing the aspect ratio, the surface area for heat transfer increases thus expediting charging / melting time. Pahamli et.al [31] studied about the effect of inclination angle on melting of cylindrical NEPCM and found that increasing the angle value increases thermal conductivity and expedites melting. In summary, certain container geometry

provides better phase change performance in comparison to conventional geometry i.e. trapezoidal and wavy shape. Even if a conventional shape is used, the performance can be enhanced by changing its aspect ratio or inclination angle.

Table 1. Summary of numerical simulations for NEPCM solidification & melting

Author	NEPCM	Particle size (nm)	Particle Volume (%)	Geometry	Numerical Technique	Findings
Khodadadi & Hosseinizadeh [6]	H_2O + Cu	10	0, 10, 20	Square	EPM - FVM	↑ **Particle fraction results in:** ↑ heat release rate & k ↓ latent heat of fusion
Sebti et.al [10]	H_2O + Cu	Not reported	0, 2.5, 5	Annulus	EPM - FVM	↑ **Particle fraction results in:** ↑ heat transfer rate & k ↑ ΔT: ↓solidification time
Elbahjaoui et.al [11]	n-Octadecane + Cu	45	0, 2, 8	Rectangle	EPM - FVM	↑ **Particle fraction results in:** ↑ solidification & heat discharge rate
Arasu et. al [12]	Paraffin + Al_2O_3	Not reported	0, 2, 5, 10	Annulus	EPM - FVM	↑ **Particle fraction results in:** ↑ charge-discharge rate ↑ heat transfer rate & k ↓ energy storage capacity
Kashani et. al [13]	n-Hexadecane + Cu	10	0, 3, 8	Square	EPM - FVM	↑ **Particle fraction results in:** ↑ heat transfer rate & k + ↓wall T =↑solid fraction
Sharma et.al [14]	H_2O + Cu	10	0, 10, 20	Trapezium	EPM - FVM	↑ **Particle fraction &ΔT results in:** ↑ solidification & heat transfer rate ↑ **Incline angle results in:** ↑ solidification rate
Kashani et.al [29]	H_2O + Cu	1	0, 5, 10	Wavy	EPM - FVM	↑ **Particle fraction & waviness:** ↑ solidification rate
Fan et. al [15]	Eicosane + CNT	Not reported	0, 2, 10	Rectangle	EPM -FVM	↑ **Particle fraction results in:** ↑ melting rate & k ↑ viscosity ↓ latent heat of fusion
Sushobhan & Kar [16]	n-octadecane + CuO	Not reported	0, 2.5, 5	Square	EPM - FVM	↑ **Particle fraction results in:** ↑ melting rate & k ↑ viscosity
Hosseini et.al [17]	RT 50 + Cu	1	0, 3, 5	Annulus	EPM - FVM	↑ **Particle fraction results in:** ↑ melting rate ↑ melt front penetration ↑ liquid fraction

Author	NEPCM	Particle size (nm)	Particle Volume (%)	Geometry	Numerical Technique	Findings
Sebti et. al [18]	Paraffin + Cu	Not reported	0, 2.5, 5	Square	EPM - FVM	↑ **Particle fraction results in:** ↑ melting rate & k ↑ **ΔT results in:** ↓melting time
Bechiri et.al [19]	NaNO3+ Al_2O_3	Not reported	0, 3, 6	Circle	FVM	↑ **Particle fraction results in:** ↑ melting rate & k
Parsazadeh and Duan [20]	Paraffin + CuO	10	0 – 7	Shell-tube	EPM - FVM	↑ **Particle fraction results in:** ↑ melting rate ↑ viscosity
Feng et.al [21]	H_2O + Cu	1	0, 5, 10	Square	EB - LBM - DDF	↑ **Particle fraction results in:** ↑ melting rate & k
Darzi et.al [22]	H_2O + Cu	100	0, 1, 2, 3	Square	EB - LBM - DDF	↑ **Particle fraction results in:** ↑ melting rate & k ↓ latent heat of fusion
Jourabian et. al [23]	H_2O + Cu	100	0, 2, 4	Cylinder	EB - LBM - DDF	↑ **Particle fraction results in:** ↑ melting rate & k ↓ latent heat of fusion
Jourabian and Farhadi [24]	H_2O + Cu	100	0, 2, 4	Semi circle	EB - LBM - DDF	↑ **Particle fraction results in:** ↑ melting rate & k ↓ latent heat of fusion ↑ viscosity
El Hasadi & Khodadadi [25]	H_2O + Cu	2, 5	10 (wt%)	Square	EPM - FVM	↓ **Size particles results in:** ↑ thermal conductivity ↓ frozen region thickness ≠ phase interface shape
Mostafavinia et.al[26]	Paraffin + Al_2O_3	Not reported	0, 2, 5	Square	EPM - FVM	**Source-sink at dedicated walls and hot wall on top results in:** = Highest melting rate
Ebrahimi & Dadvand [27]	Paraffin + Al_2O_3	Not reported	0, 2, 5	Square	EPM - FVM	**Source-sink with alternate location & order on vertical wall results in:** = Highest melting rate
Arici et. al [28]	Paraffin + CuO	29	0, 1, 3	Square	EPM - FVM	**Bottom heating results in:** ↑ melting rate ↑ stored energy
Elbahjaoui and Qarnia [30]	P116 + Al_2O_3	45	0, 2, 6, 8	Rectangle	EPM - FVM	↑ **Aspect ratio results in:** ↑ area for heat transfer ↑ melting / charging rate
Pahamli et. al [31]	RT 50 + CuO	1	0, 2, 4 (wt)	Annulus	EPM - FVM	↑ **Particle fraction results in:** ↑ melting rate & k ↑ **Inclination angle:** ↑ melting rate

Note:
EPM-FVM: Enthalpy-Porosity Method coupled with Finite Volume Method
EB-LBM-DDF: Enthalpy Based-Lattice Boltzmann Method with Double Distribution Function

3. NEPCM Solidification & Melting Modelling

The system of equations for natural convection coupled with phase change is widely used to simulate NEPCM solidification and melting. Assuming the mixture is incompressible, behaves as a Newtonian fluid; the continuity, momentum and energy conservation equations are written as follows:

Continuity equation:

$$\frac{\partial u}{\partial x}+\frac{\partial v}{\partial y}=0 \tag{1}$$

X-momentum equation:

$$\frac{\partial u}{\partial t}+u\frac{\partial u}{\partial x}+v\frac{\partial u}{\partial y}=\frac{1}{\rho_{nf}}\left[-\frac{\partial P}{\partial x}+\mu_{nf}\left(\frac{\partial^2 u}{\partial x^2}+\frac{\partial^2 u}{\partial y^2}\right)+Source\ term\ /external\ force\right] \tag{2}$$

Y-momentum equation:

$$\frac{\partial v}{\partial t}+u\frac{\partial v}{\partial x}+v\frac{\partial v}{\partial y}=\frac{1}{\rho_{nf}}\left[-\frac{\partial P}{\partial y}+\mu_{nf}\left(\frac{\partial^2 v}{\partial x^2}+\frac{\partial^2 v}{\partial y^2}\right)+Source\ term\ /external\ force\right] \tag{3}$$

Energy equation:

$$\frac{\partial h}{\partial t}+u\frac{\partial h}{\partial x}+v\frac{\partial h}{\partial y}=\frac{k_{nf}}{(\rho C_p)_{nf}}\left(\frac{\partial^2 h}{\partial x^2}+\frac{\partial^2 h}{\partial y^2}\right)\pm Source\ term\ /external\ force \tag{4}$$

Subscript *nf* denotes nanofluid (NEPCM). Source term/external force are added to complement the problem to be solved. Three distinct regions will be present: a solid region, a liquid region, and a mushy region consisting of liquid and solid.

3.1. Enthalpy-Porosity Method (EPM)

This approach is the most widely used method as shown in Table 1 to model NEPCM solidification and melting. It can be coupled with any numerical techniques available such as Finite Volume Method (FVM) or Finite Element Method (FEM). In this method, the basic conservation equations have to be solved throughout a single calculation domain and the NEPCM is assumed as a single-phase substance. Details on the theory of the method is available [7, 8].

In a system going through a phase change via heat transfer, the total enthalpy is indicated as:

$$H=h+\Delta H \tag{5}$$

where it is the sum of sensible enthalpy, h and latent heat, ΔH. The latent heat component is temperature dependent and the phase change process should occur within a temperature range since NEPCM is a non-pure substance:

$$\Delta H=\varepsilon(T)=\begin{Bmatrix} L\ for\ T>T>T_L \\ L(1-\varepsilon)\ for\ T_L>T>T_S \\ 0\ for\ T<T_S \end{Bmatrix} \tag{6}$$

where a parameter known as liquid fraction, ε is introduced to indicate the fraction of liquid in all cells and T_L and T_S is the melting and freezing temperature respectively while L is the latent heat of fusion. The liquid fraction is defined as follows:

$$\varepsilon=\frac{\Delta H}{L} \tag{7}$$

For linear phase change which should occur for NEPCM (non-pure substance):

$\varepsilon = 0$ if $T > T_S$ $\varepsilon = 1$ if $T < T_L$ (8)

For $T_S < T < T_L$, liquid fraction, ε is

$$\varepsilon = \frac{T-T_S}{T_L-T_S} \quad (9)$$

This parameter is calculated for each cell in the domain at each iteration, based on an enthalpy balance. The mushy zone is a region in which the liquid fraction lies between 0 and 1. The mushy zone is modelled as a pseudo porous medium in which the porosity increases from 0 to 1 or decreases from 1 to 0 as the NEPCM liquefies or solidifies. Porosity decreases to zero in the solid region and therefore the velocity in the region will also drop to zero as indicated in the next source term.

3.1.1. Source term S_x and S_y in x and y-momentum equation

$$S_x u = -C\frac{(1-\varepsilon)^2}{\varepsilon^3+b}u \qquad S_y v = -C\frac{(1-\varepsilon)^2}{\varepsilon^3+b}v \quad (10)$$

The condition that all velocities in solid regions are zero is accounted for by defining the source term S_x and S_y. The velocity value is gradually reduced from a finite value in liquid to zero in full solid for solidification process and vice versa for melting over the control volume that are changing phase. The cell is assumed to behave as a porous cell with porosity, ε = liquid fraction. Value of C depends on the morphology of the porous media while b is a small computational constant to avoid division by zero.

3.1.2. Source term S_b in y-momentum equation

$$S_b = (\rho\beta)_{nf} g\left(h - h_{ref}\right) \quad (11)$$

Since the phenomena is natural convection in the liquid region, buoyancy is represented by Boussinesq Approximation.

3.1.3. Source term S_h in energy equation

$$S_h = \frac{\partial[\rho\Delta H]}{\partial t} + \frac{\partial[\rho\Delta H]}{\partial x} + \frac{\partial[\rho\Delta H]}{\partial y} \quad (12)$$

The latent heat source term is the rate of change of volumetric latent heat. This source term must be accompanied by positive sign for melting and a negative sign for solidification.

3.2. Enthalpy Based Lattice Boltzmann Method (EB-LBM)

This method uses enthalpy approach to model phase change while LBM is used to solve the equations. LBM predicts the evolution of particles displacement due to collision and streaming in mesoscale point of view which is represented by distribution functions. Particles behavior are pre-averaged or only particle distributions that live on the lattice nodes are traced, rather than all the individual particles. Distribution functions are used to calculate the macroscopic variables by taking moment to the distribution function thus satisfying macroscopic equations or conservation equations. Details on the theory for this method is available [9, 32].

The Boltzmann equation discretized in space and time is given as follows:

$$f_i(x + c_i\Delta t, t + \Delta t) - f_i(x,t) = -\frac{f_i - {f_i}^{eq}}{\tau_f} + F_i \quad (13)$$

$$g_i(x + c_i\Delta t, t + \Delta t) - g_i(x,t) = -\frac{g_i - {g_i}^{eq}}{\tau_g} \quad (14)$$

where distribution function f is used to calculate density and velocity fields while distribution function g is used to calculate temperature field thus the term Double Distribution Function (DDF). The right side of equation is collision term represented by Bhatnagar-Gross-Krook (BGK) approximation. *F* is the external force field and τ is the relaxation time for each equation. The equilibrium distribution functions are defined so that they satisfy macroscopic equation (Navier-Stokes equations) through Chapman-Enskog expansion:

$${f_i}^{eq} = \omega_i\rho(1 + 3(c_i.u)/c^2 + 4.5(c_i.u)^2/c^4 - 1.5u^2/c^2) \quad (15)$$

$${g_i}^{eq} = \omega_i\rho(1 + 3c_i.u + 4.5(c_i.u)^2 - 1.5u^2) \quad (16)$$

The values of weight ω_i is decided as per the chosen lattice model. As example, some researchers used D2Q9 model (refer Figure 1) for their models therefore $\omega_1 = \frac{4}{9}, \omega_{2,3,4,5} = \frac{1}{9}, \omega_{6,7,8,9} = \frac{1}{36}$

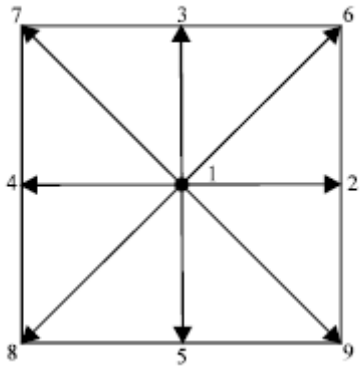

Figure 1. D2Q9 LBM Model

Macroscopic variables such as density, velocity and temperature are calculated by taking moment to the distribution functions:

$$\rho(x,t) = \sum_i f_i(x,t)\ , \rho u(x,t) = \sum_i c_i f_i(x,t)\ , \rho T(x,t) = \sum_i c_i g_i(x,t) \quad (17)$$

The time relaxation and effective viscosity are as follows:

$$\upsilon = \frac{1}{3}\left(\tau_\upsilon - \frac{1}{2}\right) \quad (18)$$

To solve phase change problem, the similar principle applied for enthalpy formulation is applied at mesoscale level.

4. Conclusion

The factors influencing the numerical simulations results for Nano-Enhanced Phase Change Materials (NEPCM) solidification and melting have been summarized in this review. The degree of nanoparticle influence to the NEPCM mostly depend on its fraction. Higher fraction of its presence will enhance

the thermal conductivity of the whole mixture thus improving solidification/melting process. This fact has been proven again and again while remained as the most investigated factor. Although it is agreed that using proper concentration of nanoparticle will improve performance of NEPCM during solidification and melting, care must be taken as not to diminish the convective effect in liquid region when the viscosity is too high. Another factor which have received less attention is nanoparticle size. Research conducted reported that with the same nanoparticle fraction/concentration, the performance can be further enhanced if a smaller nanoparticle size is utilized. This factor does not only improve the performance but also affects the physical appearance of the final result. The authors would recommend more investigation to be done to study the effect of nanoparticle size and shape on NEPCM phase change process. Such studies should be conducted by numerical simulations as the diameter can be modified easily with relatively minimum effort. Other factors affecting the NEPCM performance such as boundary conditions and geometry of container can also be easily modified in numerical simulations with minimum effort compared to experimental works.

Numerical models for simulation of solidification and melting process of NEPCM have been established mainly by applying enthalpy-porosity formulation coupled with finite volume method. Most models solved a single macroscale domain at each iteration by assuming NEPCM as a single-phase substance which resulted in no attempt to model it as a two-phase substance which requires a relatively more complex multidomain approach. Such dilemma is avoided when mesoscale methods are applied because the value predictions are based on evolution of particles displacement due to collision and streaming at a different smaller scale. The authors recommend that further investigation is done on mesoscale methods application in NEPCM phase change process simulations.

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