Numerical Analysis of Coalescence Characteristics of Low Melting Point Alloy Fillers Using a Non-Equilibrium Phase Field Model

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A non-equilibrium phase field model (NPFM) is proposed to examine the three-dimensional coalescence characteristics of low melting point alloy (LMPA) fillers, widely used for self-organized interconnection applications. This model sufficiently considers the non-equilibrium process during phase transitions, resulting in accurate prediction of interfaces between two different phases with a high density ratio. The preliminary simulation is carried out for validation of the present model and numerical predictions are in good agreement with analytical solutions for a one-dimensional solidification problem. The proposed NPFM successfully predicts the phase transition, the heat transfer from solid to liquid, and the coalescence behaviors of LMPA fillers, whereas the conventional enthalpy method fails to describe the non-equilibrium phase transition. Moreover, the results show that there exists grid dependency in the mush zone, suggesting that special care should be taken of the mush zone for better prediction of interfaces between different phases. [doi:10.2320/matertrans.MF200915]

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1. Introduction

In recent years, much attention has been paid to eco-friendly alternative bonding material to conventional lead-tin solders being of toxicity and environmental hazard for the self-organized interconnection using anisotropic conductive adhesive (ACA) with low melting point alloy (LMPA) fillers.¹⁻³ In a self-organized interconnection procedure, as shown in Fig. 1, conductive fillers are initially in solid phase but during the reflow (thermal and compression) process, they become molten and coalesced with adjacent fillers, and finally some of larger fillers make conduction paths, resulting in forming interconnection. For designing the optimal ACA process, we should understand detailed physics behind phase change and coalescence characteristics. In fact, because of limitations in time and space for current measurement, computational modeling offers a promising way to get the detailed information on transient behavior of phase change and the corresponding coalescence among fillers. Unfortunately, few studies have been reported for simulating the coalescence of LMPA fillers for interconnection process.

For a simulation of conduction path formation in the self-organized interconnection of ACA, the two phase flow including molten fillers and resin should be first studied. Figure 2 illustrates the formation of different interfaces during phase transition. For two-phase flows, a time evolution of distinct interfaces between two different materials seems difficult to be described because of complexity of related phenomena. In this case, the surface tension becomes more dominant than the gravity force. The analysis of the mush zone at the phase boundary affects the mass conservation. Lee et al.⁴ performed a two-dimensional simulation on the self-organized interconnection using ACA with LMPA fillers and analyzed wetting and coalescence characteristics during the process as seen in Fig. 3.

However, their results were obtained for non-thermal single liquid phase with a high density ratio.

Generally, the surface tension makes pressure and velocity discontinuous and thus treating the phase boundary of the flows in the case of high density changes is important. The present study adopts the cubic interpolated propagation (CIP) method⁵⁻¹⁰ using a semi-Lagrangian method for a multi-phase flow problem. This method consists of the advection part. Welland et al.¹¹ applied the phase field function to a two phase conduction problem considering the phase change with the treatment of the numerical mush zone. Shyy et al.¹² used the phase field value as a function of temperature using the enthalpy method. In addition, Takada et al.¹³ investigated the coalescence of single liquid drop into a liquid film in a gas under gravity with a high density ratio in three dimensions.
As a matter of fact, the present study can be regarded as the first step towards developing a computational model for the self-organized interconnection process. The non-equilibrium phase field model (NPFM) is proposed in the present study for analyzing the three-dimensional coalescence characteristics including phase transition. Unlike the previous works, the thermal-induced phase change is simulated with the use of the CIP method and the implicit phase field method. The numerical solutions using the proposed method are validated by being compared with the analytic solutions for the one-dimensional solidification problem. In particular, the present study compares the NPFM with the conventional enthalpy method which has been widely utilized in many commercial codes, and discusses the difference between two models. The numerical results for the phase change from solid to liquid and subsequent coalescence of two different LMPA fillers are explained in detail.

2. Theoretical Approach and Mathematical Representation

2.1 Non-equilibrium phase field method (NPFM)

Typically, the molten process of solid fillers undergoes clearly non-equilibrium phase change for a very short time. However, the conventional enthalpy method assumes that phase change takes place under the thermodynamic equilibrium. The present study utilizes the non-equilibrium phase field method (NPFM) associated with the Gibbs free energy for phase transition. This study solves the mass, momentum (including pressure), and energy conservation equations by using the C-CUP (CIP-Combined Unified Procedure) method.

\[
\frac{\partial \rho}{\partial t} + u \cdot \nabla \rho + \rho \nabla \cdot u = 0
\]  
\[
\frac{\partial u}{\partial t} + u \cdot \nabla u = - \nabla p + \nabla f + \frac{\nabla \cdot k}{\rho} \nabla T
\]

where \( p \) represents the density, \( u \) is the velocity, and \( p \) is the pressure. \( g, \sigma, \kappa, \) and \( c_i \) indicate the gravitational acceleration, the surface tension coefficient, the curvature, and the speed of sound, respectively. The symbol \( \alpha \) is defined as the phase-field parameter which indicates the diffusive interface profile in the phase-field method and its range is \( 0 < \alpha < 1 \). If the value of \( \alpha \) is set to unity, the cell is composed of \( \alpha \) phase. In the present study, the smoothed phase-field parameter \( f \) is also used to avoid the singularity appearing at the interface. \( k \) and \( C_p \) are the thermal conductivity and the specific heat, respectively. In the mush zone, the value of the sound of speed \( c_i \) inside the mush zone is interpolated linearly between constants of solid fillers and liquid fillers. The same interpolation rule is applied to the viscosity, \( \mu \), the thermal conductivity, \( k \), and the specific heat, \( C_p \). The force term \( Q_{\alpha} \) represents the viscous force. The present model calculates the surface tension by using the method of continuum surface tension by Brackbill.

\[
\frac{\partial q}{\partial t} + u \cdot \nabla q = M_q \left[ \frac{1}{T} \left( \frac{\partial q(\alpha)}{\partial \alpha} \Delta G_{\alpha\gamma} + \frac{\partial K_{m}(\alpha)}{\partial \alpha} \right) - \epsilon^2 \nabla^2 q \right]
\]

where \( \Delta G_{\alpha\gamma} \) is the difference of Gibbs free energy \( g_{\alpha} - g_\gamma \), and the smoothing function \( q(\alpha) \) and \( K_{ma} \) are obtained as follows:

\[
q(\alpha) = \alpha^3(6\alpha^2 - 15\alpha + 10)
\]

\[
K_{ma}(\alpha) = \frac{W_o}{\partial(1 - \alpha^2)}
\]

The function \( K_{ma} \) describes the excess surface energy of the materials on the boundary between solid and liquid. The constants, \( W_o, \varepsilon_a, \) and \( M_a \), are determined from

\[
W_o = 6\sigma/\delta
\]

\[
\varepsilon_a = 12\sigma/\delta
\]

\[
M_a = \frac{k}{\delta} \left( \frac{T_{sat}}{\delta H_{sat}} \right)^2
\]

where \( \delta \) is the mush zone thickness, \( T_{sat} \) is the saturation temperature, \( \Delta H_{sat} \) is the latent heat, representing enthalpy difference between \( \alpha \) and \( \gamma \) phases.

2.2 Numerical algorithm

The governing equations are divided into the convection term and the non-convection term. The equations presenting the convective term are solved by using the CIP method as follows:

\[
\frac{\partial \rho}{\partial t} + u \cdot \nabla \rho = -\rho \nabla \cdot u
\]

\[
\frac{\partial T}{\partial t} + u \cdot \nabla T = -\nabla \cdot \left( \frac{k}{ho C_p} \nabla T \right)
\]

\[
-\left( \frac{\partial q(\alpha)}{\partial \alpha} \right) \frac{\Delta H_{\alpha\gamma}}{\rho C_p} + \frac{K_{ma}(\alpha)}{\rho C_p} \alpha
\]

where \( \rho \) represents the density, \( u \) is the velocity, and \( p \) is the pressure. \( g, \sigma, \kappa, \) and \( c_i \) indicate the gravitational acceleration, the surface tension coefficient, the curvature, and the speed of sound, respectively. The symbol \( \alpha \) is defined as the phase-field parameter which indicates the diffusive interface profile in the phase-field method and its range is \( 0 < \alpha < 1 \). If the value of \( \alpha \) is set to unity, the cell is composed of \( \alpha \) phase. In the present study, the smoothed phase-field parameter \( f \) is also used to avoid the singularity appearing at the interface. \( k \) and \( C_p \) are the thermal conductivity and the specific heat, respectively. In the mush zone, the value of the sound of speed \( c_i \) inside the mush zone is interpolated linearly between constants of solid fillers and liquid fillers. The same interpolation rule is applied to the viscosity, \( \mu \), the thermal conductivity, \( k \), and the specific heat, \( C_p \). The force term \( Q_{\alpha} \) represents the viscous force. The present model calculates the surface tension by using the method of continuum surface tension by Brackbill.

\[
\frac{\partial q}{\partial t} + u \cdot \nabla q = M_q \left[ \frac{1}{T} \left( \frac{\partial q(\alpha)}{\partial \alpha} \Delta G_{\alpha\gamma} + \frac{\partial K_{m}(\alpha)}{\partial \alpha} \right) - \epsilon^2 \nabla^2 q \right]
\]

Fig. 3 Two-dimensional coalescence and wetting characteristics for an only liquid phase with the high density ratio.
where $\rho^*$, $u^*$, $p^*$, $T^*$, and $\alpha^*$ indicate the predicted values from the CIP method, whereas $\rho^n$, $u^n$, $p^n$, $T^n$, and $\alpha^n$ mean the old values obtained at the previous time. The time step $\Delta t$ is $t^{n+1} - t^n$. On the other hand, the non-advection term can be described as

$$\frac{\rho^{n+1} - \rho^n}{\Delta t} = -\rho^* \nabla \cdot u^{n+1}$$

$$\frac{u^{n+1} - u^n}{\Delta t} = -\nabla p^{n+1} + Q^*_u + g + \sigma \alpha^* \frac{\nabla f^*}{\rho^*}$$

$$\frac{p^{n+1} - p^n}{\Delta t} = -\rho^* c_s^2 \nabla \cdot u^{n+1}$$

$$\frac{T^{n+1} - T^n}{\Delta t} = -T^* \nabla \cdot u^{n+1} + \frac{1}{\rho^*} \nabla \cdot \left( k \frac{\nabla T^{n+1}}{\rho_c C_p} \right) - \left( \frac{\partial q(\alpha^{n+1})}{\partial \alpha} \frac{\Delta H_{av}}{\rho_c C_p} + \frac{K_{sw}(\alpha^{n+1})}{\rho_c C_p} \right) \alpha^{n+1}$$

$$\frac{\alpha^{n+1} - \alpha^n}{\Delta t} = -\alpha^* \nabla \cdot u^{n+1} - M_a \left[ \frac{1}{T^{n+1}} \left( \frac{\partial q(\alpha^*)}{\partial \alpha} \Delta G_{av} + \frac{\partial K_{sw}(\alpha^*)}{\partial \alpha} \right) - \alpha^* \frac{1}{\sigma} \nabla \cdot \alpha^{n+1} \right]$$

By taking divergence of eq. (17) and substituting it into eq. (18), we obtain consequently the pressure equation as follows:

$$\nabla \cdot \left( \frac{1}{\rho^*} \nabla \delta p \right) = \frac{1}{\rho^* c_s^2 \Delta t} \delta p + \frac{1}{\Delta t} \nabla \cdot u^{n*}$$

$$\frac{u^{n*} - u^n}{\Delta t} = -\nabla p^n + Q^*_u + g + \sigma \alpha^n \frac{\nabla f^n}{\rho^n}$$

$$\delta p = p^{n+1} - p^n$$

3. Results and Discussion

3.1 Analysis for one-dimensional solidification

In order to validate the proposed model, the present study solves the well-known one-dimensional solidification problem for water as shown in Fig. 4. In this problem, water is changed to ice and their interface moves along the direction of solidification with the phase boundary. The mesh is a structured uniform grid. The grid system consists of $5 \times 5 \times 50$ and the minimum mesh size is $0.02 \text{ m}$. The time step of the simulation is $0.01 \text{ s}$. Because the exact solution can be obtained from the equilibrium assumption, the one-dimen-

Fig. 4 The schematic of the one-dimensional solidification problem.
3.2 Phase transition and coalescence characteristics of alloy fillers

In the real circumstance, solid fillers undergoing phase changes are in the non-equilibrium state and when the melting process is completed, larger liquid drops are yielded from the coalescence process. Due to the complicated phase change process, most of researchers did not directly include the thermal-induced phase change process in their simulation, and instead assumed that solid fillers have been already molten. Consequently, the liquid phase has been only treated in their simulation. Unlike the previous works, the present study concentrates mainly on the coalescence process and the phase change of LMPA fillers.

Figure 6 illustrates the schematic of the coalescence simulation and shows that two solid fillers are initially located in the liquid resin. Using the structured uniform grid, the computational grid system consists of \(20 \times 20 \times 20\) with the minimum mesh size of 5 \(\mu\)m. The time step of simulation is taken as \(1 \times 10^{-9}\) s. Symmetric boundary conditions are applied for the momentum conservation equation. Constant temperature conditions are used at two boundaries in the \(x\) direction, and their temperatures equally are 535 K, which is higher than the melting temperature of 506.2 K. In addition, the adiabatic condition is used for other boundaries. The physical properties required are listed in Tables 1 through 3.

Figure 7 depicts the normalized mass predicted by the enthalpy method with respect to time during the phase change. The normalized mass can be determined by the normalization of the mass of each phase with the total mass. When the mass conservation is satisfied, the normalized mass of sum of both phases should be equal to unity. The solid phase does not completely change to the liquid phase and the mass fractions of two phases are nearly equal, which indicates such a compulsive equilibrium state. Note that the phase equilibrium assumption in the enthalpy method violates the real existence of the non-equilibrium state during the phase change.

Figure 8 shows the transient behaviors of two different fillers undergoing the phase transition. An initial solid state begins to change toward mixed phase state, indicating that liquid and solid phases coexist as time goes on. The melting process is completed after 1.2 \(\mu\)s, when two different liquid phases of molten fillers and resin simultaneously exist with the high density ratio.

A mush zone is defined to be an artificial region having mixed different phases. Note that the thickness of the mush zone may be an important factor to obtain precise solutions of multiphase problems of interest. Figures 9–11 show the variation of the predicted normalized mass during the phase change for different mush zone thicknesses, i.e., 0.5, 1.0,
and 1.5 times of unit mesh size, respectively. Compared to the enthalpy method, the NPFM can predict the complete phase change from solid to liquid. The enthalpy method with the equilibrium assumption determines the phase field value from only the temperature field, which excludes the diffusion term \( \frac{M e}{C_1} \), whereas the NPFM includes the diffusion effect as well as the surface tension effect at the phase boundary by estimating the rate of change in \( \frac{M e}{C_1} \) by using eqs. (5) and (20). Thus, the enthalpy method would be inappropriate for the simulation of the self-organized interconnection process where the transient phase change becomes substantially important, in which the non-equilibrium state should be considered for these kinds of calculations. From the results, the times to reach the phase equilibrium are estimated 0.16, 0.42, and 0.84 \( \mu s \) for the mush zone thicknesses of 0.5, 1.0 and 1.5 times the unit mesh size, respectively. For the mush zone thickness equal to 1.0 and 1.5 times the unit mesh size, the ratios of increase in final filler mass to initial filler mass are estimated 37.8% and 33.7%, respectively, compared to 2.75% for the case of 0.5 times the mesh size. These results indicate the normalized mass of molten fillers greater than unity. These unreasonable results obtained show that the predictions of the phase change highly depends on the numerical mush zone thickness.

From the sensitivity analysis with respect to the mush zone thickness, the mass conservation is violated, while the normalized mass must be unity in the ideal case. However, it turns out that the results from the NPFM simulation are not unity in cases of the mush zone thicknesses of 1.0 and 1.5 times the unit mesh size. This is due to the basic assumption related to the derivation of \( W e, e_d, \) and \( M e \). The constants, \( W e, e_d, \) and \( M e \), related to the mush zone thickness in eq. (5), were derived by the assumption of sharp interface. By using the sharp interface assumption, the left part of eq. (25) is ignored and the constants are derived.
\[
\frac{1}{M_a} \left( \frac{\partial \alpha}{\partial t} + \mathbf{u} \cdot \nabla \alpha + \alpha \nabla \cdot \mathbf{u} \right)
= -\left[ \frac{1}{F} \left( \frac{\partial p(\alpha)}{\partial \alpha} \Delta G_{\alpha r} + \frac{\partial K_{\text{wall}}(\alpha)}{\partial \alpha} \right) - \sigma_\alpha^2 \nabla^2 \alpha \right]
\]

(25)

However, if the \( \frac{1}{M_a} \approx \delta^2 \) is big, the left part of eq. (25) cannot be neglected and this may result in dissatisfaction of the conservation. To solve this problem, as the future work, the material derivative term of the phase field parameter in eq. (25) should be included and treated carefully for finding the optimal solution for mush zone thickness.

4. Conclusions

This article describes the simulation of coalescence and phase transition characteristics of low melting point alloy (LMPA) fillers by using non-equilibrium phase field method with the use of the Navier-Stokes equation. This work has great meaning as the first step towards simulating three-dimensional self-organized interconnection process including the ACA process. The conclusions are summarized as follows: First, the proposed non-equilibrium phase field model (NPFM) successfully predicted the thermal-induced phase change from solid to liquid, whereas the enthalpy method failed to describe the non-equilibrium phase change. The proposed model was validated for the one-dimensional solidification problem, and the corresponding predictions were in quite good agreement with the analytic solutions. It turns out that the enthalpy method would be inappropriate for the simulation of the self-organized interconnection process where transient phase change becomes substantially important. Note that the nonequilibrium state during phase changes should be necessarily considered for calculations. Finally, the presence of the grid dependency on the mush zone indicates that the mush zone treatments in capturing interfaces between two different phases should be carefully made for better predictions.

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