Modelling of intermetallic layers formation during solid-liquid joining of dissimilar metallic materials

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Abstract. Joining of dissimilar metallic materials gives rise to the production of components with improved properties. At the joint interface, the formation of brittle intermetallic layers might deteriorate the component application. Therefore, its formation and growth must be well understood and carefully controlled. Numerical simulations assist in the determination of processes parameters to obtain the desired bond characteristics. In solid-liquid joining processes, the melt movement plays an important role and must be accounted in the simulations. In this paper, a mathematical model at the continuum scale is used for the description of the interface formation during solid-liquid joining. The description of convection and advection of the molten alloy and the dissolution of the solid substrate are of interest. The substrate dissolution into the melt alters the local composition of the molten alloy, leading to precipitation of intermetallic compounds on the substrate surface during the solidification process. To account for these phenomena, convection heat transfer with phase transformation and diffusion and reaction at the solid-liquid interface are considered in the mathematical model that is based on conservation equations of mass, momentum, energy and species. The different scale problem is solved by applying a dynamic mesh refinement at the interface to accurately solve the intermetallic layer growth. The model was implemented in OpenFoam® and simulation results for the intermetallic layer thickness as a function of solid substrate temperature ($T_s$) for compound casting between initially liquid Al and a solid Cu substrate are presented. In general, for $T_s$ initial value ranging from 400 to 700 °C while keeping the initial Al melt temperate at 750 °C, the intermetallic layers thickness increases as $T_s$ increases.

1. Introduction

The use of materials with improved properties is essential to attend the technological and industrial advances. However, the material may not have the desired properties to perform a specific function on its own. Therefore, the possibility of joining different materials to obtain a component with better mechanical, physical and chemical properties can overcome this problem. Depending on the required application of the component, the combination of different materials can be achieved by different techniques, such as coating, welding or casting, among others. The interest of this paper is on the joining of dissimilar materials by compound casting. In general, it consists of liquid metal being cast onto a solid metallic substrate. The high temperatures promote the dissolution of the solid substrate into the
melt that starts to solidify. Under the right process parameters, a controlled dissolution of the solid material into the melt occurs and a sound metallurgical bond is achieved. However, if the materials are not completely soluble in each other, intermetallic compound layers (ICL) may form in between, weakening the bond and, consequently, the performance of the component. Over the years, the compound casting has become sophisticated and new challenges have been faced, such as its application in continuous casting [1, 2], which makes this technique economically competitive.

Much effort is put into determining the correlation between process parameters and formation and growth of ICL so that an optimum metallurgical bond for a given component can always be achieved. In most cases, this optimization is done via several trial-and-error experimental investigations and researches focus on the characterization of the interface, while its formation mechanism is still controversial. To overcome the necessity of expensive experimental work and to help to understand the interface formation, numerical simulations are performed. Nevertheless, numerical techniques have also been improved over the years to better describe the physical phenomena and to help to define the optimal process parameters for general metallurgical processes. One of the applications of numerical modelling is to help understand the behaviour that is not yet fully understood through an analysis of experiments, such as the formation mechanism of ICL during solid-liquid joining, as occurs in compound casting. Among the physical phenomena associated with compound casting, convection heat transfer with phase transformation and diffusion-reaction at the interface between materials are highlighted, which makes a comprehensive modelling of this complex phenomena challenging.

The modelling of compound casting resembles the modelling of solidification problems with the addition of interfacial diffusion and reaction effects. The necessity of realistic models applicable to high scale processes is therefore of interest. Depending on the degree of detail that is desired, the problem can be studied in different scales. In order to understand the application of different scales for the study of solidification, the reader is referred to the work of Stefanescu [3]. In this article, we focus on the formation and growth of ICL and how it affects and is affected by the solidification process during the compound casting.

In the literature, dissimilar joint problems are approached in different scales, such as molecular simulations [4] or mesoscale simulations, where usually phase-field techniques [5–7] or lattice-Boltzmann simulations [8] are applied. These methods, however, lack in providing the correlation between process parameter and the ICL in the compound casting framework due to the usual very small numerical domain. Thus, its applicability to industrial scale is impractical. In a trial to overcome these problems, we present a continuum approach for the compound casting with the prediction of thickness and composition of the ICL. For instance, the conventional conservation equations are applied and a local dynamic mesh refinement at the interface is used to properly solve the diffusion and reaction problem.

The modelling of formation and growth of intermetallic phases at the solid-liquid interface during solidification based on a continuum-scale requires a deep understanding of the relevant physical mechanisms in order to be described into model equations. Different methods must be combined to obtain a comprehensive model for ICL formation and growth. These methods involve thermodynamic modelling, reaction kinetics, heat transfer and fluid flow. This multifaceted approach is presented in this paper within the mathematical description. The current model is, however, a simplified approach to assist in the determination of input parameters, such as pouring temperature and preheating of the substrate, for the compound casting process. Figure 1 schematically shows the problem address in this paper. The solid-liquid diffusion and reaction are coupled with the enthalpy-porosity method to simulate the compound casting process. For this, the complex phenomena observed in the process are translated into a simplified mathematical model that is based on conservation equations of mass, momentum, energy, and species. The model description is an adaption from [9].

2. Mathematical model
The compound casting is physically understood as a solidification process with diffusion and reaction taking place at the interface between both base materials. For the modelling of the solidification process,
the enthalpy-porosity method was used to track the solid-liquid interface [10]. To track the formation and growth of the ICL, a source term is added to the species transport equation. The equations are presented under the assumption of laminar Newtonian fluid flow in Cartesian coordinates. To comprehension of the model, the solidification modelling and the formation and growth of ICL modelling are discussed separately, as follows.

![Figure 1. Schematic compound casting problem. Substrate dissolution at the moment melt wets the solid surface and diffusion and reaction are considered under the influence of heat transfer and fluid flow.](image)

2.1. Solidification modelling
In the enthalpy-porosity formulation, it is considered that the solidified structures adjacent to the solid-liquid interface form a mushy zone that is represented as a porous medium, where the porosity of the medium is given by the liquid fraction ($f_l$) and it decreases as solidification progresses. The governing equations are the conservation of mass, momentum and energy, respectively given by [11]:

\[
\frac{\partial (\rho)}{\partial t} + \nabla \cdot (\rho \mathbf{u}) = 0
\]  
\[
\frac{\partial (\rho \mathbf{u})}{\partial t} + \nabla \cdot (\rho \mathbf{u} \mathbf{u}) = -\nabla p + \nabla \cdot (\mu \nabla \mathbf{u}) + \mathbf{b} + S_M
\]  
\[
\frac{\partial (\rho h)}{\partial t} + \nabla \cdot (h \rho \mathbf{u}) = \nabla \cdot (k \nabla T) + S_H
\]

where $\rho$ is the density, $\mathbf{u}$ is the velocity field, $\mu$ is the viscosity, $h$ is the enthalpy, $T$ is the temperature and $S_M$ and $S_H$ are source terms that include the effects of solidification in the model. The natural convection effect is accounted for in the body force term, $\mathbf{b}$, in which the Oberbeck-Boussinesq approximation is applied [12], given by:

\[
\mathbf{b} = \rho g \left[ f - \beta (T - T_{ref}) \right]
\]

where $g$ is the gravity vector, $\beta$ is the thermal expansion coefficient and $T_{ref}$ is the reference temperature.

$S_M$ is added in the momentum equation, equation (2), to account for the fluid velocity drop due to the decrease of liquid fraction during the evolution of the mushy zone and it is given by:

\[
S_M = \frac{\mu}{K} u
\]

where $K$ is the permeability of the porous media, formulated here from the Karman-Cozeny equations, as follows:

\[
K = B \frac{(1-f_s)^3}{f_s^2 + \varepsilon}
\]
where $B$ is a constant that describes the morphology of the porous medium, $\varepsilon$ is a small number to avoid division by zero and $f_\varepsilon$ is the solid fraction, related with $f_i$ by equation (7). It can be seen from equations (5) and (6) that $S_M$ is modelled so that tends to zero in the liquid region ($f_s = 0$), while becoming dominant in the mushy zone ($0 < f_s < 1$), in such a way that the momentum equation approaches D’Arcy’s law, which is the equation that governs fluid flow in porous media. Because of its function, $S_M$ is also called D’Arcy source term [13].

$$f_s = 1 - f_i$$

(7)

In the energy equation, conveniently written in the enthalpy form [14], the added source term $S_H$, represents the release of latent heat during solidification, given by:

$$S_H = \frac{\partial(Lp)}{\partial t} + \nabla (L\rho u f_i)$$

(8)

where $L$ is the latent heat.

To determine the evolution of the mushy zone the solid fraction must be calculated. In the current model, the solid fraction is determined with the help of the software Pandat, using Scheil-Gulliver calculations, as described in [15]. The results depend on the local temperature and solute distribution in the mushy zone. The composition distribution near the interface is highly affected by the dissolution of the substrate material into the melt and must be considered. Therefore, the redistribution of the chemical solution near the interface is accounted in the model of ICL formation and growth, described below, while details for the enthalpy-porosity method can be found in [10] and [14].

2.2. Intermetallic compound layer formation and growth modelling

The chemical composition is a compulsory quantity to predict not just the solid fraction required in the solidification model, but it is also an input parameter to determine if a reaction might take place for the formation of a new compound. The local mass fraction of each species, $Y_i$, evolved in the system is predicted within the conservation of species equation [11], given by:

$$\frac{\partial (\rho Y_i)}{\partial t} + \nabla (\rho u Y_i) = -\nabla (J_i) + R_i$$

(9)

where $J_i$ is the diffusion flux of species $i$, and $R_i$ is the reacting source term. In a binary diffusion system, $J_i$ can be related to the species concentration using Fick’s first law [16]. However, for a multi-component mixture, the Maxwell-Steefen equations are recommended [11]. Regardless, the diffusion flux depends on the diffusion coefficient, $D$. This is an important parameter that describes the diffusivity of one species into another and it is usually determined via experimental analyses. It is important to note that the summation of equation (9) over all the $i$ elements recovers equation (1), which in the case is the equation of continuity for the mixture [11].

The formation and growth of a compound are considered in equation (9) in the reaction source term $R_i$, which for a non-reversible reaction is given by [17]:

$$R_i = M_{w,i} \sum_{r=1}^{N_R} (v_{i,r}^i \cdot v_{i,r}^n) k_{fr} \prod_{j=1}^{N_R} (C_{j,i})^{\delta_{ij}}$$

(10)

where $M_{w,i}$ is the molecular weight of species $i$, $N_R$ is the number of reactions that the species participate in, $v_{i,r}^i, v_{i,r}^n$, are the stoichiometric coefficients for the reactant and the product of the $i$ specie, $M_i$, in the reaction $r$, as generally given by equation (12) [17]. $k_{fr}$ is the forward reaction rate constant, computed using the Arrhenius expression, given by equation (11).

$$k_{fr} = A_i T^{\beta_i} \exp \left( \frac{E_r}{RT} \right)$$

(11)
\[ \sum_{i=1}^{N_k} \nu_{i,r}' M_i \rightarrow_{\beta_r} \sum_{i=1}^{N_k} \nu_{i,r}'' M_i \]  

where \( A_r \) is the pre-exponential factor, \( \beta_r \) is the temperature power exponent, \( E_r \) is the activation energy, and \( R \) is the universal gas constant.

In metallurgy, the phases to precipitate are closely related to the phase diagram of the representative components of the liquid melt and solid substrate, as demonstrated in [18]. As an example, figure 2 shows the Al - Cu binary phase diagram with a diffusion scheme and the specific reactions written in the form of equation (12). In the present model, the precipitation and evolution of each phase is tracked by equation (9).

### 2.3. Coupling between models

Although the ICL modelling and the solidification modelling were presented separately, they are naturally coupled together. The composition information obtained with the ICL model is used as input to determine the solid fraction in the solidification model, while the velocity and temperature fields calculated in the solidification model are used as input in the species transport equation. The interface between materials is coupled considering that at the interface between the solid and the liquid/semi-solid, the temperature must be the same, as well as the heat flow between them, which are usual boundary conditions applied in conjugate heat transfer problems [14, 19]. At the solid state, the diffusion reaction problem is simplified to a one-dimensional problem.

### 3. Numerical implementation

For the numerical simulation, the conservation equations were discretized using the finite volume method. The gradient compilation was done via least squares method. For the approximation of advective terms, the QUICK scheme was applied, while the discretization of diffusion fluxes was done via Gauss method with linear interpolation scheme. A first order implicit scheme was applied for time discretization.

The implementation was done using OpenFOAM® environment, as described in [9]. Due the very small scale needed to solve the ICL model, mesh refinement is applied to achieve greater accuracy at the interface to evaluate the different compound layers. In this manner, the size of the most refined volume at the interface reaches the order of 0.1 µm.

### 4. Model application to Cu-Al compound casting

The model presented in this paper was applied to determine the ICL between Cu and Al. The problem consists of pouring Al melt over a solid Cu substrate. Dissolution from Cu substrate surface into the Al melt and the precipitation reaction of Cu-Al compounds are simulated. In the continuum modelling, the composition profile cannot be calculated, unless all the diffusion coefficients are known as a function of temperature and concentration. For the present work, we consider the diffusion of Al and Cu between each other and within the compounds. The diffusion of Al melt into the solid Cu substrate is neglected.

Another important simplification is regarding the order of phase’s appearance. Following [18], the phases to appear are considered as in the phase diagram, going from the solid substrate in the direction of higher temperatures. Based on these assumptions, the reactions are listed in figure 2(c). They are based on the phase diagram of figure 2(a). Since the \( \delta_1 \) phase is usually not observed in experimental results, [20, 21] it was omitted in the calculation for simplification. Also, adjacent the Al\(_2\)Cu layer, a eutectic layer composed of Al\(_2\)Cu and Al solid solution is usually observed [20]. The model, however, does not consider the eutectic reaction.

For the simulation, diffusion coefficients and activation energies were taken from the literature. The values not available were determined experimentally as described in [9]. The simulation boundary conditions and geometry resembles the experimental work described in [21]. The influence of the initial
temperature of the solid substrate is investigated. The liquid Al initial temperature is kept at 750 °C while the solid substrate temperature varies from 400 to 700 °C.

\[ 9\text{Cu} + 4\text{Al} \rightarrow \text{Al}_4\text{Cu}_9 \]
\[ 11\text{Al} + 4\text{Al}_4\text{Cu}_9 \rightarrow 9\text{Al}_3\text{Cu}_4 \]
\[ 1\text{Al} + 1\text{Al}_3\text{Cu}_4 \rightarrow 4\text{AlCu} \]
\[ \text{Al} + \text{AlCu} \rightarrow \text{Al}_2\text{Cu} \]

(a) Al-Cu binary phase diagram and (b) a diffusion scheme and (c) the specific reactions.

4.1. Results

The phases on the interface are expressed in terms of their volume fraction, \( \varphi_i \). Figure 3 shows the result of \( \varphi_i \)'s profile on the interface obtained for \( T_s = 600 ^\circ \text{C} \). The curves in figure 3 overlap, so the diffusion and reaction model used does not reproduce a clear interface between the precipitated phases. To obtain a quantitative result, the length of a phase was determined considering only the length where this phase has its largest volume fraction, as exemplified in figure 3 for the AlCu phase.

For the reaction order considered in figure 2(c), first \( \text{Al}_4\text{Cu}_9 \) is formed at the interface and it is rapidly consumed for the formation of \( \text{Al}_3\text{Cu}_4 \). It is considered then, that as soon as \( \text{Al}_3\text{Cu}_4 \) forms, it reacts with Al and forms AlCu phase. With the formation of AlCu, the \( \text{Al}_2\text{Cu} \) phase starts to form. This reaction order is a significant simplification to the model. However, it proved to be enough to determine the size of each intermetallic layer.

The model was validated using experimental data from the literature, as shown in table 1. The numerical results are within the value range of the experimental results. This shows that the model
applied, and the interpretation of the results presented here, are quantitively comparable with experimental data. Simulations for initial substrate temperature, $T_s$, higher than the pouring temperature, $T_p$, were unstable and no convergence was achieved. The model is considered successful for $T_s < T_p$.

Table 1. Numerical results for intermetallic layers size obtained for comparison with experimental data from the literature.

| Phase | Phase length [µm] |
|-------|-------------------|
| $T_p = 800 \degree C / T_s = 506\degree C$ | Numerical results (present work) | Experimental [20] |
| $Al_4Cu_9$ | 0.2 | <0.5 |
| $Al_2Cu_4$ | 8.1 | 7.2–8.1 |
| $AlCu$ | 1.7 | 1.8–2.4 |
| $Al_2Cu$ | 6.2 | 5.7–11.3 |

For different substrate preheating temperatures, the calculated size of the intermetallic layers is shown in figure 4. In general, the intermetallic layers thickness increases with increasing $T_s$. The $Al_4Cu_9$ layer is the least present at the interface. Its size barely exceeds 0.2 µm for $T_s = 700 \degree C$. $Al_2Cu$ and $Al_4Cu_9$ layers are of comparable sizes for $T_s = 400, 500 \text{ and } 600 \degree C$. For $T_s = 700 \degree C$ $Al_2Cu$ is significantly larger than $Al_4Cu_9$. It shows how the $Al_4Cu_9$ layer is the most affected by substrate temperature, it increases much faster as the temperature rises compared to the layers of other compounds. These finds corroborate experimental results from the literature [20, 21].

![Figure 4](image_url)

**Figure 4.** Intermetallic layers thickness as a function of the Cu substrate temperature. Initial Al liquid temperature was kept at 750 °C.

5. **Conclusion**

This paper presented a numerical model for compound casting simulation. The model considers the formation of intermetallic layers during the contact between a solid substrate and the melt with subsequent solidification. It was implemented in the OpenFoam environment, adjusting existing solvers.

The continuum model ability to predict the intermetallic layers thickness in solid-liquid joining problems, depends on the quality of the mobility data and the thermodynamic properties available, which makes necessary an adjustment of the model for each system of interest. Results applied for the Al-Cu compound casting problem demonstrates the use of the model.

Improvements in modelling are still needed, especially regarding nucleation of a compound at the interface, so that the order in which the compounds appear in the intermetallic layers is not an input, but
rather a result. This would assist in understanding the reason for the lack of a compound in the ICL, which stills a controversial subject in the literature. The model application into higher-order systems, such as ternary systems, are also necessary to obtain more realistic results.

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