

## ON THE MATHEMATICAL MODELING OF COPPER DISSOLUTION AND IMC GROWTH IN LIQUID SOLDER

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### ABSTRACT

Soldering is the default joining process in the electronic packaging industry. Solder joints are obtained by interaction of the substrate (mostly copper) with the molten solder. The dissolution kinetics of copper and the growth of intermetallic compound (IMC) during soldering process are concurrent processes. The dissolution of the metal from the substrate components is even more critical due to ever increasing miniaturization of the electronic components. The assessment of base metal lost during the process is another important subject being investigated by the researchers in the last two decades. Extensive experimentation is done to quantify the IMC thickness and metal dissolution for various combinations of substrate metal and solders. As a matter of fact these experiments are time consuming and expensive. Moreover, the experimental results have the application to a limited domain. Various models have been suggested by researchers to quantify the substrate thickness lost and IMC growth during soldering. Most of the existing models to estimate the IMC growth and metal dissolution during the soldering process are heavily based on the experimental data. These models do not depict the actual physics of the process. In the present study a review of the modeling techniques has been done and a model has been proposed which is based on the diffusion of and interface kinetics during soldering process. The proposed one-dimensional model has been solved and compared with the experimental data. The present model has a good potential to be extended in two and possibly three dimensions and incorporation of scalloped IMC in order to capture the complete physics of the process.

**KEYWORDS:** Copper Dissolution, IMC Modeling, Interface Kinetics, Lead, Free Solder, Non-Equilibrium Melting, Soldering

### NOMENCLATURE

$r_o$	thickness of substrate (m)	$r$	spatial coordinate normal to substrate (m)
$R_o$	thickness of solder matrix (m)	$V_i$	interface velocity (m/s)
$C$	solute concentration in solder (wt %)	$a$	constant to set the coordinate system
$C_L^i$	solute conc. at the solder side of interface (wt %)	$V^*$	normalized interface velocity
$C_{eq}$	solute solubility in the solder matrix (wt %)	$C^*$	normalized solute concentration
$D_L$	solute diffusivity in liquid solder ( $m^2/s$ )	$r^*$	normalized spatial coordinate
$t$	time (s)	$t^*$	normalized time
<b>Greek Symbols:</b>			
$\mu$	interface reaction kinetics coefficient (m/s wt%)		

### INTRODUCTION

Substrate dissolution and intermetallic compound (IMC) formation during reflow of solder joints are critical issues for joint reliability. Most of studies in the literature aimed at the coarsening and growth of the IMC layer of the

solder joints during service and only limited data is available for the wet phase of soldering process. This is particularly true for lead-free solders, which are still in the developing stage [1-2]. During soldering process, the substrate material reacts with solder and eventually dissolves into the solder matrix. The intense substrate-solder interaction also favors quick depletion of substrate material [3]. Rapid depletion of substrate is contributing to dewetting and early failure of the joint. Dissolution of substrate in lead-free solders is observed to be much higher than lead-bearing solders [4]. As the thickness of substrate is small and limited, the substrate material must be preserved and kept under control during soldering process [3]. Therefore, the loss of copper is of particular importance to the integrity of solder joints and the overall reliability of electronic devices. As a result, extensive research has been performed in the last decade on solder/Cu interface reactions [5-8].

Considering the wet soldering reaction as an isothermal process, as the molten solder comes in contact with the substrate, the substrate material starts to dissolve. Let us assume that the substrate is made up of  $\alpha$  phase and the solder as  $\beta$  phase. It is further assumed that  $\alpha$  phase has a limited solubility at equilibrium in the  $\beta$  matrix. This equilibrium concentration, at a particular temperature, can be calculated from the corresponding binary phase diagram. Due to the soldering reaction, the concentration of the solute ( $\alpha$  phase) increases at the interface. If the original concentration of solute in the  $\beta$  phase matrix is less than the equilibrium concentration at the given temperature,  $\alpha$  phase will have a tendency to diffuse in the bulk solder matrix. Substrate dissolution is assumed to proceed by a sequential mechanism involving two separate processes. The first process involves the crossing of the substrate element through the interface between  $\alpha$  phase and  $\beta$  matrix followed by diffusion of substrate element in the solvent matrix. Each of the two mechanisms consumes time. The slowest mechanism controls the rate of the entire process. The first process is known as interface controlled mechanism and is governed by the interface reaction rate while the second is called the long-range diffusion process. The dissolution process of  $\alpha$  phase is governed both by the interface reaction and long-range diffusion. If the interface reaction mechanism proceeds sufficiently fast with respect to the long-range diffusion, then the concentration of the solute at the interface will be maximal and equal to the equilibrium concentration of the dissolving phase. Therefore, long-range diffusion controlled mechanism implies a local equilibrium at the interface. However, due to the finite rate of reaction at the interface, especially during the initial stage of the dissolution process, the concentration of  $\alpha$  phase at the interface will not be in equilibrium. Consequently, non-equilibrium condition exists at the interface during early stages of dissolution process.

Dissolution of metals or their compounds in the solvent matrix has been an important phenomenon in the metallurgical processing of metals. Researchers long ago have devised various numerical models to understand the process [12-17]. While majority of these models assume an equilibrium state at the moving interface [12-14], some of the models consider non-equilibrium conditions also [15-17]. Almost all of these models deal with the dissolution of metal compounds or precipitates into solid metal matrix.

The focus of the present study is to develop a mathematical model to simulate soldering process and to understand the dissolution behavior of substrate material. Lead-free solders are the much desired industry standard soldering materials in recent and coming years. Almost all the potential candidate materials are high tin-containing solders. In an attempt to setup a more accurate yet simple model for the copper dissolution, we have not considered the formation of intermetallic compound (IMC) at the copper-solder interface. However, the formation of IMC is an important phenomenon observed in such processes. We will incorporate the compound formation in future models. The present study aims at the relative

importance of the interface kinetics reactions and long-range diffusion mechanisms during copper-tin interaction and the subsequent dissolution of copper in Sn matrix during the wet phase of soldering process.

## MODELING HISTORY

The copper-tin interaction and the dissolution of copper in the solder during wet soldering reaction is a complicated process. Due to this complex nature of the reaction process, most of the previous models for the quantitative analysis of the copper dissolution are based on phenomenological notions. These empirical models do not deal with the actual physics involved in the process.

### Arrhenius Model

The most widely cited model in the literature is better known as a power law relation. The model is inherently applied to quantify the products of chemical reaction. The experimental data follow the power law relationship. If  $x$  is the thickness of the copper layer dissolved in the reflow time,  $t$  at a reflow temperature,  $T$ , then the power law can be written as:

$$x(T, t) = Dt^n \quad (1)$$

where  $D$  and  $n$  are the two fitting parameters. Usually,  $D$  is called the diffusion coefficient. Then for each curve, with a given value of  $n$ , one can fit the data with equation (1) to estimate a value for  $D$ , which is now a function of the reflow temperature. The plot of experimentally estimated  $D$  values with respect to the corresponding reflow temperature in a logarithmic scale, exhibits a linear relationship. It is then suggested that the diffusion coefficient  $D$  could be related to the reflow temperature through an Arrhenius type equation as follows:

$$D = D_o e^{\left(\frac{-Q_a}{RT}\right)} \quad (2)$$

where  $D_o$  is a pre-exponential temperature-independent constant,  $Q_a$  the apparent activation energy, and  $R$  the universal gas constant. By fitting the data with Eq. (2), one can estimate two important kinetics parameters,  $D_o$  and  $Q_a$ , for IMC growth and/or copper dissolution in the solder. These empirical models do not specifically deal with the interface kinetics or diffusive mechanisms involved in the dissolution of copper during soldering process.

Chada et al. [24] introduced an enhancement in the Arrhenius model. The analysis was done for the reflow of eutectic Sn-Ag solder on a Cu substrate. The improved method took into account the dissolution and precipitation of the intermetallic phase during the thermal cycle of the reflow process. To take into consideration the effect of non-isothermal behavior they divided the relevant temperature-time profile of reflow cycle into practically small isothermal regions. Kinetic parameters were calculated numerically for each isothermal portion of the cycle. A simultaneous dissolution of the IMC was considered until the time when solder is saturated with Cu. Net growth of the IMC in each time step was taken as the growth minus dissolution prior to solder saturation and growth plus precipitation after the solder was saturated with Cu. The model was primarily based on the power law relationship, and required the experimentally determined data to calculate parameters for the equation.

### Analytical Models Using Mass Balance

The diffusivity parameter appearing in equation (1) is not the real diffusivity of the solder, rather it has been cited

in literature as the apparent diffusivity of solute. In an attempt to crystallize the subject of diffusivity, Ghosh [25] analyzed the coarsening kinetics of  $\text{Ni}_3\text{Sn}_4$  scallops during the interfacial reaction between liquid solders and Cu, Ni, Pd and Ni-Pd substrates. Eutectic Sn-Ag, Sn-Bi and Sn-Pb solders were reflowed over the substrates for various times and at different temperatures. Ghosh suggested the shape and kinetics of the IMC on Ni substrate similar to that of  $\text{Cu}_6\text{Sn}_5$ . A similar analysis by previous researchers [25, 26] was done and the kinetic parameters were estimated. Coarsening kinetics of  $\text{Ni}_3\text{Sn}_4$  scallops was rigorously investigated in this study. It was demonstrated that in the radial growth of scallops the grain boundary diffusion might be playing a dominant role. A relation was presented as follows for connecting various diffusivity values involved in the coarsening process.

$$D_{app} = D_{bk} + \frac{\kappa\delta D_{gb}}{d} \quad (3)$$

where  $D_{app}$ ,  $D_{bk}$  and  $D_{gb}$  are apparent, bulk and grain boundary diffusivity respectively. Grain boundary width,  $\delta$  and a geometric parameter,  $\kappa$  are also involved in the process. With the assumption that the thickening of  $\text{Ni}_3\text{Sn}_4$  scallops is controlled by the grain boundary diffusivity only, the equation was simplified after dropping the first term on the right hand side of equation.

In an attempt to estimate the rate of consumption of copper in a soldering process [27], they analyzed the reflow process of eutectic Sn-Pb solder on Cu substrate. With the assumption that the amount of copper lost from the substrate is the sum of the copper present in the Cu-Sn compound and in the liquid solder, they presented a mass balance equation. When the molten solder gets saturated with Cu, the consumption rate of Cu depends only on the change of volume of Cu-Sn compounds. Kim and Tu demonstrated that the experimental data for Cu consumption closely followed equation the proposed mass balance equation. But their analysis was good for the case where the amount of solder is very small and the results will be valid only after the solder is saturated with Cu.

In another study Kim and Tu [28] investigated the growth of IMC during the reflow reaction of Sn-Pb alloy solder on Cu substrate. The focus of the study was to assess the mechanism involved behind the growth of scallops of  $\text{Cu}_6\text{Sn}_5$  compound. They found, like other researchers, that the scallops of the IMC compound grow larger but fewer with time. The basic mechanisms involved behind the growth were proposed to be the ripening reaction among the scallops and the interfacial reaction between the scallops and the substrate. However, they suggested that the ripening is not a constant volume process since it is also accompanied with the soldering reaction at the Cu/IMC interface. The ripening flux was calculated following the Gibbs-Thomson effect and the Cu concentration in the molten solder at the surface of the IMC grain was suggested following an exponential equation. Another flux, contributing to the growth of the IMC phase, called the interfacial reaction flux was calculated with the assumption that the total Cu lost from substrate was equal to the total amount of Cu dissolved through the open channels, and was given through an equation. By making use of the Gauss' theorem, the two fluxes were combined together to give a final relation for the growth of IMC scallops with time. One term in the final equation represented ripening and the other term pertained to the interfacial reaction. The analysis is good only when the solder is saturated with solute.

### Fick's Law

The assumption of a constant mean radius of the IMC scallop as given in [28] was relaxed in another study by Gusak and Tu [29] in 2002. In this analysis of the growth of IMC scallops with time, they considered a bell-shaped size

distribution of the spheroids. The total flux of Cu contributing to the growth of these scallops was again considered to be the sum of the ripening and interfacial reaction. A mathematically tedious analytical solution was presented and it was concluded that the ripening process in the IMC growth is a flux driven ripening (FDR) process. However, the analysis was limited to the condition when the solder is saturated with solute. The application of the study was stated to be marginal or nil prior to the saturation stage. For the dissolution of the solute in the molten solder, a relation by Dybkov [30] is widely cited in literature. The equation is given as follows:

$$\frac{dc}{dt} = k^* \frac{S}{V} (c_s - c) \quad (4)$$

where  $c_s$  is solute solubility in liquid solder at the reflow temperature,  $c$  is concentration of solute in the solder at any time,  $t$ ,  $k^*$  is dissolution rate constant,  $S$  is the surface area of the intermetallic compound in contact with the solder and  $V$  is volume of the liquid solder. It is important to note that the dissolution rate constant,  $k^*$  is an experimentally determined parameter and represents the overall rate of dissolution and does not particularly reflect any reaction kinetics at the dissolving interface. It can easily be demonstrated that equation (4) follows immediately from Fick's equations on the assumption of a quasi-stationary distribution of the concentration of solute components within the diffusion boundary layer. This means that the solute distribution in the diffusion boundary layer is close to linear. However, this assumption is not always true especially at the beginning of the dissolution process and one should use the Fick's law of diffusion to address the problem in a concise manner.

### Combined Thermodynamic and Kinetic Model

The modeling attempt to simulate the IMC growth through combination of thermodynamics and kinetics is probably the most accurate approach to deal with the complicated physics of the process. Through this approach one can capture the phase stability, growth rate of reaction products and driving force for the chemical reactions. The one dimensional combined thermodynamic – kinetic model was suggested by Ronka et al. [31] and Kivilahti [32] with the assumption of IMC growth as purely a diffusion controlled process. However, the disadvantage of such approach is that the model is still dependent of the experimentally determined growth kinetic parameter for a particular material system. A very extensive database for the lead-free system is needed in order to model the problem with this approach.

### Phase Field Model

Phase field modeling is the most promising technique to evaluate the evolution of the irregular morphology of the IMC in liquid solder. The effect of grain boundary diffusion, solder/IMC interface energy and the solder composition etc. can all be considered through this approach. However, the precise data for these parameters is still not known for the lead-free solder system.

### Present Mathematical Model

To demonstrate the basic physics of the non-equilibrium process of copper dissolution, a better understanding of the relative importance of interface kinetic reaction and long-range diffusion at various stages of soldering process is important. In the present simulation, we propose a more mathematically rigorous model involving interface kinetics and diffusion. The model is based on the fundamental governing equations. Considering the soldering process as isothermal, the one-dimensional mass diffusion equation can be written as follows:

$$\frac{\partial C_L}{\partial t} = \frac{1}{r^a} \frac{\partial}{\partial r} \left( r^a D_L \frac{\partial C_L}{\partial r} \right) \quad (5)$$

where the symbols have the usual meanings as given in nomenclature and the constant  $a$  can have the values 0, 1, or 2 respectively for linear, cylindrical or spherical coordinate systems.

At the copper-solder interface, after neglecting the diffusion in solid, we have a mass conservation condition as:

$$C_L^i V_i = - \left( D_L \frac{\partial C_L}{\partial r} \right)_i \quad (6)$$

It is to be noted that  $V_i$  is negative because the interface moves in the negative direction of the spatial coordinate during dissolution. In addition, at the interface, the following kinetic relationship can be obtained [17, 23]:

$$V_i = \mu (C_L^i - C_{eq}) \quad (7)$$

$C_{eq}$  is the equilibrium solute concentration in the solder in wt% at the given operating temperature and can be found from the relevant phase diagram. The problem involves three unknowns, viz. interface concentration, interface velocity and solute concentration in the matrix. Equations (5) through (7) can be solved simultaneously to determine these unknowns at any instant of time. When the interface reaction constant,  $\mu$  is infinitely high, Eqn (7) tends to approach the equilibrium conditions at the interface.

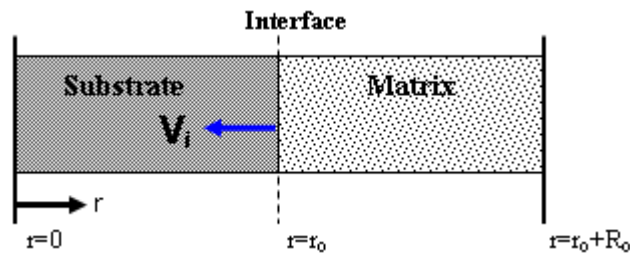


Figure 1: Coordinate System for the Moving Interface

Refer to the coordinate system Figure 1, it is assumed that the boundaries at the two ends of the domain are isolated with the surroundings, so the following boundary conditions are valid

$$\frac{\partial C}{\partial r} = 0 \text{ at } r = 0 \text{ and } r = (r_0 + R_0) \quad (8)$$

It is also assumed that the solder has no copper concentration prior to the onset of dissolution process, so the following initial conditions exist

$$C(r, t=0) = 100 \quad \text{for } r = 0 \text{ to } r_0 \quad (9a)$$

$$C(r, t=0) = 0 \quad \text{for } r = r_0 \text{ to } R_0 \quad (9b)$$

Equations (5) through (9) constitute a mass diffusion problem with a moving boundary and classically known as the Stefan-problem. In the present model the problem is also associated with a non-equilibrium condition at the interface.

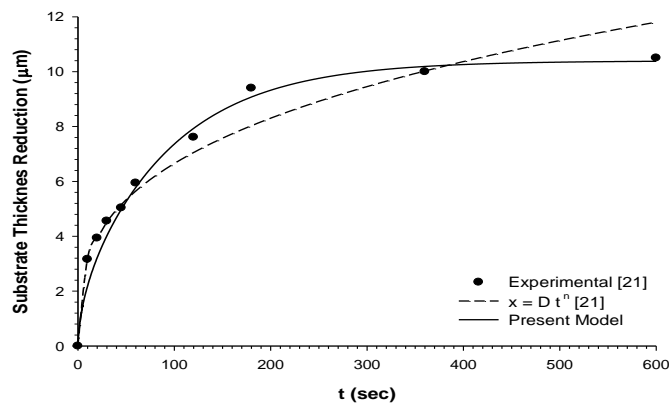
Analytical solutions for such problems are available in literature [22]. These analytical solutions suffer through the assumptions of infinite domain and the equilibrium condition at the moving interface.

For the present study, the above system of equations was solved numerically using a control volume approach. A coordinate transformation technique was employed in order to immobilize the moving interface in computational domain. A detailed description of the numerical scheme can be found in our previous papers [19]. A non-uniform grid was used near the interface on the solder side to accurately calculate the large gradient of the solute concentration at the moving boundary.

## RESULTS AND DISCUSSIONS

In the present model, one can identify that there are two rate-controlling parameters, which play a critical role in the overall progression of the dissolution of substrate material in the solvent matrix. These two parameters are the interface reaction kinetics coefficient,  $\mu$  and the diffusion coefficient,  $D_L$ . We consider that the soldering process is isothermal and proceeds at 250°C. At this temperature, the solder (pure Sn, Melting Point = 232°C) will be in liquid state. Copper was taken as the substrate material. It has been reported in literature that in liquid state the diffusivity is not very sensitive to the temperature [11]. Therefore, for the present study we take  $D_L = 10^{-9} \text{ m}^2/\text{sec}$  [18]. It is further assumed that the diffusivity is independent of solder composition.

To study the relative effect of the interface reaction and long-range diffusion on the overall dissolution behavior, concentration and location of the interface together with the dissolution rate were calculated for different values of  $\mu$  for constant diffusivity. A rigorous parametric analysis was carried to establish the rate controlling parameters for the present model. The results of this analysis will be presented in future.



**Figure 2: Comparison of Substrate Dissolution with Experimental Data and Other Model [21]**

The present model was tested to estimate the amount of copper dissolved for a practical case of a copper substrate dissolving in Sn matrix during actual reflow process at 250°C. Results were compared with experimental data Figure 2 and the existing power law model. The phenomenological model [21] well predicted the dissolved thickness for the initial part of the reflow process, but failed to predict the results accurately for higher reflow times. The present model underpredicted the copper dissolution in the early stage of the reflow process, when compared with the experimental data. The underprediction was attributed to the fact that in the present model, the development of intermetallic compound (IMC) at the interface was not considered. For a copper-tin interaction, IMC ( $\text{Cu}_6\text{Sn}_5$ ) is developed at the substrate-solder interface. The IMC can combine more Cu molecules with Sn, thereby increasing the copper solubility in the Sn matrix.

It was obvious to note that if the copper from substrate could precipitate into the IMC, it would be possible to dissolve more copper before the solder got saturated with solvent. It is envisaged that the incorporation of IMC in the present model will improve the prediction of the dissolved substrate thickness.

## CONCLUSIONS

A critical review of the existing models in literature to model the IMC formation and dissolution of copper in liquid solder had been done. Most of these models are found based on phenomenological approach. A mathematical model based on the basic physics of the copper dissolution in liquid solder is proposed for estimating the copper dissolution of planar substrate. The problem was solved numerically by using the coordinate transformation technique. It was observed that the interface reactions kinetic and solder diffusivity played a critical role in the overall dissolution of base metal. The relative effects of these parameters were meticulously analyzed. The present model was tested to estimate the consumed thickness of a planar substrate in liquid pure tin solder for actual reflow process. The results matched very well with the experimental data. Based on the present analysis, value for the interface reaction coefficient for copper-tin interaction has been proposed as  $7 \times 10^{-7}$  m/s wt%.

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