Research Article

Integration of machine learning with phase field method to model the electromigration induced Cu₆Sn₅ IMC growth at anode side Cu/Sn interface .

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Currently, in the era of big data and 5G communication technology, electromigration has become a serious reliability issue for the miniaturized solder joints used in microelectronic devices. Since the effective charge number (Z^*) is considered as the driving force for electromigration, the lack of accurate experimental values for Z^* pose severe challenges for simulation-aided design of electronic materials. In this work, a data-driven framework is developed to predict the Z^* values of Cu and Sn species at the anode based LIQUID, Cu₆Sn₅ intermetallic compound (IMC) and FCC phases for the binary Cu-Sn system undergoing electromigration at 523.15 K. The growth rate constants (k_{em}) of the anode IMC at several magnitudes of applied low current density ($j=1 \times 10^6$ to 10×10^6 A/m²) are extracted from simulations based on a 1D multi-phase field model. A neural network employing Z^* and j as input features, whereas utilizing these computed k_{em} data as the expected output is trained. The results of the neural network analysis are optimized with experimental growth rate constants to estimate the effective charge numbers. For negligible increase in temperature at low j values, effective charge numbers of all phases are found to increase with current density and the increase is much more pronounced for the IMC phase. The predicted values of effective charge numbers Z^* are then utilized in a 2D simulation to observe the anode IMC grain growth and electrical resistance changes in the multi-phase system. As the work consists

the aspects of experiments, theory, computation and machine learning, it can be called the four paradigms approach for study of electromigration in Pb-free solder. Such a combination of multiple paradigms of materials design can be problem-solving for any future research scenario that is marked by uncertainties regarding determination of material properties.

Key words: Phase field method; Artificial neural network; Intermetallic compound; Current density; Synchrotron radiation

1. Introduction

In this era of big data and miniaturized microelectronic devices, the growing trend for the use of 5G communication technology in artificial intelligence (AI) applications, has provided the microelectronics industries an impetus to promote 2.5D integrated circuits (2.5D IC) devices into mass production ^[1]. Compared to 2D IC devices, 2.5D IC devices require one more redistribution layer (RDL) and, subsequently, one more level of solder joints owing to the use of vertical interconnects. This makes the solder joints of the 2.5D devices more vulnerable to electromigration induced failures. Electromigration is defined as the process in which materials or species transport occurs due to a gradient in electric potential ^[2]. With the advancements in the joining technology, the continuous downscaling of solder joints has become a major paradigm in electronic packaging industries (in accordance to Moore's law), and this miniaturization of the joints has caused electromigration to be a major reliability concern ^[3-8]. In context of sub-100 nm structures, the current density during device operation may reach several $\frac{MA}{cm^2}$ ^[3]. The localized temperature raise caused by Joule heating, can then become high enough to melt some parts of the Sn-based solder joint. For example, localized spots with anode temperatures above 250 °C ^[9] have been experimentally observed for a Cu/30 μ m-Sn/Cu joint stressed with a current density of 1.3×10^4 - $1.5 \times 10^4 \frac{A}{cm^2}$. In this sense, characterization and prediction of the electromigration behavior in liquid solder has become a necessity in reliability studies.

The driving force for electromigration F_{em} is expressed using a dimensionless parameter, the effective charge number (Z^*) ^[10], as follows

$$F_{em} = eZ^*\rho j \quad (1)$$

In Eq. 1, e is the charge of an electron, E is the electric field intensity, ρ is the electric resistivity of the phase, and j is the current density. Actually, the effective charge number is the sum of the electrostatic charge number (Z^e) and the electron wind charge number (Z^w) ^[11]. Accordingly, the

electromigration driving force is comprised of an electrostatic and an electron wind contribution [10, 12-16].

$$F_{em} = e(Z^e + Z^w)\rho j \quad (2)$$

Generally, Z^e represents the direct electrostatic force on the moving ion, and its value is equal to the nominal valence of the ion. The contribution Z^w accounts for the electron wind force, which is generally the dominant contribution during practical electromigration in metallic joints and interfaces.

The electromigration parameters, such as electric resistivity and current density can be measured. However, there is no relevant work till now which ascertains accurate values of effective charge numbers (Z^*) in binary, ternary or, in general, multicomponent alloys. This induces an uncertainty in the quantification of the overall electromigration driving force for phase evolution. For a multiphase system consisting of more than one component undergoing electromigration, the major difficulty is in quantifying the electron wind charge number (Z^w) [13, 15-16].

For the binary Cu-Sn system consisting of Cu-rich, Sn-rich and intermetallic phases, a multiphase field formalism [17] has been utilized to investigate the evolution of interfacial intermetallic compound (IMC) layers under electromigration conditions [18- 20]. The research works [18- 20] utilize a free energy density term to describe the electromigration driving force on phase evolution, and this term includes the effective charge number. As highlighted earlier, it is not clearly understood how the magnitude of Z^* has been assigned in these works. In the work by Zhou and Johnson [18], the magnitudes of effective charge number for generalized species A and B are respectively taken as 90 and 0, but these values are constants in all the three phases considered. Park et al [19] have assigned a constant magnitude of 50 for effective charge number, implying that both Cu and Sn are considered to have the same value Z^* for all of the FCC, IMCs and BCT phases. Though Attari et al [20] took different values of the effective charge numbers in different phases, they assumed that the numbers for Cu and Sn are of a same magnitude in the same phase. Chao et al [10] stated that for the binary Cu-Sn system, in general, the effective charge numbers of Cu and Sn are not equal and are also different in different phases.

It can be expected that the growth behavior of the intermetallic layer at the anode Cu/Sn interface is primarily affected by interdiffusion and electromigration [10]. Hence, if the diffusion parameters are well known, the effective charge number can be inferred back by comparing experimentally determined growth rates of the IMC layer with growth rates obtained from simulations combining interdiffusion and electromigration and assuming a range of electromigration parameter values. An important difficulty is then that, at higher current densities,

the temperature of the Cu-Sn system increases considerably due to Joule heating, resulting in considerable fluctuations in the value of the interdiffusion coefficients, complicating an accurate determination of the electromigration parameters. However, at lower current density ($< 10 \times 10^6$ A/m²), the temperature rise is found to be negligible [2], and constant values of the diffusivities in the different phases can therefore be assumed in the procedure to predict the electromigration parameters.

On the one hand, multi-phase field methods can integrate multi-physics data of electromigration and interdiffusion parameters to uncover mechanisms that explain the emergence of anode Cu₆Sn₅ IMC growth kinetics. On the other hand, machine learning can integrate multimodality and multifidelity data, thus helping to reveal correlations between intertwined phenomena, such as growth kinetics and the effective charge number of the different elements in the different phases. In this regard, machine learning and phase field method can naturally complement each other and will enable us to create strong predictive models which integrate the underlying physics and explore massive material design spaces including the effect of electromigration ^[21]. Moreover, multiphysical models (finite element analysis), when coupled with experimental observation, can be utilized to construct the dataset for machine learning model. It has been demonstrated in ^[22] that finite element method generated (FEM-generated) data for given laser soldering experiments can be used to train the neural network , for prediction of the morphology of interfacial intermetallic compound (IMC).

In the sector of materials design and discovery, data-driven materials science has emerged as the fourth scientific paradigm following the first three paradigms-experiments, theory and computation ^[23]. In our present work and as presented in Fig. 1, we employ these four paradigms - (I) Experiments, (II) Theory, (III) Computation, and (IV) Machine learning in a combined way, to understand the materials science of anode interface in Cu-Sn binary system undergoing electromigration. With the relevant phases outlined through in-situ experimental imaging, phase field (PF) simulation will be performed in this work to integrate the data of electromigration parameters -j and, phase and composition dependent Z^* with the spatio-temporal evolution of anode IMC phase. The work will focus on cases with low current density to avoid the complexities associated with Joule heating at high current density. For the phase field simulations generated datasets, a neural network analysis will be done to predict the growth rate constant at given input values of j and Z^* . The growth rate constant is then optimized with available experimental data obtained from thickness measurement of cross-sectional image of the anode interface. Using inverse design, the neural network is then used to estimate the optimum effective charge numbers for Cu and Sn in different phases. These Z^* values will be used in a 2D phase field model to study

IMC grain growth due to interdiffusion and electromigration. For better performance of electronic devices and solar photovoltaic modules, the interconnections and joints utilized in these devices and systems must be characterized with good electrical and thermal conductances [24-25]. Since the intermetallics grains have different electrical resistivity compared to pure Sn and pure Cu metals, the accelerated growth of IMC phases due to electromigration causes the change in the electrical resistance of the overall system, and a method to compute the change in electrical resistance due to the microstructural evolution of phases is also included in the present work.

2. Experimental methods for data preparation

In context of experimental electromigration tests, the interfaces that have been characterized ex-situ can be utilized as a source growth kinetics data, but for the validation of the interfacial phases required by the multiphase field model, in-situ experiments are the proper choices. The in-situ synchrotron radiation (SR) X-ray imaging technology offers a promising strategy to observe in real-time and precisely distinguish exact phases formed and evolved as a result of diffusion and electromigration in experimental sample. In-situ synchrotron radiography experiments (Fig. 2) for the electromigration test of present work were performed in the beamline BL13W1 of Shanghai Synchrotron Radiation Facility (SSRF), Shanghai, China. As shown in Fig. 2(i), liquid Sn is sandwiched between two Cu substrates at the ambient temperature maintained at 523.15 K. The two Cu substrates are then connected by electrical wires to the DC voltage source. The Cu electrode connected to the negative terminal of the battery is anode whereas another Cu substrate linked to the positive terminal is the cathode (illustrated in Fig. 3 (a)). The Sn-rich liquid media of length 450 μm across interfaces, has larger electric resistivity than the electrodes, and is the conducting media for current flowing from anode to the cathode terminal. The thickness of liquid Sn (maintained by ceramic supports) and Cu electrodes was designed as 100 μm , so that the X-ray beam passing through these thin films can produce images of high contrast between the phases. Energy of the monochromatic X-ray beam was set as 28 keV. A charge coupled device (CCD) camera was kept at a distance of 200 mm from the sample, and the parameters of the CCD camera such as resolution ratio and exposure time were set as 0.74 $\mu\text{m}/\text{pix}$ and 4 s, respectively. More details of the experimental parameters associated with the synchrotron radiation imaging technology for the electromigration test at constant current density of $5.6 \times 10^6 \text{ A}/\text{m}^2$ is described in [26]. The images shown in Fig 2(ii) depict the asymmetrical growth of interfacial intermetallic compound (IMC) phases at the two FCC(Cu-rich phase)/LIQUID (Sn-rich phase) interfaces. In context of image (a) corresponding to $t = 5 \text{ min}$, the IMC layers are not noticeable at either interfaces. At $t = 10 \text{ min}$, the anode side interface's IMC, yet difficult to measure, is visible whereas

the IMC layer at cathode interface is still not visible. However, in image (d) at $t=45$ min, it can be seen that the anode based IMC is asymmetrically larger, and still the cathode side IMC layer is not noticeable. The bubble present at the vicinity of anode interface being progressively engulfed with the IMC grains as time proceeds from 10 min to 25 min validates the electromigration influenced rapid growth of the IMC layer therein. At 45 min, the bubble is merged deep within the IMC layer.

It is interesting to note from Fig. 2 (ii) that the phases observed in the electromigration experiment at $t=45$ min are FCC (both at anode and cathode), Cu_6Sn_5 IMC at the anode interface and LIQUID. Though the formation of Cu_3Sn IMC between FCC phase and Cu_6Sn_5 IMC at anode interface is also thermodynamically favorable, the diffusion based growth kinetics of Cu_3Sn IMC (interfaced between two solid state phases) is quite negligible compared to Cu_6Sn_5 IMC (remaining in contact with the LIQUID phase too). However, at lower temperatures, when both Cu substrate and Sn solder are in solid states, and the experiment is performed for a few days, the layer thickness of both Cu_3Sn and Cu_6Sn_5 IMCs are comparable [27]. In this present study, when the solder is in LIQUID state and the experiment is performed for 1 h, the growth of only one IMC, namely, Cu_6Sn_5 IMC, is considered at the anode (Ref. Fig. 3(b)), and thus Cu_3Sn IMC phase has been ignored. These experimentally observed three phases at anode interface - FCC, Cu_6Sn_5 IMC (mentioned as IMC hereafter) and LIQUID, are used for setting up the initial condition for the multiphase field model of Section 3.

Apart for the use in determination of the initial phases for multiphase field model, the experimental data are crucial for the validation of the results of phase field model. Ex-situ characterization of the anode interface's cross-section using scanning electron microscopy (SEM), can assist in rendering the growth kinetics data of anode IMC. Experimentally determined growth kinetics data of anode Cu_6Sn_5 are obtained from Feng et al. [28], and these values are further utilized in Section 5 to calculate the error in growth constant rates of phase field generated datasets. In other words, the experimental data and observations are integrated with the computational and the machine learning models to estimate optimized physical quantities associated with electromigration.

3. Multi-phase field model for Cu-Sn system undergoing electromigration

In context of Cu/Sn/Cu joint subjected to electric current stressing as shown in Fig. 3(a), Cu substrate (FCC phase) connected to the positive terminal of the battery is called anode whereas the Cu substrate attached to the negative terminal is defined as cathode. The system temperature is 523.15 K. In line with the available experimental data, electromigration studies at low current

density values (1.0×10^2 to 1.0×10^3 A/cm²) are considered for the current study. Experimentally, for a Cu/Sn/Cu system undergoing electric current stressing and after a certain amount of time, Cu₆Sn₅ intermetallic compound (IMC) at the anode interface is relatively thick, while that at the cathode interface is negligibly small [2, 26]. As the experimental thickness measurement is more precise for a growing intermetallic compound (IMC) and difficult for a dissolving one [26], the computational domain is confined to the 3 phases at the anode interface of the system as indicated in Fig. 3.

For the anode interface, the system consists of two components (Cu and Sn), and three phases (LIQUID, Cu₆Sn₅ intermetallic compound (IMC) and FCC). In multiphase field models, the different phases in the microstructure are represented by a set of non-conserved order parameters (η_i). For this system, we consider $\eta_1 = 1$ for FCC, $\eta_{2,j} = 1$ for IMC (with j the grain number) and $\eta_3 = 1$ for Liquid. Each order parameter (η_i) has its minimum and maximum values at 0 and 1, respectively. Furthermore, a conserved variable (c) is used to represent the local mole fraction of Sn in the binary Cu-Sn system. Since the mole fraction of Cu, c_{Cu} , can be obtained as $c_{Cu} = 1 - c$, it is sufficient to only consider the evolution of the mole fraction of Sn. The mole fraction of Sn in FCC phase is denoted by c^{fcc} . Similarly, c^{imc} and c^{liq} are the respective mole fraction of Sn in IMC and LIQUID phases. Moreover, for the symbols of Gibbs free energy, free energy density, the coefficients in the parabolic free energy density, mobilities and diffusivities the phase identification will be specified in superscript.

The non-conserved order parameters are functions of time (t) and spatial co-ordinates(x) [29]. In multi-phase field models, the material properties at the interface region are interpolated in a thermodynamically consistent way using the function ($h(\eta_i)$) defined in [30], with

$$h_i = \frac{\eta_i^2}{\sum_{k=1}^N \eta_k^2} \quad (3)$$

for LIQUID and FCC, which have only 1 grain, and

$$h_i = \frac{\sum_{j=1}^p \eta_{i,j}^2}{\sum_{k=1}^N \eta_k^2} \quad (4)$$

for the IMC phase, where more than one grain (in this case p grains) can be present. In these equations, N is the total number of order parameters of the system, including the possibility of having more than one grain in the IMC phase meaning that N can be larger than the number of phases. Although an IMC grain is chemically identical another IMC grain, they have difference in structural orientation [31], and thus the p grains of IMCs need to be represented by p order parameters.

The total free energy (F_{total}) of a thermodynamic system can be expressed as the sum of bulk,

interfacial and electric free energy as follows

$$F_{total} = F_{bulk} + F_{int} + F_{elec} = \int_V (f_{bulk} + f_{int} + f_{elec}) dV \quad (5)$$

where the three major contributing factors are outlined as follows

$$f_{bulk} = \sum_i h_i f_{chem}^i = \sum_i h_i \frac{G_m^i}{V_m} \quad (6)$$

$$f_{int} = m f_o + \frac{\kappa}{2} \sum_i (\eta_i)^2 \quad (7)$$

$$f_{elec} = \sum_i h_i f_{elec}^i = N_A e \phi \sum_i h_i \frac{Z_{eff}^* c^i}{V_m} \quad (8)$$

The first term inside the integral sign of Eq. 5, namely the bulk free energy density part, is described by Eq. 6. In Eq. 6, G_m^i is the molar Gibbs free energy of a phase. The molar volume (V_m) for all the phases is assumed to be equal to $16.29 \text{ cm}^3 \text{ mol}^{-1}$ [29]. Parabolic composition dependence is assumed for the molar Gibbs free energy functions. Following the methodology outlined in [32], the coefficients in the parabolic molar Gibbs free energy functions were fitted to the CALPHAD expressions for FCC, LIQUID and IMC (CU6SN5) in the Cu-Sn system from Moon et al [33]. Although, the Cu₆Sn₅ IMC is modeled as a stoichiometric compound in this CALPHAD description, a parabolic composition dependence is included for the IMC phase in the phase field model to be able to extract a first and second derivative of the free energy density with respect to molar fraction, required in the diffusion equation. The parabolically fitted molar Gibbs free energies for the three phases at 523.15 K are shown in Fig. 4. The expression of the molar Gibbs free energy of phase i is given as

$$G^i = 0.5A^i(c^i - c_{eq}^i)^2 + B^i(c^i - c_{eq}^i) + C^i \quad (9)$$

The coefficients A^i , B^i and C^i of the three phases are provided in Table 1.

Application of the common tangent rule to the fitted molar Gibbs free energy functions gives the following equilibrium compositions: $c=0.15$ in the FCC phase and $c=0.946$ in the LIQUID phase when in equilibrium with the IMC phase. The equilibrium composition of the IMC phase when in equilibrium with the FCC and LIQUID phases is 0.436 and 0.460, respectively.

In the interfacial free energy density part (Eq. 7), f_o is a fourth-order Landau polynomial of the phase field variables [32, 34] defined as :

$$f_o = \sum_i \left(\frac{(\eta_i)^4}{4} - \frac{(\eta_i)^2}{2} \right) + \gamma \sum_i \sum_{j \neq i} \eta_i^2 \eta_j^2 + \frac{1}{4} \quad (10)$$

where we take $\gamma = 1.5$. The diffuse interface width (δ [m]) is considered as a numerical parameter [29] and is related to the model parameters m and κ of Eq. 7 as :

$$m = \frac{6\sigma}{\delta} \quad (11)$$

$$\kappa = \frac{3\delta\sigma}{4} \quad (12)$$

where, σ [J/m²] is the interfacial energy.

The third term of Eq. 5, namely f_{elec} is the electrostatic free energy density [18, 19, 20, 35]. In Eq. 8, N_A is the Avogadro number, e is the natural charge of an electron and ϕ is the electric potential. For a phase i , Z_{eff}^* is composition dependent and is expressed as

$$Z_{eff}^* = Z_{Sn}^i + \frac{1-c^i}{c^i} Z_{Cu}^i \quad (13)$$

In the formula for the composition dependent effective charge number, Z_{Sn}^i and Z_{Cu}^i are respectively the effective charge numbers of Sn and Cu in i^{th} phase. In [19], Z^* is considered as a constant and does not vary with the phases, whereas in [20] it is considered as a phase dependent property. Within a phase, it is possible that Z^* has a different value for Cu and Sn [10]. However, there are only few experimental works outlining the composition dependent effective charge numbers for the individual phases in the Cu-Sn system at 523.15 K. Kumar et al [13] have found that the effective charge number of pure LIQUID Sn is approximately 2. They have highlighted that for pure metals, the effective charge number is lower for the LIQUID than for its solid phase. By solving the inverse problem of intermetallics growth at 423.15 K, Chao et al [10] have predicted the Z^* values for Cu and Sn in the Cu₆Sn₅ phase. The values for the effective charge numbers of Cu and Sn in BCT (Sn rich) have been provided at the same temperature. In case of the FCC (Cu rich) phase, only the effective charge number of Cu is provided. The values of Z^* are estimated to be in the range 1-100. In the work of [10], all of the Cu-rich, Sn-rich and Cu₆Sn₅ phases are in solid state, whereas in the present study, the Sn-rich phase is in LIQUID state. The inferences from [10, 13] will be utilized as a reference to form the initial arrays of effective charge number in the training of the neural network.

$$\bar{Z} = \{Z_{Cu}^{fcc}, Z_{Sn}^{fcc}, Z_{Cu}^{imc}, Z_{Sn}^{imc}, Z_{Cu}^{liq}, Z_{Sn}^{liq}\} \quad (14)$$

In Eq. 14, the superscript within the elements of the set Z , namely FCC, IMC, LIQUID represent the phases, whereas the subscripts Cu and Sn represent the components. For convenience, the $*$ symbol will not be used with Z , when there are subscripts, superscripts or overline associated with it.

The evolution of the conserved variable (c) is expressed by the following equation:

$$\frac{\partial c}{\partial t} = \nabla \cdot (\sum h_i M^i) \nabla \left(\frac{\partial f_{bulk}}{\partial c^i} + \frac{\partial f_{elec}}{\partial c^i} \right) \quad (15)$$

In Eq. 15, M^i ($= M(\eta_i)$) is the mobility^[36], which is related to the diffusivity or diffusion coefficient (D^i) of i^{th} phase^[37] and the grain boundary diffusion mobility M_{gb} by the following expression:

$$h_i M^i = h_i \left(\frac{D^i}{\frac{\partial^2 f_{bulk}}{\partial c_i^2}} + \sum_{j \neq i} h_j M_{gb} \right) \quad (16)$$

M_{gb} represents the additional contribution to the diffusion mobility via the grain boundary between grains i and j . The grain boundary diffusion mobility is related to the grain boundary diffusivity as follows:

$$M_{gb} = 3 \frac{D_{gb}}{h_i A^i + h_j A^j} \frac{\delta_{gb}}{\delta} \quad (17)$$

In this work, all the grain boundaries are assumed to have the same diffusion coefficient (D_{gb}) and grain boundary width (δ_{gb}). The magnitude of δ_{gb} is chosen to be equal to 0.5 nm^[36]. For the parabolic free energy curves given by Eq. 9, the second derivative of the free energy density of a phase with respect to the molar fraction of Sn, gives the constant A^i , thereby making the bulk diffusion mobility independent of composition in this model.

The evolution of the non-conserved order parameter is described by the equation provided below:

$$\frac{\partial \eta_i}{\partial t} = L(\eta_1, \dots, \eta_N) \left(\frac{\partial f_{bulk}}{\partial \eta_i} + \frac{\partial f_{elec}}{\partial \eta_i} - \nabla \cdot \frac{\partial f_{int}}{\partial \nabla \eta_i} \right) \quad (18)$$

In Eq. 18, $L(\eta_1, \eta_2, \dots, \eta_N)$ is given as following^[30, 38].

$$L(\eta_1, \dots, \eta_N) = \frac{\sum_{i=1}^N \sum_{j>1}^N L_{ij} \eta_i^2 \eta_j^2}{\sum_{i=1}^N \sum_{j>1}^N \eta_i^2 \eta_j^2} \quad (19)$$

where L_{ij} is the interfacial mobility, and for binary Cu-Sn system, its value is provided as:

$$L_{ij} = \frac{4m}{\kappa \xi_{ij}} \quad (20)$$

In Eq. 20, the reciprocal of ξ_{ij} is defined as:

$$\frac{1}{\xi_{ij}} = \frac{0.5(M^i + M^j)}{(c_{i,eq,j}^i - c_{j,eq,i}^j)(c_{i,eq,k}^i - c_{k,eq,i}^k)} \quad (21)$$

Fig. 4 can be referred for understanding about the term $(c_{i,eq,j}^i - c_{j,eq,i}^j)(c_{i,eq,k}^i - c_{k,eq,i}^k)$ in Eq. 21. When the two common tangents, one between IMC and FCC phases (i and j phases), and another between IMC and LIQUID phases (i and k phases), are projected on the composition axis (c_{sn} , or simply, c), the product of their absolute lengths represents the denominator at the right

hand side of the equation. The value of $(c_{i,eq,j}^i - c_{j,eq,i}^j)(c_{i,eq,k}^i - c_{k,eq,i}^k)$, as obtained from the measurement of these projection lines is 0.15.

The distribution of the electric potential in the domain is governed by the following laplacian of the potential variable:

$$\nabla \cdot \lambda \nabla \phi = 0 \quad (22)$$

where, λ is the phase dependent electric conductivity, and is the reciprocal of electric resistivity, i.e. $\lambda = \frac{1}{\rho}$. The interpolation function is utilized to express the phase dependence of electric resistivity as given by the following equation.

$$\rho = \sum_i h_i \rho^i \quad (23)$$

Thus, each phase has a resistivity ρ^i and the interface resistivity is interpolated with the use of h_i .

3.1. Material properties and parameters

The material properties and model parameters required for solving the phase field and static current conduction equations for the considered Cu-Sn system are listed in Table 2. The values of m and κ for a given value of interfacial energy are dependent on the values of diffuse interface width (δ_{gb}). As illustrated in the table, the diffusion coefficient for LIQUID phase is several times larger than the IMC and FCC phases. The grain boundary diffusion coefficient (D_{gb}) is assumed to be 200 times the diffusivity of the IMC phase as has been considered in one of the models of Hektor et al. [29]. The interfacial energy between the phases is considered to be uniform everywhere in the computational domain. The values for electrical resistivities of FCC (Cu rich) and IMC phases are taken from [19], whereas that of the Sn-rich LIQUID phase is taken from [20]. It must be noted that ρ^{fcc} and ρ^{liq} are taken to be equal to that of pure Cu and Sn respectively, and are represented as ρ_{Cu} and ρ_{Sn} in the table.

3.2. Boundary conditions

For both the conserved variable (c) and non-conserved order parameters (η_i), no-flux boundary conditions are applied at the edges/sides consisting solely of either FCC or LIQUID. Periodic BCs are applied elsewhere. In context of the electric field equation (Eq. 22), the following Neumann BCs were adopted at the FCC and LIQUID ends respectively:

$$(\nabla \phi)_{fcc} = -j \cdot \rho_{Cu} \quad (24)$$

$$(\nabla \phi)_{liq} = -j \cdot \rho_{Sn} \quad (25)$$

In the equations, j (A/m^2) is the constant current density imposed on the materials system. With the abovementioned material properties and boundary conditions, the set of Eqs. 15, 18 and 22 were solved using finite element method (FEM) in Multiphysics Object Oriented Simulation Environment (MOOSE) Framework [39-44]. Within MOOSE Framework, the solutions of residuals is obtained using the Preconditioned Jacobian-free Newton-Krylov (PJFNK) method. Kim-Kim-Suzuki (KKS) model is implemented in this binary Cu-Sn system involving multiple phases (3 phases). In accordance to KKS model [45], (i) the chemical potential between any two adjacent phases is assumed to be equal, and (ii) the global composition of Sn is expressed as $\sum_i h_i c^i$.

3.3. Determination of IMC layer growth rate under electromigration

With the methodology outlined in previous sections, a total number of 71 one dimensional (1D) phase field simulations were performed to model the IMC growth under electromigration. The current density was varied from 1.0×10^6 to $1.0 \times 10^7 \text{ A/m}^2$. The effective charge number of Cu, Sn in the different phases i were varied over the 71 simulations, considering the information available in the works [10, 13, 19-20]. The total length of the computational domain is taken as $1.5 \text{ }\mu\text{m}$. The Cu-rich FCC phase at the left edge has an initial length of $0.550 \text{ }\mu\text{m}$, and the Sn-rich LIQUID phase is $0.90 \text{ }\mu\text{m}$. The IMC phase with an initial length of 50 nm ($0.05 \text{ }\mu\text{m}$) is interfaced between the FCC and LIQUID phase. The thickness of the anode Cu_6Sn_5 layer at each time step is determined from the simulated η_{imc} profiles. For $x^{i,j}$ and $x^{j,k}$ being the positions of FCC-IMC interface and IMC-LIQUID interface, respectively, the IMC layer thickness y at time (t), is obtained as the difference between the positions of its two interfaces, i.e. $y(t) = x^{i,j} - x^{j,k}$. The interface positions of IMC phase with other phase (LIQUID or FCC), can be located at a given time by finding the grid positions where $0 < \eta_{imc} < 1$. The growth rate for anode IMC undergoing electromigration is observed to be linear, and thus, the growth rate constant (k_{em}) for this case can be determined as the slope of the y - t curve. For evaluating the growth kinetics of the IMC phase, 1D simulation is sufficient, and this provides us the benefit of higher computational speed. With this advantage, the length of the simulation domain can be increased to the scale of micrometers and the simulated time to the scale of a minute. The growth kinetics determined for longer duration of simulation time tend to be better from the viewpoint of error minimization.

4. Synergistic effect of j and Z^* on the growth kinetics of anode IMC layer

To gain more understanding in the combined or synergistic effect of the electromigration parameters on the growth kinetics (and growth rate constant) of the anode Cu₆Sn₅ IMC layer, the results of 10 (out of the total of 71 simulation performed) 1D phase field simulations are chosen for a comparative study in this section. In these 10 phase field simulations, two values of current densities - 1.0×10^6 A/m² and 10.0×10^6 A/m² are considered and the following sets of effective charge numbers for Cu and Sn in the different phases are selected:

$$\bar{Z}_1 = \{4.00, 4.00, 26.0, 36.0, 2.00, 4.00\} \quad (26)$$

$$\bar{Z}_2 = \{8.00, 8.00, 52.0, 72.0, 4.00, 4.00\} \quad (27)$$

$$\bar{Z}_3 = \{2.00, 2.00, 13.0, 18.0, 2.00, 2.00\} \quad (28)$$

$$\bar{Z}_4 = \{10.0, 5.00, 20.0, 15.0, 5.00, 10.0\} \quad (29)$$

$$\bar{Z}_5 = \{20.0, 20.0, 20.0, 20.0, 20.0, 20.0\} \quad (30)$$

In the list of $\bar{Z}_n$ above, the values of the effective charge numbers of Cu and Sn in the different phases are listed in the same order as in Eq. 14. Figure 5(a) shows the evolution of intermetallic compound thickness as a function of time for $\bar{Z}_1$, $\bar{Z}_2$, $\bar{Z}_3$, $\bar{Z}_4$, $\bar{Z}_5$ at $j = 1.0 \times 10^6$ A/m². A linear dependence of IMC thickness with time (i.e. (thickness = $k_{em} \times t$) is clear from the graphs, which is in agreement with the experimental reports in [26, 28]. For $\bar{Z}_2$ most of the effective charge numbers are double of the corresponding effective charge number in $\bar{Z}_1$. For $\bar{Z}_3$, all the effective charge numbers, except Z_{Cu}^{liq} , are half of the corresponding effective charge numbers in $\bar{Z}_1$. In none of the simulations, the value for Z_{Cu}^{liq} has been taken below 2, such that the effective charge number of an element is always equal to or greater than its valency. In $\bar{Z}_4$, the effective charge number of Cu and Sn in each phase is assigned a different value. A constant effective charge number of 20, that is independent of both phase and composition, has been utilized in case $\bar{Z}_5$. For an initial IMC thickness of 50 nm (0.05 μ m), the highest growth rate is attained for $\bar{Z}_2$, with the effective charge numbers of Cu and Sn within the IMC phase equal to $Z_{Cu}^{imc} = 52.0$ and $Z_{Sn}^{imc} = 72.0$, respectively, and the Z values for Sn and Cu both equal to 4 in the LIQUID phase and both equal to 8 in the FCC phase. With a growth rate constant (k_{em}) of 0.337 μ m/min, the Cu₆Sn₅ layer attains a thickness of 1.125 μ m at the end of the simulation after 200 s

of growth. For the case with $\bar{Z}_1$, having all of the effective charge numbers except Z_{Sn}^{liq} , equal to half of that of $\bar{Z}_2$; the growth rate constant is around 0.282 $\mu\text{m}/\text{min}$, and the IMC layer is 0.94 μm at the end of the simulation. The growth rate constants for the cases with $\bar{Z}_3$ (one-fourth of $\bar{Z}_2$ for FCC and IMC phases), $\bar{Z}_4$ and $\bar{Z}_5$ are 0.255, 0.252 and 0.229 $\mu\text{m}/\text{min}$ respectively. The lines corresponding to $\bar{Z}_3$ and $\bar{Z}_4$ have nearly equal slopes (i.e. the growth constant is almost equal). After 200 s, the IMC layer corresponding to $\bar{Z}_3$ reaches 0.85 μm and that corresponding to $\bar{Z}_4$ obtains a thickness of 0.84 μm . It appears that the sum of effective charge number in the IMC phase being larger in case of $\bar{Z}_4$ would favour the IMC phase growth kinetics as compared to $\bar{Z}_3$, but again the larger effective charge numbers in the LIQUID and FCC phase in case of $\bar{Z}_4$ would simultaneously favour the growth of other phases at the expense of IMC. This counter effect phenomena originated from Z values of other phases, lower the IMC growth rate. This explains the lower net growth rate of IMC in case of $\bar{Z}_4$. It can be inferred that larger values of IMC effective charge number favour the anode Cu_6Sn_5 growth rate, but in the similar way, the larger value of effective charge number of other phases counterbalance its growth. The most notable counter effect phenomena is exhibited through the case with constant effective charge numbers over all phases ($\bar{Z}_5$), as it has the lowest IMC growth rate constant.

In experiments, the application of current density is always noted with an accelerated increase of anode IMC over other phases, and this helps in deducing the fact that anode IMC phase's effective charge numbers are in general larger than that of other phases, for driving this overall increment in growth kinetics. This fact is supported by the numerical results presented in [10]. Since, the application of current can also alter the effective charge numbers of LIQUID and FCC phases, the problem becomes more complicated. But again with the experimental observation of anode IMC phase's dominance over LIQUID and FCC, it can be generalized that the rate of increment in Z values in these phases is significantly smaller than for the IMC phase. This observation is an important principle in designing effective charge numbers and current density as the input features of neural network in section 5. The results obtained for intermetallic compound thickness for a current density of $10.0 \times 10^6 \text{ A/m}^2$ are presented in Fig. 5(b). Compared to Fig. 5(a), the thickness curves in Fig. 5(b), become more divergent as the time proceeds. The effect of effective charge number on IMC growth rate is thus more pronounced at larger current density. The growth rate constants at a current density of $10.0 \times 10^6 \text{ A/m}^2$, for effective charge numbers

$\bar{Z}_1$, $\bar{Z}_2$, $\bar{Z}_3$, $\bar{Z}_4$ and $\bar{Z}_5$, are 0.662, 0.989, 0.459, 0.442 and 0.236 $\mu\text{m}/\text{min}$, respectively. The case $\bar{Z}_2$ for the larger current density (Fig. 5(b)) shows a growth rate constant of about 2.93 times that of the case with the same set of effective charge numbers for the lower current density (Fig. 5(a)). It has to be noted that the thickness value for the case with $\bar{Z}_5$ in Fig. 5(b) is 0.315 μm at 76 s. Thus, when the current density is increased by 10 times, and the effective charge number for all phases/components is maintained 20, the growth rate constant increases only by 1.03 times. This shows again the importance of the difference in magnitude of the effective charge numbers for the IMC and that of those of the other phases. Both, current density and variation in the value of the effective charge number among components and different phases, affect the growth kinetics of the IMC phase.

5. Prediction of Z^* using machine learning

As discussed in the previous section, the choice of the value of effective charge numbers of Cu and Sn for the three phases, can alter the growth kinetics of IMC at a given current density. Moreover, the growth rates are amplified with higher current density. It is important to optimize the $\bar{Z}$ values as functions of phase, composition and strength of electric field, and this can be done with machine learning.

5.1. Regression based feed-forward neural network (RFFNN)

The growth rate constants – k_{em} obtained from phase field simulations using different sets of Z values are utilized to build the output features in the datasets with 71 observations. On the other hand, the electromigration parameters, namely, the effective charge numbers (Z_{Cu}^{fcc} , Z_{Sn}^{fcc} , Z_{Cu}^{imc} , Z_{Sn}^{imc} , Z_{Cu}^{liq} , Z_{Sn}^{liq}) and current density (j) as the input features. Since the output feature is a continuous function of the input values, this is not a classification problem, but clearly a regression task. Regression based feed-forward neural network (RFFNN) can be used with fair accuracy even for small datasets [46- 48]. A regression based feed-forward neural network shown in Fig. 6, is constructed to predict the relationship between the electromigration parameters (input) and growth rate constant (output). The component and phase dependent effective charge numbers ($\bar{Z}$) and current density (j), used respectively as material properties and boundary conditions in the finite element method (FEM) based phase field simulations, are fed into the input layer of the artificial neural network (ANN). The growth rate constant ($k_{em,nn}$), predicted at the output layer of the neural network for each set of $\bar{Z}$ and j , is compared and optimized against the phase field calculated

growth rate constants. It is known that a lower number of hidden layers (HLs) is more appropriate when analysing small datasets ^[49]. Moreover, the number of neurons in the hidden layer also affects the predictability of the artificial neural network ^[50]. For this study consisting of 71 observations, neural networks with 2, 3 and 4 hidden layers and different number of neuron per layers were tested for their performance. Among them the network with 3 hidden layers, and possessing respectively 7, 10, 8, 8, and 1 neurons in input layer, first HL, second, HL, third HL and output layer, has been observed with the lowest mean square error (MSE). This neural network is chosen for the data analysis and its MSE is presented in Sec. 5.1.3. Rectified linear unit (ReLU) is used as the activation function in all three hidden layers, whereas linear activation function is used for the output layer.

5.1.1 Data visualization and analysis

In order to get insight, not only on the distribution of a single machine learning variable, but also on the interrelationship between electromigration parameters and the subsequent intermetallic compound growth rate constant (k_{em}), the dataset has been represented with pairplots using seaborn visualization library ^[51] in Figs. 7 and 8. In the data visualization and machine learning procedure, current density and growth rate constants have been presented using the units of $\frac{A}{mm^2}$ ($1 \frac{A}{mm^2} = 10^6 \frac{A}{m^2}$) and $\frac{nm}{s}$ ($1 \frac{nm}{s} = 0.06 \frac{\mu m}{min}$), respectively, so that the numerical values of all the parameters will be of a similar order of magnitude. The distributions of effective charge numbers of the Cu component in each phase, have been plotted along with the growth rate constant of the IMC phase, for two ranges of current density, namely a lower ($1-4 \frac{A}{mm^2}$) and higher range ($5-10 \frac{A}{mm^2}$), in Fig. 7. Similarly, Fig. 8 presents the pairplot corresponding to the Sn component. From the scatter plots in both the figures, the data points for high current density (orange colored markers) have on average a greater growth rate than the data for lower current density (blue colored points). The diagonal images in the pair plot reveal the kernel density estimates (kde) for the distribution of each parameter. In case of both Cu and Sn, the kde plots for effective charge numbers (Z parameters) at both ranges of current density overlap, suggesting that these parameters have a similar distribution for both ranges of the current density. Neglecting the small fluctuations in the probability distribution function of the effective charge numbers due to current density, the peak values of the kde area plots of Z_{Cu}^{fcc} , Z_{Cu}^{imc} , Z_{Cu}^{liq} , Z_{Sn}^{fcc} , Z_{Sn}^{imc} , Z_{Sn}^{liq} are 4.01, 25.9, 2.14, 3.75, 36.55, 4.29, respectively. Their respective probabilities are 0.21, 0.04, 0.23, 0.14, 0.03, 0.25. On the other hand, the kde plot of the k parameter, is different for the two ranges of current density.

The probability distribution function of this parameter for the higher range of current density (orange color) has its mean value shifted to the right compared to the distribution for lower current density (blue color), meaning that the former has a larger mean value of growth rate constant. The peak of the blue colored kde area plot corresponds to the growth rate constant of 4.75 nm/s with the probability value of 0.5, whereas the peak of orange colored area plot has its peak at 8.55 nm/s with a probability value of 0.11.

5.1.2. Normalization of the database

In general, for a hidden layer of a neural network, an activation function transforms received input values in such a way that the output values obtained from the hidden layer are well within the manageable range. For any input x , ReLU activation function is mathematically defined as $\text{ReLU}(x) = \max\{0, x\}$. Any negative valued input x processed through $\text{ReLU}(x)$ becomes 0 at output, whereas a positive value of x at input of the function comes out unchanged at the output. The real dataset of effective charge numbers, current density, and k_{em} , even after changing units, vary beyond the range 0-1. For such a dataset consisting of parameters covering different ranges, the parameter with the largest range will have more dominance over the results ^[48, 52], and in order to avoid such dominance, a process called normalization is utilized. If X_i is an element of a column array consisting of one of the input or expected output, then the formula for normalization is given as follows:

$$\bar{X}_s = \frac{X_i - X_{min}}{X_{max} - X_{min}} \quad (31)$$

where, $\bar{X}_s$ is the normalized (scaled) value of a parameter data point corresponding to the actual feature's value X_i . X_{max} and X_{min} are respectively the maximum and minimum values of the feature over the whole dataset. With the normalization procedure, all the values of effective charge number, current density, predicted growth-rate constant and expected growth rate constant, will be in the range of 0-1. In order to find the optimized growth rate constants for the given input network features in the physical units, Eq. 31, can be utilized again to calculate X_i from $\bar{X}_s$.

5.1.3. Cross-validation and performance evaluation of the ANN

Next it is necessary to investigate on how the ANN model for electromigration, performs for data outside the sample (training data), when applied to a new dataset (validation data). This procedure of assessing the prediction performance or robustness of the neural network is called cross-validation. In order to initiate the cross-validation task, the original dataset consisting of 71 observations, has been divided into training and validation datasets in the ratio 4:1, and this

splitting or categorization of the observations data is performed randomly . Mean square error (MSE) is a mathematical parameter to evaluate the performance loss during the model run, and its value can be utilized to quantify the robustness of the ANN model. The lower the value of MSE, the better or more robust is the neural network model.

For N datasets, MSE value between the prediction value (y_p) of the network, and the target value (k_{em}) [53, 54], is defined by the following mathematical expression:

$$MSE = \frac{1}{2n} \sum_{n=1}^N (y_p - k_{em})^2 \quad (32)$$

In the definition of the performance function MSE, the target value is the growth-rate constant of anode IMC determined from multi-phase field simulations and thus, k_{em} is FEM-generated output for each observation. With MSE as the cost function of the neural network, its performance can now be evaluated for the training and validation data. Utilizing adam optimizer, the neural network was built and compiled in tensorflow [55] with keras [56] front end. In neural network analysis, the number of epochs is understood as a hyperparameter which denotes the total number of times a learning algorithm shall work throughout the entire training dataset. The result for MSE of the training and validation data for 1000 epochs are presented in Fig. 9. As the error is reduced with the increment of epoch, the neural network can be assumed to have a good performance.

5.2. Deduction of the Z^* values reproducing experimental data

After the successful training and validation of the dataset with the regression based ANN, the model can be used to make generalized predictions of the growth rate constant of the anode IMC layer, provided the input electromigration parameters are well within the distribution functions given in the Figs. 7 and 8. The growth rate constants determined from the neural network model can then be compared against the constraints provided by the experimental results on electromigration.

Experimental thickness measurements for anode based Cu₆Sn₅ in Cu/liquid-Sn/Cu have been presented in Ma et al. [26] and Feng et al. [28]. The growth kinetics of anode IMC has been described for only a single value of current density ($j=5.60 \times 10^6$ A/m²) at 523.15 K in Ma et al. [26], whereas in case of Feng et al. [28] the growth rate constant has been presented for two different current density values and two different working temperatures. For example, the growth rate constants corresponding to $j=1.0 \times 10^6$ A/m² at 533.15 K and 573.15 K have been mentioned respectively as 0.107 and 0.152 $\mu\text{m}/\text{min}$ in Feng et al. [28]. When the current density is increased to 2.0×10^6 A/m²

at 533.15 K, the growth rate constant rose to 0.212 $\mu\text{m}/\text{min}$. Thus, it has been inferred in Feng et al. [28], that for experiments at lower current densities, the increase in current density by twice the original value, also would increase the anode IMC growth rate constant by double of its reference value. With this information available for different current densities and temperatures, fitting has been done in the present work to obtain the experimental growth rate constants at 523.15 K for different current densities. As an illustration, the growth rate constant for 1.0×10^6 and 2.0×10^6 A/m^2 at 523.15 K, are 0.098 and 0.194 $\mu\text{m}/\text{min}$, respectively.

Now, running the same neural network as a prediction model, with the current density fixed to the experimental value, the most realistic set of effective charge numbers ($\bar{Z}$) is the one, which produces the predicted growth rate constant (y) having the least difference with the experimentally obtained growth rate constant (k_{em}) for a given temperature. The neural network model, that has been previously trained and validated, is utilized to predict the optimum growth rate constant for 2000 different input parameters (with current density varying from 1×10^6 - 10×10^6 A/m^2). At each magnitude of current density, about 200 different sets of effective charge numbers were fed into the network. In context of IMC phase, in which the range of effective charge numbers is quite large, the initial guess values of Z_{Cu}^{imc} and Z_{Sn}^{imc} , for current density above 2.0×10^6 A/m^2 , are chosen by neglecting the values after decimal point. This would mean that the predicted values of effective charge numbers of Cu in Sn in IMC phase for $j \geq 3.0 \times 10^6$ A/m^2 would have only 0 after decimal. The results from the prediction model (machine learning) are presented in Fig. 10 and Table 3. As shown in the curves of Fig. 10(a)-(c), the effective charge numbers of Cu and Sn in all phases increase with the rise of current density. The trend is most pronounced for the IMC phase, whereas it is least pronounced for the LIQUID phase. As an illustration, at $j = 1 \times 10^6$ A/m^2 , Z^* values of Cu and Sn in the IMC phases are 2.1 and 2.25 respectively, whereas these values in the LIQUID phases are 2.02 and 2.03 respectively. However, when the current density is increased 10 times, Z_{Cu}^{imc} and Z_{Sn}^{imc} are raised to magnitudes of 58 and 68 respectively. At $j = 10 \times 10^6$ A/m^2 , Z_{Cu}^{liq} and Z_{Sn}^{liq} increase to just 5.8 and 5.9, respectively. The results for growth rate constant shown in Fig. 10(d), verifies the consistency between the results of machine learning and the experimental data.

Above effective charge numbers have been predicted via machine learning at a constant temperature of 523.15 K, for which the diffusion coefficients of the phases (D^{fcc} , D^{imc} , D^{liq}) and of grain boundary diffusion (D_{gb}) are kept constant. This assumption is valid for experiments or applications at lower current densities ($\leq 10.0 \times 10^6$ A/m^2). In our previous work [2], the maximum

increase in temperature (due to Joule heating) for electromigration experiments performed at $j = 5.6 \times 10^6 \text{ A/m}^2$ and 523.15 K was found to be not more than 1 K, and for another experiment at the same temperature but at $j = 30.0 \times 10^6 \text{ A/m}^2$, the maximum increment in temperature was around 4 K. At experiments beyond $100.0 \times 10^6 \text{ A/m}^2$, the Joule heating induced temperature raise will be significant enough to alter the phase diffusivities by a large amount. In other words, the electromigration induced anode IMC growth rate, as an output parameter of the neural network, is proportional to Z^* and j , at lower j ($\leq 10.0 \times 10^6 \text{ A/m}^2$) and, proportional to the product of phase diffusivity and effective charge number ($D^i \times Z^*$) and j , at higher j ($\geq 100.0 \times 10^6 \text{ A/m}^2$) [21]. Thus, when the temperature fluctuations due to Joule heating are not large enough to alter diffusivities, as in the context of the present study, the input parameters for neural network can be sufficiently designed with effective charge numbers (Z^*) and current density (j) alone. At current densities beyond $100.0 \times 10^6 \text{ A/m}^2$, where temperature fluctuations are large, the input features of the neural network (NN) must include the phase diffusivities (D^i), apart from Z^* and j .

It is important to note that the effective charge numbers predicted through neural network analysis in the present work must be used for the current density values lower than $10.0 \times 10^6 \text{ A/m}^2$. Future work is required to predict the effective charge numbers for electromigration experiments at higher current densities.

6. IMC grain growth at anode interface under electromigration

With the effective charge numbers determined through machine learning for a given current density, the growth of IMC grains at the anode Cu/Sn interface is simulated using 2D phase field simulation for $j =$ (i) $2 \times 10^6 \text{ A/m}^2$, and (ii) $10 \times 10^6 \text{ A/m}^2$. As mentioned in Table 3, at $2 \times 10^6 \text{ A/m}^2$ the NN predicted effective charge numbers for Cu in FCC, IMC and LIQUID are 3.7, 8.7 and 2.8, whereas for Sn in FCC, IMC and LIQUID phase they are 3.8, 12.2 and 2.9. Similarly, at $10 \times 10^6 \text{ A/m}^2$, the effective charge numbers for Cu and Sn are 11.5, 58, 5.8 and 11.75, 68, 5.9 respectively in FCC, IMC and LIQUID phase. These optimized effective charge numbers are used to simulate the growth of anode IMC grains in 2D.

For a square domain of area $300 \text{ nm} \times 300 \text{ nm}$, corresponding to $t=0$ of Fig. 11, the initial geometries of Cu-rich FCC phase is represented by a blue colored rectangular strip of height = 300 nm and width = 100 nm. At the interface of Sn-rich LIQUID phase (red colored region in the figure), two square grains of Cu₆Sn₅ IMC phase (brown colored) are placed. The smaller grain of area $50 \text{ nm} \times 50 \text{ nm}$ is denoted as Gr 1, and the larger one of size $100 \text{ nm} \times 100 \text{ nm}$ is referred to

as Gr 2. For the temperature being maintained at 523.15 K (implying constant D^i), the grains at $t=0.51$ s corresponding to current stressing with $j = 10 \times 10^6$ A/m², are significantly larger than those corresponding to $j=2 \times 10^6$ A/m² even at a later time ($t = 0.59$ s). When the initially square IMC grains are evolving towards a curvature geometry, the driving force for grain growth due to electric field potential is different for different magnitudes of j and Z^* (Z^* of IMC phases predicted from machine learning to increase with j). Thus, with an increase in j , the corresponding rapid increase in effective charge numbers of Sn and Cu in IMC phase, results in a cumulative effect on the growth kinetics. This will not only affect the size of the final grains (Gr 1 and Gr 2), but also their orientations and morphological evolution.

Fig. 12 shows the distribution of electric field potential (ϕ) for the IMC grains evolution corresponding to $j=2 \times 10^6$ A/m² at $t=0.59$ s. Since, the electric resistivities are also phase dependent, the drop in potential across the phases is dependent on the areas of the grains. Since constant current density BC, is implemented at the left and right side of the domain, these criteria must be met at the expense of a voltage drop when the area changes at the junctions between phases/grains ^[57]. It is to be noted that a larger voltage drop is noticed at the vicinity of a smaller grain (Gr 1).

In Fig. 13, the evolution of the individual grains is depicted by means of a graphical presentation of the function $\sum_i (\eta_i)^2$. As the summation of the square of the order parameters leads to unity within the bulk of phases and is less than unity at the interfaces between phases/grains, the graphical plot of $\sum_i (\eta_i)^2$ can be utilized to monitor the changes in morphologies of the two connected grains Gr 1 and Gr 2. The two initially square grains - Gr 1 and Gr 2 of initial sizes of 50 nm $\times$ 50 nm, and 100 nm $\times$ 100 nm respectively (Fig. 11 (at $t = 0$)) are shown to evolve at a different rate, when subjected to current densities of two different magnitudes namely $j=2 \times 10^6$ A/m² (in Fig. 13(a)) and $j=10 \times 10^6$ A/m² (in Fig. 13(b)). Such effects on transient morphology and sizes of the individual grains, can definitely alter the ostwald ripening kinetics ^[58] between Gr 1 and Gr 2.

7. Computation of change in electrical resistance for the multiphase system

From engineering and technological applications viewpoint, the major topic of interest is the overall electrical resistance of the interconnection used in microelectronic devices ^[59] or of the PV ribbon ^[60] used in connecting the Solar Photovoltaic cells. The interconnection and joints with lower electrical resistance are preferable for use in these applications. Although it is possible to

measure the electrical resistance of an overall joint or ribbon, it is quite difficult to experimentally quantify the resistance at the precise interfaces with highly irregular microstructure. The difficulty in the experimental measurement lies in the determination of length and area in the formula $R = \rho \frac{\text{length}}{\text{area}}$, for the irregular and dynamic morphology of interfacial Cu_6Sn_5 compound. This problem can be solved by using the interpolation function of the current phase field model to capture the exact geometrical fraction of a phase at a given time.

The interpolation function defined by Eq. 3 is a function of the square of order parameters, and thus it simply positions a particular phase with respect to all of the phases. For a 2D simulation, computation of the integral area of a phase is the necessary factor for subsequent estimation of phase resistance. Area (S) of a phase i, can be computed in the finite element model using the following formula:

$$S_i = \int h_i dS \quad (33)$$

Initially, the geometrical areas of FCC, IMC and LIQUID phases for the computational domain are 3.0×10^4 , 1.25×10^4 and $4.75 \times 10^4 \text{ nm}^2$ respectively. The temporal evolution of the geometrical areas of the phases are computed from the multiphase field model using Eq. 33 and the results for the two current densities (2×10^6 , and $10 \times 10^6 \text{ A/m}^2$) are presented in Fig. 14. It is obvious that the area of intermetallic compound increases at the expense of areas of LIQUID and FCC phases; and after comparing Fig. 14(a) and 14(b), it can be inferred that the higher the current density, the greater is the rate of IMC area increment. The faster consumption of LIQUID and FCC phase at higher current density causes the IMC to grow at an accelerated manner. For example at $t = 0.8 \text{ s}$ and when $j = 2 \times 10^6 \text{ A/m}^2$, the respective areas of FCC, IMC and LIQUID phases are $1.1681 \times 10^4 \text{ nm}^2$ (decreased), $4.6390 \times 10^4 \text{ nm}^2$ (increased), and $3.1939 \times 10^4 \text{ nm}^2$ (decreased). In context of $j = 10 \times 10^6 \text{ A/m}^2$, when $t = 0.8 \text{ s}$ (same time), the IMC phase has already attained an area of $7.7219 \times 10^4 \text{ nm}^2$, whereas FCC and LIQUID phases are reduced to possess the respective areas of 66 nm^2 and $1.2715 \times 10^4 \text{ nm}^2$. Thus, at higher current density the growth or shrinkage of the phases at anode occurs at a faster rate. This has a severe implication on the electrical resistance of the overall joint.

For the computational domain mentioned in Sec. 6, let R be the total resistance of the multiphase system at time 0 and t respectively. For the current passing from anode to cathode and considering the two IMC grains as a single material system or phase, the different phases can be assumed as resistances in series. So, the overall resistance R is given as:

$$R = \sum_{i=1}^3 R_i = R_{Cu} + R_{IMC} + R_{Sn} \quad (34)$$

The width of the domain $b = 100 \text{ nm}$ is a constant, and for the system let d be the unit depth normal

to the area S_i . With S_{Cu} , S_{IMC} and S_{Sn} being the initial area of FCC, IMC and LIQUID phases respectively, the resistance of the multiphase system at $t = 0$ can be expressed by the following equation:

$$R_0 = \rho_{Cu} \times \frac{S_{Cu}}{b \times d} + \rho_{IMC} \times \frac{S_{IMC}}{b \times d} + \rho_{Sn} \times \frac{S_{Sn}}{b \times d} = \frac{\rho_{Cu}S_{Cu} + \rho_{IMC}S_{IMC} + \rho_{Sn}S_{Sn}}{b^2 d} \quad (35)$$

As the area of phases are evolving, let's consider the temporal values of phase areas at time t as $\bar{S}_{Cu}$, $\bar{S}_{IMC}$ and $\bar{S}_{Sn}$. Then the total resistance at time t is given as:

$$R_t = \frac{\rho_{Cu} \bar{S}_{Cu} + \rho_{IMC} \bar{S}_{IMC} + \rho_{Sn} \bar{S}_{Sn}}{b^2 d} \quad (36)$$

The temporal increment in the overall resistance of the system with respect to the initial resistance ($\frac{\Delta R}{R_0}$) can be then defined as:

$$\frac{\Delta R}{R_0} = \frac{R_t - R_0}{R_0} = \frac{R_t}{R_0} - 1 \quad (37)$$

The incremental change in resistance with respect to initial resistance, can also be expressed in terms of phase areas as:

$$\frac{\Delta R}{R_0} = \frac{\rho_{Cu} \bar{S}_{Cu} + \rho_{IMC} \bar{S}_{IMC} + \rho_{Sn} \bar{S}_{Sn}}{\rho_{Cu} S_{Cu} + \rho_{IMC} S_{IMC} + \rho_{Sn} S_{Sn}} - 1 \quad (38)$$

In Eq. 38, it is interesting to note that under the influence of electromigration, the anode IMC grows at the expense of FCC and LIQUID phases and the sum of total phase areas ($\sum_{i=1}^3 S_i$) is a constant, and has a value of $9 \times 10^4 \text{ nm}^2$. However, as mentioned in Table 2, the electrical resistivity of IMC phase ($1.75 \times 10^{-7} \Omega\text{m}$) is larger than those of FCC ($1.70 \times 10^{-8} \Omega\text{m}$) and LIQUID ($1.10 \times 10^{-7} \Omega\text{m}$) phases. The weighted sum of electrical resistivity and geometrical areas of each phases renders the numerator to be larger than the denominator in context of the first term in the RHS of Eq. 38. Hence, at the anode interfaces the transient and rapid increment of the size IMC compound with the largest resistivity value, will cause the temporal increment in the resistance of the overall multiphase system. Though the cathode side shown in Fig. 1(ii) but not considered in the present numerical model, has no significant presence of IMC, the dissolution of FCC phase (phase with lowest electrical resistivity) into the liquid, also ensures a contribution to relative increase of the net resistance therein, and this remains the topic of future study.

In order to obtain the transient values of $\frac{\Delta R}{R_0}$, the areas of the phase microstructures are post-processed from the computational results of phase field model in accordance to Eq. 33, and these areas are utilized along with the electrical resistivities of the phases in accordance to Eq. 38. The

computed values of $\frac{\Delta R}{R_0}$ has been depicted in Fig. 15 for the current densities of 2.0×10^6 and 10.0×10^6 A/m². The positive values of the fraction in the y-axis of figure indicate that the electrical resistance always increases with time. It is important to note that the increment rate of electrical resistance is larger for higher current density than for lower one. For instance, at $t = 0.8$ s, $\frac{\Delta R}{R_0} = 0.9$ in context of model with $j = 10 \times 10^6$ A/m², whereas even at longer duration ($t = 1.4$ s), $\frac{\Delta R}{R_0} = 0.8$ in case of model with $j = 2 \times 10^6$ A/m². Thus, there is a greater probability of multiple reliability issues with microelectronic devices performing at higher current density.

8. Conclusion

The following conclusions can be derived from this study:

- i. The four paradigms of materials design, namely experiments, theory, computation and data-driven methods are employed in this work to study the interfacial dynamics at the anode side of Cu-Sn-Cu system undergoing electromigration.
- ii. A 2D multiphase field model, including an electromigration driving force, has been developed for calculation of Cu₆Sn₅ IMC layer growth kinetics at the anode solid Cu/Liquid Sn interface at $T=523.15$ K and current densities (j) in the range of $1-10 \times 10^6$ A/m². The phases for the simulation are validated using experimental in-situ synchrotron radiation imaging technology.
- iii. Random sets of Z^* (with values ranging from 2-100), combined with j in the range of $1-10 \times 10^6$ A/m², are considered in a set of 71 phase field simulations. For each set of parameter values, a growth rate constant (k_{em}) of anode side's Cu₆Sn₅ IMC, is determined from 1D simulations.
- iv. An artificial neural network, consisting of 3 hidden layers, is designed with j and Z^* as input features, and the k_{em} obtained from the phase field simulations as the output.
- v. The prediction model of this regression based ANN is then optimized with experimental data of growth kinetics of anode IMC at the same temperature. With this optimization procedure, Z^* values are determined for each j . The effective charge numbers of Cu and Sn are found to increase with j for all phases, but the increase is most pronounced for the IMC phase.
- vi. The Z^* values, predicted in this way from machine learning, are then utilized to model two

Cu₆Sn₅ IMC grains at the anode for the Cu-Sn system undergoing electromigration at current densities of 2×10^6 and 10×10^6 A/m². A cumulative effect of j and the j -dependent Z^* , on anode IMC grain growth, was clearly observed from the 2D multiphase field model performed at two values of current densities.

- vii. The phase dependent electrical resistivities are integrated with the order parameters to evaluate the incremental changes in electrical resistance of the system with time. As IMC phase has the largest resistivity, and its area is increasing at the expense of the other two phases, the rate of increment in resistance of the total system is always positive. The value of $\frac{\Delta R}{R_0}$ is larger at higher current density.
- viii. In future research, the prediction of effective charge numbers for higher current densities ($j \geq 100 \times 10^6$ A/m²), which involve larger temperature rises due to enormous Joule heating, could be aimed by considering T-dependent phase diffusivities as the additional input parameters in the ANN.

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Figure and table captions

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Table 2 Model parameters and material properties used in the multi-phase field simulation.

Table 3 Values of the neural network optimized effective charge numbers at different values of current density. The predicted and experimental growth rate constants are denoted as $k_{em,pred}$ and $k_{em,expt}$ respectively .

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Fig. 8. Pairplot for the effective charge numbers of the Sn component in each phase ($Z_{Sn}^{fcc}, Z_{Sn}^{imc}, Z_{Sn}^{liq}$) and growth rate constant (knmps, k_{em} in $\frac{nm}{s}$) for 2 ranges of current density (j in $\frac{A}{mm^2}$).

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Table list:

Table 1 Coefficients and equilibrium composition for parabolic free energies of phases.

Phase/ Properties	A^i (J/mol)	B^i (J/mol)	C^i (J/mol)	c_{eq}^i
FCC	3.1360E+05	-1.67168E+04	-2.471215E+04	0.15
CU6SN5 (IMC)	1.96604e+06	-1.67168E+04	-2.953436E+04	0.436 ^a / 0.460 ^b
LIQUID	4.65512E+04	2.054E+03	-2.8749E+04	0.9467

^aIMC in equilibrium with FCC , ^bIMC in equilibrium with LIQUID.

Table 2 Model parameters and material properties used in the multi-phase field simulation.

Properties	Values
D^{liq}	$4.95 \times 10^{-10} \text{ m}^2/\text{s}$
D^{fcc}	$3.09 \times 10^{-16} \text{ m}^2/\text{s}$
D^{imc}	$4.07 \times 10^{-15} \text{ m}^2/\text{s}$
D_{gb}	$8.0 \times 10^{-13} \text{ m}^2/\text{s}$
N_a	$6.022 \times 10^{23} \text{ mol}^{-1}$
σ	0.5 J/m^2
grid space 1D (Δx)	$1.43 \times 10^{-9} \text{ m}$
δ (1D)	$1.0 \times 10^{-8} \text{ m}$
m (1D)	$3.0 \times 10^8 \text{ J/m}^3$
κ (1D)	$5.3625 \times 10^{-10} \text{ J/m}$
grid space 2D ($\Delta x = \Delta y$)	$3.75 \times 10^{-9} \text{ m}$
δ (2D)	$2.5 \times 10^{-8} \text{ m}$
m (2D)	$1.2 \times 10^8 \text{ J/m}^3$
κ (2D)	$9.375 \times 10^{-10} \text{ J/m}$
ρ_{Sn}	$1.10 \times 10^{-7} \text{ }\Omega\text{m}$
ρ_{Cu}	$1.70 \times 10^{-8} \text{ }\Omega\text{m}$
ρ_{IMC}	$1.75 \times 10^{-7} \text{ }\Omega\text{m}$

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j (A/m ²)	Z_{Cu}^{fcc}	Z_{Sn}^{fcc}	Z_{Cu}^{imc}	Z_{Sn}^{imc}	Z_{Cu}^{liq}	Z_{Sn}^{liq}	$k_{em,pred}$ ($\mu\text{m}/\text{min.}$)	$k_{em,expt}$ ($\mu\text{m}/\text{min.}$)
1×10^6	2.1	2.15	2.1	2.25	2.02	2.03	0.11	0.098
2×10^6	3.7	3.8	8.7	12.2	2.8	2.9	0.194	0.194
3×10^6	4.3	4.9	15.0	23.0	3.3	3.4	0.3	0.294
4×10^6	5.4	5.9	26.0	36.0	3.85	4.2	0.398	0.392
5×10^6	6.6	6.9	30.0	39.0	4.3	4.7	0.47	0.49
6×10^6	8.0	8.5	36.0	46.0	4.5	4.9	0.57	0.588
7×10^6	9.75	10	42.0	52.0	5.1	5.2	0.677	0.686
8×10^6	9.9	10.1	48.0	56.0	5.2	5.2	0.791	0.784
9×10^6	10.0	10.25	52.0	62.0	5.2	5.3	0.897	0.882
10×10^6	11.5	11.75	58.0	68.0	5.8	5.9	1.0	0.98

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