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Molecular dynamics simulation of atomic diffusion in friction stir spot welded Al to Cu joints

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Abstract

Dissimilar metals joining, especially Aluminium (Al) to copper (Cu), have gained importance in batteries for electric vehicles. Although friction stir spot welding (FSSW) has recently been used for welding dissimilar materials, progress has been very slow towards understanding the effect of temperature on diffusion condition between the two materials with the same FCC crystal structure. The thermo-mechanical modelling has been used to define the trajectory of Al and Cu particles at the weld interface, but it had a limitation to quantified the diffusion coefficient. Hence, the molecular dynamics (MD) study has been used to investigate the atomic interdiffusion of Al and Cu. The transmission electron microscopy results are used to validate the MD simulation outcome to understand the formation of dislocations and intermetallic compounds. The MD results implicated the formation of $\gamma$-phase (BCC), i.e., Al$_4$Cu$_9$ IMC towards the Cu side. Further, the In-situ investigation of non-FCC phase formation at FSSW condition has also been studied.

Keywords

Modelling, dynamic, simulation, Diffusion, dislocations, friction, welding, dissimilar

1. Introduction

The friction stir spot welding (FSSW) has recently gained significant importance in overlap joining of dissimilar materials\textsuperscript{1,2}, which is a variant of solid state friction stir welding (FSW) originally developed for difficult to fusion weld materials such as Al-alloys\textsuperscript{3–8}. Among such, aluminum (Al) and copper (Cu) welded joints have potential applications in battery pack fabrication\textsuperscript{9}. The current collectors (tabs) of the Li-ion battery are welded to make a series and parallel combinations\textsuperscript{9}. The higher conductive properties of Al and Cu needed an optimum process condition to generate a sufficient amount of heat to plasticise the weld materials for good quality welds. In battery applications, the quality of weld joint is mainly asse by their strength and electrical conductivity properties\textsuperscript{10,11}. These properties depend on various aspects like good material mixing, atomic diffusion between Al and Cu, and formation of dislocations.
These dislocations are nothing but short-circuit defects and strongly influence the interdiffusion process, where the diffusion coefficient of atoms is enhanced along with these defects. The short-circuit diffusion in the dissimilar materials enhances the intermetallic compound (IMC) formation at the weld interface. The atomic interdiffusion by the solid-state leads to the formation of epitaxial Al-rich (Al$_2$Cu [002]) and Cu-rich (Al$_4$Cu$_9$ [113]) IMC. The molecular dynamics simulation (MD) is an effective method to investigate this microscopic phenomenon like diffusion and IMC formation in a system that is placed under given process conditions. Coming to the MD simulation, there are two different scenarios that have been used to apply friction stir welding conditions to a given system, i.e., 1) Al-Cu materials are placed in overlap condition with a compressive force and applied a temperature of 800 K, and 2) Al-Fe materials are placed in overlap conditions with a compressive force of 600 MPa, in addition, a temperature of 890 K and a relative sliding speed of 0.5 m/s is applied to the system. Since, in the present study, MD simulation has been carried out with scenario 1, wherein actual FSSW condition there will be no relative sliding motion will occur. The current research focused on the interaction between Al and Cu materials atoms during FSSW condition in the solid-solid interface using numerical modelling and MD simulation for microanalysis. The MD simulation has been carried out with scenario 1 because, in the actual FSSW condition, there will be no relative sliding motion between Al and Cu. Moreover, the study also focuses on the formation of non-FCC phases that have not been detailed in earlier literature.

2. Experimental procedure

In continuation, a Lagrangian implicit method has been used to create a 3-D thermo-mechanical FSSW model for Cu-Al weld in lap configuration in DEFORM 3D. The base materials (BM), i.e., Cu and Al, are assumed as two different continuum rectangular bodies of dimensions 30 mm × 30 mm × 0.4 mm each, and are referred to as a rigid viscoplastic material, as shown in Fig 1a. A total of 53,470 elements of tetrahedral were utilized in meshing each plate. The both FSSW tool and the anvil plate are defined as rigid objects, and H13 is used as a tool material. The dimensions of the tool are 10 mm shoulder diameter, 0.4 mm tool pin length, and with a 5 mm tool pin diameter.

The material undergoes from solid to viscous state; therefore, flow stress is determined for Cu based on strain rate, temperature, and flow stress, as given in Equation 1. The flow stress, stress-strain behaviour is defined at different strain rates (4–40/s), and temperatures (20–899 °C) from DEFORM material library are used as input to the simulation for Cu.

\[ \sigma = \bar{\sigma}(\varepsilon, \dot{\varepsilon}, T) \]  

where, \( \sigma, \varepsilon, \dot{\varepsilon}, T \) are flow stress, strain, strain rate, and temperature.
To determine the flow stress of the Al, the Sellers Tegart material model was employed. It is expressed in Equation 2. It depends on both, i.e., temperature and strain rate. In the expression, Z denotes the Zener-Hollomon parameter, which is expressed in Equation 3.

$$\bar{\sigma} = \frac{1}{\alpha} \ln \left( \frac{(Z/\alpha)^{1/n}}{A} \right) + \left[ 1 + \left( \frac{Z/\alpha}{A} \right)^{1/n} \right]$$  \hspace{1cm} (2)

$$Z = \dot{\varepsilon} \exp \left( \frac{\Delta H}{RT} \right)$$  \hspace{1cm} (3)

where, $\bar{\sigma}$, $\dot{\varepsilon}$, $T$, $\Delta H$, $R$ are effective stress, effective strain rate, temperature, activation energy, and gas constant respectively. The values of material constants, i.e., $\Delta H$, $R$, $\alpha$, $n$ are tabulated in Table 1.

The Al and Cu micro sheets are welded by using a friction stir welding process in lap configuration. Fig 1b represents a weld sample cross-section with a process parameter condition, i.e., a tool rotational speed ($\omega$) of 1500 rpm, the tool plunges into the Al of 0.2 mm, and dwelling time of 15 sec. The simulation weld morphology is validated with the experimental scanning electron microscopy result, as shown in Fig 1c. At the same time, Figs 1b, c shows a slope at the joint interface just beside the pin diameter, which is due to the extrusion of plastic material while the tool is plunging into the workpiece. The rotating and forging actions of a tool over the workpiece generate frictional heat and diffusion of atoms occurs, respectively, which leads to the joining of two base materials. The cross-section of the weld interface (nugget zone) of sample is subjected to investigated under transmission electron microscope (TEM), in order to identify the formation of intermetallic compounds and dislocations. In the welding process, generated frictional heat is modelled and depicted in Figs 1d, and e. The maximum temperature is generated at the weld nugget zone (NZ), i.e., in a temperature range of [486–535] C, and the simulation result is validated with the experimental work by using a thermal imager. The observed temperature value is at 498 C, as shown in Fig 1f. The obtained peak temperature has been fed into the MD simulation for further analysis.

The molecular dynamics simulation study is performed by using LAMMPS packages. The interatomic interaction of Al and Cu is defined by using an embedded atom method potential (EAM). The simulation box size is 51.6 A$^0 \times 108.46$ A$^0 \times 108.46$ A$^0$, where the x plane is considered a shrink-wrapped boundary, and the y-z plane is a periodic boundary. In the solid-solid interface, the heating process in each simulation is done by using a Nose-Hoover thermostat at a specific temperature of 300 K and a time step of 1 fs. The initial velocity of Al and Cu atoms are set to obey the Gaussian distribution. The system is modelled with FSSW conditions, i.e., the gradual increase in temperature at a rate of 10 K/ps from 300 K to the maximum temperature of 771K using canonical ensemble moles (N), volume (V), and temperature (T) (N-V-T), and after reaching 771K a compressive strain rate of 1E-3 s$^{-1}$ is applied to the total system. The diffusion system is cooled from 771 K to 300 K with the same rate of 10 K/ps.
3. Results and discussion

Based on the above-mentioned applied conditions, the mutual diffusion of atoms is explored near the Al and Cu interface. The diffusion is considered along the perpendicular direction to the interface, i.e., x-direction, as depicted in Fig 2a, b. The intense plastic deformation during the FSSW may lead to several crystal defects like vacancy and dislocations. The dislocation analysis for the MD simulation has been studied by using the OVITO software. Fig 2c illustrate the presence of dislocations in the NZ of the Al-Cu weldment. The grain refinement in the NZ increases the grain boundary, which simultaneously decreases the depth of diffusion facilitating the formation of intrinsic dislocations as depicted in Fig 2d through bright-field transmission electron microscopy (TEM). Due to further extreme plastic deformation, dislocation multiplications accelerate to the dislocation loops, as shown in Fig 2d.

The material mixing plays an important role in the diffusion of the FSSW. Hence, it is traced by considering two points in the same plane along the thickness direction, shown in the left-hand corner of Fig 3a for both Al and Cu. Among them, one point is located between the tool pin (T_{pa}) and workpiece, and another point is just beside the tool pin contact area. During the plunging of a tool, points in the scope of the contact area are displaced towards Al. The point located in the T_{pa} and Cu thickness plane, i.e., the Cu_P1 point, is displaced more than the Al_P1. The higher displacement is due to more forging action of the tool over the Cu workpiece than over Al during the plunging of the tool.

In contrast, the trajectory of a point located beside the T_{pa}, in the Al thickness plane, i.e., Al_P2, observes less displacement during plunging time. However, as the dwelling starts, displacement of Al_P2 becomes higher as compared to Cu_P2. This is because the Al atom has a larger atomic radius (1.43 Å) than that of the Cu atom (1.27 Å). The temperature generated during the dwelling phase can easily break the bond of an Al atom from its neighbour, and trigger diffusion leads to the formation of θ-Al_{2}Cu at Al side. Sometimes, the short-circuit diffusion enhances the Al atoms to penetrate into the Cu side and leads to the formation of γ-Al_{4}Cu_{9} at the Cu-side. The growth of IMC with respect to time and temperature in the Al-Cu system is represented in the form of a time-temperature transformation (TTT) curve, shown in Fig 3b. With the diffusion of Al [0\overline{1}\overline{1}] and Cu [1\overline{1}\overline{2}], the nose of the C-curve determines the start position of the IMC formation. Fig 3b shows the formation of 0-Al_{2}Cu [002] and \gamma-Al_{4}Cu_{9} [113] IMCs at a temperature range of 350 °C to 420 °C, which is lower temperature according to the present experimental condition, i.e., 498 °C. Hence, these two IMC are developed in the system. The nose of 0-Al_{2}Cu appears early, which concludes early formation in the Al-Cu welding system. The crystal structure of the IMC 0-Al_{2}Cu has 22 atoms (i.e., 12 Al and 10 Cu), and \gamma-Al_{4}Cu_{9} has 52 atoms (i.e., 16 Al and 36 Cu) as illustrated in Fig 3b.

The mean square displacement (MSD) of Al and Cu is calculated using LAMMPS to quantify the mobility of atoms during FSSW. MSD data is collected for every 1000 steps to get the mutual diffusion of atoms at the interface. The MSD is computed by Equation 4.  

\[ MSD = \langle |r(t) - r(o)|^2 \rangle, \]  

(4)
where \( r(o) \) and \( r(t) \) are the positions of Al and Cu atoms at time \( t = 0 \) and \( t = t \), respectively.

The diffusion coefficients of Al and Cu atoms are computed by using Newton’s equation of motion with a leap-frog algorithm \(^{36}\) based on their respective positions as a function of time, as shown in Equation 5 \(^{35}\).

\[
D = \frac{1}{6} \lim_{t \to \infty} \frac{dMSD}{dt}
\]  

The MSD of Al and Cu atoms exists due to the formation of a coherent interface \(^{37}\), where Al and Cu have similar FCC crystal structures. Fig 3c shows the MSD curves for both Al and Cu, where the change in slope of the curves is non-zero. This is due to the transfer of energy that takes place due to the interaction between the atoms. Accordingly, the diffusion coefficient is calculated and found that the \( D_{Al} \) and \( D_{Cu} \) are \(3.65 \times 10^{-8} \text{ cm}^2/\text{s} \) and \(1.505 \times 10^{-8} \text{ cm}^2/\text{s} \), respectively. However, the Al atoms diffuse more profoundly into the interior of Cu at a lower ratio due to the lesser atomic radius of Cu, leading to the formation of \(\gamma\)-Al\(_4\)Cu\(_9\) IMC. The bright-field TEM images of Fig 3d, and e depicted the shape of the \(\theta\)-Al\(_2\)Cu and \(\gamma\)-Al\(_4\)Cu\(_9\) IMCs, and conclude that the size of the crystal structure of \(\theta\)-Al\(_2\)Cu is smaller than that of \(\gamma\)-Al\(_4\)Cu\(_9\), where \(\gamma\)-Al\(_4\)Cu\(_9\) is a quasicrystalline IMC \(^{38}\), also known as a complex metallic alloy, with excellent mechanical strength, electrical conductivity, and corrosion resistance \(^{39}\).

A modifier adaptive common neighbour analysis (a-CAN) in OVITO software can distinguish the crystalline structure \(^{40}\) of Al and Cu during the FSSW conditions. All Al and Cu atoms are of face-centred cubic (FCC) structure in the initial state, as shown in Fig 4a. The eutectoid reaction of the Al-Cu system takes place at higher Cu % [~ 70 to 80%] at a temperature of about ~770K resulting in the formation of metastable \(\beta\) phase. Further, the \(\beta\) phase undergoes martensitic transformation temperature on heating (MTTH) to form 3 disordered phases, i.e., \(\beta'\), \(\beta'_1\), and \(\gamma'\). \(\beta'\) phases are formed at the lower and higher Al concentrations, respectively. The martensite phases \(\beta'\) and \(\gamma'\) in the system inherit the BCC and HCP ordering, respectively \(^{32}\). Fig 4b illustrates the MTTH condition, where the initial FCC atoms undergo several disorders to form distinct non-FCC crystalline structures, i.e., BCC and HCP \(^{41}\). From Fig 4c, it is observed that more HCP crystalline structures are formed towards the Al side. \(\gamma\)-Al\(_4\)Cu\(_9\) phase is composed of two 26 atoms clusters with an ordered BCC crystal structure. Hence, after removing FSSW conditions over the system, Fig 4c shows BCC structures on the Cu materials. This concludes the Cu-rich IMC is formed towards the Cu material side. The orientation relationship (OR) models like Kurdjumov-Sachs (KS) and Nishiyama-Wassermann (NW) report a relation between FCC and BCC. According to these models, the closed-packed [111] FCC is parallel to the most densely packed [110] BCC. The OR models for the Al-Cu system are KS: [111] FCC // [110] BCC ; <110> FCC // <111> BCC and NW: [111] FCC // [110] BCC; <112> FCC // <110> BCC \(^{42,43}\). The difference between these two models is considering the direction to which parallel planes face. During the FSSW, the absence of chemical reaction between Al and Cu or crystallization leads to the formation of amorphous phases. Fig 4d depicts that the amorphous transition layer creates a boundary between Cu [112], and \(\gamma\)-Al\(_4\)Cu\(_9\) [113] IMC. The amorphous structure is confirmed using a
selective area electron diffraction (SAED) pattern, and the same is shown in Fig 4e. The SAED patterns for the Cu and $\gamma$-Al$_4$Cu$_9$ are shown in Fig 4f and g, respectively.

4. Conclusions

The results and analysis described above show that an effective thermo-mechanical model for FSSW of dissimilar materials is developed using DEFORM 3D. The temperature obtained from the macro simulation is validated and directly feed into the MD simulation. In addition to temperature, the compressive shear strain is given to the system to mimic the FSSW process parameter conditions. The intense plastic deformation may lead to the formation of dislocations, and further, it converts to a dislocation loop. The diffusion behaviour is studied along the x-direction for macro thermo-mechanical modelling and MD simulation with EAM potential. The macro simulation result shows that the small quantity of Al particles is displaced deeper into the Cu matrix. Similarly, from the MD simulation, it has been found that the diffusion coefficient of Al is 59% higher than that of Cu. The movement of Al and Cu particles during the dwelling stage is more than the plunging state. The complex metallic alloy is formed due to the diffusion of Al in the Cu matrix, i.e., the $\gamma$-Al$_4$Cu$_9$ IMC phase. However, improper diffusion may lead to the formation of amorphous crystal structures, which are unidentified phases. The BCC, $\beta'$ phase, is found towards Cu-side and concludes it is the crystal structure of $\gamma$-Al$_4$Cu$_9$.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

The MD simulation is carried out in this work by using the Supercomputing facility of IIT Kharagpur established under National Supercomputing Mission (NSM), Government of India and supported by Centre for Development of Advanced Computing (CDAC), Pune.

Data availability statement

The datasets generated during and/or analysed during the current study are available from the corresponding author on reasonable request.

Reference


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Table 1: Material constants of Sellars Tegart model

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Figures:

Fig 1. (a) The meshed assembly of Cu-Al and tool setup (side view), cross-sectional view of the weld samples (b) simulation, (c) experimental weld morphology, temperature contours (d) weld interface, (e) enlarged weld interface showing different weld zones, and (f) temperature measured at the weld nugget zone using the thermal imager.

Fig 2 (a) Initial simulation box highlighting the Cu particles at the interface, (b) diffusion of atoms at the weld interface, formation of dislocations and dislocation loop.
loops in the weld zone (c) from the MD simulation using OVITO software, (d) bright-field TEM image for the welded sample.

Fig 3 (a) Specific particles trajectory pointed on Cu and Al, (b) TTT curves and crystal structure along with the lattice indices for the IMC formed at the weld interface, (c) MSD for AL and Cu atoms, and bright-field TEM image to show IMC (d) Al$_2$Cu and (e) Al$_4$Cu$_9$ and
Fig 4 (a) Initial FCC phases of Al and Cu atoms, (b) formation of different phases during the martensitic transformation temperature on heating condition, (c) phase formation after reaching the equilibrium state, (d) representing the amorphous transition layer, selective area electron diffraction images for (e) amorphous layer, (f) recrystallized Cu, and (g) γ-Al₄Cu₉