Effect of Enhanced Atomic Diffusion on Microstructure Evolution of Heterogeneous Friction Welded Joints

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Abstract. In this paper, the dissimilar pure metal diffusion couples with different metallurgical compatibility (Cu/Ni miscible couple, Mg/Ti immiscible couple) were designed and a physical simulation apparatus of rotation friction welding was used to investigate the diffusion of the heterogeneous interface during severe plastic deformation. The experimental results showed that an interface layer which presented the gray gradient transition characteristics appeared at Cu/Ni interfaces. The diffusion rate was calculated and was $10^4$ times of that of thermally activated at the same temperature for Cu/Ni couple. The enhanced diffusion was also observed on the Mg/Ti friction welded joints. The diffusion coefficients under different strain rate were obtained and the phenomenological law between diffusion coefficient and strain rate were analysed. A linear relationship between diffusion coefficient and strain rate was obtained.

1. Introduction
The joining of dissimilar metals has always been a hot topic in modern manufacturing industry [1]. A sound joint may provide interesting combinations of structural and functional properties [2]. Friction welding (FW) is the most promising method to join dissimilar materials. In previous works, dissimilar metals such as Al/Fe [3], Al/Ti [4], Al/Cu [5], and Ti/Fe [6] of the friction welding have been carried out by many researchers. It is generally known that the materials near the joint have simultaneously encountered high temperature ($0.5 \sim 0.7 T_m$) and severe plastic deformation (SPD) (strain rate $10 \sim 1000\text{s}^{-1}$) during FW process [7]. The metallurgical interactions on the heterogeneous interface during the SPD of FW are critical for the joint strength and stability, which has become a key issue in the field of friction welding research [8-9]. In particularly, the diffusion behaviour has a very important significance on the understanding of the metallurgical bonding mechanism, since it is the smallest scale of dynamic behaviour.

The phenomenon of the deformation promoting the diffusion has been concerned by Balluffi et al in 1963 [10]. Since then, it was found in the process of the mechanical alloying [11], accumulated rolling welding and friction stir welding [12] and a series of SPD process. Experimental data show that the diffusion coefficient is $10^3 \sim 10^8$ times of the thermal activation diffusion under the condition of same temperature, which called superdiffusion in physics. There were two perspectives on the generation mechanism of the superdiffusion generally. Xu et al [13] think that the surge of vacancy defects in the process of SPD is the primary reasons of super-diffusion. Another perspective suggests that the deformation process is a ballistic relocation of atoms and independent of temperature. Bellon and Odunuga et al [14-15] proposed the analytical expressions of ballistic activated diffusion coefficient with the aid of richardson marked particle in fluid mechanics, as thus given by:
\[ D_b = \frac{C'}{3\pi} b R \varepsilon \]  

Among them, the \( D_b \), \( b \), \( R \) is the ballistic diffusion coefficient, burgers vector and the atom spacing on both sides the dislocation respectively, \( C' \) is a constant related to the system. These conclusions are well tested and verified based on molecular dynamics and monte carlo method in numerical simulation at the same time [16–17]. However, the physics mechanism of superdiffusion during the friction welding process and its influence on joint microstructure evolution has not been clarified.

In this paper, the dissimilar pure metal with different metallurgical compatibility (Cu/Ni miscible couple, Mg/Ti immiscible couple) were designed and a physical simulation apparatus of rotation friction welding was used to investigate the interface diffusion during processing of SPD provided by FW. The phenomenological law between diffusion coefficient and strain rate were analysed. The effects of superdiffusion on joint microstructure evolution were analysed.

2. Experimental procedure
Commercially available bars of Cu (99.9 wt. %), Ni(99.9 wt. %), Mg(99.95 wt. %) and Ti(99.7 wt.%) were used. The specimens were machined with a dimension of 20 mm in joining part and 14 mm in clamping part. The rotation friction welding process was carried out on a physical simulation apparatus which shown in Figure 1. During the friction welding operations, the rotation speeds of friction welding was 1900rpm for Cu/Ni and 1200rpm for Mg/Ti couple, the different axial pressure was used as 20, 28, 36, 44, 52 MPa, with friction time 5s. In the meanwhile, the interface temperature was observed by the thermocouple embedded in the fixed bar 1.5mm from the interface.

After joining, the welded sample for metallographic examination was cut perpendicular to the welding interface. The samples were subjected to metallographic characterization employing scanning electron microscopy (SEM, JEOL, JSM-6700) equipped with an energy-dispersive spectrum (EDS) and transmission electron microscope (TEM, JEM-3010).

3. Results and discussion
3.1 The morphologies of joint cross section
The defects free joints were obtained by the friction welding simulation experiments. The typical interface morphology of Cu/Ni and Mg/Ti rotary friction welding joints were shown in Figure 2. The Cu/Ni interface shows the gray gradient transition characteristics with a thickest diffusion layer, as shown in Figure 2a. This reflects the coupling effect of thermally activated and deformation activated diffusion in this miscible alloy systems. For Mg/Ti joints, the good bonding on the interface was formed although the interface transition region was not obvious as shown in Figure 2b.
3.2 The superdiffusion in the SPD process of FW

3.2.1 The atom concentration distribution. In order to obtain more detailed microstructures of the interfacial transition layer, the images of larger magnification were used and the line scan and point scanning component analysis across the interface were been detected. Figure 3 shows the typical micro-morphology and the concentration profiles of elements of the Cu/Ni joining interface. Figure 3a and 3c show the detailed microstructures by SEM and TEM. The interface shows gradient transition characteristics in gray scale with different element content. A continuous solid solution was formed between Cu and Ni element. Figure 3b and 3d show the compositions and the concentration profiles of Cu and Ni element across the interface by EDS measurements with the electron beam diameter about 25 nm. The scanning positions and path were shown in Figure 3a. The composition analysis shows an obvious interdiffusion between Cu and Ni element occurs during FW process. The compositions obtained by point scan were consistent with the concentration profiles across the interface obtained by line scan, as shown in Figure 3a. The interdiffusion layer width could be roughly calculated according to concentration profiles, with the 8~10% content of solute elements as starting position of the measurement. Figure 4 shows the typical micro-morphology and the concentration profiles of elements of the Mg/Ti joining interface. The micro-morphology of Mg/Ti couples shows obvious interfaces in SEM image, as shown in Figure 4a. Further analysis obtained by TEM proved the existence of atomic diffusion in Mg/Ti joining interfaces. The microstructures of Mg/Ti interface present more clear boundaries in Figure 4b.

Figure 2. The typical interface morphology of joint: (a) Cu/Ni; (b) Mg/Ti
3.2.2 The interface temperature and the strain rate of the plastic region. During the FW process, the temperature of the interface region during the FW process was detected by the thermocouple embedded in the fixed bar 1.5mm from the interface. The temperature curves have an unsteady ascent stage and then become relatively stable stage. The average temperatures (shown in Table 1) were regarded as the diffusion temperature used in the calculation below. The strain rate of diffusion region was calculated according to the friction welding thermal coupling analytical model in references [18].

3.2.3 The interdiffusion coefficient of heterogeneous friction welding interface. It is reasonable that the atomic mixing during the severe plastic deformation may be enhanced by the following three factors during the FW process: increase of temperature due to plastic deformation and shear friction; a reduction in atomic diffusion distance by microstructural refinement; and an increase in the density of lattice defects such as vacancies, dislocations and grain boundaries. When the surface concentration
constant in the Semi-infinite diffusion couple. The relation between the diffusion distance, $x$, and interdiffusion coefficient, $D$, is generally described for a given period of time, $t$, as Eq. (2) [19]:

$$x = \sqrt{Dt}$$  

(2)

According to the Eq (2), the interdiffusion coefficients and the strain rate under different parameters for Cu/Ni and Mg/Ti systems were calculated, as shown in Table 1.

Table 1. The diffusion coefficient, strain rate and average temperature for Cu/Ni and Mg/Ti couples

| Diffusion | Strain rate (s$^{-1}$) | Average temperature (°C) | Diffusion coefficient ($10^{-12}$m$^2$/s) |
|-----------|------------------------|--------------------------|------------------------------------------|
| Cu/Ni     | 878                    | 607                      | 1.99                                     |
|           | 1087                   | 612                      | 2.09                                     |
|           | 1429                   | 634                      | 3.24                                     |
|           | 1969                   | 645                      | 4.11                                     |
|           | 2803                   | 659                      | 6.01                                     |
|           | 708                    | 392                      | 0.79                                     |
|           | 959                    | 415                      | 1.17                                     |
| Mg/Ti     | 1180                   | 427                      | 1.36                                     |
|           | 1484                   | 438                      | 1.62                                     |
|           | 1705                   | 450                      | 2.03                                     |

It can be seen that as the strain rate increases, the interdiffusion coefficient is continuously increasing. By fitting the data in Table 1, the curve of the interdiffusion coefficient as a function of strain rate is obtained as shown in Figure 5.

By analyzing the effect of different strain rate on the thermal-mechanical coupled diffusion coefficient $D_s$ of Cu/Ni, Cu/Fe and Mg/Ti, a linear relationship of the form $y=A+Bx$ between $D_s$ and strain rate was obtained. By extrapolating this formula, the contribution of thermally activated diffusion during friction severe deformation can be estimated when the strain rate is equal to zero. In other words, the first part $A$ in the formula presents the thermally activated diffusion coefficient $D_t'$. The second part $Bx$ can be considered as the contribution of deformation activated diffusion.

Figure 5. Interdiffusion coefficients as a function of strain rate and the fitting coefficient

From the perspective of non-equilibrium thermodynamics, the thermal-mechanical coupled diffusion process can be regarded as a driven system in a nonlinear non-equilibrium state. The dislocation-related micro-regions can be dealt with as a non-equilibrium steady-state based on the local equilibrium hypothesis. The new defect in deformation results in the arrangement of atomic space more disorder, but the heat activated diffusion induce the arrangement of atoms tends to be ordered. Under the influence of this pair of contradictory and mutually offsetting factors, the moving speed of the movable defects (mainly dislocations) in the micro system is kept constant, while the
temperature and strain rate remains constant in the macro system. Therefore, the movement speed, \( v \), of defects (particularly dislocations) is proportional to the deformation activation diffusion, \( D_b \), it can be expressed as:

\[
D_b = k_1 v
\]  

(3)

where \( k_1 \) is the proportionality constant. In addition, since the relationship between \( v \) and strain rate can be expressed as [20]:

\[
\dot{\varepsilon} = b \rho_m \nu
\]  

(4)

where \( b \) and \( \rho_m \) are the Burgers vector and the moving dislocation density, respectively. The expression of \( D_b \) can be obtained by substituting Eq. (4) into Eq. (3):

\[
D_b = \frac{k_1}{b \rho_m} \dot{\varepsilon}
\]  

(5)

Comparing Eq. (1) with Eq. (6), it can be found that the deformation activation diffusion, \( D_b \), is proportional to strain rate despite the differences in the representation of the proportional constants, which shown as:

\[
D_b = k_2 \dot{\varepsilon}
\]  

(6)

where \( k_2 \) is the proportionality constant. Based on the thermal-mechanical coupled diffusion law of the Cu/Ni and Mg/Ti systems in Figure 5, it can be found that their diffusion coefficient \( D_b \) and strain rate follow the linear distribution although the systems have different metallurgical compatibility. This thermal-mechanical coupled superdiffusion actually involves thermal activation diffusion and deformation activation diffusion. And the presence of a linear additive relationship indicates that the existence of two types of diffusion in thermal-mechanical coupled superdiffusion is a linear combination. Therefore, the superdiffusion coefficient \( D_s \) can be expressed as:

\[
D_s(T, \dot{\varepsilon}) = D_s(T) + D_b = D_s(T) + k_2 \dot{\varepsilon}
\]  

(7)

4. Conclusions

The defects free joints were obtained by the friction welding simulation experiments. The interface of the Cu/Ni appears thickest diffusion layer with the gray gradient transition characteristics. The enhanced diffusion was also observed on the Mg/Ti friction welded joints. The diffusion coefficients under different strain rates were obtained.

A linear relationship between the thermo-mechanical diffusion coefficient and strain rates was obtained. This thermal-mechanical coupled diffusion actually involves thermal activation diffusion and deformation activation diffusion. A theoretical model of atom diffusion in the SPD interface of friction welding was established.

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6. References

[1] Guo W, You GQ, Yuan GY and Zhang, XL 2017 J. Alloys. Compd. 695: 3267–3277
[2] Guo YN, Attallah MM, Chiu Y, Li HY, Bray S and Bowen P 2017 Mater. Charact. 127: 342–347
[3] Taban E, Gould JE and Lippold JC 2010 Mater. Des.31(5): 2305–2311
[4] Meisnar M, Baker S, Bennett JM, Bernad A, Mostafa A, Resch S, Fernandes N and Norman A 2017 Mater. Des. 132: 188–197
[5] Avettand-Fenoël MN, Racineux G, Debeugny L and Taillard R 2016 Mater. Des. 98: 305–318
[6] Li X, Li JL, Liao ZX, Jin F, Zhang FS and Xiong JT 2016 Mater. Des. 99: 26–36
[7] Wanjara P and Jahazi M 2005 Metall. and Mater. Trans. A 36A: 2151–2164
[8] Sakai T, Miura H, Goloborodko A and Sitdikov O 2009 Acta Mater 57 (1): 153–162
[9] Morisada Y, Imaizumi T and Fujii H 2015 Scr. Mater. 106: 57–60
[10] Fukumoto S, Tsubakino H, Okita K, Aritoshi M and Tomita T 2000 Scr. Mater. 42 (8): 807–812
[11] Balluffi RW and Ruoff AL1963 J. App. Phys. 34 (6): 1634–1647
[12] Qian JW, Li JL, Xiong JT, Zhang FS and Lin X 2012 Mater. Sci. Eng. A 550: 279–285
[13] Xu Y and Chen J 2015 J. Mech. Phys. Solid 75: 45–57
[14] Bellon P, Averback RS, Odunuga S, Li Y and Krasnochtchekov P 2007 Phys. Rev. Lett. 99 (11), 110602: 1–4
[15] Odunuga S, Li Y, Krasnochtchekov P, Bellon P and Averback RS 2005 Phys. Rev. Lett. 95 (4), 045901: 1–4
[16] Schwen D, Wang M, Averback RS and Bellon P 2013 J. Mater. Res. 28 (19): 2687–2693
[17] Kuksin AY and Yanilkin AV 2015 Mech. Solid. 50 (1): 44–51
[18] Xiong JT, Li JL, Wei YN, Zhang FS and Huang WD2013 Acta Mater 61(5): 1662–1675
[19] Shewmon P 1989 Diffusion in solids. 2nd ed. Warrendale, PA: The Minerals, Metals & Materials Society.
[20] Nemat–Nasser S and Li Y 1998 Acta Mater 46 (2): 565–577