Diffusion in Gold-rich Gold-Aluminium Alloys

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The tracer diffusion coefficient of ^{198}Au in gold-rich Au-Al alloys containing up to 2 at% Al at 1023 K has been determined by the serial sectioning method. The linear enhancement factor of the solvent diffusivity has been determined to be 53 ± 9 . In addition to the above, the interdiffusion and intrinsic diffusion coefficients in gold-rich Au-Al alloys containing up to 6 at% Al at 1023 K have been determined by use of semi-infinite couples and thin-plate sandwiches with the Kirkendall marker. Using the value of self-diffusion coefficient in pure Au and the extrapolated values of interdiffusion coefficient and intrinsic diffusion coefficient of Au to the infinite dilution of Al, the vacancy flow factor has been estimated to be -0.78 . From the linear enhancement factor and the vacancy flow factor, an appropriate numerical set of the correlation factor and the jump frequency ratios in the five-frequency model for the impurity diffusion of Al in Au has been evaluated. The result suggests that the attraction between an Al atom and a vacancy in Au is highly strong and that a vacancy makes very many consecutive exchanges with an Al atom rather than with any solvent atom neighboring the vacancy.

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I. Introduction

Information on the diffusion behavior of gold-aluminium alloys is indispensable for the semiconductor technology, because the thermal bonding between gold thin wire and aluminium thin film metallized on a substrate is a fundamental technique and failure in the bonding is a serious problem. Therefore, reaction diffusion and growth kinetics of inter-metallic compounds in the Au-Al system have been studied by many authors⁽¹⁾⁻⁽³⁾. Diffusion in gold-rich Au-Al alloys is also interesting from the academic viewpoint. Diffusion of a polyvalent solute atom such as indium, tin or antimony in monovalent noble metal such as copper, silver or gold has been characterized as high diffusivity which is ten or more times as large as the solvent diffusivity and as strong correlated jumps of the solute to a vacancy in the solvent⁽⁴⁾⁽⁵⁾. According to Fogel'son and Trofimova⁽⁶⁾, the diffusion coefficient of

aluminium in gold is about ten times as large as the self-diffusion coefficient in gold. Further, the aluminium atom in gold is expected to make strongly correlated jumps to a vacancy. Application of the five-frequency model and evaluation of the jump frequency ratios for impurity diffusion of Al in Au based on the experiments of diffusion in gold-rich Au-Al alloys is very interesting; however, it has not been made so far.

The correlation factor and the jump frequency ratios in the five-frequency model for the impurity diffusion⁽⁷⁾⁽⁸⁾ can be determined experimentally using the results of the following four experiments. The first is to determine the solvent and solute diffusivities in the pure solvent. The second is to measure the enhancement factor of the solvent diffusivity by a small addition of solute atom into the solvent metal. The third is the experiment on the isotope effect for the impurity diffusion. The fourth is the determination of the vacancy flow factor based on the experiment of the Kirkendall effect in very dilute alloys. The data obtained from the above experiments contribute complementarily to the evaluation of the correlation factor and the jump frequency ratios in the five-frequency model.

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In the present work, three of the four experiments described above have been performed. The experiment of the isotope effect for the impurity diffusion of Al in Au cannot be done because no suitable pair of radioactive isotope of aluminium for diffusion experiment is available. The diffusion coefficient of ^{198}Au in gold-rich Au–Al alloys at 1023 K has been measured to determine the solute enhancement factor, b , for the solvent diffusion. Further, concentration-penetration profiles and the Kirkendall marker shifts in various interdiffusion semi-infinite couples and thin-plate sandwiches consisting of pure Au and gold-rich Au–Al alloys have been examined, and the interdiffusion coefficients and the intrinsic diffusion coefficients at 1023 K have been determined. By the aid of the value of the intrinsic diffusion coefficient of Au in Au–Al alloys extrapolated to the infinite dilution of Al, the vacancy flow factor, G , has been calculated. From the values of b and G , an appropriate numerical set of the correlation factor and jump frequency ratios for impurity diffusion of Al in Au has been calculated.

II. Experimental Procedure

1. Diffusion of ^{198}Au in gold-rich Au–Al alloys

Rods of pure Au and Au–Al alloys containing 0.3, 0.6, 1.0, 1.5 and 2.0 at% Al were made by melting gold blocks of 99.99% purity together with aluminium blocks of 99.99% purity in a high purity graphite crucible under blowing of argon gas and casting into a steel mold 8 mm in diameter. To cause grain growth, the rods of pure Au and the alloys were annealed in high purity argon gas at 1023 K for 2 days. The resultant grain size was 2 to 3 mm in the pure Au and 1 to 2 mm in the alloys. The rods were cut to make the disc specimens 10 mm in thickness. The cut surfaces were ground on abrasive papers and polished on a buff with fine alumina paste. To obtain deformation-free surface, the specimens were annealed at 773 K for 3.6 ks in high purity argon gas. The radioisotope ^{198}Au (γ -ray, 0.412 MeV; half-life, 2.69 d) was sup-

plied in a form of tetrachloride in 4 kmol m^{-3} HCl solution from Amersham International Plc, U.K. The radioisotope was electroplated on the surface of the specimen from the solution. The specimens were then sealed in quartz tubes with high purity argon gas and diffused at 1023 K for 86.4 ks (24 h) in a furnace controlled within $\pm 1 \text{ K}$.

After the diffusion, a cylindrical surface of the specimen was removed to a depth of about 0.5 mm by a precision lathe to eliminate the anxious effect of the surface diffusion along the cylindrical surface. To determine the volume diffusion coefficient of ^{198}Au , the serial sectioning method was employed. The flat surface layer of the specimen was removed successively by grinding on abrasive papers 6 cm wide and 12 cm long. After each grinding, the abrasive paper was collected together with the removed section of the specimen in the form of powder. The loss in the collection of each ground section was less than about 3%. The total γ -ray from each removed section was measured by a well-type Tl-activated NaI scintillation detector in conjunction with a 1024 channels pulse height analyzer.

2. Interdiffusion in gold-rich Au–Al alloys

In addition to the pure Au and Au–Al alloys for the radioactive tracer diffusion experiment, four Au–Al alloys containing 3.0, 4.0, 5.0 and 6.0 at% Al for the interdiffusion experiment were prepared in the same way as described above. The rods were cut to make the disc specimens 3 mm in thickness and thin plate specimens 1 mm in thickness. The flat faces of the specimens were polished in the same way as the tracer diffusion experiment. Semi-infinite couples and thin-plate sandwiches were made. The semi-infinite couple consisted of two discs. The thin-plate sandwich consisted of three thin plates, two outer thin plates of pure Au and a central thin plate of Au–Al alloy. As the Kirkendall marker, a tungsten wire of $2.4 \mu\text{m}$ in diameter was put between discs or thin plates. The semi-infinite couple was used for determination of the interdiffusion coefficient and the intrinsic diffusion coefficients by Matano's analysis⁽⁹⁾ and Darken's relation⁽¹⁰⁾, respectively. The thin-plate sandwich was used

for determination of the intrinsic diffusion coefficients in very dilute alloys by Heumann's analysis⁽¹¹⁾. The semi-infinite couple was put in a stainless steel holder with two screws, pressed by the screws and diffusion-welded by heating in a stream of argon gas at 1023 K for 3.6 ks. The couple was removed from the holder and sealed in a quartz tube with argon gas. The same procedure was used to make the thin-plate sandwich with tungsten wire markers. Diffusion anneals were carried out at 1023 K for 64.8 ks to 4.5 Ms in a furnace controlled within ± 1 K.

After the diffusion, the couple and sandwich were cut parallel to the diffusion direction, and the cut surfaces were polished on a buff with fine diamond paste to examine the concentration-penetration profile by an electron probe microanalyzer (Shimadzu ARL-II), operated under the following conditions; accelerating voltage, 20 kV; take-off angle of Al- $K\alpha$ X-ray, 0.916 rad; specimen current, 50 nA; line scanning speed, $0.8 \mu\text{m s}^{-1}$. By the aid of the standard alloy specimens the calibration curve was obtained. It was shown that the relative intensity of Al- $K\alpha$ X-ray emitted from the specimen was proportional to the mole fraction of Al. The interdiffusion coefficient was determined as a function of concentration by Matano's analysis⁽⁹⁾. In the Au-Al alloys containing Al less than 6 at%, the molar volume is proportional to the mole fraction of Al, and the partial molar volume of each component is constant⁽¹²⁾, therefore, the mole of Al per cubic meter can be used as the unit of concentration of the analysis of the diffusion. The Kirkendall marker shift in the semi-infinite couple was determined by measuring the distance between the marker and Matano interface position in the couple determined by analyzing the concentration-penetration profile. In the case of the thin-plate sandwich, the Kirkendall marker shift was determined by measuring the distance between the confronting marker positions.

III. Results

1. Concentration dependence of tracer diffusion coefficient of gold in Au-Al alloys

For one-dimensional volume diffusion of a tracer from an infinitesimally thin surface layer into a sufficiently long rod analyzed by the serial sectioning method, the solution of Fick's second law is given by

$$C(X, t) = \frac{M}{\sqrt{\pi D^* t}} \exp\left(-\frac{X^2}{4D^* t}\right), \quad (1)$$

where $C(X, t)$ is the concentration of the radioactive tracer at a distance X from the original surface, D^* the volume diffusion coefficient of the tracer, and t the time of diffusion. M is the total amount of tracer deposited on the surface before the diffusion.

Figure 1 shows the plots of $\ln C(X, t)$ vs X^2 for the diffusion of ^{198}Au in Au-Al alloys at 1023 K. The linearity observed in Fig. 1 proves that eq. (1) holds, and thus the volume diffu-

Fig. 1 Examples of penetration profiles for diffusion of ^{198}Au in Au-Al alloys at 1023 K for 86.4 ks.

Fig. 2 Concentration dependence of tracer diffusion coefficient of gold (D_{Au}^*) in Au-Al alloys at 1023 K.

sion has been concerned. The tracer diffusion coefficients of Au in Au-Al alloys at 1023 K, D_{Au}^* , thus determined from the slope of $\ln C(X, t)$ vs X^2 plots by use of eq. (1), are plotted in Fig. 2 as a function of the Al concentration. The value of the self-diffusion coefficient in Au at 1023 K determined by the present experiment, $1.1 \times 10^{-14} \text{ m}^2 \text{ s}^{-1}$, is in good agreement with those obtained by Makin *et al.*⁽¹³⁾ and Herzig *et al.*⁽¹⁴⁾ It is recognized that D_{Au}^* increases linearly with the Al content and the concentration dependence of D_{Au}^* is expressed by the least square fits as follows:

$$D_{Au}^*(N_{Al}) = 1.1 \times 10^{-14} [1 + (53 \pm 9)N_{Al}] \text{ m}^2 \text{ s}^{-1}, \quad (2)$$

where N_{Al} is the mole fraction of Al in the alloy. Thus, the linear enhancement factor, b , of the solvent diffusivity by addition of small amount of solute Al into the solvent Au has been determined to be 53 ± 9 .

2. Concentration dependence of interdiffusion and intrinsic diffusion coefficients

The interdiffusion coefficients, $\tilde{D}$, in Au-Al alloys at 1023 K determined with four different semi-infinite couples consisting of pure Au and Au-Al alloys of four different concentrations (3.0, 4.0, 5.0 and 6.0 at% Al) are in agreement with one another within the experimental error. Figure 3 shows that the logarithm of $\tilde{D}$ increases linearly with N_{Al} and is expressed by

$$\tilde{D}(N_{Al}) = 1.7 \times 10^{-13} \exp(22.5N_{Al}) \text{ m}^2 \text{ s}^{-1}. \quad (3)$$

The value of $\tilde{D}$ extrapolated to $N_{Al}=0$ is

Fig. 3 Concentration dependence of interdiffusion coefficient ($\tilde{D}$), intrinsic diffusion coefficients (D_{Au} and D_{Al}) and tracer diffusion coefficient of gold (D_{Au}^*) in Au-Al alloys at 1023 K.

calculated by eq. (3) to be $1.7 \times 10^{-13} \text{ m}^2 \text{ s}^{-1}$, which is slightly smaller than the value ($2.3 \times 10^{-13} \text{ m}^2 \text{ s}^{-1}$) obtained in the same way by Fogel'son *et al.*⁽⁶⁾

In all the interdiffusion couples and sandwiches, movement of the Kirkendall marker toward the Al-rich side has been observed. This indicates that Al atom diffuses faster than Au atom in the alloys. The Kirkendall void has not been observed so far as an optical microscope has been used. On the basis of the experimental data on the interdiffusion coefficient, the marker shift, the partial molar volumes of both components⁽¹²⁾ ($\bar{V}_{Al} = 9.43 \times 10^{-6} \text{ m}^3 \text{ mol}^{-1}$ and $\bar{V}_{Au} = 10.43 \times 10^{-6} \text{ m}^3 \text{ mol}^{-1}$, $\bar{V}_{Al}/\bar{V}_{Au} = 0.904$) and Darken's relation⁽¹⁰⁾ or Heumann's analysis⁽¹¹⁾, the intrinsic diffusion coefficients, D_{Al} and D_{Au} , for the concentration of marker position have been calculated, as listed in Table 1. The concentration

Table 1 Interdiffusion coefficient ($\tilde{D}$) and intrinsic diffusion coefficients (D_{Al} and D_{Au}) in Au–Al alloys at 1023 K.

Composition at the marker position/ at% Al	$\tilde{D}$	D_{Al}	D_{Au}	$D_{\text{Al}}/D_{\text{Au}}$
Semi-infinite couple				
3.3	3.6	3.7	1.9	1.9
2.8	3.2	3.2	2.0	1.6
2.2	2.9	2.9	2.2	1.3
1.6	2.5	2.5	2.0	1.3
1.6	2.5	2.5	1.7	1.5
Thin-plate sandwich				
0.95	2.1	2.2	1.9	1.2
0.48	1.9	1.9	1.4	1.4

Unit of all the diffusion coefficients: $10^{-13} \text{ m}^2 \text{ s}^{-1}$.

dependence of $\tilde{D}$, D_{Al} and D_{Au} in Au–Al alloys at 1023 K is shown in Fig. 3. The concentration dependence of solvent tracer diffusion coefficient D_{Au}^* is also shown in Fig. 3. The logarithms of the intrinsic diffusion coefficients also increase linearly with N_{Al} . The concentration dependence of D_{Al} and D_{Au} in the range of N_{Al} from 0 to 0.033 can be expressed by

$$D_{\text{Al}}(N_{\text{Al}}) = 1.7 \times 10^{-13} \exp(23.4N_{\text{Al}}) \text{ m}^2 \text{ s}^{-1} \quad (4)$$

and

$$D_{\text{Au}}(N_{\text{Al}}) = 1.6 \times 10^{-13} \exp(8.97N_{\text{Al}}) \text{ m}^2 \text{ s}^{-1}. \quad (5)$$

The value of $D_{\text{Al}}(0)$ and $D_{\text{Au}}(0)$ are estimated from eqs. (4) and (5) to be 1.7×10^{-13} and $1.6 \times 10^{-13} \text{ m}^2 \text{ s}^{-1}$, respectively. The value of $D_{\text{Au}}(0)$ in Au–Al system at 1023 K is about fifteen times as large as the self-diffusion coefficient of Au. Such a large difference between $D_{\text{Au}}(0)$ and $D_{\text{Au}}^*(0)$ suggests that the diffusion of gold is remarkably enhanced by a large vacancy flow effect in Au–Al system.

According to Heumann⁽⁸⁾, the vacancy flow factor, G , defined by the ratio of the phenomenological coefficients of the flux equations for $N_{\text{Al}}=0$ can be evaluated in terms of the self- and impurity diffusion coefficients in pure solvent Au, $D_{\text{Au}}^*(0)$ and $D_{\text{Al}}^*(0)$, and the intrinsic diffusion coefficient of the solvent in pure solvent, $D_{\text{Au}}(0)$, by the following equation:

$$G = (L_{\text{AuAl}}/L_{\text{AlAl}})_{N_{\text{Al}}=0} = \frac{D_{\text{Au}}^*(0)}{D_{\text{Al}}^*(0)} \left(\frac{1}{f_0} - \frac{D_{\text{Au}}(0)}{D_{\text{Au}}^*(0)} \frac{\bar{V}_{\text{Al}}}{\bar{V}_{\text{Au}}} \right), \quad (6)$$

where $f_0 (= 0.781)$ is the correlation factor for self-diffusion in pure f.c.c. metals. According to Darken's relation⁽¹⁰⁾, $D_{\text{Al}}^*(0)$ is equivalent to $\tilde{D}(0)$. Putting the values of $\tilde{D}(0) = D_{\text{Al}}^*(0)$, $D_{\text{Au}}^*(0)$ and $D_{\text{Au}}(0)$ and the value of $\bar{V}_{\text{Al}}/\bar{V}_{\text{Au}}$ into eq. (6), the vacancy flow factor, G , is determined to be -0.78 . Such a large negative value of G has been also obtained in other alloy systems such as Cu–Sn⁽¹⁵⁾ ($G = -0.84$ at 1089 K, -1.06 at 1014 K), Cu–In⁽¹⁶⁾ ($G = -0.57$ at 1089 K, -0.71 at 1005 K) and Cu–Sb⁽¹⁷⁾ ($G = -1.20$ at 1005 K) systems.

IV. Discussion

The linear enhancement factor, b , can be related theoretically to the jump frequencies, ω_i , of a vacancy neighboring a solute atom as follows⁽¹⁸⁾:

$$b = -18 + \frac{4\omega_4}{f_0\omega_0} \left(\frac{\omega_1}{\omega_3} \chi_1 + \frac{7}{2} \chi_2 \right), \quad (7)$$

where ω_i has a usual meaning in the five-frequency model for impurity diffusion, $\omega_0 - \omega_4$ being defined as follows:

- ω_1 the jump frequency of a vacancy from a nearest neighbor site of an impurity to another nearest neighbor site;
 - ω_2 the jump frequency for an impurity-vacancy exchange;
 - ω_3 the frequency for vacancy jumps from nearest neighbor to non-nearest neighbor positions;
 - ω_4 the frequency for the reverse jump of ω_3 ; and
 - ω_0 the jump frequency for all other jumps.
- χ_1 and χ_2 are the mean partial correlation factors defined by Howard and Manning⁽¹⁸⁾ and given as functions of three jump frequency ratios, ω_2/ω_1 , ω_3/ω_1 and ω_4/ω_0 , respectively. The correlation factor f_i for the impurity diffusion is related to the three jump frequency ratios by the following equations⁽¹⁹⁾:

$$f_i = \frac{1 + (7F/2)(\omega_3/\omega_1)}{1 + (\omega_2/\omega_1) + (7F/2)(\omega_3/\omega_1)} \quad (8)$$

and

$$\begin{aligned} D_{\text{Al}}^*(0)/D_{\text{Au}}^*(0) \\ = (f_i/f_0)(\omega_2/\omega_1)(\omega_1/\omega_3)(\omega_4/\omega_0), \quad (9) \end{aligned}$$

where F is a known function of the jump frequency ratio ω_4/ω_0 . Furthermore, the vacancy flow factor G can be related to the jump frequency ratios as follows⁽²⁰⁾:

$$G = \frac{-2 + 3(\omega_3/\omega_1) + (\omega_3/\omega_1)(\omega_0/\omega_4 - 1)(7 - 7F)}{1 + (7F/2)(\omega_3/\omega_1)}. \quad (10)$$

Thus, a numerical set of f_i , ω_2/ω_1 , ω_3/ω_1 and ω_4/ω_0 can be determined by putting the empirical values of b , $D_{\text{Al}}^*(0)/D_{\text{Au}}^*(0)$ and G into eqs. (7)~(10). In the present work, at first the linear enhancement factor b is calculated as a function of ω_4/ω_0 with the aid of eqs. (7), (8) and (9), where we take f_i as a parameter and use the values of χ_1 and χ_2 computed by Howard and Manning⁽¹⁸⁾ as functions of the three jump frequency ratios. The plot of b versus ω_4/ω_0 with f_i as a parameter is shown in Fig. 4, where the experimental value of b is also shown. Next, the vacancy flow factor G is calculated as a function of ω_4/ω_0 using the experimental value of $D_{\text{Al}}^*(0)/D_{\text{Au}}^*(0)$ and eqs. (8), (9) and (10). The plot of G versus ω_4/ω_0 with f_i as a parameter is shown in Fig. 5, where the experimental value of G and the relationship between f_i and ω_4/ω_0 obtained from the ex-

perimental value of b in Fig. 4 are also shown. Thus, combining the experimental value of G with that of b , we can make a graphical determination of the values of f_i and ω_4/ω_0 . Then, by putting the values of f_i and ω_4/ω_0 in eqs. (8) and (9), the values of ω_3/ω_1 and ω_2/ω_1 are calculated. The error in the calculation of the jump frequency ratios is estimated to be less than 20%. The value of f_i , ω_4/ω_0 , ω_3/ω_1 and ω_2/ω_1 determined in this way are listed in Table 2, where the relationships $f_i \ll 1$, $\omega_2 \gg \omega_1$, $\omega_4 > \omega_0$ and $\omega_3 < \omega_1$ are recognized. Those relationships suggest that the attraction between an Al atom and a vacancy in Au is highly strong and that a vacancy makes very many consecutive exchanges with an Al atom rather than with any solvent atom neighboring the vacancy.

Fig. 4 Relationship between linear enhancement factor (b) and ω_4/ω_0 with f_i as a parameter in Au-Al system at 1023 K. Experimental value of b is also shown.

Fig. 5 Relationship between vacancy flow factor (G) and ω_4/ω_0 with f_i as a parameter in Au-Al system at 1023 K. Experimental value of G and the relationship between f_i and ω_4/ω_0 determined based on the experimental value of b are also shown.

Table 2 Correlation factor (f_i) and jump frequency ratios (ω_4/ω_0 , ω_3/ω_1 and ω_2/ω_1) for impurity diffusion in Au. Experimental values of $D_B^*(0)/D_A^*(0)$, b and G are also shown.

Alloy	Temperature/K	$D_B^*(0)/D_A^*(0)$	b	G	f_i	ω_4/ω_0	ω_3/ω_1	ω_2/ω_1
Au-Al	1023	15.4	53	-0.78	0.1	4	0.6	20
Au-Sn	1059	16.4 ^a	65 ^c	-0.86	0.16 ^d	5	0.7	15
Au-In	1075	8.6 ^b	32 ^c	-0.40	0.26 ^d	3	0.8	9

A: solvent, B: solute.

a: Ref. (21), b: Ref. (22), c: Ref. (23), d: Ref. (5).

It is expected that the jumping behavior of other polyvalent solutes in Au is similar to that of Al in Au. However, only a few elements in Au have been examined⁽⁵⁾. Using the experimental values of $D_B^*(0)/D_A^*(0)$ by Heumann *et al.*⁽²¹⁾⁽²²⁾, b by Neumann⁽²³⁾ and f_i by Herzig *et al.*⁽⁵⁾ for Sn and In in Au, the values of G and jump frequency ratios are calculated in the same way as described above, and these are shown in Table 2, where the relationships $f_i \ll 1$, $\omega_2 \gg \omega_1$, $\omega_4 > \omega_0$ and $\omega_3 < \omega_1$ are also confirmed. As a matter of course, the same behavior of diffusion jumps of the polyvalent solute has been observed in Cu⁽⁴⁾.

V. Conclusion

From the experimentally determined linear enhancement factor, b , for solvent diffusivity and vacancy flow factor, G , for interdiffusion, an appropriate numerical set of the correlation factor, f_i , and the jump frequency ratios, ω_4/ω_0 , ω_3/ω_1 and ω_2/ω_1 , for the impurity diffusion of Al in Au has been determined using the theoretical equations. The result indicates that diffusion jumps of solute Al atom to a vacancy in Au are highly correlated.

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