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Effect of high pressure on interdiffusion in an Al–Mg alloy

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Interdiffusion in the Al–4.06 at% Mg alloy has been investigated under high pressure in the temperature range 690 to 877 K. The diffusion coefficients decreased with pressure. The activation volume and energy of interdiffusion were determined.

1. Introduction

Many studies of diffusion have been reported on aluminium and aluminium alloys. The contribution of monovacancies and divacancies to diffusion phenomena has been of interest in recent studies. The self diffusion in aluminium is interpreted by the large contribution of divacancies in the temperature range near the melting point of aluminium [1], or by the postulation of the temperature-dependent enthalpy of the single vacancy motion [2]. With regard to the impurity diffusion in aluminium, the small contribution of divacancies to the diffusion is indicated by the accurate measurements of the tracer-impurity diffusion over a wide temperature range [3, 4].

One of the experiments which throw light on the above problem is the high-pressure effect measurement. The activation volume of monovacancies and divacancies which contribute to the diffusion can be obtained from the pressure dependence of the diffusion coefficients [5]. In aluminium, the activation volume of the self diffusion has been determined experimentally and theoretically [6–16], but they are not consistent with each other. On the other hand, we can find no report on the activation volume of the impurity diffusion except for zinc in aluminium [17].

The several studies of magnesium diffusion in aluminium were performed under atmospheric pressure and vacuum [18–21], since magnesium is a common element in commercial aluminium alloys. Their data differ from one to another. Furthermore, the effect of high pressure on the magnesium diffusion in aluminium has not been reported yet.

The purpose of this research is to investigate the effect of high pressure on magnesium diffusion and to determine its activation volume.

2. Experimental procedures

The ingot of Al–4.06 mass % Mg alloy was prepared from 99.996 mass % Al and 99.95 mass % Mg, and then homogenized for 345.6 ksec at 793 K. In order to produce the diffusion couple for the high pressure measurements, a bar (9 mm x 9 mm x 60 mm) was cut from the alloy ingot. A hole (9 mm x 9 mm x 60 mm) was made through the centre of the 99.996 mass % Al block (30 mm x 30 mm x 60 mm). The surface of the bar and hole of the block were metallographically polished, and then chemically washed in aqua regia. The bar was inserted into the hole of the pure aluminium block. The composite metal was section-rolled at 573 K, drawn into 4.0 mm diameter at room temperature and then cut into 5 mm lengths as the diffusion couple. Since the rolling and drawing break up the oxide films at the interface of the diffusion couple and distribute them over a much larger area, the

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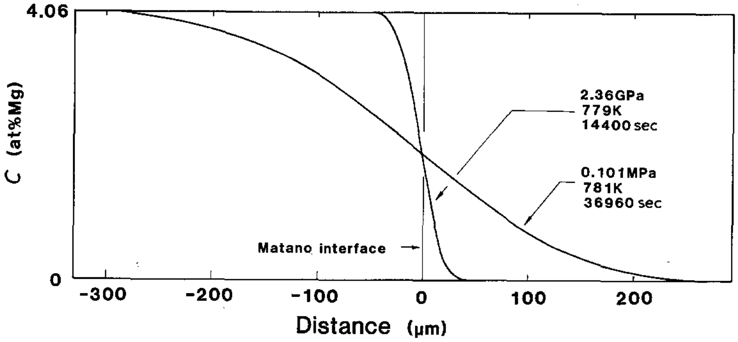


Figure 1 Concentration profiles of diffusion couples annealed under atmospheric and high pressure.

clean interface appears during the welding of the diffusion couple [22].

It was necessary for the diffusion couple under atmospheric pressure to have a larger size in order to maintain the diffusion condition (infinite), because the diffusion distance under atmospheric pressure was expected to be longer than that under high pressure (see Fig. 1). The composite metal which consists of the alloy bar and the aluminium block in the same size as the above one was rolled to a 7 mm thickness at 793 K after treatments similar to those described above. It was cut into 8 mm widths for the diffusion couples under atmospheric pressure.

Solute concentration profiles in these diffusion couples were obtained by using an electron probe microanalyser (EPMA). It was found that diffusion did not occur during the welding treatments.

A high pressure was applied to the diffusion couple by a cubic anvil apparatus with a 12 MN press (Kobe Steel Co). As shown in Fig. 2, the diffusion couple was in the BN capsule which was thermally stable and had the proper lubricity. A cubic pyrophylite ($Al_2Si_4O_{10}(OH)_2$) filled the role of the pressure medium and self-gasketing. The hydrostatic-high pressure could be obtained by the press along the three axes. The diffusion couples were annealed for times from 3.6 to 57.6 ksec at temperatures of 732 to 877 K by an internal graphite heater under high pressures of 2200 to 3450 MPa. The temperature of the diffusion couple was measured by a thermocouple located near the diffusion couple. The diffusion couple was heated to the temperature of diffusion within 60 sec, and its cooling was carried out within 10 sec. The diffusion temperatures were controlled within ±1.5 K. The calibration of temperature was carried out in a way similar to Hanneman and Strong's method [23]. The hydrostatic-high pressure was calibrated by the transition of bismuth I → II (2.55 GPa at room temperature) and bismuth II → III (2.70 GPa at room temperature).

On the other hand, the diffusion couples under atmospheric pressure were annealed in air for times ranging from 36.6 to 138.6 ksec at temperatures of 690 to 818 K.

After the diffusion annealing, the diffusion couples were ground to expose the diffusion zone where oxidation, evaporation and surface diffusion did not occur. They were metallographically polished to the diffusion direction with 0.3 μm aluminium powder and then analysed to obtain the diffusion profiles by EPMA. These intensities of characteristic X-ray reflections were converted to the concentration by Bishop [24], Springer [25] and Philibert [26] methods.

The interdiffusion coefficients were determined from the concentration profiles by means of Matano's analysis [27] and Hall's method [28]. Matano's analysis has certain disadvantages at the terminal composition of the diffusion couples, as is well known, and Hall's method enabled us to estimate accurate diffusion coefficients near the concentration extremes.

3. Results and discussion

Fig. 1 shows the concentration profiles of the diffusion couples annealed at 781 K for 369.9 ksec under 0.101 MPa and at 779 K for 14.4 ksec under 2.36 GPa. It is clear that the high pressure reduces the interdiffusion in the Al–Mg alloy. The interdiffusion coefficients are estimated at 0 and 4.06 at %Mg by Hall's method, and at 1, 2 and 3 at %Mg by Matano's analysis. They are summarized in Table I. The interdiffusion coefficients under 2.2 and 3.3 GPa which are evaluated from the above data are presented in Table II. Figs. 3a, b and c indicate that the interdiffusion coefficients under 0.101 MPa, 2.2 and 3.3 GPa increase linearly with the concentration of magnesium. This ten-

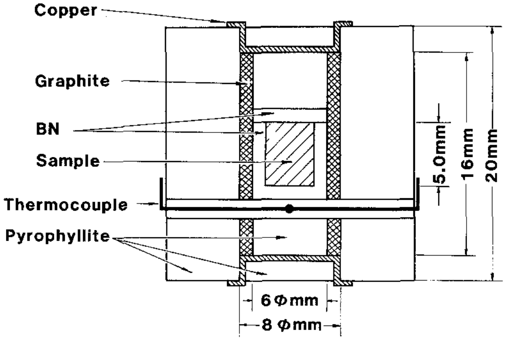


Figure 2 Sketch of assembly of high pressure and temperature cell for the hexahedral anvil apparatus.

dency is consistent with Mehl *et al.*'s [21] data measured under atmospheric pressure.

Fig. 4 shows the temperature dependence of the diffusion coefficients at 0 and 4.06 at %Mg under 0.101 MPa, 2.2 and 3.3 GPa together with the other authors' results. The data of this work fall on straight lines with little scatter, and are nearly equal to theirs under pressures less than atmospheric pressure. The diffusion coefficients satisfy the Arrhenius type equation

$$D = D_0 \exp(-Q/RT) \qquad (1)$$

where D_0 is the pre-exponential factor, Q the activation energy and R the gas constant. Table III gives a compilation of Q and D_0 evaluated using Equation 1 with other authors' results. They are in good agreement with theirs under atmospheric pressure. The expression for the diffusion under pressure in terms of the activation volume ΔV is [5]

$$D(P) = D(0) \exp(-P\Delta V/RT) \qquad (2)$$

where $D(0)$ and $D(P)$ are the diffusion coefficients at pressures of 0 and P. The activation volume of diffusion is the increase in volume as an atom goes from a normal position in a perfect crystal to an activated position. The activation energy is larger by an amount $P\Delta V$ at a pressure P than at zero pressure.

$$Q(P) = Q(0) + P\Delta V \qquad (3)$$

where $Q(0)$ and $Q(P)$ are the activation energy of diffusion at the pressures 0 and P. Fig. 5 reveals that the activation energy at 0 and 4.06 at %Mg increases linearly with pressure. This increase in pressure changes the amount of the energy necessary to form a vacancy and for a neighbouring atom to jump into the vacant site. The average value of the activation volume between 0 and 4.06 at %Mg is evaluated to be $8.1 \times 10^{-6}\,\text{m}^3\,\text{mol}^{-1}$ by Equation 3.

The activation volume of the interdiffusion can be also evaluated from the pressure dependence of the interdiffusion coefficients. According to Adda and Philibert [29]

$$\frac{\partial \ln \tilde{D}}{\partial P} = -\frac{\Delta \tilde{V}}{RT} \qquad (4)$$

TABLE I Experimental data of interdiffusion coefficients under atmospheric and high pressure in Al–Mg alloy

Pressure (MPa)	Temperature (K)	$\tilde{D}$ (m² sec⁻¹)				
		0 at %Mg	1 at %Mg	2 at %Mg	3 at %Mg	4.06 at %Mg
0.101	818	4.53×10^{-13}	4.81×10^{-13}	5.45×10^{-13}	5.66×10^{-13}	5.98×10^{-13}
	788	2.18×10^{-13}	2.35×10^{-13}	2.65×10^{-13}	2.58×10^{-13}	2.84×10^{-13}
	781	1.72×10^{-13}	1.82×10^{-13}	1.97×10^{-13}	2.10×10^{-13}	2.22×10^{-13}
	761	1.12×10^{-13}	1.15×10^{-13}	1.23×10^{-13}	1.29×10^{-13}	1.34×10^{-13}
	747	7.56×10^{-14}	8.23×10^{-14}	8.41×10^{-14}	9.12×10^{-14}	1.03×10^{-13}
	731	4.72×10^{-14}	5.07×10^{-14}	5.72×10^{-14}	6.28×10^{-14}	7.03×10^{-14}
	706	2.29×10^{-14}	2.47×10^{-14}	2.63×10^{-14}	2.91×10^{-14}	3.30×10^{-14}
	690	1.51×10^{-14}	1.63×10^{-14}	1.72×10^{-14}	1.89×10^{-14}	1.98×10^{-14}
2390	843	3.54×10^{-14}	3.72×10^{-14}	4.16×10^{-14}	4.17×10^{-14}	4.32×10^{-14}
2310	812	1.72×10^{-14}	2.09×10^{-14}	2.26×10^{-14}	2.01×10^{-14}	2.13×10^{-14}
2360	779	7.82×10^{-15}	8.37×10^{-15}	9.14×10^{-15}	9.27×10^{-15}	9.70×10^{-15}
2200	732	2.15×10^{-15}	2.30×10^{-15}	2.45×10^{-15}	2.40×10^{-15}	2.54×10^{-15}
3370	877	2.96×10^{-14}	3.25×10^{-14}	3.47×10^{-14}	3.43×10^{-14}	3.50×10^{-14}
3360	848	1.21×10^{-14}	1.37×10^{-14}	1.57×10^{-14}	1.52×10^{-14}	1.60×10^{-14}
3450	818	5.96×10^{-14}	6.46×10^{-15}	7.34×10^{-15}	7.89×10^{-15}	8.41×10^{-15}
3360	783	2.45×10^{-15}	2.61×10^{-15}	2.85×10^{-15}	2.99×10^{-15}	3.03×10^{-15}
3380	737	5.15×10^{-16}	5.45×10^{-16}	6.01×10^{-16}	6.00×10^{-16}	7.37×10^{-16}

T A B L E II Calculated values of diffusion coefficients under 2.2 and 3.3 GPa from data in Table I

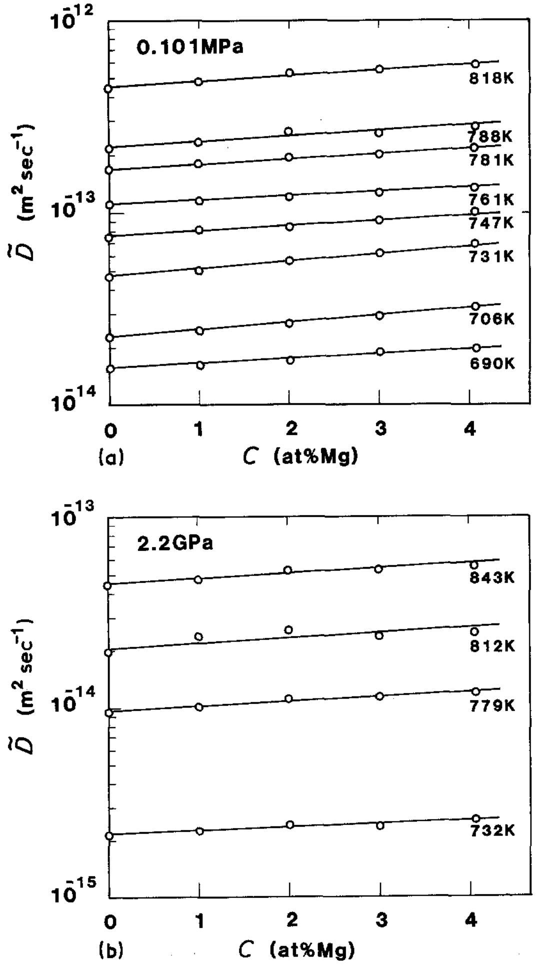
Pressure (MPa)	Temperature (K)	$\tilde{D}$ (m^2 sec^{-1})				
		0 at %Mg	1 at %Mg	2 at %Mg	3 at %Mg	4.06 at %Mg
2200	843	4.52×10^{-14}	4.75×10^{-14}	5.32×10^{-14}	5.32×10^{-14}	5.53×10^{-14}
	812	2.00×10^{-14}	2.42×10^{-14}	2.61×10^{-14}	2.46×10^{-14}	2.48×10^{-14}
	779	9.63×10^{-15}	1.03×10^{-14}	1.23×10^{-14}	1.14×10^{-14}	1.20×10^{-14}
	732	2.16×10^{-15}	2.30×10^{-15}	2.45×10^{-15}	2.40×10^{-15}	2.55×10^{-15}
3300	877	3.22×10^{-14}	3.52×10^{-14}	3.77×10^{-14}	3.72×10^{-14}	3.80×10^{-14}
	848	1.30×10^{-14}	1.47×10^{-14}	1.69×10^{-14}	1.63×10^{-14}	1.72×10^{-14}
	818	7.16×10^{-15}	7.76×10^{-15}	7.81×10^{-15}	9.44×10^{-15}	1.01×10^{-14}
	783	2.65×10^{-15}	2.82×10^{-15}	3.08×10^{-15}	3.23×10^{-15}	3.27×10^{-15}
	737	5.76×10^{-16}	6.10×10^{-16}	6.73×10^{-16}	6.73×10^{-16}	8.24×10^{-16}

where $\Delta\tilde{V}$ is the activation volume of the interdiffusion. Fig. 6 shows the pressure dependence of the interdiffusion coefficients which are evaluated from the Arrhenius equation in Table III. The interdiffusion coefficients decrease with pressure. The activation volume of the interdiffusion was evaluated to be $8.5 \sim 8.7 \times 10^{-6}\,\text{m}^3\,\text{mol}^{-1}$ from Equation 4. These values are in good agreement with one evaluated using Equation 3 mentioned above.

Jeffery and Lazarus [30] have proposed that the activation volume of the tracer diffusion is given by

$$\Delta V^* = \frac{\partial G}{\partial P} = \frac{-RT\,\partial \ln D^*}{\partial P} + r_G K_T RT \quad (5)$$

where ΔV^* is the activation volume of the tracer diffusion D^*, r_G the Grüneisen constant and K_T thermal compressibility. Darken [31] has established the relationship between the interdiffusion



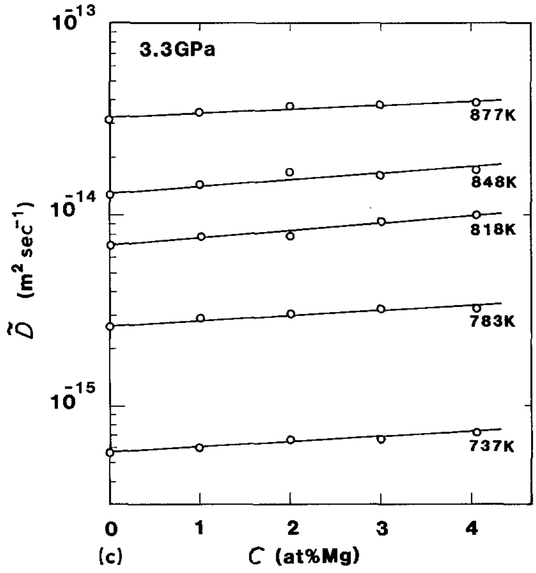


Figure 3 Interdiffusion coefficients as a function of magnesium concentration for Al–4.06 at %Mg alloy under (a) atmospheric pressure, (b) 2.2 GPa, and (c) 3.3 GPa.

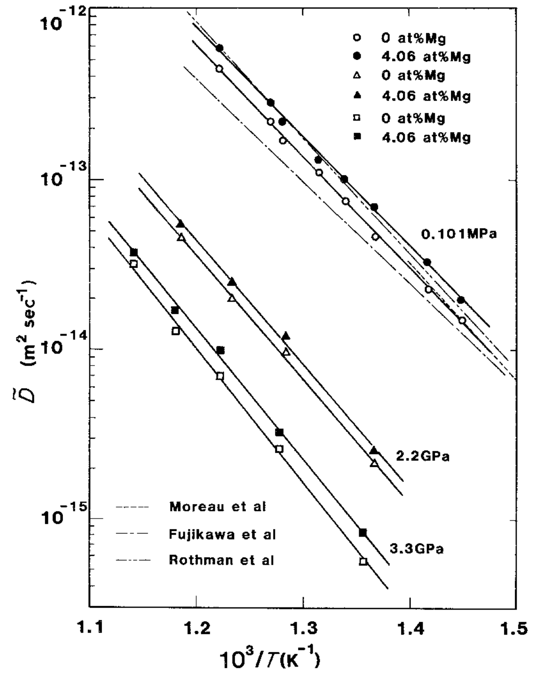


Figure 4 The interdiffusion coefficients under pressure as a function of reciprocal absolute temperature.

coefficient and the tracer coefficient. In Al–Mg alloy

$$\tilde{D} = (N_{Mg}D^*_{Al} + N_{Al}D^*_{Mg})\left(\frac{\partial \ln \gamma_{Mg}}{\partial \ln N_{Mg}} + 1\right) \quad (6)$$

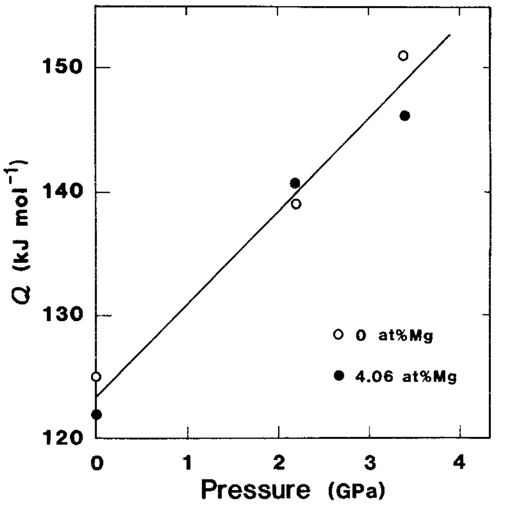


Figure 5 Pressure dependence of activation enthalpy for interdiffusion.

where N_{Mg} and N_{Al} are the atomic fractions of magnesium and aluminium, and γ_{Mg} the activity coefficient of magnesium atoms in the Al–Mg alloy. In the case of $N_{Mg} = 0$, Equation 6 becomes

$$\tilde{D} = D^*_{Mg} \quad (7)$$

From Equations 4, 5 and 7,

$$\Delta V^*_{Mg} = \Delta \tilde{V} + r_G K_T RT \quad (8)$$

TABLE III Diffusion parameters for diffusion in aluminium and Al–Mg alloy

Source (pressure)	Mg (at %)	D_0 ($m^2 sec^{-1}$)	Q(kJ mol^{-1})
This work (0.101 MPa)	0	4.2×10^{-5}	125
	1	4.9×10^{-5}	124
	2	6.1×10^{-5}	127
	3	3.2×10^{-5}	122
	4.06	4.5×10^{-5}	122
This work (2.2 GPa)	0	1.9×10^{-5}	139
	1	2.5×10^{-5}	141
	2	3.7×10^{-5}	142
	3	3.1×10^{-5}	142
	4.06	2.9×10^{-5}	141
This work (3.3 GPa)	0	3.0×10^{-5}	151
	1	4.4×10^{-5}	153
	2	4.4×10^{-5}	153
	3	4.1×10^{-5}	152
	4.06	1.9×10^{-5}	146
Moreau *et al.* [20] (0.101 MPa)	0	1×10^{-4}	130
Fujikawa and Hirano [18] (< 0.101 MPa)	0	6.23×10^{-6}	115
Rothman *et al.* [19] (< 0.101 MPa)	0	1.24×10^{-4}	130

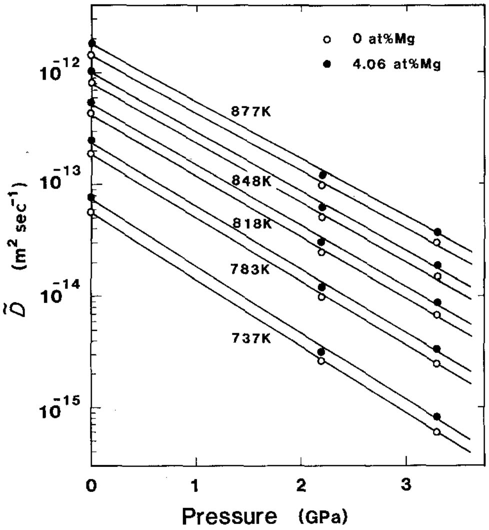


Figure 6 Pressure dependence of interdiffusion coefficients in Al–4.06 at %Mg alloy.

Furthermore, $D^*_{Mg}N_{Al} \gg D^*_{Al}N_{Mg}$ in the dilute Al–Mg alloy. In Al–4.06 at %Mg,

$$\tilde{D} = 0.9594D^*_{Mg}(\partial \ln r_{Mg}/\partial \ln N_{Mg} + 1) \quad (9)$$

Differentiating Equation 9 with respect to the pressure P,

$$\frac{\partial \ln \tilde{D}}{\partial P} = \frac{\partial \ln D^*_{Mg}}{\partial P} + \frac{\partial(\partial \ln \gamma_{Mg}/\partial \ln N_{Mg} + 1)}{\partial P}$$

$$= \frac{\partial \ln D^*_{Mg}}{\partial P} + \left(\frac{N_{Mg}}{RT}\right)\left(\frac{\partial \bar{V}_{Mg}}{\partial N_{Mg}}\right) \quad (10)$$

where $\bar{V}_{Mg}$ is the partial molar volume of magnesium. The second term on the right-hand side of Equation 10 is about $1.5 \times 10^{-11}\,m^3\,mol^{-1}$ and can be neglected. Therefore, also in the dilute Al–Mg alloy,

$$\Delta V^*_{Mg} = \Delta\tilde{V} + r_G K_T RT \quad (11)$$

The activation volume of the tracer diffusion is estimated to be $8.8 \sim 9.0 \times 10^{-6}\,m^3\,mol^{-1}$ from Equation 8 using the values $\Delta\tilde{V}$, r_G [32] and K_T [33].

The difference in the activation volume between the impurity and self diffusion is evaluated by differentiating the difference in the activation enthalpy between them with pressure P [17]. The difference in their activation enthalpy is theoretically evaluated with the model of Lazarus [34] and LeClaire [35], using an inter-ion potential given by Deplante and Blandin [36] as reported by Anand and Agarwala [37].

$$\Delta Q = Q_i - Q_0 = H_f + H_m - K\,\partial \ln F_i/\partial(1/T)$$
$$= 2(-3/2e)V[r = (11/16)r_0] \quad (12)$$
$$V(r) = e\alpha_0 \cos(2K_f r + \phi)/(4\pi K_f^2 r^3)$$

where Q_i and Q_0 are the activation energies of the impurity and the self diffusion, H_f the difference in the energy required to form a vacancy next to an isolated solute atom in the solvent and in the pure solvent, ΔH_m the difference in the jump energy between a vacancy-solute exchange and a vacancy-solvent exchange in the pure solvent, F_i the correlation factor for the impurity atom, r the distance from the impurity atom, r_0 the inter-atomic distance, K_f the Fermi wave number for the solvent, α_0 and K are constants, and ϕ the phase angle.

If it is assumed that F_i does not change when there is no change in the diffusion mechanism with pressure, the differentiation of Equation 11 with respect to the pressure P yields,

$$\Delta(\Delta V) = \Delta V_i^* - \Delta V_0$$
$$= \partial\{\Delta H_f + \Delta H_m - [\partial \ln F_i/\partial(1/T)]\}/\partial P$$
$$= \Delta(\Delta V_f + \Delta V_m)$$
$$= \frac{2(-3/2)\alpha_0 K_T e^2 \sin(2K_f(11/16)r_0 + \phi)}{6\pi K_f((11/16)r_0)^2} \quad (13)$$

where ΔV_i^* and ΔV_0 are the activation volume of impurity and self diffusion, and K_t the isothermal compressibility. α_0 is 1.2, ϕ is 0.66 [18], r_0 is $2.86 \times 10^{-10}\,m$, K_f is $1.75 \times 10^{10}\,m^{-1}$ and K_T is $1.9 \times 10^{-11}\,m^2\,N^{-1}$ for the magnesium diffusion in aluminium. The value of $\Delta(\Delta V)$ is evaluated to be $5.0 \times 10^{-7}\,m^3\,mol^{-1}$, and then the activation volume of the self diffusion is $9.3 \sim 9.5 \times 10^{-6}\,m^3\,mol^{-1}$.

The activation volume of diffusion is summarized with other authors' results in Table IV, where V_0 is the molar volume of aluminium at atmospheric pressure, and superscripts 1 and 2 refer to monovacancy and divacancy respectively. In the case where diffusion occurs by the vacancy mechanism, the activation volume of diffusion is the sum of the migration volume ΔV_m and formation volume ΔV_f [5],

$$\Delta V = \Delta V_m + \Delta V_f$$

or

$$\Delta V^1 = \Delta V^1_m + \Delta V^1_f,\ \text{and}\ \Delta V^2 = \Delta V^2_m + \Delta V^2_f \quad (14)$$

TABLE IV Summary of activation volume for diffusion in aluminium and aluminium alloys

Quantity	Method	Temperature (K)	$\Delta V/V_0$ ($V_0 = 10.0 \times 10^{-6}\ \mathrm{m^3\,mol^{-1}}$)					Source
			0 at %Mg	1 % at Mg	2 at %Mg	3 at %Mg	4.06 at %Mg	
$\Delta\tilde{V}$	Equation 3	737 ~ 877	0.78	0.85	0.78	0.90	0.75	This work
$\Delta\tilde{V}$	Equation 4	737	0.85	0.85	0.85	0.86	0.85	This work
		783	0.86	0.85	0.86	0.87	0.86	
		818	0.86	0.86	0.86	0.86	0.86	
		848	0.87	0.86	0.86	0.86	0.87	
		877	0.87	0.86	0.87	0.85	0.87	
ΔV^*_{Mg}	Equation 8	737	0.88					This work
		783	0.89					
		818	0.90					
		848	0.90					
		877	0.90					
ΔV_{Al}	Equation 13	737	0.93					This work
		783	0.94					
		818	0.95					
		848	0.95					
		877	0.95					
$\Delta\tilde{V}$ in Al–Zn	Interdiffusion	767 ~ 881	0.86					Minamino *et al.* [17]
ΔV_{Al}	NMR	667 ~ 772	0.70					Engardt and Barnes [6]
	Creep	0.3 ~ 0.5 T_{m}	0.74					Mc Cormick and Ruoff [9]
		0.5 ~ 0.6 T_{m}	0.87					
	Dislocation annealing	~ 562	0.44 ~ 0.87					Norris [10]
	Creep	536 ~ 563	1.36					Buescher and Emrick [13]
	Tracer diffusion	713 ~ 883	1.29					Beyeler and Adda [8]
	Thermodynamic model		0.85					Sherby *et al.* [12]
ΔV^1_{f}	Thermoelectric power	R.T. ~ 900	0.54					Bourassa *et al.* [14]
ΔV^2_{f}		R.T. ~ 900	0.96					
ΔV^1_{f}	Resistivity	600 ~ 700	0.60					
ΔV^2_{f}		800 ~	1.45					
ΔV^1_{f}	Quenched in resistivity	693	0.62					Emrick and McArdle [15]
ΔV^1_{f}	Pseudopotential	20 ~ 900	0.55 ~ 0.66					Audit and Gilder [16]
ΔV_{f}	Pseudopotential		0.519					Straub [11]
ΔV_{m}			0.343					
ΔV^1_{m}	Annealing of excess quenched resistivity	604	0.18					Buescher and Emrick [13]
ΔV^2_{m}		853	0.17					

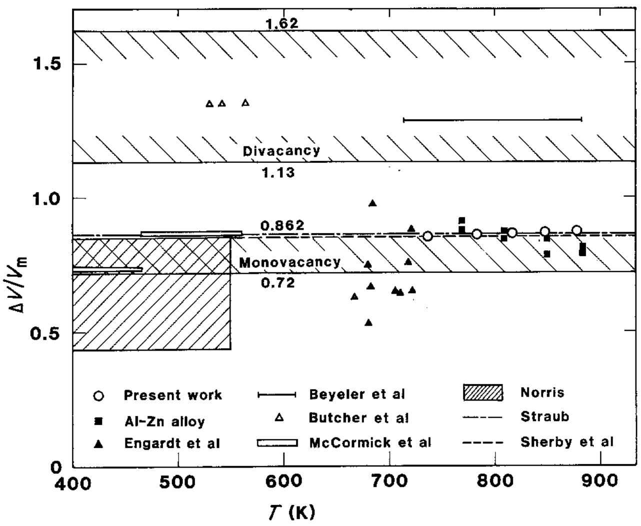


Figure 7 Ratio of activation volume of diffusion in aluminium to molar volume of aluminium as a function of absolute temperature.

The activation volume for the monovacancy mechanism in aluminium $(0.72 \sim 0.80) V_0$, and that of the divacancy is $(1.13 \sim 1.62) V_0$ from the data listed in Table IV. Straub [11] calculated the activation volume of the self diffusion in aluminium to be $0.862 V_0$ for the single vacancy mechanism by a pseudopotential method.

Fig. 7 shows the activation volume of diffusion as the ratio $\Delta V/V_0$ with other authors' results. Although the data of the activation volume are very scattered and not consistent with one another, it is found that the activation volume separates into two groups which are in the regions of the monovacancy and divacancy diffusion mechanisms respectively. The activation volume of this work is in the neighbourhood of expectation for the monovacancy diffusion mechanism. Similar results are found in the Al–Zn alloy [17]. Bartdorff and Reimers [38] reported the isotope effect for the diffusion of silver in aluminium, and Peterson and Rothman [39] did the same for zinc and copper in aluminium. It is concluded that the divacancy contribution to silver diffusion in aluminium can be neglected, or that the diffusion of zinc and copper in aluminium appears to take place via weakly bound single vacancies. The results of this work are consistent with their conclusions.

4. Summary

The interdiffusion coefficients are decreased by high pressure in the Al–4.06 at % Mg alloy. The activation energy increases with pressure. The activation volume of the interdiffusion is between $0.81 V_0$ and $0.87 V_0$.

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