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Experimental Diffusion Research on BCC Ti-Al-Sn Ternary Alloys

Qiaojun Wu[1,2] · Jingya Wang[3,4] · Yuanyu Gu[1,2] · Yanhua Guo[1] · Guanglong Xu[1] · Yuwen Cui[1,2]

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Abstract The interdiffusion behavior of BCC alloys of Ti-Sn binary and Ti-Al-Sn ternary was investigated at 1473 K (1200 °C) by the diffusion-couple technique. The extracted interdiffusion coefficients of Ti-Sn binary was found to gradually increase with increasing the Sn content, whereas those of Ti-Al-Sn ternary, either main or cross, greatly enhance as the content of diffusing species increases. By a way of comparison, Sn diffusion in Ti-Al-Sn ternary was discerned to occur by the vacancy mechanism.

Keywords BCC Ti-Al-Sn alloys · diffusion couple · impurity diffusion · interdiffusion behavior

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✉ Guanglong Xu
guanglongxu@njtech.edu.cn

✉ Yuwen Cui
ycui@unizar.es

[1] Tech Institute of Advanced Materials and School of Materials Science and Engineering, Nanjing Tech University, Nanjing 210009, People's Republic of China

[2] Instituto de Ciencia de Materiales de Aragon, Zaragoza 50009, Spain

[3] Department of Materials Science, Polytechnic University of Madrid, 28040 Madrid, Spain

[4] IMDEA Materials Institute, C/Eric Kandel 2, Getafe, 28906 Madrid, Spain

1 Introduction

Titanium and titanium alloys are excellent materials for aerospace applications owing to their high strength-to-density, higher yield strength, good corrosion resistance and high-temperature resistance.[1] The main application of titanium alloy in the aerospace industry is high-temperature titanium alloy used at temperatures not higher than 900 K (627 °C). Many of typical high-temperature titanium alloys that have been applied as compressor disks and blades of gas turbines for advanced jet engines, e.g. IMI834,[2] Ti-1100,[3] BT36,[4] and Ti600,[5] have common elements like Al and Sn and others. The practical α-stabilizing element of Al increases tensile strength and creep resistance while reducing alloy density,[6] and the neutral element Sn is used as solid-solution strengthening element, often in conjunction with Al to achieve higher strength without embrittlement.[7] Sn can be also considered as the α-stabilizing element because it can replace Al in the hexagonal ordered Ti_3Al phase (α_2).[8]

The microstructures and mechanical properties, such as strength, ductility, creep resistance and fracture toughness, of high-temperature titanium alloys, are markedly affected by thermomechanical treatment and aging via such complex processes as recovery, recrystallization, grain size growth, phase transformation, and precipitation, which are all related to diffusion and its interaction. Diffusion of some β-stabilizing elements in binary and ternary Ti-based alloys in BCC region have been performed previously, including Ti-Al-Cr,[9] Ti-Al-V,[10] Ti-Al-Co,[11] Ti-Al-Fe,[12] Ti-Al-Mo,[13] and Ti-Al-Ni.[14] Diffusion of Sn in BCC-Ti and Ti-Sn alloys have been researched,[15–20] however, that in BCC Ti–Al–Sn ternary alloys have not been addressed. Accordingly, the objectives of this work are to determine the diffusion properties of BCC Ti-Sn

binary and Ti–Al–Sn ternary, and by a way of comparison to unveil diffusion mechanism of Sn in titanium alloys.

2 Experimental Procedure

2.1 Diffusion Couples and Composition Profiles

Nine ingots including seven binary and two ternary alloys were prepared with pure metals of 99.9 mass% sponge Ti, 99.99 mass% Al and the master alloy of Ti-75% Sn (mass percent) by levitation melting in the argon atmosphere. As listed in Table 1, the nominal and actual compositions of Ti-Al, Ti-Sn binary and Ti-Al-Sn ternary alloys all locate in BCC region at 1473 K (1200 °C), according to the accepted Ti–Al,[21] Ti–Sn,[22] and Ti–Al–Sn[23] phase diagrams. The samples were re-melted six times to attain a homogeneous composition, and then solid-solutioned at 1473 K (1200 °C) for 18 h under vacuum, finally followed by water quenching. This results in the average grain size of the ingots larger than several millimeters so that the grain boundary diffusion effect can be negligible. The ingots were cut into small cylinder samples of $\varphi 15 \times 5$ mm in size by wire-electrode cutting. The surfaces of alloy disks were polished by the standard metallographic techniques to obtain the mirror-like surface. All the diffusion couples were assembled with appropriate pairs of the small disks under vacuum at 1173 K (900 °C) for 90 min with a load of 10 MPa on a vacuum diffusion bonding machine. The diffusion couples were sealed into quartz capsules with argon as shield gas and finally annealed at 1473 K (1200 °C) for 10 h followed by water quenching.

After diffusion treatment, the samples were cut to parallel to the diffusion direction, and the cut surface was polished by standard metallographic techniques. The composition profiles were analytically measured by electron microprobe analysis (EPMA) on JEOL JXA 8230.

2.2 Extraction of Diffusion Coefficients

To avoid the errors in determining the so-called Matano interface, the Sauer-Friese method[24] was accepted to extract the binary interdiffusion coefficient with a composition dependence. It introduces a relative composition variable, namely $Y = \frac{x-x_L}{x_R-x_L}$ (x_L and x_R represent the compositions at the far left and far right ends, respectively), to solve inverse composition profiles of binary system. As such, a binary interdiffusion coefficient can be yielded via Eq 1.

$$\tilde{D}(Y^*) = \frac{V_m}{2t(dY/dz)_{Z^*}}\left[(1-Y^*)\int_{-\infty}^{Z^*}\frac{Y}{V_m}dz + Y^*\int_{Z^*}^{+\infty}\frac{1-Y}{V_m}dz\right], \quad \text{(Eq 1)}$$

where $V_m$is the molar volume; t is the diffusion time and z denotes diffusion distance. Due to the lack of experimental data of the variation of $V_m$with the composition, it is regarded as a constant of 1E−5 m^3/mol during the interdiffusion coefficients calculations. Errors introduced by the neglect of molar volume change are considered to be well within the analytical error and may therefore be ignored.[25]

By introducing a set of similar normalized composition variables, Whittle and Green[26] devised a scheme to extract the interdiffusion coefficients of ternary systems using a pair diffusion couples whose diffusion paths have an intersection composition. For the Ti-Al-Sn ternary system with Ti as the solvent, it yields Eq 2:

$$\frac{1}{2t}\left(\frac{\partial z}{\partial Y_{\mathrm{Al}}}\right)_{Z^*}\left[(1-Y_{\mathrm{Al}})\int_{-\infty}^{Z^*}Y_{\mathrm{Al}}dz + Y_{\mathrm{Al}}\int_{Z^*}^{+\infty}(1-Y_{\mathrm{Al}})dz\right] = \tilde{D}^{\mathrm{Ti}}_{\mathrm{AlAl}} + \tilde{D}^{\mathrm{Ti}}_{\mathrm{AlSn}}\frac{\partial x_{\mathrm{Sn}}}{\partial x_{\mathrm{Al}}} \quad \text{(Eq 2a)}$$

Table 1 Terminal Compositions of Ti-Sn binary and Ti-Al-Sn Ternary Diffusion Couples

Couple name	Nominal composition, at.%	Actual compositions, at.%
G1	Ti/ Ti-9Al-5.5Sn	Ti/Ti-9.4Al-5.3Sn
G2	Ti/Ti-14Al-3.5Sn	Ti/Ti-14.4Al-3.6Sn
G3	Ti-9.0Al-5.5Sn/Ti-15.0Al	Ti-8.5Al-5.6Sn/Ti-14.7Al
H1	Ti-2.5Sn/Ti-5.0Al	Ti-2.6Sn/Ti-5.4Al
H2	Ti-5.5Sn/Ti-5.0Al	Ti-5.6Sn/Ti-5.1Al
H3	Ti-5.5Sn/Ti-15.0Al	Ti-5.5Sn/Ti-14.1Al
H4	Ti-8.5Sn/Ti-10.0Al	Ti-8.3Sn/Ti-9.6Al
H5	Ti-8.5Sn/Ti-15.0Al	Ti-8.6Sn/Ti-15.7Al
H6	Ti-8.5Sn/Ti-20.0Al	Ti-8.3Sn/Ti-19.6Al
J1	Ti/Ti-8.5Sn	Ti/Ti-8.5Sn

$$\frac{1}{2t}\left(\frac{\partial z}{\partial Y_{Sn}}\right)_{Z^*}\left[(1-Y_{\mathrm{Sn}})\int_{-\infty}^{Z^*} Y_{\mathrm{Sn}}dz + Y_{\mathrm{Sn}}\int_{Z^*}^{+\infty}(1-Y_{\mathrm{Sn}})dz\right] = \tilde{D}^{\mathrm{Ti}}_{\mathrm{SnSn}} + \tilde{D}^{\mathrm{Ti}}_{\mathrm{SnAl}}\frac{\partial x_{\mathrm{Al}}}{\partial x_{\mathrm{Sn}}}, \quad \text{(Eq 2b)}$$

where $\tilde{D}^{\mathrm{Ti}}_{\mathrm{AlAl}}$ and $\tilde{D}^{\mathrm{Ti}}_{\mathrm{SnSn}}$ are the main interdiffusion coefficients; $\tilde{D}^{Ti}_{AlSn}$ and $\tilde{D}^{\mathrm{Ti}}_{\mathrm{SnAl}}$ are the cross interdiffusion coefficients. Two main and two cross interdiffusion coefficients can be extracted by solving a homogeneous system of four linear equations at the cross composition of two independent diffusion profiles.

The impurity diffusivity, referring to the infinite dilution of solute diffusion coefficient in pure metal or a homogeneous binary alloy, was extracted by the Hall method[27,28] for the Ti-Sn binary and by its generalized version for the Ti-Al-Sn ternary.[28] It is in the methods that the composition profile is rewritten as $\mu = \mathrm{erf}^{-1}(2Y-1)$ versus $\lambda = x/\sqrt{t}$ and then fitted with a linear relation$\mu = h\lambda + k$. The impurity diffusivity for the left and right composition can be determined by Eq 3a and 3b, respectively,

$$\tilde{D}(x') = \frac{1}{4h_1^2}\left[1 + 2k_1\pi^{1/2}\exp(\mu^2)\times Y(x')\right], \quad \text{(Eq 3a)}$$

$$\tilde{D}(x') = \frac{1}{4h_2^2}\left\{1 - 2k_2\pi^{1/2}\exp(\mu^2)\times [1-Y(x')]\right\}, \quad \text{(Eq 3b)}$$

with the linear fit coefficients h_1 and k_1 for the left side, and h_2 and k_2 for the right side. Regarding the Ti-Al-Sn ternary, the impurity diffusivities of Al in Ti-Sn and Sn in Ti-Al were computed from the profiles of diffusion couples H1–H6 at the terminal compositions.

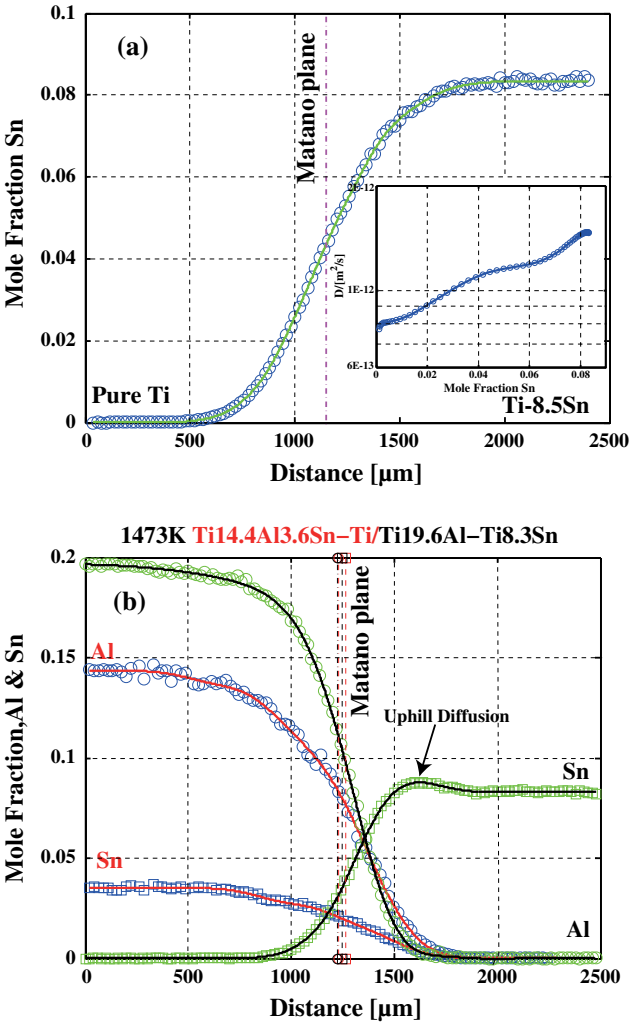

Fig. 1 The composition profiles, (a) the J1 Ti-Sn binary couple, (b) the G2-H6 Ti-Al-Sn ternary couples, annealed at 1473 K for 10 h. The symbols are the EPMA data

3 Results and Discussion

3.1 Diffusion Paths and Interdiffusion Coefficients

The composition profile measured in the diffusion zone is shown in Fig. 1(a) for the Ti/Ti-8.5 at.% Sn binary couple and in Fig. 1(b) for the representative G2 and H6 Ti-Al-Sn ternary couples, all annealed at 1473 K (1200 °C) for 10 h. The diffusion distances for all diffusion couples are typically around 2500 microns, which is significantly larger than that (by a rough estimate, a few microns) arising from the diffusion bonding at 1173 K (900 °C) for 90 mins. Note that the experimental EPMA profiles were represented by the error function expansion[29] to provide the profiles in high-accuracy analytical form for the extraction of diffusion coefficients. It is in the inset of Fig. 1(a) that the extracted Ti-Sn binary interdiffusion coefficients by the Sauer-Friese method are displayed versus the content of Sn. As expected by an empirical supposition,[30] the binary Ti-Sn diffusion rate is gradually enhanced by adding the Sn solute that has a lower melting temperature, in good accord with the previous studies.[18–20]

While it learns from the composition profiles of the ternary couples that the penetration depth of Al, as wide as about 1-2 mm, is comparable to that of Sn, revealing that Al diffuses comparably to Sn in BCC Ti-Al-Sn system at 1473 K. The fact is further justified by the diffusion paths obtained from all ternary diffusion couples at 1473 K (1200 °C) when plotted on the Ti-Al-Sn ternary isotherm in Fig. 2. As shown, all the paths are slightly S-shaped with a minor bent, suggesting a small difference between the diffusion rates for Al and Sn at 1473 K (1200 °C). The extracted ternary interdiffusion coefficients of the Ti-Al-Sn BCC alloys at 1473 K (1200 °C) are listed in Table 2, and our examinations show that they all satisfy the

thermodynamic constraints.[31] It can be seen that all the coefficients are positive and that the main coefficients $\tilde{D}^{Ti}_{AlAl}$ and $\tilde{D}^{Ti}_{SnSn}$ are indeed comparable for most of the compositions. The average values, summarized in Table 2, indicates that $\tilde{D}^{Ti}_{AlAl}$(9.2E−13 m^2/s) is approximately 1.5 times greater than $\tilde{D}^{Ti}_{AlSn}$ (5.9E−13 m^2/s) while $\tilde{D}^{Ti}_{SnSn}$(8.2E−13 m^2/s) is about 3.5 times than $\tilde{D}^{Ti}_{SnAl}$ (2.4E−13 m^2/s), also revealing that diffusion of Sn is much more affected by Al.

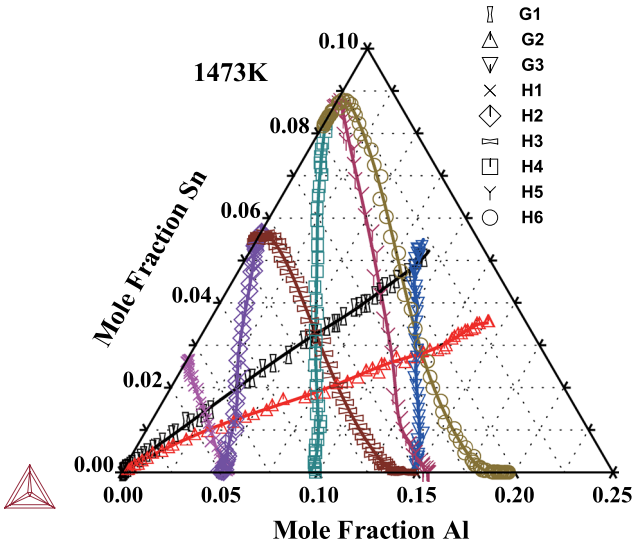

Fig. 2 Diffusion paths for all the Ti-Al-Sn ternary diffusion couples annealed at 1473 K for 10 h

3.2 Composition and Temperature Dependence of Diffusion Coefficients

The impurity diffusivities of Al in Ti-Sn and Sn in Ti-Al binary alloys, i.e. $D^{*}_{Al(Ti-Sn)}$ and $D^{*}_{Sn(Ti-Al)}$ were determined from Sn and Al profiles at the terminal compositions in six ternary couples (H1–H6) by the generalized Hall's method, respectively. As listed in Table 3, $\tilde{D}^{*}_{Al(Ti-Sn)}$ and $\tilde{D}^{*}_{Sn(Ti-Al)}$ decrease respect to adding Sn and Al contents respectively to the binary alloys. It is well known that, in i-j-k ternary, the interdiffusion coefficients satisfy three constraints in Eq 4,[32]

$$\lim_{C_j \to 0} \tilde{D}^{k}_{ii} = \tilde{D}^{\text{binary}}_{ik} \quad \text{(Eq 4a)}$$

$$\lim_{C_i \to 0} D^{k}_{ii} = \tilde{D}^{ik^*}_{i} \quad \text{(Eq 4b)}$$

$$\lim_{C_i \to 0} \tilde{D}^{k}_{ij} = 0 \quad \text{(Eq 4c)}$$

stating that the limits of $\tilde{D}^{k}_{ii}$ and $\tilde{D}^{k}_{ij}$ in the vicinity of the binary are retarded to binary interdiffusion coefficient or the impurity of the third element in binary alloy or zero. Together with the binary ($\tilde{D}_{Ti-Al}$ from Ref 33 and $\tilde{D}_{Ti-Sn}$ from this work) and ternary interdiffusion coefficients have

Table 2 Interdiffusion coefficients in BCC Ti-Al-Sn Alloys at 1473 K

Diffusion couples	Composition, at.%		Interdiffusion coefficients, $10^{-13} m^2/s$				Position of Matano plane, μm			
	Al	Sn	$\tilde{D}^{Ti}_{AlAl}$	$\tilde{D}^{Ti}_{AlSn}$	$\tilde{D}^{Ti}_{SnSn}$	$\tilde{D}^{Ti}_{SnAl}$	G-Al	G-Sn	H-Al	H-Sn
G1-H1	2.4	1.4	6.3	0.8	6.5	0.5	1236	1251	1292	1278
G1-H2	3.4	2.0	5.5	1.9	6.9	0.3	1236	1251	1242	1243
G1-H3	5.8	3.3	8.2	4.6	9.0	2.8	1236	1251	1261	1256
G1-H4	5.8	3.3	8.3	4.4	9.3	2.7	1236	1251	1257	1259
G1-H5	7.8	4.3	8.1	5.0	9.2	3.1	1236	1251	1238	1242
G1-H6	8.5	4.7	7.3	3.5	9.6	3.2	1236	1251	1223	1261
G2-H1	3.2	1.0	7.3	1.8	7.2	0.7	1232	1263	1292	1278
G2-H2	4.2	1.2	7.9	2.0	7.3	0.9	1232	1263	1241	1243
G2-H3	8.3	2.1	11.1	8.2	7.4	1.5	1232	1263	1261	1256
G2-H4	7.6	1.9	9.9	7.5	8.1	1.2	1232	1263	1257	1259
G2-H5	10.5	2.6	15.0	14.0	8.3	2.8	1232	1263	1238	1242
G2-H6	11.8	2.8	14.5	14.0	8.3	2.8	1232	1263	1223	1261
G3-G2	11.5	2.8	15.5	13.5	9.8	2.9	1242	1242	1232	1263
G3-H5	14.5	0.4	8.4	4.4	7.3	0.8	1242	1242	1238	1242
G3-H6	10.6	3.5	8.6	4.9	7.1	2.2	1242	1242	1223	1261
H3-H4	5.8	3.3	8.2	4.2	10.1	3.3	1261	1256	1257	1259
H6 (a)	0.5	8.8	5.3			8.8				
Average	7.2	2.9	9.2	5.9	8.2	2.4				

(a) "Darken-type" couple,[34] local maximum of Sn

Table 3 Impurity coefficients of Al in Ti-Sn and Sn in Ti-Al Binary Alloys at 1473 K

Composition	Impurity diffusivity, m^2/s	Composition	Impurity diffusivity, m^2/s
$\tilde{D}^*_{Al(Ti-0Sn)}$	5.8×10^{-13}[33]	$\tilde{D}^*_{Sn(Ti-0Al)}$	6.4×10^{-13}[18]
$\tilde{D}^*_{Al(Ti-2.6Sn)}$	5.2×10^{-13}	$\tilde{D}^*_{Sn(Ti-5.1Al)}$	5.9×10^{-13}
$\tilde{D}^*_{Al(Ti-5.5Sn)}$	3.7×10^{-13}	$\tilde{D}^*_{Sn(Ti-9.6Al)}$	5.6×10^{-13}
$\tilde{D}^*_{Al(Ti-8.6Sn)}$	2.8×10^{-13}	$\tilde{D}^*_{Sn(Ti-15.7Al)}$	4.6×10^{-13}
		$\tilde{D}^*_{Sn(Ti-19.6Al)}$	2.5×10^{-13}

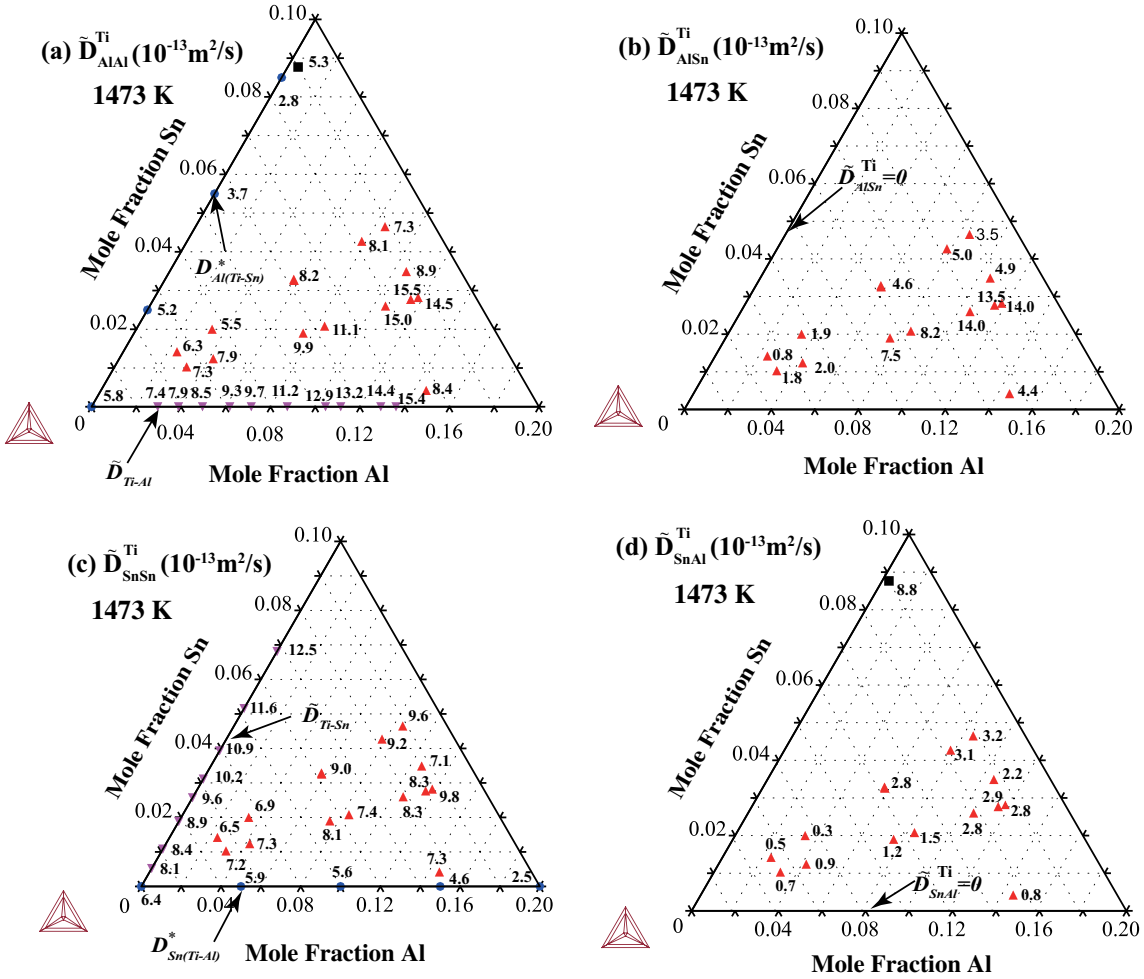

Fig. 3 The main and cross interdiffusion coefficients of the Ti–Al–Sn BCC ternary alloys at 1473 K (a) $\tilde{D}^{Ti}_{AlAl}$; (b) $\tilde{D}^{Ti}_{AlSn}$; (c) $\tilde{D}^{Ti}_{SnSn}$; and (d)$\tilde{D}^{Ti}_{SnAl}$, together with the impurity diffusion coefficients$\tilde{D}^*_{Sn(Ti-Al)}$ and $\tilde{D}^*_{Al(Ti-Sn)}$, and the binary interdiffusion coefficients $\tilde{D}_{Ti-Al}$ obtained from literature[33] and $\tilde{D}_{Ti-Sn}$by the present work

come the mapping of all diffusion rates on the Ti-Al-Sn ternary isotherm in Fig. 3. It is apparent that the main interdiffusion term of the Ti-Al-Sn ternary, either $\tilde{D}^E_{AlAl}$ or $\tilde{D}^{Ti}_{SnSn}$, profoundly enhances as the content of diffusing species increases while weakly depends on that of the others, and this more or less holds for the two cross terms as well. Close inspection of the variations of ternary coefficients versus the composition of diffusing species was illustrated in Fig. 4. A rising linear trend-line was best fit for all four types of interdiffusion data.

The inspection also allows anomalous uphill diffusion to be identified, e.g. Sn diffusion in the couple H6 where Sn is high in content, causing that the Sn profile exhibits a maximum at the marked arrow, i.e. the "Darken-type" couple.[34] In this case, one of the main and one of the cross interdiffusion coefficients at the extreme position can be

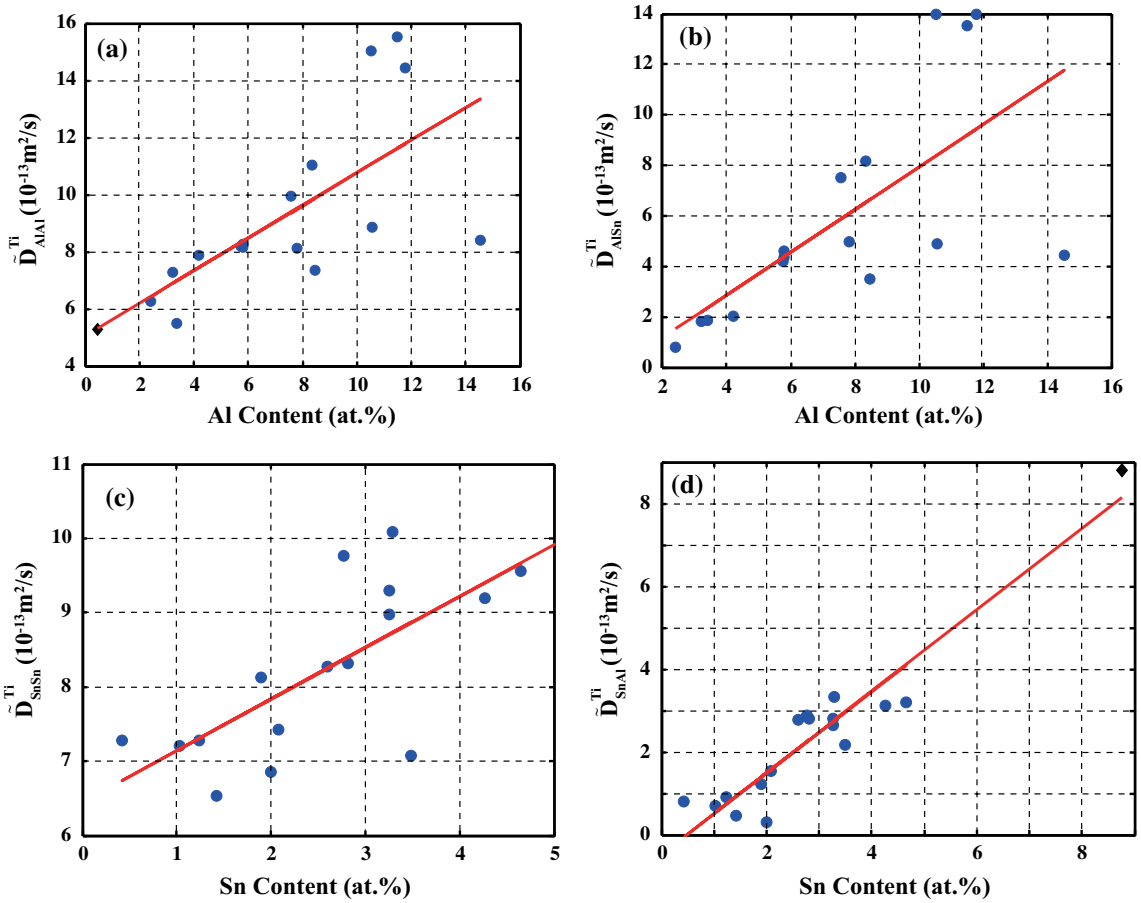

Fig. 4 The variation of ternary interdiffusion coefficients with the compositions. (a) $\tilde{D}^{Ti}_{AlAl}$ with Al and (b) $\tilde{D}^{Ti}_{AlSn}$ with Al; (c) $\tilde{D}^{Ti}_{SnSn}$ with Sn and (d) $\tilde{D}^{Ti}_{SnAl}$ with Sn

Table 4 Average interdiffusion coefficients in Ti-Al-X (Co, Fe, Ni, Cr, V, Mo, and Sn) ternaries at 1473 K

	Average $\tilde{D}^{Ti}_{AlAl}$, m^2/s	Average $\tilde{D}^{Ti}_{XX}$, m^2/s	Average $\tilde{D}^{Ti}_{AlX}$, m^2/s	Average $\tilde{D}^{Ti}_{XAl}$, m^2/s
Ti-Al-Co[11]	1.3×10^{-12}	1.9×10^{-11}	3.3×10^{-13}	2.5×10^{-12}
Ti-Al-Fe[12]	1.3×10^{-12}	1.2×10^{-11}	1.8×10^{-13}	1.1×10^{-12}
Ti-Al-Ni[14]	1.9×10^{-12}	2.2×10^{-11}	2.8×10^{-13}	-0.5×10^{-12}
Ti-Al-Cr[9]	6.8×10^{-13}	4.1×10^{-13}	2.4×10^{-13}	1.4×10^{-13}
Ti-Al-V[10]	7.4×10^{-13}	1.6×10^{-12}	2.8×10^{-14}	2.3×10^{-13}
Ti-Al-Mo[13]	4.5×10^{-13}	1.5×10^{-13}	7.6×10^{-14}	1.4×10^{-14}
Ti-Al-Sn	9.2×10^{-13}	8.2×10^{-13}	5.9×10^{-13}	2.4×10^{-13}

obtained, for H6 those are $\tilde{D}^{Ti}_{AlAl}$(5.3E−13 m^2/s) and $\tilde{D}^{E}_{SnAl}$(8.8E−13 m^2/s) at (0.5 at.% Al, 8.8 at.%Sn).

A systematic comparison of diffusion coefficients of seven Ti-Al-X (Co, Fe, Ni, Cr, V, Mo, and Sn) ternaries at 1473 K was made in Table 4. The average values of interdiffusion coefficients show that Co, Fe, and Ni diffusions are one or two order(s) of magnitude faster than the other four, manifesting themselves with the ultra-fast diffusion anomalies. This is because diffusion of Co, Fe and Ni in BCC-Ti are predominantly of interstitial nature or a mixed mechanism,[11,12,14,35] and the presence of these alloying elements markedly enhances the Al diffusion as well. However, the much lower diffusions in Ti-Al-(Cr, V, Mo, and Sn) occur by the normal vacancy mechanism. Comparing the main interdiffusion terms with the cross ones, the effect of the Al composition gradient on the diffusion of Sn is more significant than that of Co, Fe, V, and Mo.

4 Conclusion

In brief, the interdiffusion behavior of BCC alloys of Ti-Sn binary and Ti-Al-Sn ternary was investigated at 1473 K (1200 °C) by the solid-state diffusion couple technique. The inter and impurity diffusion coefficients at 1473 K (1200 °C) were extracted by the Sauer-Friese, Whittle–Green and Generalized Hall methods, respectively. The interdiffusion coefficient of the Ti-Sn binary was revealed to gradually increase with increasing the Sn content, whereas the main interdiffusion term of the Ti-Al-Sn ternary, either $\tilde{D}^{\mathrm{Ti}}_{\mathrm{AlAl}}$ or $\tilde{D}^{\mathrm{Ti}}_{\mathrm{SnSn}}$, profoundly enhances as the content of diffusing species increases while weakly depends on that of the others. In addition, comparison with Ti-Al-X (Co, Cr, Ni, Fe, Mo, and V) systems, diffusion in Ti-Al-Sn was found to occur by the vacancy mechanism, which is similar to Cr, V, and Mo.

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