Interlayer growth at interfaces of pure titanium/aluminium alloys and pure titanium/pure aluminium friction weld joints

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ABSTRACT

It was clarified that the effect of heat treatment on the intermediate layer (interlayer) growth consisted with intermetallic compound phases at the interfaces of pure titanium (Ti)/aluminium-manganese (Al-1%Mn) alloy and pure Ti/aluminium-magnesium (Al-4.6%Mg) alloy friction weld joints heated up to 873K for up to 180ks (600oC-50H). The mechanism of the interlayer growth was also clarified for pure Ti/pure Al friction weld joint. The followings are concluded. The interlayer growth rate of Ti/Al-4.6%Mg joint was much faster than that of Ti/Al-1%Mn one. The interlayer was consisted with Al_3Ti for Ti/Al-Mn joint, and it was consisted with $Al_{18}Mg_3Ti_2$ for Ti/Al-Mg one. While the interlayer grew from Al alloy substrate to Ti side for Ti/Al-1%Mn joint, it grew from Ti substrate to Al alloy side for Ti/Al-4.6%Mn one. Neither linear nor parabolic time-dependence relation could be applied to the interlayer growth rate for both joints. The interlayer growth stopped for several hours on heating of approximately every 36ks (10H). The direction of the interlayer growth of Al_3Ti for Ti/Al joint was close to <001> and <111> crystal directions. It can be indicated that nucleation and growth of nuclei are necessary for the interlayer growth. These phenomena described above may be the reason for the occurrence of the plateau region and the interlayer growth rate being depended on neither linear nor parabolic heating time-dependence.

INTRODUCTION

Dissimilar welding operations have several severe problems in the industrial usage. One of these occurs when dissimilar welding joints are operated at high temperature environment. That is, intermediate layer (hereafter called as interlayer) consisting of brittle intermetallic compound phases grow at the interface of dissimilar joint, and they gives a detrimental damage on the mechanical and metallurgical properties of the joint. There were a lot of and wide range of studies investigated for diffusion phenomena and interlayer growth at interfaces of dissimilar weld joints or diffusion couples up to date [1-12].

Generally speaking, the relation between interlayer growth rate and heating time is according to parabolic time-dependence, i.e. square root relation (parabolic theory) when the interlayer growth occurs due to mutual diffusion of each element in both substrates joined. However the parabolic theory could not be applied to some combinations of dissimilar joints. For example, it could not be fit to the relation of interlayer width versus heating time of a diffusion couple between pure titanium (Ti) and pure aluminium (Al). In this material combination, the interlayer (Al3Ti) increased with a heating time linearly [1,9,10]. One of the reasons may be the generation of Kirkendall voids at the interface, etc [10]. The authors think that another reason is due to the experimental method. That is, most of researches were carried out to observe the interlayer growth at room temperature with an optical microscope etc., i.e. specimens were cooled after heat treatment. Therefore, the data were intermittent (discontinuous), so that true diffusion phenomena and interlayer growth by long heating time could not be clarified for dissimilar joints because of fracture occurring in interlayer. One of the authors, Fuji, had studied the interlayer growth of pure Ti/pure Al friction weld joint by using in-situ and continuous observation system [13]. Fuji had indicated that one of the reasons is as follows. The interlayer growth was not in accordance with neither linear nor parabolic-time dependence was due to nucleation and growth of nuclei of Al-Ti binary intermetallic phases at the interface [13].

By the way, manganese (Mn) and magnesium (Mg) are ones of the most important elements for Al alloy as industrial usage. However, there are few studies how both elements affect interlayer growth at interface of pure Ti and Al alloy friction weld joints up to date. In this study, the authors aim to clarify the effect of Mn and Mg on the interlayer growth of pure Ti/Al alloy dissimilar friction weld joints during post-weld heat treatment. Furthermore, the authors also intend to study interlayer growth at interface of pure Ti/pure Al friction weld joint by an Orientation Imaging Microscopy (OIM) in order to approve the discussion for interlayer growth described in the former study [13].

EXPERIMENTAL PROCEDURE

The material used for this study was commercially pure Ti. Experimental Al with 1.03mass%Mn (Al-Mn) and Al with 4.63mass%Mg (Al-Mg) were used as foreign metals to examine the effect of Mn and Mg in Al substrates on interlayer growth during post-weld heat treatment. All materials were 17mm in diameter. Polishing with a buff was carried out to finish faying (contacting) surfaces of all materials to decrease the effect of surface contamination on joining. Brake-type friction welding equipment was employed through friction welding operations. The following conditions were kept constant: rotational speed was 25revolution per second (1500revolution per minute); friction pressure was 50MPa; friction time was 2s; upsetting pressure was 50MPa; and upsetting time was 6s. Hereafter, for example, the friction joint of pure Ti/Al-Mn is called as "Ti/Al-Mn joint".

Figure 1 shows the preparation method of specimens to heat treat for (a)conventional vacuum furnace and (b)in-situ observation furnace. Some specimens with 3mm in diameter and 3mm in thickness of Ti/Al-Mn joint for in-situ and continuous observation were extracted perpendicular to the interface including the interface in as-welded joint by a Wire-Electric Discharge Machining. The specimens of Ti/Al-Mn joint were heat treated at 853K for 360ks (580oC-100H) with the same in-situ observation furnace, and the interlayer growth was observed during heat treatment continuously. The observational method was the same as the former report [13]. Other specimens including Ti/Al-Mn joint were heat treated by a conventional vacuum type furnace. All specimens were observed at a half of the radius, i.e. the mid portion between the center axis and the periphery surface of all joints. The heating temperatures were from 773-873K (500-600oC) and heating times were up to 180ks (50H) for Ti/Al-Mn joint. The heating temperatures were from 673-773K (500-600oC) and heating time were up to 180ks (50H) for Ti/Al-Mg joint. SEM analysis was carried out to analyze the chemical compositions of intermetallic compound phase at interface regions.

To clarify the interlayer growth direction, the specimens that were marked at the interface between Ti substrate and Al alloys in as-welded condition by using a micro-Vickers hardness machine were heated with a conventional vacuum furnace. Succeeding to heating, those specimens were polished and the interfaces were observed by SEM.

An Orientation Imaging Microscopy (OIM) was used to obtain crystallographic orientation images in order to distinctly observe grain structure of Al3Ti interlayer at the interface of Ti/Al joint. The sample including approximately 25microns wide Al3Ti was polished with buff. Succeeding to it, colloidal silica solution that contained several-nanometer size SiO2 was used for final polishing. Crystallographic data collection by OIM was performed in a JEOL JSM-6500F scanning electron microscopy (SEM), operating at 25kV under step size of 0.1 to 0.3mm.

RESULTS AND DISCUSSION

Figure 2 shows the relation between heating time and width of interlayer at the interface of Ti/Al-Mn joint by in-situ observation method. Figure 2(a) shows the result by using linear scale for heating time (horizontal axis). On the other hand,

Fig.2 (b) shows that by square root scale. The interlayer growth rates were depended on neither parabolic nor linear relation for heating time. The interlayer almost saturated up to approximately 80microns in width for both joints. It is most important to note that the interlayer growth stopped for a while (several hours) on approximately every 36ks (10H). Neither linear nor parabolic time-dependence relation could be applied to the interlayer growth rate. That is, several plateaus appeared, and these phenomena are almost same as that of pure Ti/pure Al joint reported before [13]. The relation between heating temperature and interlayer growth of Ti/Al-Mn joint heated for 18, 36, 72 and 180ks (5, 10, 20 and 50hours) is shown in Fig.3. The interlayer grew with increasing of heating temperature. Figure 4 shows the relation between heating time and width of interlayer at interface of Ti/Al-Mn joint heated for 773, 823 and 873K (500, 550 and 600oC) by conventional vacuum furnace. Figure 4(a) shows the result with linear scale as heating time and 4(b) shows it with square root scale as it. Neither linear (Fig.4(a)) nor parabolic (Fig.4(b)) time-dependence relation could be exactly applied to the interlayer growth rate.

Figure 5 shows the effect of heating temperature on width of interlayer at the interface of Ti/Al-Mg joint heated for 18, 36, 72 and 180ks (5, 10, 20 and 50hours) by conventional heating method. The interlayer grew with increasing of heating temperature. Figure 6 shows the relation between heating time and width of interlayer at interface of Ti/Al-Mg joint heated for 773, 823 and 873K (500, 550 and 600oC) by conventional vacuum furnace. Figure 6(a) is the result with linear scale as heating time and 6(b) is with square root scale as it. Neither linear (Fig.6(a)) nor parabolic (Fig.6(b)) time-dependence relation could be exactly applied to the interlayer growth rate. It is important to note that the interlayer growth rate of Ti/Al-Mg joint was much faster than that of Ti/Al-Mn joint. For example, while the interlayer width of Ti/Al-Mg joint was approximately 20microns heated at 773K for 180ks (Fig.6(a)), that of Ti/Al-Mn joint was up to 2microns for same heating condition (Fig.4(a)). Even though the heating temperature was higher for Al-Mn joint than that for Al-Mg joint, the layer growth rate of Al-Mn joint was much faster than that of Al-Mg joint.

An example of SEM-microstructure and chemical compositions across the interface region of Ti/Al-Mn joint heated at 873K for 72ks (600oC-20H) is shown in Fig.7. As the chemical compositions of the interlayer were approximately (64-85)mol%Al-(36-17)mol%Ti- 2mol%Si for Ti/Al-Mn joint by SEM-EDS analysis, it can be thought that the interlayer was consisted with Al3Ti including Si, that is, (Al,Si)3Ti. However, there was no Si concentration area occurred at the interface between Ti and (Al,Si)3Ti interlayer that could be observed at the interface between them of pure Ti/industrial pure Al friction welding joint [13]. Figure 8 shows an example of SEM-microstructure and chemical compositions across the interface region of Ti/Al-Mg joint heated at 773K for 72ks (500oC-20H). The chemical compositions of the interlayer were approximately (78-80)mol%Al-(9-8)mol%Ti-(14-14.5)mol%Mg, so that it can be estimated that the interlayer was consisted with Al18Mg3Ti2 [16]. While the width of the interlayer was approximately 10microns for Ti/Al-Mn joint (Fig.7), that of Ti/Al-Mg joint was about 25microns (Fig.8). Si concentration area occurred at Ti substrate adjacent to Al18Mg3Ti2 interlayer. There is crack occurred at the interface between the interlayer and Ti substrate in Ti/Al-Mg joint (Fig.8). This crack was due to thermal stress generated by the differences of thermal expansion between Ti substrate and the interlayer during heating and cooling stages. Si concentrated at Ti substrate adjacent to the interlayer/crack, and the content was approximately 11mol.%. This is same as pure Ti/industrial pure Al friction weld joint [13]. By the way, the chemical compositions of the white particles observed in the Ti/Al-Mg substrate (Fig.8) were approximately 87mol%Al-14mol%Mn by chemical analysis. Therefore, the particle could be thought as the inclusions consisting with Al6Mn that are conventionally precipitated in Ti/Al-Mn alloy during fabricating.

We can see that the interlayer growth rate of Ti/Al-Mg joint was much faster than that of Ti/Al-Mn joint. This reason has not been clarified now because less research is reported for the mutual diffusion coefficients between Ti and Mg. The detail binary phase diagram between them has not been also clarified [e.g.,14]. However, the authors think the reason as follows. Generally speaking, several conditions are necessary that an interlayer consisting with intermetallic compound phase can grows fast [15]. That is: (1)diffusion rate of each element is fast in interlayer, (2)the rate is also fast in layers or substrates adjacent to interlayer, (3)chemical compositions of interlayer are similar to those of substrates neighboring to the interlayer, (4)there is no barrier that decrease diffusion of each element, (5)free energies (minus value) of interlayer are small, and (6)crystal structures of interlayer are close to those of substrates, etc. In this experiment, Al18Mg3Ti2 interlayer occurs at the interface of Ti/Al-Mg joint and Si concentration area existed at Ti substrate adjacent to Al18Mg3Ti2 interlayer (Fig.8). Even though the Si concentration area existed, maybe, the lower free energy of Al18Mg3Ti2 compound than that of Al3Ti one accelerate the layer growth for Ti/Al-Mg joint.

Figure 9 shows the interlayer growth direction at interfaces of Ti/Al-Mn and Ti/Al-Mg joints. After a micro-Vickers hardness machine makes the square marks at the interfaces of both specimens in as-welded condition, the specimens were heat-treated. While the interlayer grew from Al alloy substrate to Ti substrate for Ti/Al-Mn joint, it grew from Al alloy to Ti for Ti/Al-Mg one. The interlayer growth direction of Ti/Al-Mn joint is same as that of Ti/Al joint [8-13]. However, the interlayer of Ti/Al-Mg joint apparently grew in the opposite direction to them.

This reason has not been clarified because no research is reported for the mutual diffusion coefficients and their binary phase diagram between Ti and Mg. However, the authors estimate that the diffusivity and solubility between Ti and Mg affect the layer growth direction. Further investigation must be necessary to clarify this phenomenon.

Figure 10 shows the microstructure of the interlayer at the interface of pure Ti/pure Al joint obtained by Orientation Imaging Microscopy (OIM). The crystal size of Al substrate was very fine, that is, sub-micron orders. However, that of Al3Ti interlayer is approximately 1-5microns, and much larger than Al ones. While the size of crystals that are neighbor to Al substrate is several microns, that of Ti side is 1-2microns or less. Figure 11(a) shows the crystallographic orientation image, that is, the pole figures of Al3Ti interlayer at interface of pure Ti/pure Al joint obtained by OIM. Figure 11(b) shows the inverse pole figure to growth direction of Al3Ti. Figure 14 shows the schematic illustration of the relation between joint direction and interlayer growth direction of Al3Ti crystal structure. The interlayer was consisted with Al3Ti for Ti/Al joint, and the growing direction of Al3Ti was close to its <001> and <111> crystal directions. The crystal direction of the interlayer is to special direction, i.e., <001> and <111> means that the interlayer grows like "epitaxial growth". One of the authors, Ikeuchi indicated that the interlayer growth of Al3Ti was depended on linear time-dependence, not square root one, and this is due to the generation of Kirkendall voids at the interface [10]. However, another reason is can be thought that the nucleation and growth of nuclei is necessary for the interlayer growth. The period of nucleation may be corresponded to the plateau regions shown in Figs.2(a) and (b).

CONCLUSIONS

The effect of post-weld heat treatment (PWHT) on the intermediate layer (interlayer) growth of pure Ti/Al-1%Mn alloy (Ti/Al-Mn) and pure Ti/4.6%Mg alloy (Ti/Al-Mg) friction weld joints was clarified. The mechanism of interlayer growth during PWHT for pure Ti/pure Al (Ti/Al) one was also clarified. The followings are concluded.

(1) The interlayer growth rate of Ti/Al-Mg joint was much faster than that of Ti/Al-Mn one. The interlayer was consisted with Al3Ti for Ti/Al-Mn joint, and it was consisted with Al18Mg3Ti2 for Ti/Al-Mg one.

(2) While the interlayer grew from Al alloy substrate to Ti side for Ti/Al-Mn alloy joint, it grew from Ti substrate to Al alloy side for Ti/Al-Mn one.

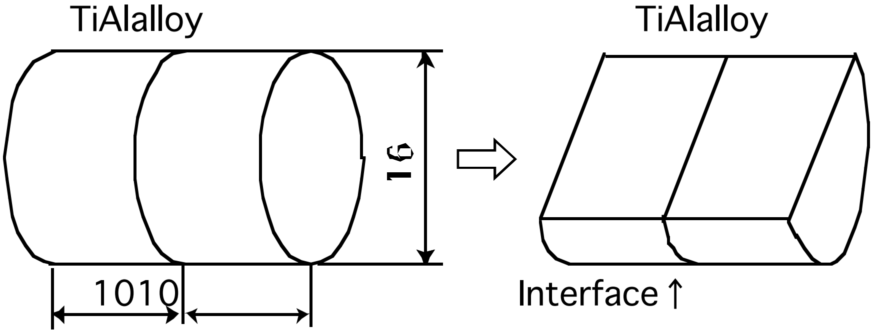
(3) Neither linear nor parabolic time-dependence relation could be applied to the interlayer growth rate for both joints. The interlayer growth stopped for several hours on heating of approximately every 36ks (10H). The nucleation and growth of nuclei is necessary for the interlayer growth.

(4) The direction of the interlayer growth of Al3Ti for Ti/Al joint was close to <001> and <111> crystal directions.

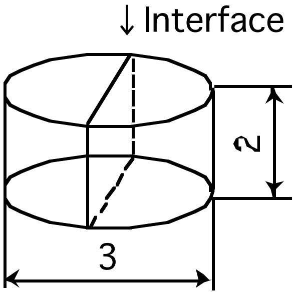
(5) These phenomena described in (4) were the reason for the occurrence of the plateau region and the interlayer growth rate being depended on neither linear nor parabolic heating time-dependence.

REFERENCE

1) F.J.J.van Loo and G.D.Rioeck : *Acta Metall.*, 21 (1973), p.61.
2) F.J.J.van Loo and G.D.Rioeck : *Acta Metall.*, 21 (1973), p.73.
3) D.Goold : *J. of Institute of Metals*, 88 (1959-60), p.444.
4) S.Tsuji : *Trans. of Japan Institute of Metals*, 14 (1973), p.377.
5) Y.Wakamatsu, M.Ohnishi and T.Shimozaki : *J. of Japan Institute of Metals*, 53 (1989), 4, p.378.
6) H.Nakajima and M.Koiwa : *Bulletin of Japan Institute of Metals*, 30 (1991), 6, p.526.
7) J.G.Luo and V.L.Acoff : *Welding Journal*, 79 (2000), 9, p.329s.
8) T.Enjo, K.Ikeuchi, M.Kanai and T.Maruyama : *J. of Japan Welding Society*, 46 (1977), 2, p.83.
9) T.Enjo, K.Ikeuchi and T.Akikawa : *J. of Japan Welding Society*, 50 (1981), 10, p.977.
10) T.Enjo, K.Ikeuchi and T.Horinouchi : *J. of Light Metals (in Japanese)*, 36 (1986), 3, p.143.
11) A.Fuji, K.Ameyama, M.Futamata and Y Shimaki : *The Quarterly J. of Japan Welding Society*, 14 (1994), 1, p.101.
12) T.Hamajima, K.Ameyama and A.FUji : *J. of the Society of Materials Science Japan*, 44 (1995), October, p.1424.
13) A.Fuji : *Science and Technology of Welding and Joining*, 7 (2002), 6, p.414.
14) M.Hansen and K.Anderko : Constitution of Binary alloys (second edition), McGraw-Hill Book Company, 1991, p.923.
15) T.Nishizawa and A.Chiba : *J. of Japan Institute of Metals*, 34 (1970), 6, pp.629.
[The original references are "W. Seith: *Diffusion in Metallen*, Springer-Verlag, Berlin, (1955), p. 177, and G.V.Kidson: *J.Nuc.Mat.*, 3 (1961), p.21."]
16) Villars, A.Prince and H.Okamoto : *Hand book of Ternary Alloy Phase Diagrams*, The Materials information Society, No.4, p.3924.



(a)Forconventionalvacuumfurnace



(b)Forin-situovservation furnace

[unit:mm]

Fig.1 Preparation method for specimen for heat treatment with a conventional vacuum furnace and b in situ observation furnace.

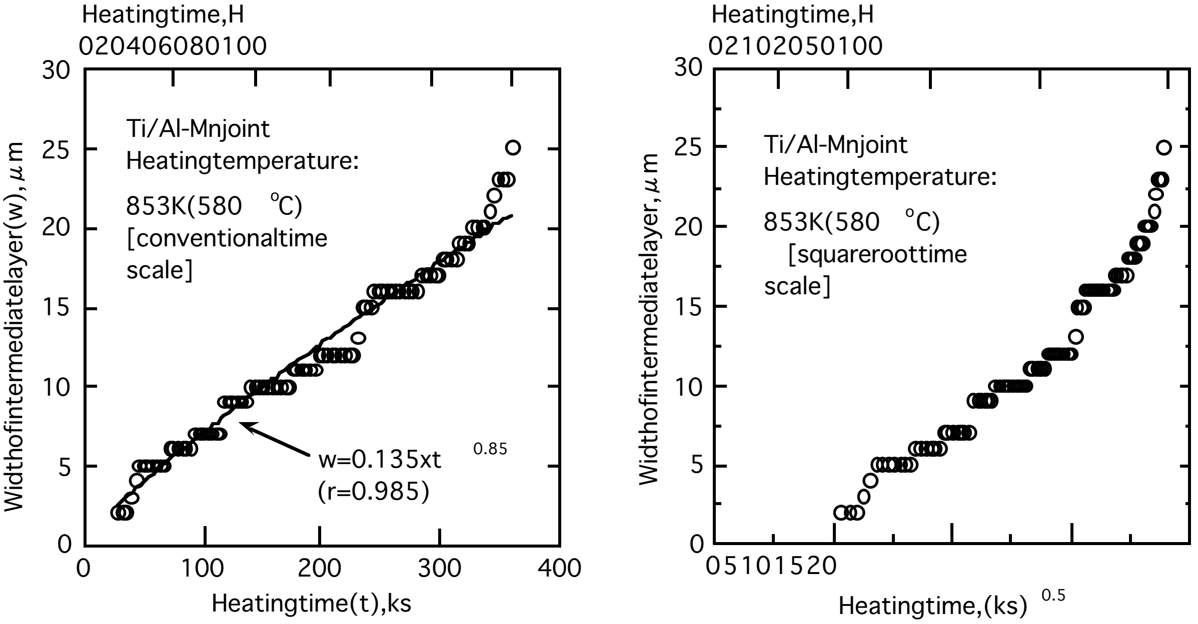


Fig.2 Relationship between heating time and width of intermediate layer at interface of Ti/Al−ﾐMn joint by in situ observation method; a with linear scale for heating time (horizontal axis) and b with square root scale for heating time.

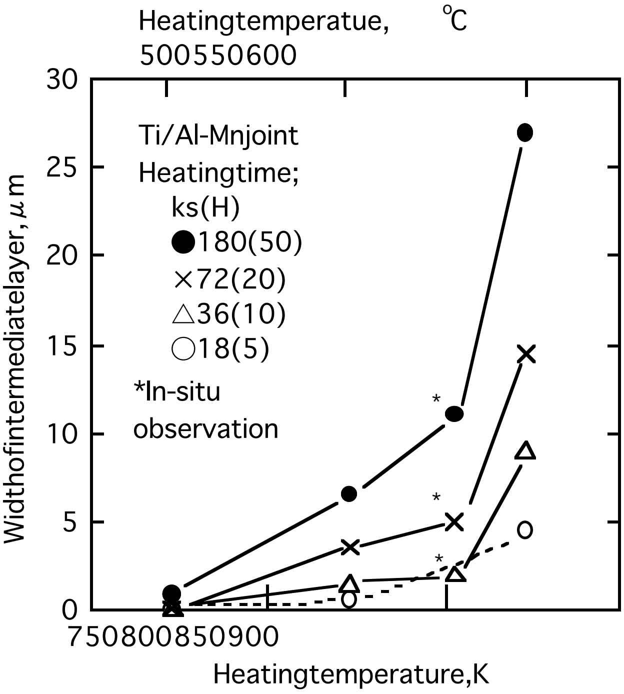


Fig.3 Effect of heating temperature on width of intermediate layer at interface of Ti/Al–Mn joint observed by conventional heating method.

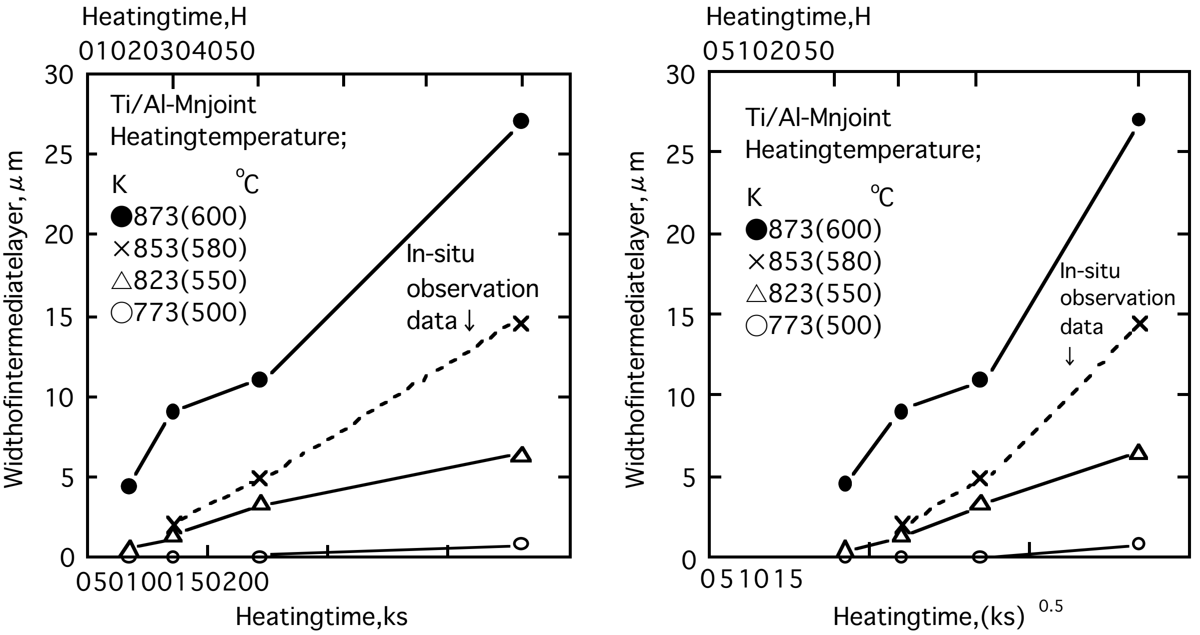


Fig.4 Relation between heating time and width of intermediate layer at interface of Ti/Al–Mn joint by conventional vacuum furnace; a with linear scale for heating time and b with square root scale for heating time.

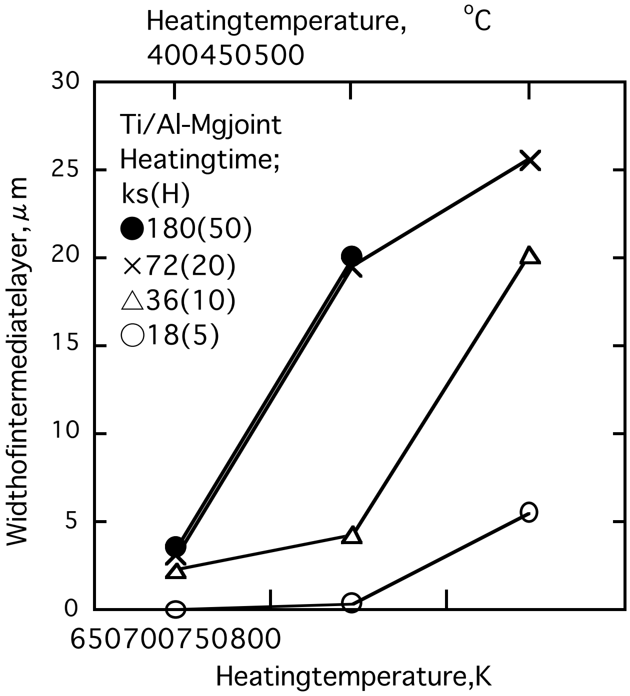


Fig.5 Effect of heating temperature on width of intermediate layer at interface of Ti/Al–Mg joint observed by conventional heating method.

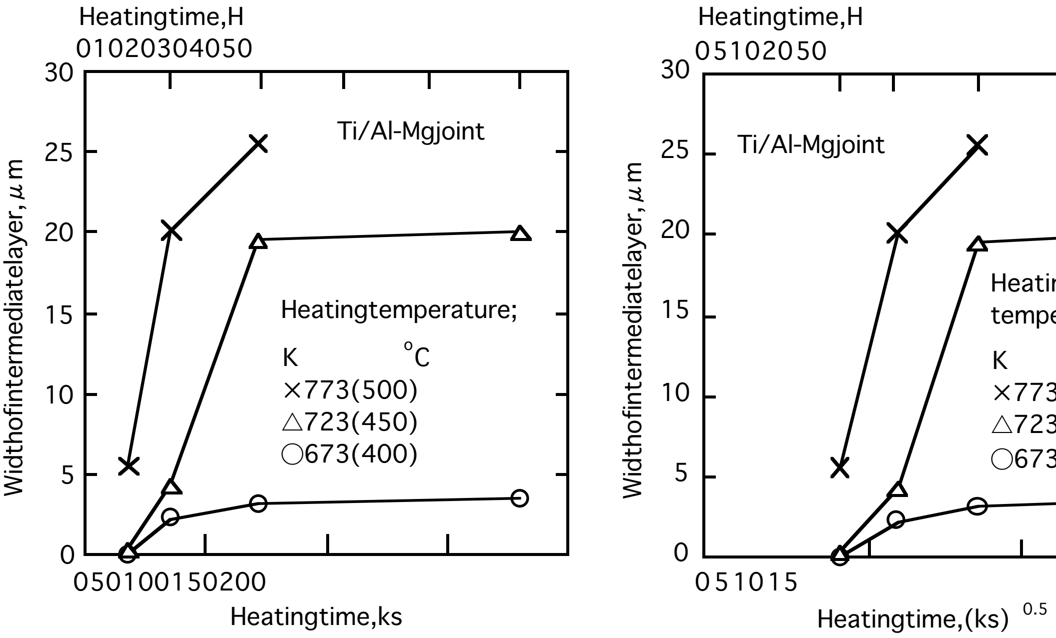
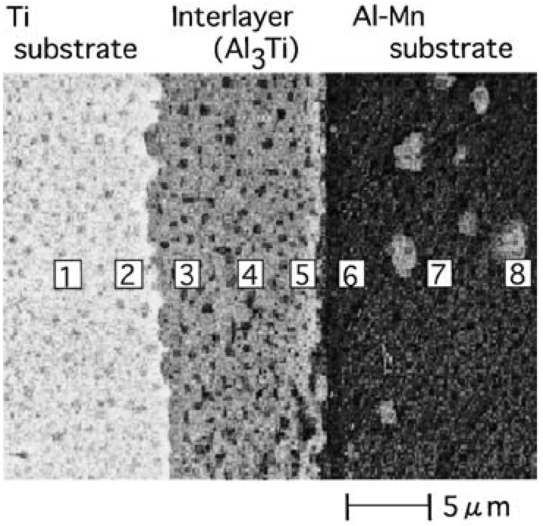


Fig.6 Relationship between heating time and width of intermediate layer at interface of Ti/Al–Mg joint by conventional vacuum furnace; a with linear scale for heating time and b with square root scale for heating time.

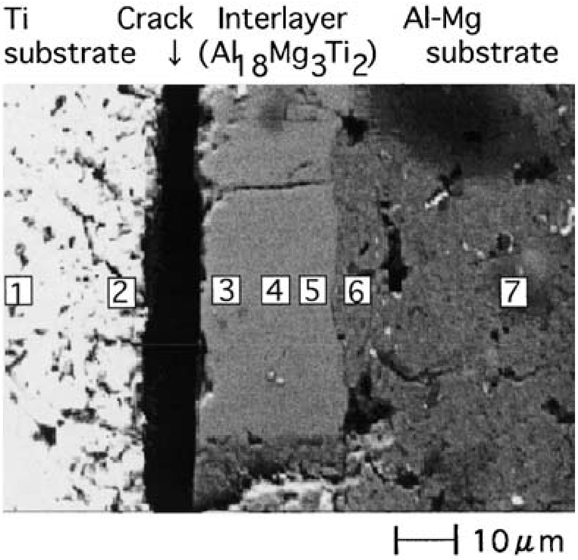


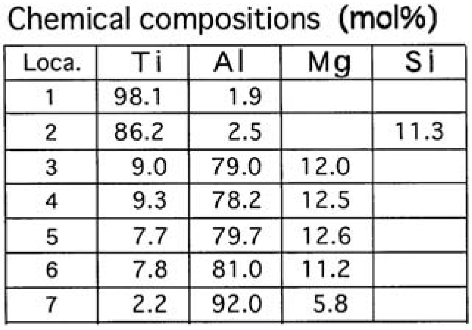
[White particle in Al-Mn is Al_6Mn.]

Chemical compositions (mol.%)

Loca.	Ti	Al	Mn	Si
1	(100)			
2	(100)			
3	36.1	63.9		
4	26.7	71.1		2.2
5	15.0	85.0		
6	11.7	88.3		
7		(100)		
8		98.8	1.1	

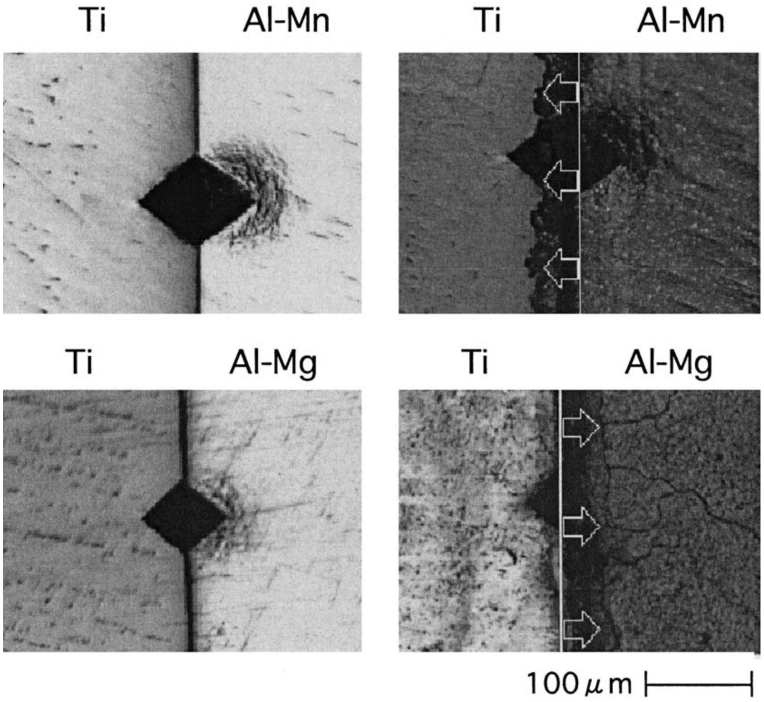
Fig.7 Example of intermediate layer at interface of Ti/Al–Mn joint heated at 873 K for 72 ks (600uC, 20 h)




Chemical compositions (mol%)

Loca.	Ti	Al	Mg	Si
1	98.1	1.9		
2	86.2	2.5		11.3
3	9.0	79.0	12.0	
4	9.3	78.2	12.5	
5	7.7	79.7	12.6	
6	7.8	81.0	11.2	
7	2.2	92.0	5.8	

Fig.8 Example of intermediate layer at interface of Ti/Al–Mg joint heated at 773 K for 72 ks (500uC, 20 h).



(a) AW (b) Heat treated

Fig.9 Growing direction of intermediate layer at interfaces of Ti/Al–Mn and Ti/Al–Mg joints.

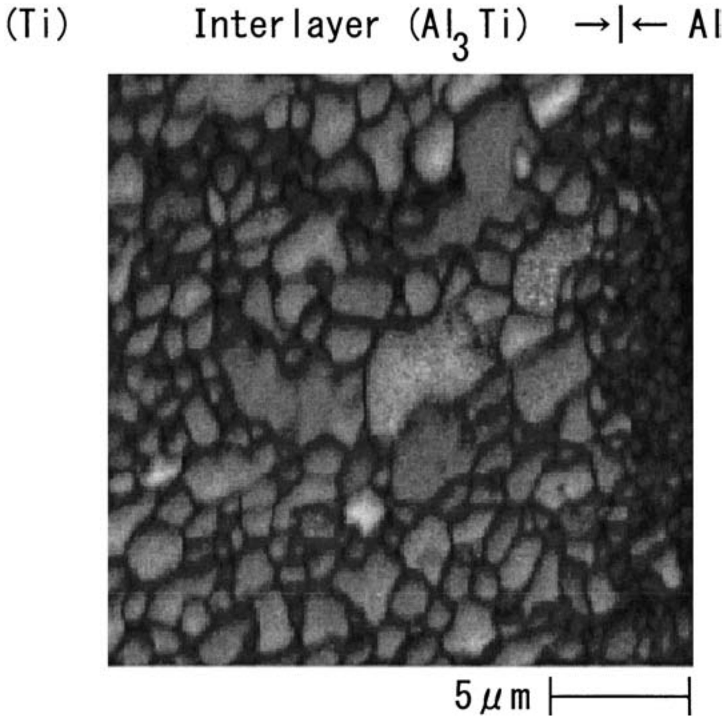


Fig.10 Microstructure of intermediate layer at interface of pure Ti/pure Al joint obtained by Orientation Imaging Microscopy (OIM).

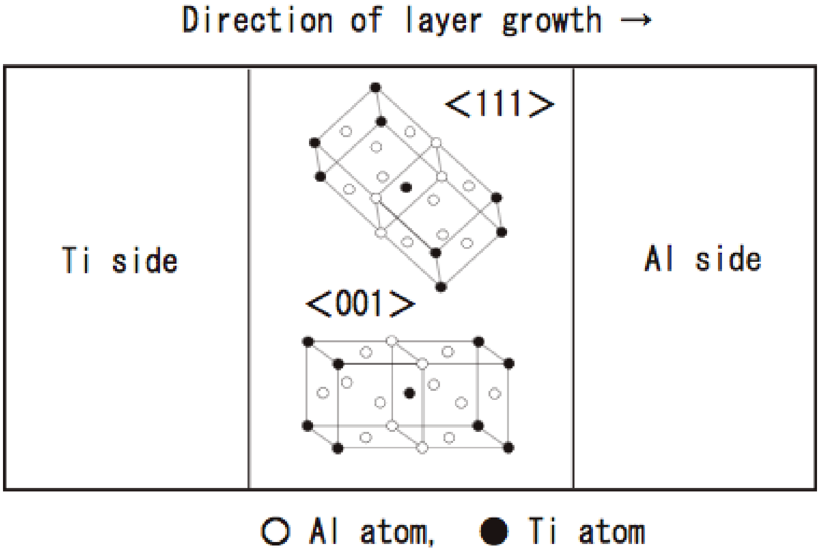


Fig.11 Schematic illustration of relationship between joint direction and growing direction of Al3Ti crystal structure by OIM.