



Title	Effect of Oxide Morphology on Bond Strength of Diffusion-Bonded Interfaces of Al-Alloys(Materials, Metallurgy & Weldability)
Author(s)	Kotani, Keiko; Jung, Jae-Pil; Ikeuchi, Kenji et al.
Citation	Transactions of JWRI. 1999, 28(2), p. 27-37
Version Type	VoR
URL	https://doi.org/10.18910/6852
rights	
Note	

The University of Osaka Institutional Knowledge Archive : OUKA

<https://ir.library.osaka-u.ac.jp/>

The University of Osaka

Effect of Oxide Morphology on Bond Strength of Diffusion-Bonded Interfaces of Al-Alloys†

Keiko KOTANI*, Jae-Pil JUNG**, Kenji IKEUCHI ***, and Fukuhisa MATSUDA****

Abstract

The oxides at the diffusion-bonded interface of a pure aluminum and Al binary alloys (Al-Mg(0.6-2%), Al-Si(1%), Al-Mn(0.5%), Al-Zn(1%), and Al-Cu(1%)) have been investigated by transmission electron microscopy in order to examine the effect of their morphologies on the bond strength of the joint interfaces. As the bonding temperature and Mg content were increased, the strength of the joint interfaces of the Al-Mg alloys increased to levels not less than that of the base metals, and the interfacial oxide altered from continuous amorphous films to dispersed crystalline particles of Al_2MgO_4 and MgO. For the pure aluminum and the alloys, other than those of the Al-Mg system, the strength of the joint interfaces was much lower than that of the base metals at all bonding temperatures employed, and the interfacial oxide remained as amorphous films. These results suggest that the amorphous oxide film prevents an increase in the strength of the diffusion-bonded interface of the Al alloy and that strengths comparable to those of the base metal can be obtained, when almost all amorphous films alter to crystalline particles. When the foils of the Al-Mg alloys were applied as an intermediate layer, the bond strength of the joints of the pure aluminum and the alloys, other than the Al-Mg alloys, was improved significantly, and the oxides at the interface of the intermediate layer to the base metal became crystalline in the same way as those observed in the joints of the Al-Mg alloys. Considering that the morphological change of the interfacial oxide described above can be explained as a consequence of the reductive reaction with Mg, as reported in a previous paper, it can be concluded that the addition of an element having a strong affinity to oxygen, like Mg, into the intermediate layer as well as the base metal can improve the bond strength of the joint interface by changing the morphology of the interfacial oxide from amorphous film to crystalline particle.

KEY WORDS:(Diffusion Bonding)(Oxide Film)(TEM Observation)(Al-Mg Alloy)(Al-Si Alloy)
(Al-Mn Alloy)(Al-Zn Alloy)(Al-Cu Alloy)

1.Introduction

In the diffusion bonding of aluminum and its alloys, the stable superficial oxide film, which remains at the interface of the joint bonded even at temperatures close to the melting point of aluminum, is generally accepted as the most detrimental factor that prevents the metal-to-metal contact and the increase in the joint strength^{1,2)}.

Through TEM observation of the joint interface of commercial aluminum alloys and Al-X binary alloys

(X=Mg, Si, Mn, Zn, Cu)²⁻⁹⁾, we have found that (1) the morphology of the oxide at the joint interface is significantly affected by the alloying element, (2) within the alloying elements investigated, Mg reacts with the interfacial amorphous oxide film to form the crystalline particles of Al_2MgO_4 and MgO depending on its content and temperature, (3) alloying elements of Si, Mn, Zn, and Cu at concentrations less than 1% have almost no influence on the interfacial oxides, and (4) the formation of Al_2MgO_4 and MgO can be explained thermodynamically by assuming that these crystalline

† Received on November 29, 1999

* Joint Researcher (Tokoname Ceramic Research Institute of Aichi Prefecture)

** Foreign Research Fellow (University of Seoul)

***Professor

**** Professor Emeritus (Presently at Japan Power

Engineering and Inspection)

Transactions of JWRI is published by Joining and Welding Research Institute of Osaka University, Ibaraki, Osaka 567-0047, Japan.

Effect of Interfacial Oxide on Joint Strength of Al-Alloy

Table 1 Chemical compositions of base metals used (at%).

Alloy	Mg	Si	Fe	Cu	Mn	Cr	Zn	Ti	Ca	Al
4N pure Al	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	Bal.
Al-0.06%Mg	0.06	<0.01	<0.01	0.00	0.00	<0.01	0.00	0.00	0.00	Bal.
Al-0.1%Mg	0.11	<0.01	<0.01	0.00	0.00	<0.01	0.00	0.00	0.00	Bal.
Al-0.6%Mg	0.56	<0.01	<0.01	0.00	0.00	<0.01	0.00	0.00	0.00	Bal.
Al-0.9%Mg	0.89	<0.01	<0.01	0.00	0.00	<0.01	0.00	0.00	0.00	Bal.
Al-1.1%Mg	1.09	<0.01	<0.01	0.00	0.00	<0.01	0.00	0.00	0.00	Bal.
Al-1.3%Mg	1.30	<0.01	<0.01	0.00	0.00	<0.01	0.00	0.00	0.00	Bal.
Al-1.6%Mg	1.62	<0.01	<0.01	0.00	0.00	<0.01	0.00	0.00	0.00	Bal.
Al-2.0%Mg	2.03	<0.01	<0.01	0.00	0.00	<0.01	0.00	0.00	0.00	Bal.
Al-Si (1%)	0.00	0.98	<0.01	0.00	0.00	<0.01	0.00	0.00	0.00	Bal.
Al-Mn (0.5%)	0.00	<0.01	<0.01	0.00	0.51	<0.01	0.00	0.00	0.00	Bal.
Al-Zn (1%)	0.00	<0.01	<0.01	0.00	0.00	<0.01	1.02	0.00	0.00	Bal.
Al-Cu (1%)	0.00	<0.01	<0.01	0.97	0.00	<0.01	0.00	0.00	0.00	Bal.

Table 2 Diffusion-bonding parameters, where, T_w , t_w , and P_w are the bonding temperature, time, and pressure, respectively.

Alloy	T_w (K)	t_w (ks)	P_w (MPa)
4N pure Al	793-903	1.8	1.0(at $T_w=793$ K)- 0.5(at $T_w=903$ K)
Al-0.06%Mg	793-873	1.8	1.2(at $T_w=793$ K)- 0.7(at $T_w=873$ K)
Al-0.1%Mg	793-873	1.8	1.2(at $T_w=793$ K)- 0.8(at $T_w=873$ K)
Al-0.6%Mg	793-873	1.8	1.2(at $T_w=793$ K)- 1.0(at $T_w=873$ K)
Al-0.9%Mg	793-873	1.8	1.2(at $T_w=793$ K)- 1.0(at $T_w=873$ K)
Al-1.1%Mg	793-873	1.8	1.2(at $T_w=793$ K)- 1.0(at $T_w=873$ K)
Al-1.3%Mg	793-873	1.8	1.2(at $T_w=793$ K)- 1.0(at $T_w=873$ K)
Al-1.6%Mg	793-873	1.8	1.2(at $T_w=793$ K)- 1.0(at $T_w=873$ K)
Al-2.0%Mg	653-873	1.8	5.0(at $T_w=653$ K)- 1.0(at $T_w=873$ K)
Al-Si (1%)	873	1.8	0.8
Al-Mn (0.5%)	873	1.8	1-2
Al-Zn (1%)	873	1.8	0.2
Al-Cu (1%)	873	1.8	1

oxides are formed through the reductive reactions of the interfacial amorphous film of aluminum oxide by Mg.

The present study has been undertaken to examine the effects of these interfacial oxides on the bond strength of the joint interface of the Al alloy. The effects of the Al-Mg alloy intermediate layer on the interfacial oxide and the joint strength have also been investigated in order to establish a thermodynamic approach to the alloy - design for the intermediate layer of the Al alloy joint.

2. Experimental Details

The base metals employed were Al-Mg binary alloys with Mg contents from 0.06 to 1.6at%, 99.99% pure Al(abbreviated as 4N pure Al), and Al-X binary alloys (X=Si, Mn, Zn, Cu) with chemical composition as shown in Table 1. The specimen for diffusion bonding was a round bar 17 mm in diameter and 37 mm in length. The faying surface was finished by electropolishing subsequent to grinding a turned surface on abrasive papers. The electropolishing was carried out in an ethanol solution containing 10vol% perchloric acid at 253 K and 18 V. The diffusion bonding was carried out in a vacuum of 10^{-2}

Pa using an apparatus similar to that reported in a previous paper¹⁰. Bonding temperatures T_w and bonding pressures P_w employed are listed in Table 2. Bonding time t_w was 1.8 ks unless otherwise stated. The bonding pressure was determined so that the bonding zone was deformed by 1-2% (estimated from the increase in the cross-sectional area) during the bonding process.

The intermediate layers employed are thin foils about 50 μm in thickness prepared by rolling the Al alloys of 0.1-2.0at% Mg. The surfaces of the intermediate layer were also finished by electropolishing similar to those of the base metals.

The specimen for the TEM observation was prepared as follows. A thin plate ~ 1 mm in thickness involving the joint interface was cut at angle of 90° with the joint interface, and then ground on 1500 grade emery paper to a thickness of about 100 μm . The plate was further thinned by electropolishing using a twin-jet polishing apparatus. The electrolyte used was a solution of 5vol% perchloric acid in methanol. The electropolishing temperature and voltage were 253 K and 18 V, respectively. The plate was finally thinned by Ar ion milling. TEM observations and EDS analyses were carried out at an acceleration voltage of 200 kV. The diameter of the electron beam for the EDS analysis was about 3nm at the specimen surface.

3. Experimental Results and Discussion

3.1 Tensile strength and fracture morphology of Al alloy joints bonded without intermediate layers

The tensile strengths of the joints of the Al-X alloys ($X=\text{Si, Mn, Zn, Cu}$) bonded at 873 K are shown in Fig.1. All the joints were fractured at the interfaces at stresses no more than 25 MPa. The fracture morphology of these joints is shown in Fig.2. As can be seen from Fig.2(a), the fractured surface of the Al-Si alloy joints presented a mostly featureless appearance without obvious dimple patterns. The fractured surfaces of the joints of the other alloys shown in Figs.2(b)-2(d) also presented appearances even more featureless than that of the Al-Si alloy joint. These fracture morphologies shown in Fig.2 indicate the low bond strength of the joint interface in accord with the results from the tensile test shown in Fig.1.

The tensile strengths of the joints of the Al-Mg alloys bonded at 793K and 873 K are shown as a function of Mg content in Fig.3. In this figure, the

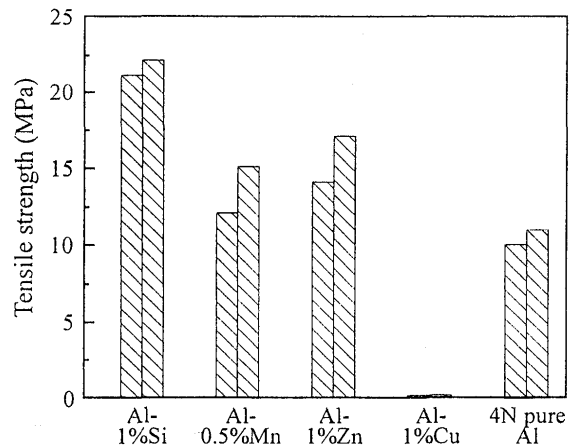


Fig. 1 Tensile strength of the joints of the Al-X alloys ($X=\text{Si, Mn, Zn, Cu}$) and 4N pure aluminum produced by diffusion bonding at 873 K for 1.8 ks ($P_w=0.8$ MPa for Al-1%Si, $P_w=1$ MPa for Al-0.5%Mn, $P_w=0.2$ MPa for Al-1%Zn, $P_w=1$ MPa for Al-1%Cu, and $P_w=0.5$ MPa for 4N pure Al). All the joints were fractured at the joint interface.

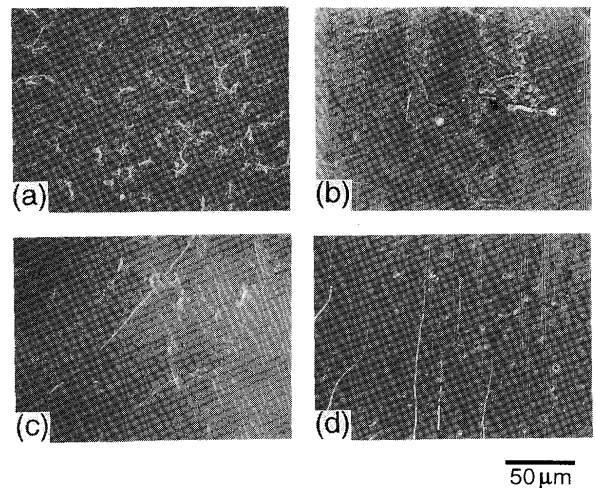


Fig. 2 Fractured surface of the joints of the Al-X alloys ($X=\text{Si, Mn, Zn, Cu}$) ($T_w=873\text{K}$): (a) Al-1%Si ($P_w=0.8\text{MPa}$), (b) Al-0.5%Mn ($P_w=1\text{MPa}$), (c) Al-1%Zn ($P_w=0.2\text{MPa}$), and (d) Al-1%Cu ($P_w=1\text{MPa}$).

morphology of the interfacial oxide observed with a TEM⁸) is also shown for comparison. At a bonding temperature of 793 K, the tensile strength of the joints increased sharply, as the Mg content was increased from 0.6 to 1.1 at%, and at higher Mg contents, the

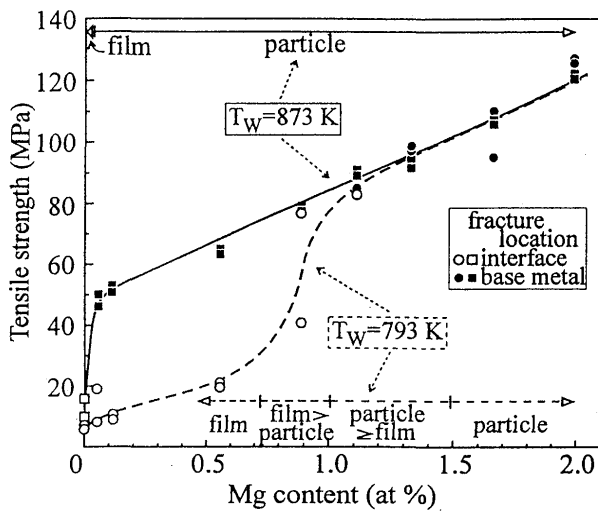


Fig. 3 Tensile strength vs. Mg content for the joints of the Al-Mg alloys bonded at 793 K ($P_w=1-1.2$ MPa) and at 873 K ($P_w=0.6-0.7$ MPa).

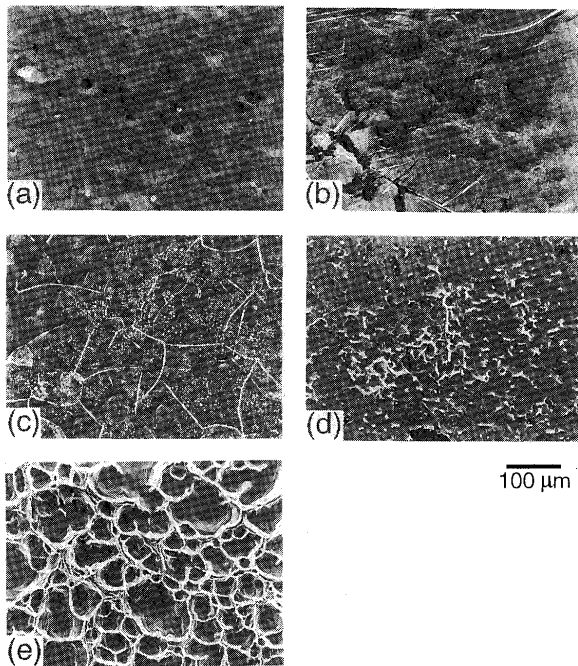


Fig. 4 Fractured surfaces of the joints of the Al-Mg alloys bonded at 793 K: (a) 4N pure Al ($P_w=1$ MPa), (b) Al-0.06% Mg ($P_w=1.2$ MPa), (c) Al-0.6% Mg ($P_w=1.2$ MPa), (d) Al-0.9% Mg ($P_w=1.2$ MPa), and (e) Al-1.1% Mg ($P_w=1.2$ MPa).

tensile strength was not less than that of the base metal. At bonding temperature of 873K, on the other hand, all the joints, except that of the 4N pure Al, showed tensile strength not less than those of the base metals.

The fracture morphologies of the joints of the 4N

pure Al and the Al-Mg binary alloys bonded at 793K are shown in Fig. 4. The fractured surface of the pure Al joint presented a featureless appearance over almost the whole area, as can be seen in Fig. 4(a). A similar appearance was also observed on the fractured surface of the Al-0.06at% Mg alloy joint (Fig. 4(b)). On the fractured surface of the Al-0.6at% Mg alloy joint, small islands showing a ductile morphology were distributed over the featureless areas (Fig. 4(c)). As shown in Fig. 4(d), the ductile island was enlarged, as the Mg content was increased, and ridges, which were probably formed by linkage of the ductile islands, were also observed at a Mg content of 0.9at%. At a Mg content of 1.1at%, dimple patterns were observed over almost the whole fractured surface, although they were smaller and shallower than those observed on the fractured surface of the base metal (Fig. 4(e)). As can be seen from Fig. 4, the morphology of the fractured surface of the joint became more ductile with the rise in Mg content.

The tensile strength of the Al-0.6at% Mg alloy joints is shown as a function of bonding temperature in Fig. 5. As can be seen from Fig. 5, the tensile strength of the joint increased with the rise in the bonding temperature; i.e., the tensile strength was increased from 20 MPa to a level comparable to that of the base metal, as the bonding temperature was increased from 793 to 853K.

The fracture morphologies of the joint of Al-0.6at% Mg alloy bonded at 793-853K are shown in Fig. 6. As shown in Fig. 6(a), the fractured surface of the joint bonded at 793 K presented a mostly featureless appearance, and very small ductile islands were distributed in it. On the fractured surface of the joint bonded at 813 K, ductile areas indicated by 'B' and 'C' became comparable to featureless areas indicated by 'A' as can be seen from Figs. 6(b) and 6(c). At a bonding temperature of 853 K, dimple patterns occupied almost the whole fractured surface as shown in Fig. 6(d), although they were considerably smaller and shallower than those observed on the fractured surface of the base metal. As a whole, the morphology of the fractured surface altered from featureless appearance to ductile appearance with the rise in bonding temperature, as was the case when the Mg content was increased.

As shown in Fig. 7, a similar relation between the tensile strength and bonding temperature was also observed for the joints of Al-2.0at% Mg alloy.

3.2 Relation between tensile strength and interfacial oxide morphology

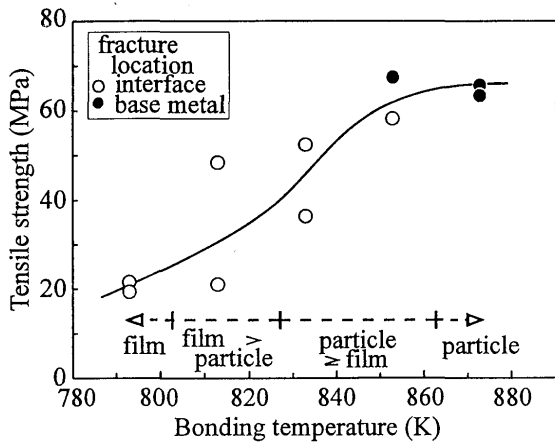


Fig. 5 Tensile strength vs. bonding temperature for the joint of the Al-0.6% Mg alloy ($P_W=1-1.2$ MPa).

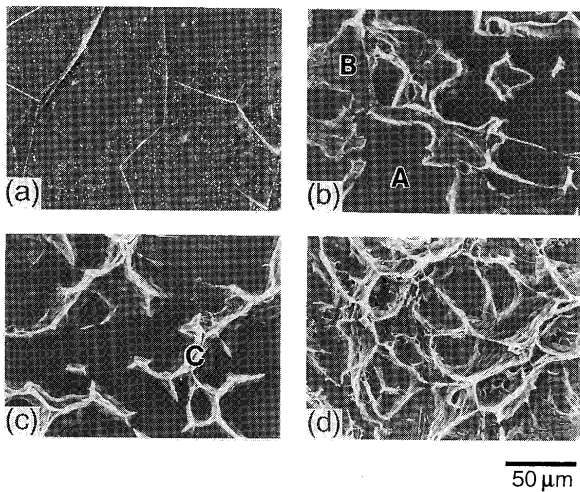


Fig. 6 Fractured surfaces of the joints of the Al-0.6% Mg alloy: (a) $T_W=793K$, $P_W=1.2$ MPa, (b) $T_W=813K$, $P_W=1$ MPa, (c) $T_W=813K$, $P_W=1$ MPa, and (d) $T_W=853K$, $P_W=1$ MPa.

As described in § 3.1, in the diffusion bonding of the Al alloys except the Al-Mg alloys, the tensile strength of the joints was very low compared with those of the base metals even at bonding temperatures ~ 900 K, and almost the whole areas of the joint interfaces were occupied by the continuous amorphous oxide film at all bonding temperatures employed⁹⁾.

In contrast, TEM observations of the joint interfaces of the Al-Mg alloys revealed that (1) at Mg contents not less than 0.06at%, the interfacial oxide altered gradually from the amorphous film to the

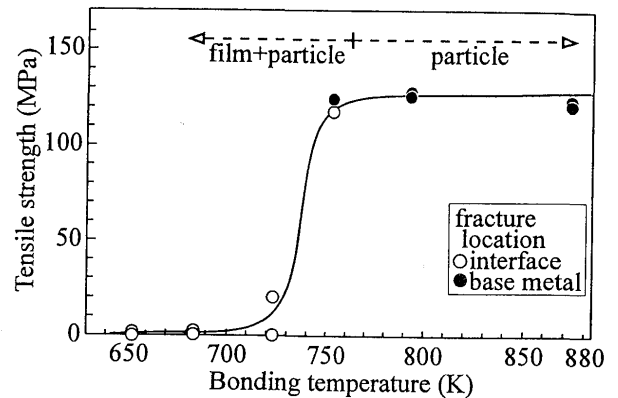


Fig. 7 Tensile strength vs. bonding temperature for the joint of the Al-2.0% Mg alloy ($P_W=1-5$ MPa).

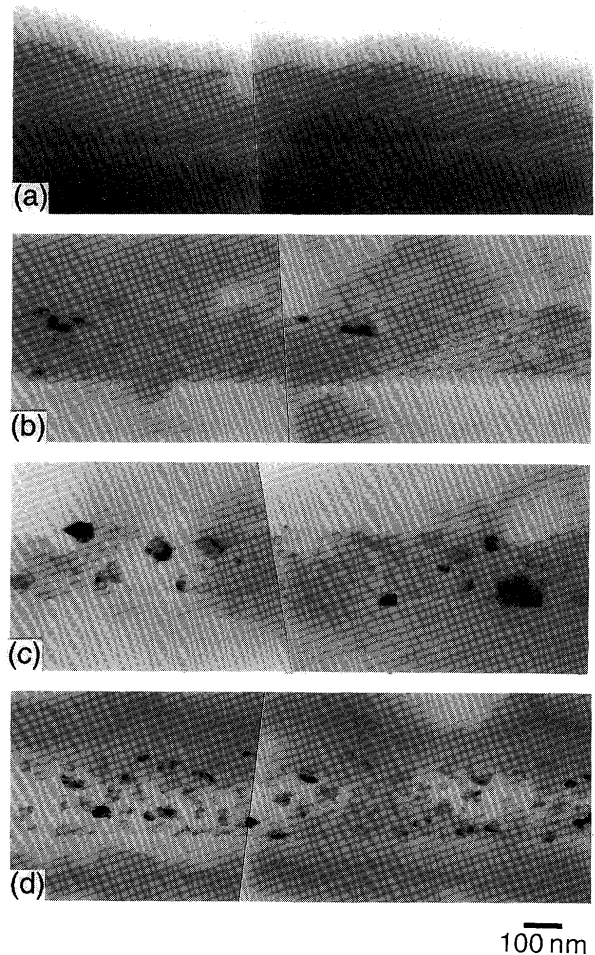

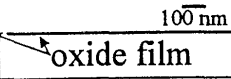

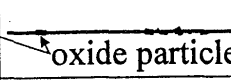
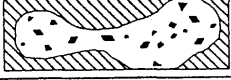
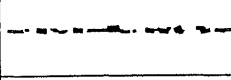

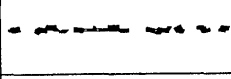

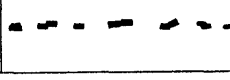


Fig. 8 Electron micrographs of the joint interfaces of the Al-Mg alloys ($T_W=793K$, $P_W=1.2$ MPa): (a) Al-0.1% Mg, (b) Al-0.9% Mg, (c) Al-1.3% Mg, and (d) Al-2.0% Mg.

Stage	Appearance of joint interface		C_{Mg}	T_W	T.S.	F.L.	F.S.
	plane-view	vertical-view					
I			low	low	low		brittle
II							
III							
IV					high		
V			high	high		base metal	ductile

C_{Mg} : Mg content T_W : bonding temperature
 T.S., F.L., F.S. : tensile strength, fracture location,
 and fractured surface of tensile test.

Fig.9 Schematic representation of the evolution of the interfacial oxide morphology with Mg content and bonding temperature, and resulting changes in the tensile properties for the diffusion-bonded joint of the Al-Mg alloy.

crystalline particle, as the bonding temperatures was increased, and (2) the amorphous oxide film was annihilated at lower bonding temperatures, as the Mg content was increased⁸⁾. The morphological change of the interfacial oxide with the Mg content is shown in Fig.8 as an example.

From the comparison of the interfacial oxide morphology shown in Fig.8 with the tensile strength of the Al-Mg alloy joints (Fig.3), it can be considered that the substantial increase in the tensile strength of the Al-Mg alloy joint was accompanied by the annihilation of the amorphous oxide film, and the tensile strength became comparable to that of the base metal, when the amorphous oxide film was annihilated. In accordance with the morphological change of the interfacial oxide from the amorphous film to the crystalline particle (see Fig.4), the fracture morphology of the joint also changed from a featureless appearance to a ductile appearance. At a bonding temperature of 873 K, the amorphous oxide film was annihilated, and all the joints of the Al-Mg alloys were fractured at the base metal in the tensile test (Fig.3).

The tensile strength and the interfacial oxide morphology of the Al-0.6at% Mg and Al-2.0at% Mg alloys joints also exhibited similar dependences on the

bonding temperature.

The relation between the morphology of the interfacial oxide and the bond strength of the interface is schematically illustrated in Fig.9 as a function of the Mg content and bonding temperature. From Fig.9, it can be concluded that the interfacial amorphous oxide film is the most detrimental factor that lowers the tensile strength, and the increase in the bond strength is accompanied by the annihilation of the interfacial amorphous oxide film, which is enhanced by the increase in the Mg content and by the rise in the bonding temperature. Similar relationship between the morphology of the interfacial oxides and the tensile strength of the joints was also found in the diffusion bonding of the commercial Al alloys^{6,7)}.

3.3 Interfacial oxide of Al-X (X=Si, Mn, Zn, Cu) alloys bonded with intermediate layer of Al-Mg alloy

In order to take advantage of the beneficial effect of the Mg addition on the bond strength of the diffusion-bonded interface of the Al-alloy, foils of Al-Mg alloys were applied as intermediate layers to the joint of an Al alloy to which Mg was not added.

The bright field images of the joint interfaces of the 4N pure Al bonded with Al-Mg alloy intermediate

layers are shown in Fig. 10. The joint interface was tilted by 35° to the incident beam. As shown in Fig.10(a), continuous amorphous oxide films less than 10 nm in thickness covered almost the whole joint interface of the 4N pure Al, when the intermediate layer of the Al-0.1at% Mg alloy was applied. When the Mg content was increased to 0.6at%, crystalline oxide particles of 20-100 nm in size coexisted with continuous amorphous oxide films (Fig.10(b)). With a further increase in the Mg content of the intermediate layer, the crystalline oxide particles became predominant as shown in Fig.10(c). Thus, as the Mg content in the intermediate layer was increased, the oxide observed at the interface of the intermediate layer to the base metal also underwent a morphological change similar to that observed in the direct bonding of the Al-Mg alloy to itself.

In the direct bonding of the Al-Mg alloy to itself, Mg atoms were concentrated in the amorphous oxide film prior to the morphological change into crystalline particles, as reported in a previous paper⁹⁾. The amorphous oxide film remaining at the interface of the intermediate layer to the base metal was analyzed by EDS in order to investigate whether its composition changed similarly to that observed in the direct bonding of the Al-Mg alloy. As shown in Figs.11(a) and 11(b), the peaks of O and Mg as well as Al were clearly observed in the EDS spectra from the amorphous oxide films. The observed peak heights of Mg in these spectra were much higher than those detected in the base metal and the intermediate layer. These results indicate that the formation of the crystalline oxide particle at the interface of the intermediate layer to the base metal is also preceded by a concentration of Mg atoms in the amorphous oxide film, similar to that observed in the direct bonding of the Al-Mg alloy.

The crystal structures of the oxide particles observed at the joint interface of the 4N pure Al bonded with the intermediate layers of the Al-Mg alloy at a bonding temperature of 873 K were identified by electron diffraction patterns. As shown in Fig.12, the oxide particle at the joint interface of the 4N pure Al bonded with the Al-0.1at% Mg intermediate layer was identified as Al_2MgO_4 . Similarly, the oxide particle at the joint interface bonded with the Al-0.6at% Mg intermediate layer was also identified as Al_2MgO_4 . At the joint interface bonded with the Al-2.0at% Mg intermediate layer, however, MgO coexisted with Al_2MgO_4 as shown in Fig.13.

In the direct bonding of the Al-Mg alloy, the interfacial amorphous oxide film was annihilated at a

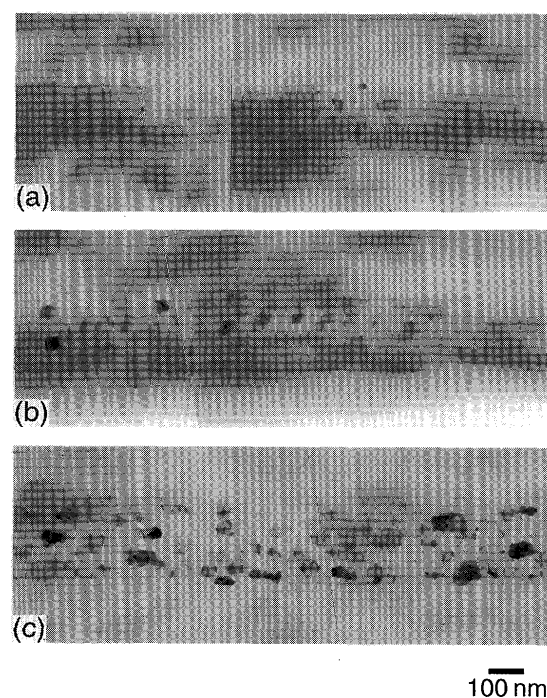


Fig. 10 Electron micrographs of the joint interfaces of the 4N pure Al bonded with the intermediate layers of 0.1% Mg alloy (a), Al-0.6% Mg alloy (b), and Al-2.0% Mg alloy (c) ($T_w=873$ K, $P_w=0.5$ MPa).

bonding temperature of 873 K, when the Mg content of the alloy was not less than 0.06at%⁸⁾. In contrast, when the pure Al was bonded through the Al-Mg intermediate layer, the interfacial amorphous oxide film remained even at a Mg content of 0.6at% as shown in Fig.10. These results suggest that the Mg content of the intermediate layer required to annihilated the amorphous oxide film is higher than that required for the direct bonding of the Al-Mg alloy.

In a previous paper⁸⁾, we have suggested that the concentration of Mg in the interfacial amorphous oxide film is a prerequisite for the morphological change of the interfacial oxide from the amorphous film to the crystalline particle. In case of the direct bonding of the Al-Mg alloy, Mg atoms are supplied from both sides of the base metal, while in case of the bonding of the pure Al with the Al-Mg intermediate layer, Mg atoms are supplied only from the intermediate layer. Therefore, in the bonding of the pure Al with the Al-Mg intermediate layer, it will take a longer bonding time to supply a sufficient amount of Mg atoms to the interfacial amorphous oxide film to initiate its morphological change to the crystalline particle. When Mg atoms are supplied only from the intermediate layer, the Mg atoms must diffuse over a

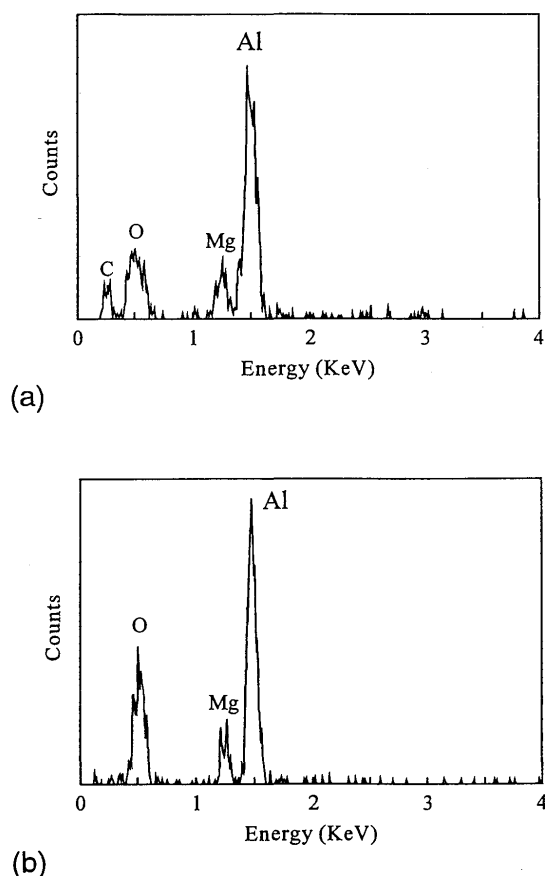


Fig.11 TEM-EDS analyses of the amorphous oxide films at the joint interfaces of the 4N pure Al bonded with intermediate layers of Al-0.1% Mg alloy (a), and Al-0.6% Mg alloy (b) ($T_w=873$ K, $P_w=0.5$ MPa).

distance twice as great as that required in the direct bonding of the Al-Mg alloy in order to supply the same amount of Mg atoms into the interfacial oxide. Generally, the diffusion distance is proportional to the square root of diffusion time; accordingly, the annihilation of the amorphous oxide film by the Mg atom in the intermediate layer will take a bonding time four times as long as that required in the case of the direct bonding of the Al-Mg alloys. This reasoning probably explains why the annihilation of the amorphous oxide film occurs at higher Mg contents of the Al-Mg intermediate layer than in the direct bonding of the Al-Mg alloy to itself. In fact, when the bonding time was increased to 3.6 ks, the amorphous oxide film observed at the joint interface of the pure Al bonded with the Al-0.6at% Mg intermediate layer (see Fig.10(b)) was annihilated to form crystalline particles of Al_2MgO_4 (see Fig.14).

The bright field images of the joint interfaces of the Al-X alloys bonded with the Al-0.6at% Mg intermediate layer are shown in Fig.15. The joint interfaces were tilted by 35° to the incident beam. As

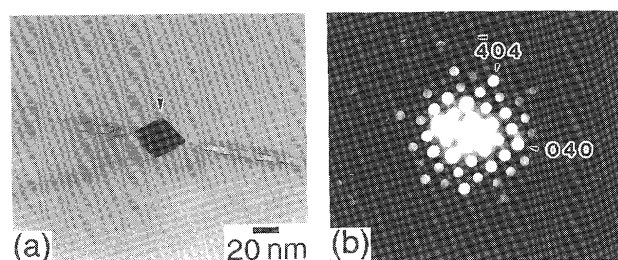


Fig.12 (a) Enlarged micrograph of a crystalline oxide particle at a joint interface of 4N pure Al bonded with the intermediate layer of the Al-0.1% Mg alloy ($T_w=873$ K, $P_w=0.5$ MPa). (b) Electron diffraction pattern from the particle in (a) (beam//[101] Al_2MgO_4).

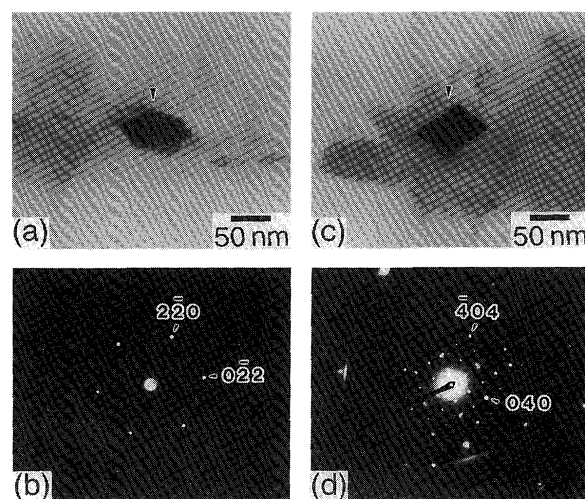


Fig.13 (a),(c) Enlarged micrographs of crystalline oxide particles at a joint of 4N pure Al bonded with the intermediate layer of the Al-2.0% Mg alloy ($T_w=873$ K, $P_w=0.5$ MPa). (b) Electron diffraction pattern from the particle in (a) (beam//[111] MgO). (d) Electron diffraction pattern from the particle in (c) (beam//[101] Al_2MgO_4).

shown in Fig.15(a), almost no continuous amorphous oxide film was observed, and crystalline oxide particles of 20-100 nm in size dominated in the joint interface of the Al-Si alloy bonded with the Al-0.6at% Mg intermediate layer. Similar morphologies of the interfacial oxides were observed at the joint interface of the Al-X ($X=Mn, Zn, Cu$) alloys bonded with the Al-0.6at% Mg intermediate layer. Thus, although the intermediate layer and bonding parameters employed were the same, the amorphous oxide film remained at the joint interface of the pure Al, while it was almost entirely annihilated at the joint interface of the Al-X alloys. The reason for this is not obvious. A possible explanation is that the characteristics of the



Fig.14 Electron micrographs of joint interfaces of 4N the pure Al bonded with the Al - 0.6% Mg intermediate layer ($t_w=3.6$ ks, $T_w=873$ K, $P_w=0.5$ MPa).

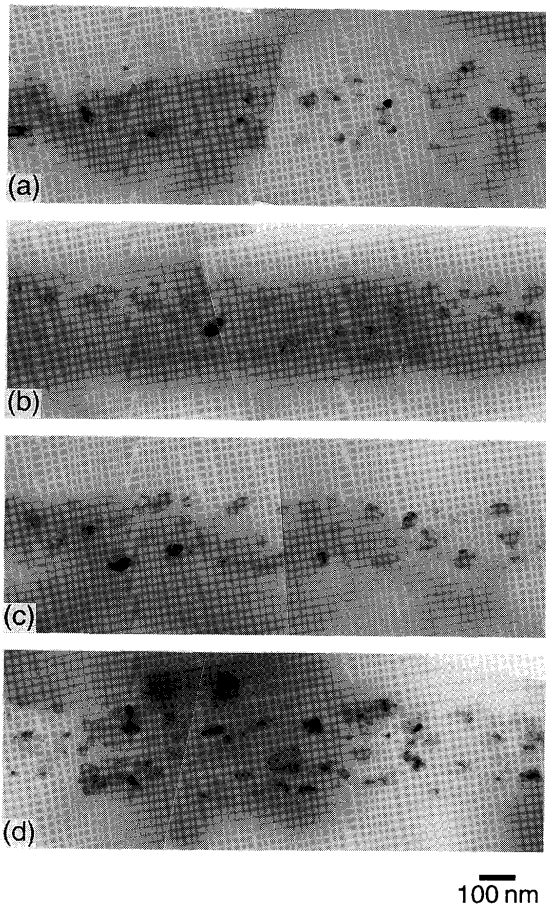


Fig.15 Electron micrographs of the joint interfaces of Al-X alloys (X=Si, Mn, Zn, Cu) bonded with the Al-0.6% Mg intermediate layer ($T_w=873$ K): (a) Al-1% Si ($P_w=0.8$ MPa), (b) Al-0.5% Mn ($P_w=2$ MPa), (c) Al-1% Zn ($P_w=0.8$ MPa), and (d) Al-1% Cu ($P_w=1$ MPa).

superficial oxide film of the Al-X alloys are different from those of the 4N pure Al; i.e, the superficial oxide film of the 4N pure Al seems to be more tenacious than those of the Al-X alloys.

The crystalline oxide particles observed at the joint interface of the Al-X alloys bonded with the Al-0.6at%Mg intermediate layer at 873 K were identified by electron diffraction patterns. As shown in Fig.16, the oxide particle at the joint interface of the Al-Si alloy was identified as Al_2MgO_4 . Similarly, the oxide particle at the joint interface of the other Al-X alloys were also identified as Al_2MgO_4 . Thus, the crystalline oxide particles formed at the joint interfaces of the Al-0.6at% Mg intermediate layer at 873 K were of the same kind as those formed at the joint interface of the 4N pure Al, although their morphologies are slightly different.

3.4 Tensile strength and fracture morphology of joints of 4N pure Al and Al-X (X =Si, Mn, Zn, Cu) alloys bonded with Al-Mg alloy intermediate layer

The results obtained in § 3.3 suggest that the application of an Al-Mg alloy intermediate layer can improve the bond strength of the diffusion-bonded joint interface of the Al alloy by annihilating the interfacial amorphous oxide film.

The tensile strength of the joints of the pure Al and the Al-X alloys bonded with the Al-Mg alloy intermediate layer at a bonding temperature of 873 K is summarised in Fig.17. All these joints showed tensile strength much higher than those bonded without an intermediate layer, and they were fractured at the base metals, except that of the pure Al bonded with the Al-0.1at% Mg intermediate layer.

These results indicate that the relation between the morphology of the interfacial oxide and the bond strength of the joint interface described in § 3.2 holds also in the bonding of the Al alloy with the intermediate layer of the Al-Mg alloy.

Therefore, it can be concluded that an alloying element that fulfills the following requirements can improve the bond strength of the diffusion-bonded joint of the Al alloy by altering the interfacial oxide from the amorphous film to the crystalline particle:

- (1) Its standard free energy of formation of the oxide is lower than that of the aluminum oxide, and
- (2) Its activity in Al solid solution is high enough to permit, thermodynamically, the reduction of the Al oxide film under the bonding conditions.

This effect of the alloying element is also valid,

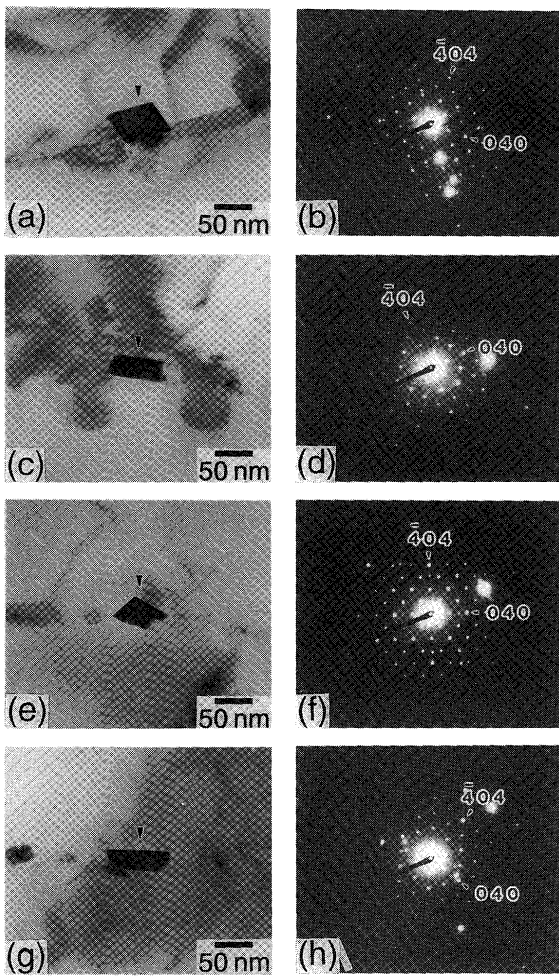


Fig. 16 Enlarged micrographs of crystalline oxide particles at the joint interfaces of the Al-1%Si alloy ($T_w=873$ K, $P_w=0.5$ MPa) (a), the Al-0.5% Mn alloy ($T_w=873$ K, $P_w=2$ MPa) (c), the Al-1%Zn alloy ($T_w=873$ K, $P_w=0.8$ MPa) (e), and the Al-1% Cu alloy ($T_w=873$ K, $P_w=1$ MPa) (g) bonded with the Al-0.6% Mg intermediate layer. (b), (d), (f), and (h) Electron diffraction patterns from the particles in (a), (c), (e), and (g) (beam//[101]Al₂MgO₄).

when it is added to the intermediate layer.

4. Conclusions

The relation between the morphology of the interfacial oxide and the tensile strength of the diffusion-bonded joint has been investigated for the 4N pure Al and Al alloys containing 0.06-2.0at% Mg, 1.0at% Si, 0.5at% Mn, 1.0at% Zn, and 1.0at% Cu. The effects of the Al-Mg alloy intermediate layer on the oxide morphology and the bond strength of the joint interfaces of the alloys have also been investigated to define the intermediate layer

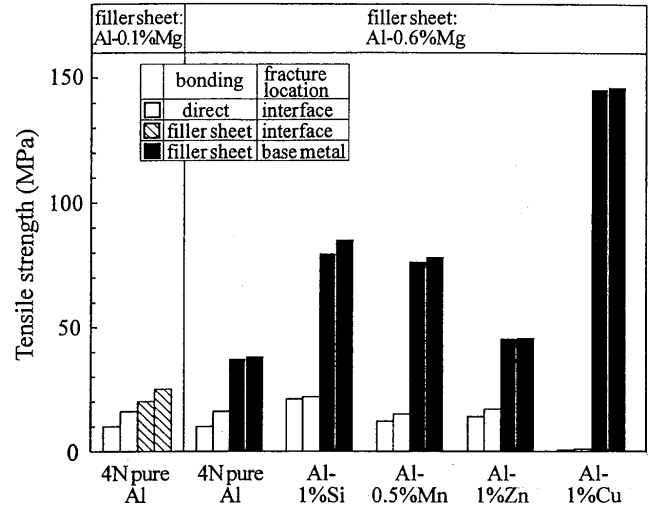


Fig.17 Effects of the Al-Mg intermediate layer on the joint strength of the 4N pure Al and Al-X alloys (X=Si, Zn, Mn, Cu) ($T_w=873$ K). Bonding pressures P_w were 0.5 MPa for the 4N pure Al, 0.8 MPa for the Al-1% Si, 2 MPa for the Al-0.5% Mn, 0.8 MPa for the Al-1% Zn, and 1 MPa for the Al-1% Cu.

characteristics for improving joint strength. The results obtained are summarized as follows:

- (1) In the case of the direct bonding without an intermediate layer, the joints of the pure Al and Al-X alloys (X=Si, Mn, Zn, Cu) showed tensile strengths much lower than the base metals, and were fractured at the interfaces. On the other hand, the tensile strength of the Al-Mg alloy joint was increased with the Mg content, and became comparable to that of the base metal even at bonding temperatures lower than those employed for the bonding of the pure Al and the other alloys.
- (2) From the comparison of the morphology of the interfacial oxide with the bond strength and fracture morphology of the joint, it can be considered that the amorphous oxide film at the joint interface is the most detrimental factor to the bond strength of the interface. The joint strength became comparable to that of the base metal when most of the interfacial amorphous oxide film altered to the crystalline particles through the reductive reaction with Mg.
- (3) When the Al-Mg alloy is applied as an intermediate layer, the reductive reaction of the interfacial amorphous oxides film by Mg can also proceed at the joint interface of the pure Al and Al-X alloys (X=Si, Mn, Zn, Cu). The tensile strength of the pure Al and Al-X alloys can be significantly improved by the application of the Al-Mg alloy intermediate layer.

Thus, the addition of Mg into the intermediate layer as well as the base metal can improve the bond strength of the diffusion-bonded interface of the Al alloy. Considering results from thermodynamic analyses reported in a previous paper ⁸⁾, this effect can be expected if the alloying element satisfies the following conditions: (1) Its standard free energy of the oxide formation is lower than that of the aluminum oxide, and (2) Its activity in the Al solid-solution is high.

References

- 1)T. Enjo, K. Ikeuchi, and N. Akikawa: Trans. JWRI, **10**(1981), 45
- 2)T. Enjo, K. Ikeuchi, and K. Furukawa: Trans. JWRI, **14**(1985), 115.
- 3)T. Enjo, K. Ikeuchi, and H. Fujita: Trans. JWRI, **15** (1986), 117.
- 4)T. Enjo, K. Ikeuchi, and H. Yoshikawa :IIW Doc. No.IX-1561-89,(1989).
- 5)K. Ikeuchi, K. Kotani, and F. Matsuda: Quart. J. Japan Weld. Soc., **14**(1996),122.
- 6)K. Kotani, K. Ikeuchi, and F. Matsuda: Quart. J. Japan Weld. Soc., **14**(1996),382.
- 7)K. Kotani, K. Ikeuchi, and F. Matsuda: Quart. J. Japan Weld. Soc., **14**(1996),389.
- 8)K. Kotani, K. Ikeuchi, and F. Matsuda: Quart. J. Japan Weld Soc., **14**(1996),551.
- 9)K. Kotani, J-P. Jung, K. Ikeuchi, and F. Matsuda: Quart. J. Japan Weld. Soc., **15**(1997), 352.
- 10)T. Enjo, K. Ikeuchi, and T. Horinouchi : Trans. JWRI, **15** (1986),61.