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Bonding of Al_2O_3 to SS41 Steel with LPC Plasma Sprayed Cu-Ti Two-layer Coatings†

Akira OHMORI*, Zhan ZHOU, Akio SUZUMURA***, Yoshiaki ARATA****, Katsunori, INOUE**** and Nobuya IWAMOTO*******

Abstract

Bonding of alumina to SS41 mild steel was investigated by using insert materials of Cu-Ti two-layer coatings and Cu-22wt% Ti composite coating which sprayed at low atmospheric pressure, compared with Cu-22wt% Ti mixed paste and Cu-Ti two-layer foils. The mean shear strength of the joints bonded at 900°C and 0min using Cu-Ti two-layer coatings showed 173MPa. The maximum strengths of the joints bonded using Cu-22wt% Ti composite coating and Cu-Ti two-layer foils were obtained at the bonding time of 30min, and the strength of the joints using Cu-22wt% Ti mixed paste was very low. The strengths of joints bonded after bonding time of 30min using all kind of insert materials decreased with bonding time.

It was recognized that bonding strength of joints were controlled by the eutectic reaction of Ti and Cu in the inter layer and the formation of brittle metallic compound at the interface near SS41 mild steel.

KEY WORDS : (Bonding) (Alumina) (SS41 Steel) (Cu-Ti Two-layer Coatings) (Cu-22wt% Ti Composite Coating) (Cu-Ti Two-layer Foils) (Cu-Ti Powders)

1. Introduction

Ceramics material have been widely used in the various applications for the characteristics of their superior heat and corrosion resistance and etc. However the composite materials of ceramics and metal for producing precise and complex parts are important due to the inferior deformability of ceramics resulted from their brittleness and high hardness. The various methods have been reported and applied to joining of ceramics to metal, such as the well known methods of the diffusion bonding[1-3] and brazing (active metal method) by using eutectic alloys of Ti, Zr active metals with Cu, Ni, Ag, Sn metals[4-6] and so on. As active metal method, the insert materials of various kinds of forms of Cu and Ti have been used for joining of ceramic and metal and they are mainly classified into (1)Cu and Ti eutectic alloy, (2)Cu and Ti mixed powders, (3)two-layer of Cu and Ti foils[7] and (4)Cu-TiH_2 mixed powders. In the case of (1), the foil of amorphous filler metal is used because it is generally difficult to make an alloy into foil when it contains much Ti which induces brittleness. When the foil is used[8-9], bonding temperature of about 1000°C is ncessary for joining of stronger strength. For the case of (2), the eutectic reaction is prevented because the oxides are formed on the surface of fine powders[10]. For the (3), enough temperature and time for reaction between the elements are necessary since the reaction between foils of Cu ant Ti is slow. And for the (4), the decomposition of TiH_2 becomes a important when it is heated.

In this paper, an insert material of two-layer coatings of Cu and Ti which produced by low pressure plasma spraying was used in the joining of Al_2O_3 ceramic and SS41 steel. To solve those problems as mentioned above and obtain a better joint, the bondability and the factors which influences the bonding strength of Al_2O_3 and SS41 steel were examined by using the insert material of Cu-Ti two-layer coatings, compared with mixed paste powders and Cu-Ti two-layer foils.

2. Experimental Procedures and Materials

As ceramics, a sintered alumina (SSA-S: Nihontogyo Co) of purity 99.7wt% was used. Al_2O_3 is 15mm in diameter and 4mm in thickness. As bonding metal, SS41 steel was used, which is 10mm in diameter 4mm in thickness. As bonding insert material, two types of industrial grade pure copper and titanium were used. The particle size of Cu and Ti powders for the spraying are 45-90 μ m and 10-44 μ m. Foils of the pure copper and

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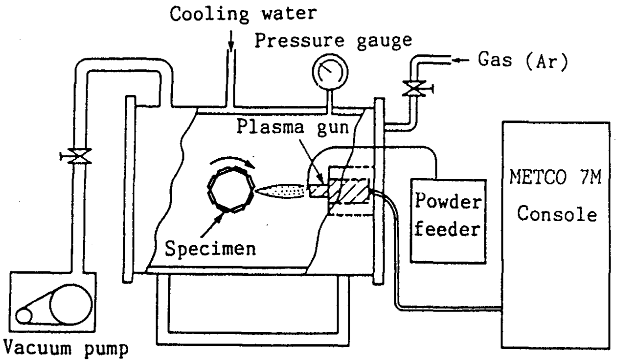

Fig. 1 Schematic diagram of the apparatus of low pressure spraying.

Table 1 Low pressure plasma spraying conditions.

Spray apparatus		METCO 7MB
Spray atmosphere (Ar)		100 Torr
Plasma gas (Ar)	pressure	7 kg/cm^2
	flow rate	42.7 l/min
Auxiliary gas (H_2)	pressure	7 kg/cm^2
	flow rate	6.4~11.8 l/min
Plasma output		15~25 KW
Spray distance		300 mm
Rotation of workpiece holder		200 rpm
Feed rate of stroker		4~10 mm/min

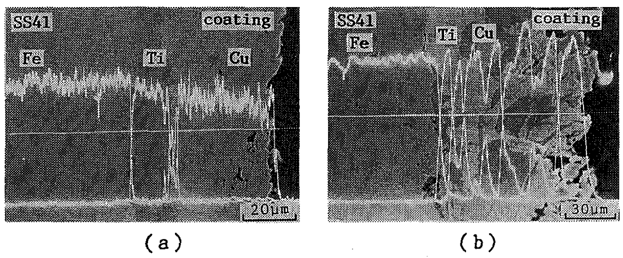

Fig. 2 SEM image and EDX line analysis results at a cross-section of Cu and Ti coating sprayed by LPC on SS41 mild steel. (a), Cu-Ti two-layer coatings; (b), Cu-22wt% Ti composite coating.

titanium of 20 μ m and 10 μ m respectively in thickness were used. Further, Nicrobraz Cement for paste mixed powders was used.

The Cu-Ti two-layer coatings for insert material were sprayed onto sand blasted surface of SS41 mild steel with a low pressure plasma spraying apparatus as shown schematically in **Fig. 1.** During spraying, the sample was attached to a sample holder which was set to rotating and traversing along vertical rotation-axis to plasma jet. The thickness of the coating was adjusted by stroking speed. The spray conditions were shown in **Table 1.** Spraying was carried out under a Ar atmospheric pressure of 100Torr.

The bonding surfaces of Al_2O_3 and inserts were polished with # 600 emery paper, and degreased in acetone with an ultrasonic bath, before the bonding. In the case of inserts of two layer coatings of Cu-Ti, thickness of Cu and Ti coatings were 40 μ m and 25 μ m respectively. Assembly of Al_2O_3-insert-SS41 steel for bonding was set in a vacuum furnace. Then the assembly was heated under a vacuum of 1×10^{-5}Torr to 900°C (Cu-Ti eutectic; 882°C) at the heating rate of 50°C/min. After keeping at the temperature of 900°C for bonding time of 0, 10, 30 or 60min, the assembly was cooled in the furnace at the cooling rate of 1.5°C/min.

Cross section and fracture surface of the obtained joints were investigated by means of optical microscope, scanning electron microscope, EDX analyzer and X-ray diffractometer. And hardness of the cross section was measured. Shear strength of the joints was measured at tensile rate of 0.5mm/min.

3. Results and Discussion

3.1 Effect of bonding conditions on the bonding strength

Figure 2 shows microstructures and EDX analysis results of a Cu-Ti two-layer coatings and a Cu-22wt% Ti composite coating sprayed under the conditions shown in Table 1. It can be recognized that the Ti coating is much dense and oxidation of Ti during spraying is suppressed compared with atmospheric spraying. It is considered that titanium of the coating sprayed under controlled atmosphere will be much active because of suppression of oxidation. Clearly, the coatings also adhered well to the substrate, as shown in Fig. 2. All those characteristics will be beneficial during bonding.

Figure 3 (a), (b), (c) and (d) shows the effect of bonding time on the shear strength of joints obtained with Cu-22wt% Ti powder pastes, Cu-22wt% Ti composite coating, Cu-Ti two-layer coatings and Cu-Ti two-layer foils as insert materials, respectively. It was recognized that bonding of Al_2O_3 with mild steel is difficult and the shear strength of the joints was lower comparing with the other kinds of insett materials, when Cu-22wt% Ti paste were used. It is considered that because the surface of powders was already oxidized before bonding, the eutectic reaction of Cu-Ti in the paste occurred partially and this leads to occurrence of pores and partial reaction at the interface to be bonded as shown in **Fig. 4.** Therefore, the joints showed a low shear strength.

When Cu-22wt% Ti composite coatings were used as the insert material, as shown in Fig. 3(b), the shear strength of the joints increases with the bonding time and reached to the maximum of 165MPa at a joint time of

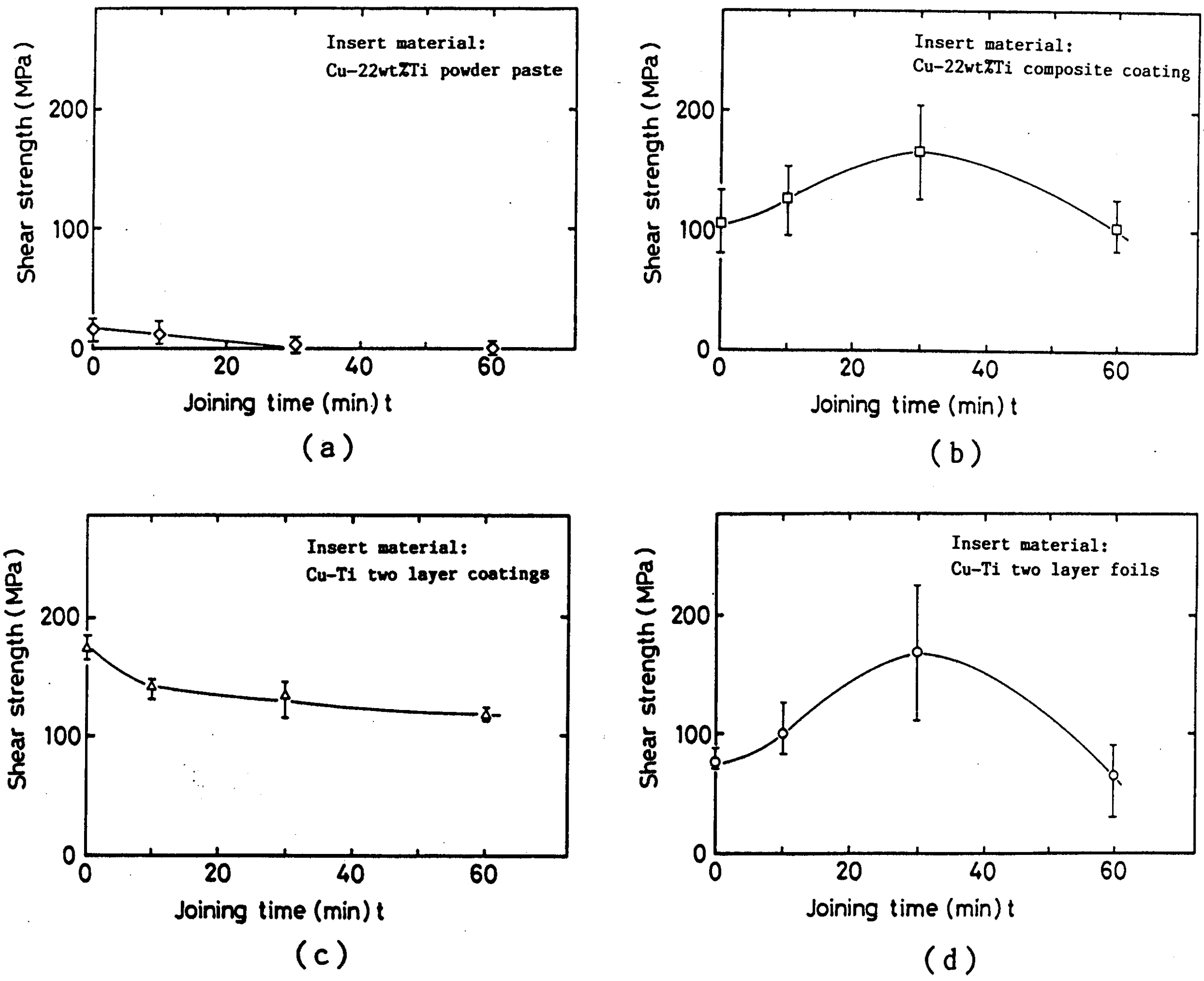

Fig. 3 Effect of joining time on the shear strength of the joints of Al_2O_3 with SS41 mild steel bonded at the temperature of 900°C with various Cu and Ti inster materials.

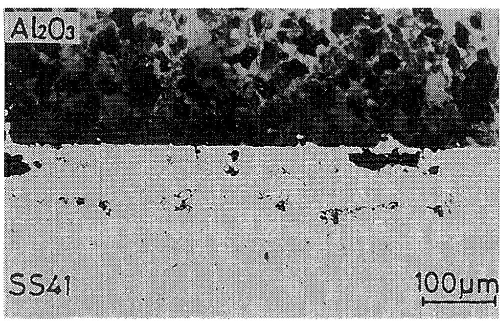

Fig. 4 Optical microstructure of a cross-section of a joint of Al_2O_3 and SS41 steel bonded with using a paste of Cu-22wt% Ti mixed powders as the inster material at 900°C and Omin.

30min. However, with increases of joint time to 60min, a decrease in the strength was recognized. The time dependence on the shear strength was also recognized when the Cu-Ti two-layer foils used.

However, when the Cu-Ti two-layer coatings were used as the insert materials, the maximum bonding strength of joints was obtained at bonding time of 0min. With an increase in joint time, the bonding strength of a joint tended to decrease. When composite coating and two-layer foils were used, the value of the bonding strength showed a large scatter. It is considered that this is due to non-uniform distribution of titanium and copper particles in the composite coatings which was brought on during the deposition because of difference in specific gravity between Cu and Ti. Such a non-uniform distribution will give a low reactivity and low reproducibility of the bonding strength. When Cu-Ti two-layer foils are used, a good ashesion at the interfaces will be difficult without a load during bonding, so that shear strength showed a large scatter. However, the strength of joints bonded with two-layer coatings which keep good contact at the interface showed a little scatter.

From the above results, it is considered that the change of the strength of a joint with bonding time depends largely on the degree of eutectic reaction occurred at the interfaces and insert material part during bonding. The examination of fracture position and analysis results of fractured surfaces with EDX and X-ray diffraction showed that the fractured morphology of a joint changed with kinds of insert materials and strength of joint. The fracture morphologies of joints of alumina and mild steel after shear strength test can be divided into four types as shown schematically in **Fig. 5**, except the joints bonded with Cu-Ti powder paste. A-type shows that fracture occurred at the interface between SS41 mild steel and insert material, where the reaction between the insert material and mild steel was insufficiently. B-type and C-type showed that the fracture occurred inside the insert part and alumina themselves, respectively. From D-type the fracture occurred form alloy-layer composed of Fe-Ti metallic compounds formed at the interface between mild steel and insert meterial. The fracrture at the interface between alumina and insert materials was not recognized. This suggests that the reaction between the alumina and insert material occurs easily for all kinds of insert materials used, which leads to strong bonding. Therefore, the bonding strength of a joint will be controlled mainly by element behavior at the other bond parts of a joint.

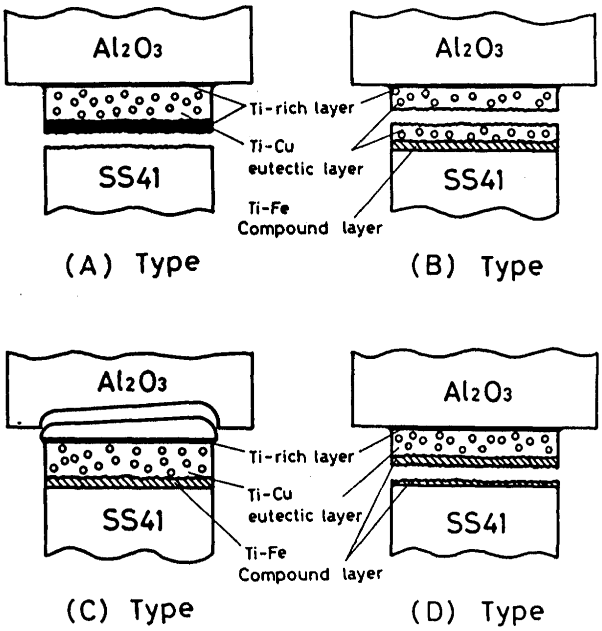

Fig. 5 Schematic diagram of fractural morphologies of joints of Al_2O_3 with SS41 steel bonded with different types of Cu-Ti insert material during the shear test.

3.2 Effect of insert materials on element behavior of joint part

Figure 6 (a), (b) and (c) shows the microstructure and EDX analysis results of alumina-mild steel joints bonded at 900°C, 0min, with Ti-Cu two-layer foils, Cu-Ti two-layer coatings and Cu-Ti composite coating, respectively. As shown in Fig. 6 (a), in the case of Cu-Ti two layer foils, the bond showed two layers of Ti-rich foil and Cu-rich foil. This fact indicates that the diffusion of Cu to inside of Ti foil and eutectic reaction of Cu and Ti did not sufficiently occur. Moreover, it is noted that at interface between mild steel and the insert material, only the

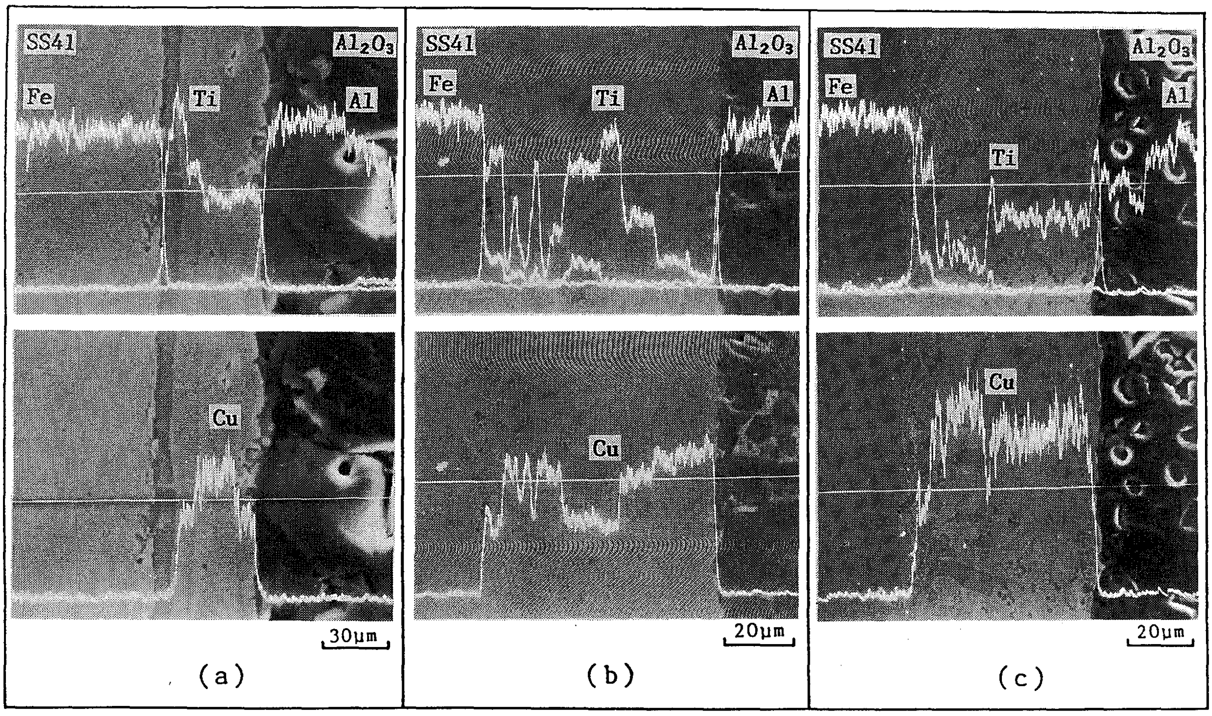

Fig. 6 SEM microstructure and EDX line analysis results of a cross-section of the joints of Al_2O_3 and SS41 steel bonded with using various Cu and Ti insert materials at 900°C and 0min. (a), Cu-Ti two-layer foils; (b), Cu-Ti two-layer coatings; (c), Cu-22wt% Ti composite coating.

diffusion of Ti and Fe at solid state occurred without the diffusion of Cu. However, Ti-rich layer at the interface near alumina was recognized.

On the other hand, in the cases of Cu-Ti two-layer and composite coatings as shown by Fig. 6 (b) and (c), in spite of insufficient eutectic reaction of Cu and Ti, Ti-rich layer near the interface of Al_2O_3-insert and dissolution of Fe into the insert were recognized, by which sufficient bonding was obtained. It was considered that Ti-rich layer at the interface near Al_2O_3 side was composed of Ti oxide and metallic compound of Al and Ti etc, as shown in previous reports[8,11]. However, as shown in **Fig. 7**, many pores were observed at the alloy layer of joint bonded with Cu-Ti composite coating. This is because of non-uniform diffusion of Cu and Ti at a short bonding time owing to the initial nonuniform distribution of Cu and Ti particles in the coating.

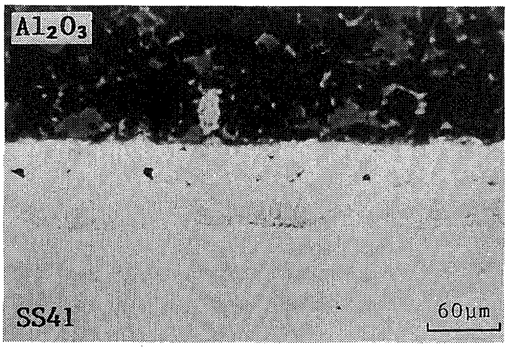

Fig. 7 Optical microstructure of a cross-section of the joint bonded at 900°C and 0min by using Cu-22wt% Ti composite coating.

As shown in Fig. 4, although the bonding at the interface of alumina-insert layer was recognized when using Cu-Ti powder paste as an insert, many pores were observed. This is because oxidation of powder surface suppressed the eutectic reaction.

Based on the above results, it is considered that when the Cu-Ti two-layer coatings is used, the activity of Ti will promote the eutectic reaction between Cu and Ti, so that reaction of Cu-Ti eutectic with both alumina and mild steel become easier. Therefore, even at a short bonding time of 0min, a joint of high bonding strength was obtained by using Cu-Ti two-layer coatings, compared with the other kinds of insert mateial.

Figure 8 (a), (b) and (c) shows SEM micorstructure and EDX analysis results of joint bonded at 900°C and 30min with Cu-Ti two-layer foils, two-layer coatings and composite coating, respectively. Compared with those of joints bonded at 900°C and 0min, it was recognized that the eutectic reaction progressed further at 30min. Furthermore, the diffusion of Fe towards the insert at the interface near mild steel can also be observed even in the case of Cu-Ti two-layer foils. In particular, at the interface near mild steel, alloy layer of Fe-Ti-Cu formed in the joint bonded with two layer and composite coatings. In the joint bonded with two-layer foils,

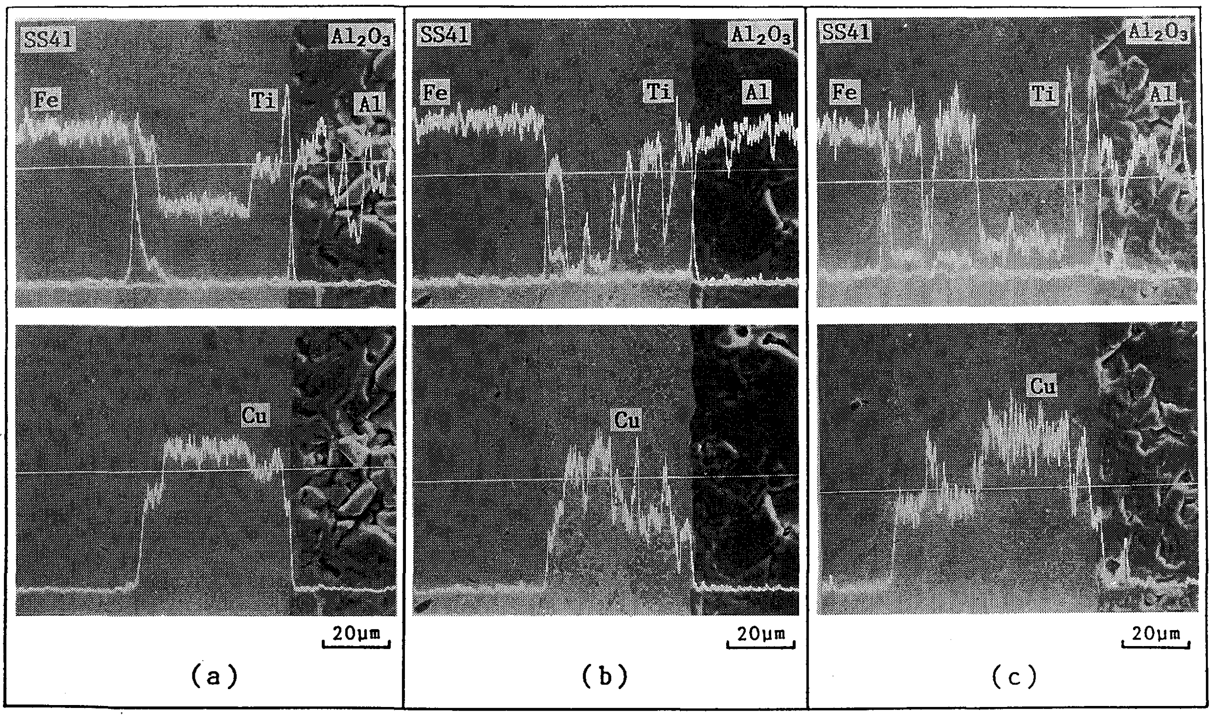

Fig. 8 SEM microstructure and EDX line analysis results of a cross-section of the joints of Al_2O_3 and SS41 steel bonded with using various Cu and Ti insert materials at 900°C and 30min. (a), Cu-Ti two-layer foils; (b), Cu-Ti two-layer coatings; (c), Cu-22wt% Ti composite coating.

formation of Cu-Ti alloy layer in the joint and Ti-rich layer at the interfaces of Al_2O_3-insert and mild steel-insert were observed. As shown in Fig. 8 (b) and (c), the microsturcture became more homogeneous.

Figure 9 (a), (b) and (c) shows the microstructure and EDX analysis results of the joints bonded at 900°C and 60min with Cu-Ti two-layer foils, two-layer and composite coatings. Compared with microstructure of joints bonded at 0min and 30min, the eutectic reaction progressed more completely and the penetration of Fe from mild steel to the insert reached to over half of bonded layer to form Fe-Ti-Cu alloy layer. On the other hand, in the joint bonded with two-layer foils, formation of Ti-Fe alloy layer was recognized remarkably at the interface of insert-mild steel.

From the above results, by using Cu-Ti two-layer coatings as an insert material, the bonding of Al_2O_3 and mild steel was carried out easily at a short bonding time because the eutectic reaction of Cu-Ti and reaction of the eutectic with both Al_2O_3 and mild steel took place more easily, compared with the other kinds of insert materials.

3.3 Factors controlling the shear strength of joints

As shown in Fig. 3, the shear strength of the joint bonded at 900° and 0min with Cu-Ti two-layer coatings was about 173MPa, which is much higher, compared with the joints bonded with the other kinds of insert materials. The joints bonded by using two-layer coatings, showed fracture of C-type on inside of Al_2O_3 and gave a high bonding strength because the reaction took place easily at both interfaces near Al_2O_3 and mild steel side. However, the joints bonded with composite coatings showed B-type fracture and gave a lower shear strength, becasue formation of pores in insert layer of the joint.

On the other hand, when Cu-Ti two-layer foils were used, the eutectic reaction of Cu-Ti was slower, as shown in Fig. 6 (a). The joint showed A-type fracture at the interface near mild steel and lower bonding strength. Therefore, the degree of eutectic reaction affected largely the bonding strength of the joint at short time bonding of 0min. With the increase in the bonding time, the eutectic reaction proceeded and bonding strength of the joints increased. The bonding strength reached maximum value at 30min and the joint showed C-type fracture.

While the composite coatings were used the pores in insert layer in the joint decreased with an increase in bonding time. Therefore, the bonding strength of the joint also increased with bonding time, and reached maximum value at 30min and the joint showed C-type fracture.

However, at a joint time of 60min, the strength of the joint decreased largely compared with that at 30min and all joints bonded with each type of insert materials showed D-type fracture. As shown in Fig. 9, at each bonded joint, Ti-rich layer at the interface near Al_2O_3

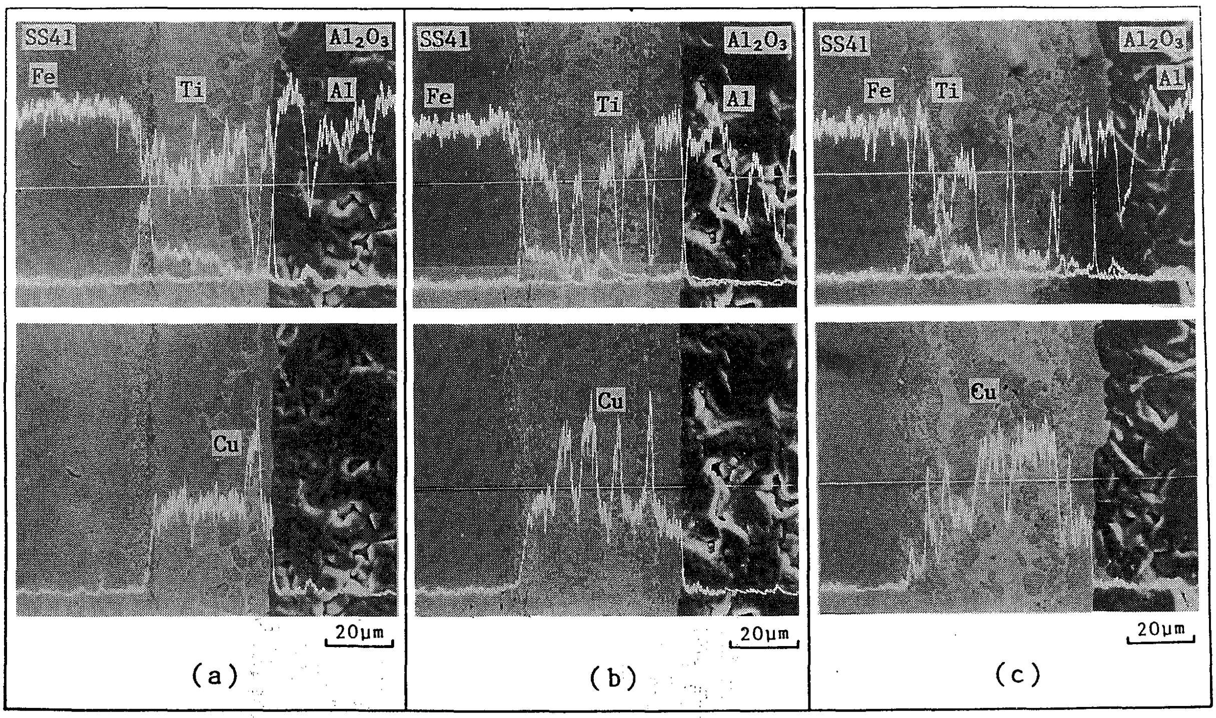

Fig. 9 SEM microstructure and EDX line analysis results of a cross-section of the joints of Al_2O_3 and SS41 steel bonded with using various Cu and Ti insert materials at 900°C and 60min. (a), Cu-Ti two-layer foils; (b), Cu-Ti two-layer coatings; (c), Cu-22wt% Ti composite coating.

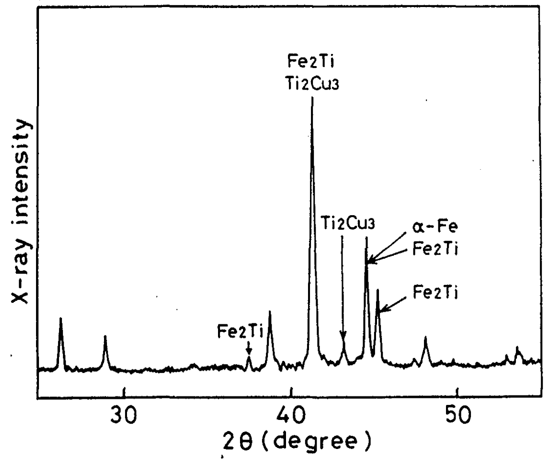

Fig. 10 X-ray diffraction pattern for a fractured surface of the joint of Al_2O_3 and SS41 steel with Cu-Ti two layer foils as an insert material at 900°C and 60min.

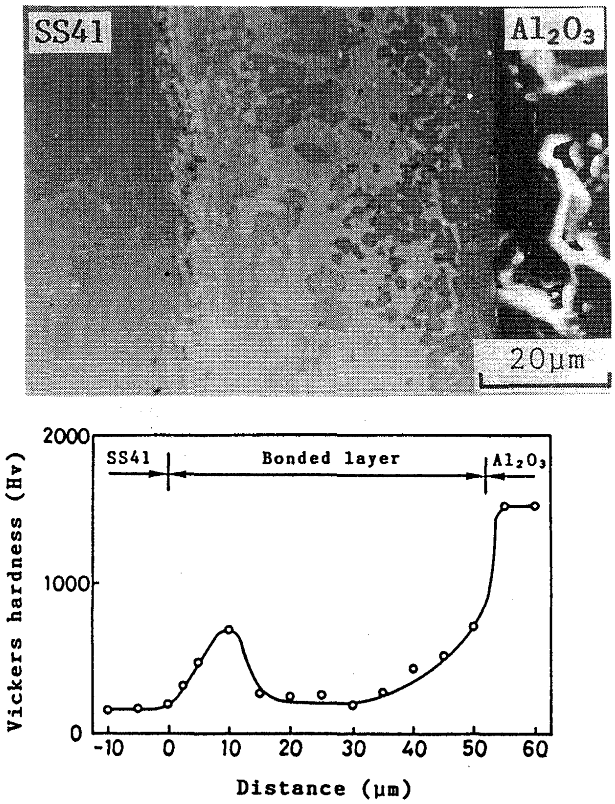

Fig. 11 SEM image and distribution of Vickers hardness across the joint between Al_2O_3 and SS41 steel bonded with Cu-Ti two layer coatings at 900°C and 60min.

side formed, but at the interface near mild steel the diffusion of Fe into the insert layer became intensive to form Fe-Ti alloy layer.

Figure 10 shows the result of X-ray diffraction of fractured surface of the joint bonded with Cu-Ti foils, of which the decrease of bonding strength was most remarkable. It can be recognized that there exists a brittle metallic compound of Fe_2Ti at fractured surface. **Figure 11** shows the distribution of Vickers hardness of cross section of the joint bonded at 900°C and 60min with Cu-Ti two-layer coatings with the microstructure. At the vicinity of the interface near mild steel, a layer of high hardness of 700Hv, which is about four times of Cu-Ti eutectic alloy, was clearly seen. By taking the result of X-ray diffraction of Fig. 10 into consideration, it is considered that the layer of high hardness appeared in Fig. 11 is mainly composed of brittle metallic compound. The occurrence of such metallic compound caused the decrease of the bonding strength and fracture of D-type.

From the above results, during the bonding with two-layer coatings, the eutectic reaction progressed and Fe-Ti compound layer also formed with increase of time. When bonding time exceeded 10min, all the joints showed D-type fracture and the decrease of bonding strength. Therefore, at a short bonding time, the joint of highest bonding strength was obtained.

In order to suppress the formation of Fe-Ti compound near mild steel, Cu-Ti-Cu multilayer foils were used as insert material. The bonding strength of the joint bonded at 900°C and 60min with Cu-Ti-Cu multilayer foils was shown in **Fig. 12** compared with that of a joint bonded with Cu-Ti two-layer foils. The former was about two times large than the latter. This is because that owing to the existence of Cu foil contacted directly with mild steel the direct reaction of Fe with Ti, which leads to the easy formation of Fe-Ti metallic compound, was suppressed. Therefore, under condition of long time bonding, the formation of brittle metallic compound of

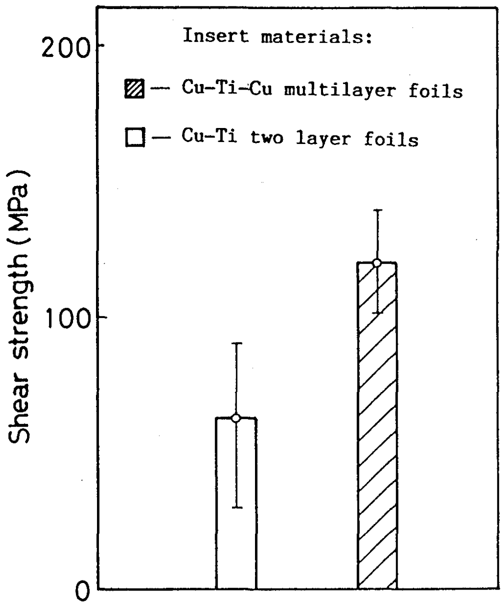

Fig. 12 Comparison of the shear strength of joints of Al_2O_3 with SS41 steel bonded at 900°C and 60min by using Cu and Ti multilayer foils in different assembling orders.

Fe-Ti at the vicinity of the interface near mild steel with control the bonding strength of the joint of Al_2O_3-mild steel.

4. Conclusions

The bonding of Al_2O_3 and SS41 mild steel was investigated by using insert material of Cu-Ti two-layer coatings, compared with paste mixed powder and Cu-Ti two-layer foils. From the bonding strength of joint and elements behavior in the joint part, the factors controlling the shear strength of the joint were examined. The results obtained in the present study are summarized as follows.

(1) The bonding of Al_2O_3 with mild steel by using the Cu-Ti two-layer coatings was carried out at short time and low temperature and the joint showed mean bonding strength of 173MPa.

(2) It was recognized that the bonding of Al_2O_3 and SS41 mild steel by using Cu-Ti two-layer coatings was more efficient owing to easiness of the eutectic rection compared with the Cu-Ti two-layer foils and the Cu-22wt% Ti mixed paste.

(3) It was defined that the bonding strength of joint was controlled by the eutectic reaction of Cu and Ti and the formation of brittle metallic compound at the vicinity of the interface near SS41 mild steel.

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