Friction Bonding of Ceramics to Metal with Intermediate Layer†

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Abstract

Intermediate layers of various metals ranging from active metals to noble metals have been applied to the friction bonding of ceramics to metal, and their influences on the bondability have been systematically investigated. The ceramics specimens to be bonded were SiC (pressureless-sintered silicon carbide) and ZrO_2 (partially stabilized zirconia including 3mol% MgO), and the metal specimen was Cu (oxygen free copper). When foils of active metals Al, Ti, Zr, and Nb were applied as an intermediate layer, the bondability of Cu to both SiC and ZrO_2 was improved remarkably, and bonded joints of Cu to SiC and ZrO_2 could be obtained. In contrast to this, when intermediate layers of Fe, Ni, and Ag were applied, the bondability was not influenced significantly by the intermediate layer, i.e., similarly to the case of bonding without an intermediate layer, the SiC and ZrO_2 specimens separated from the Cu specimen immediately after the bonding operation without application of an external load. In Cu next to the ceramics-metal interface, a region extending over ~10^2 µm from the interface was formed in which the intermediate layer was mixed mechanically with Cu, but no reaction layer between the ceramics and metals could be found.

KEY WORDS: (Friction Bonding) (Metal-Ceramics Bonding) (SiC) (ZrO2) (Copper) (Intermediate Layer) (Active Metal) (Noble Metal)

1. Introduction

There have been many investigations reported of the diffusion bonding and brazing of ceramics to metal[1-6]. An important result from these investigations is that the bondability of ceramics to metal is improved remarkably by the application of an intermediate layer (or a filler) containing active elements such as Ti, Zr, Nb, and Cr. The effect of the addition of an active element on the bondability has been ascribed to the formation of an interfacial layer resulting from the reaction between the active metal and ceramics. The interfacial reaction layer has been taken to act as a binder between the ceramics and metal, having atomic bonds and a crystal structure of a completely different type.

On the other hand, there have been few papers about the bond strength obtained by bringing very clean surfaces of a metal and ceramics into contact with each other at room temperatures in an ultra high vacuum[7,8]. They have suggested that more active metals such as Al, Fe, and Ni were bonded more strongly to ceramics than Cu and Ag. In this case, since no reaction layer can form during the contacting process at room temperatures, the contact strength of the interface may reflect directly the strength of the atomic bond between metal and ceramics surfaces. This is supported theoretically by papers that calculated the binding energy of metal atoms on the ceramics surface[9,10].

The friction bonding has been widely applied to joining of dissimilar metals[11]. However, to our knowledge, there has been little amount of information about the friction bonding of ceramics to metal[12,13]. A prominent feature of the friction bonding is that the formation of the observable intermetallic compound layer during the bonding process can be suppressed by suitable choice of bonding parameters, even when the system of the dissimilar metals forms intermetallic compounds according to the phase diagram. Therefore, it can be expected that a ceramics-to-metal joint with a very thin reaction layer or without a reaction layer can be obtained by the friction bonding. On the other hand, little attention has been paid to the application of the intermediate layer to the friction bonding, since a wide variety of dissimilar metal combinations can be

successfully friction-bonded by a suitable choice of bonding parameters. We think, however, an appropriate intermediate layer will be required to friction-bond dissimilar materials, such as ceramics and metal that have completely different properties[13].

Therefore, we have applied intermediate layers of various metals ranging from active metal to noble metal to the friction bonding of ceramics to metal, and have investigated the following:

(1) the effects of the intermediate layer of active metals on the bondability,

(2) the roll of the interfacial reaction layer in achieving high joint strength, and

(3) the effect of the friction of the ceramics surface with the metal on the bondability.

For this, we have employed an oxygen free copper (Cu), a kind of noble metal, as a metal specimen to be bonded so that the effect of the intermediate layer of active metal can be observed distinctly. Ceramics specimens employed are a silicon carbide (SiC) and partially stabilized zirconia (ZrO$_2$) that have excellent resistance to thermal shock, considering the rapid heating and cooling rates of the thermal cycle of friction bonding. Intermediate layers employed are foils of Al, Ti, Zr, Nb, Fe, Ni, and Ag.

2. Experimental Details

The SiC specimen used was produced through pressureless-sintering at 2273 K with the aid of C, and the ZrO$_2$ specimen contained 3mol% MgO and received HIP treatment at 1723 K at 200 MPa. The Cu specimen contained 5 ppm O and was annealed at 773 K. The ceramics and metal specimens were rods 16 mm in diameter. The intermediate layers were foils of Al 10 μm thick, Ti 20 μm thick, Zr 20 μm thick, Nb 25 μm thick, Fe 30 μm thick, Ni 20 μm thick, and Ag 15 μm thick. The surface of the Cu specimen was finished to JIS 3S by turning in a lath, and the surface of the ceramics specimen was finished by grinding. These surfaces were degreased by washing in acetone just before the bonding. The friction bonding was carried out with a brake-type machine by pressing a Cu specimen to a turned ceramics specimen. The following two friction-bonding conditions were employed: (I) rotation speed N = 40 s^{-1}, friction time t_1 = 3 s, friction pressure P_1 = 20 MPa, forge time t_2 = 6 s, and forge pressure P_2 = 70 MPa, and (II) N = 53 s^{-1}, t_1 = 6 s, P_1 = 20 MPa, t_2 = 6 s, and P_2 = 70 MPa. Condition II was employed to investigate the effect of the friction of the ceramics surface with the metal on the bondability, by increasing the rotation speed and friction time. In order to avoid

rapid oxidation of the intermediate layer during friction bonding, the bonding zone was sealed from the atmosphere by pouring liquid nitrogen. The joint efficiency was estimated from tensile strength. The as-bonded joint was subjected to the tensile test without machining. A device was developed to allow the tensile test on the as-bonded joint.

3. Results and Discussion

3.1 Friction bonding of silicon carbide to oxygen free copper

The joint strength of SiC to Cu and the longitudinal deformation of Cu during the bonding process were influenced strongly by the intermediate layer as shown in **Fig. 1**. When the intermediate layers of Fe, Ni, Cu (direct bonding), and Ag were applied, the Cu specimen separated from the SiC at the interface immediately after the bonding operation without applying an external load. In contrast to this, when the intermediate layers of active
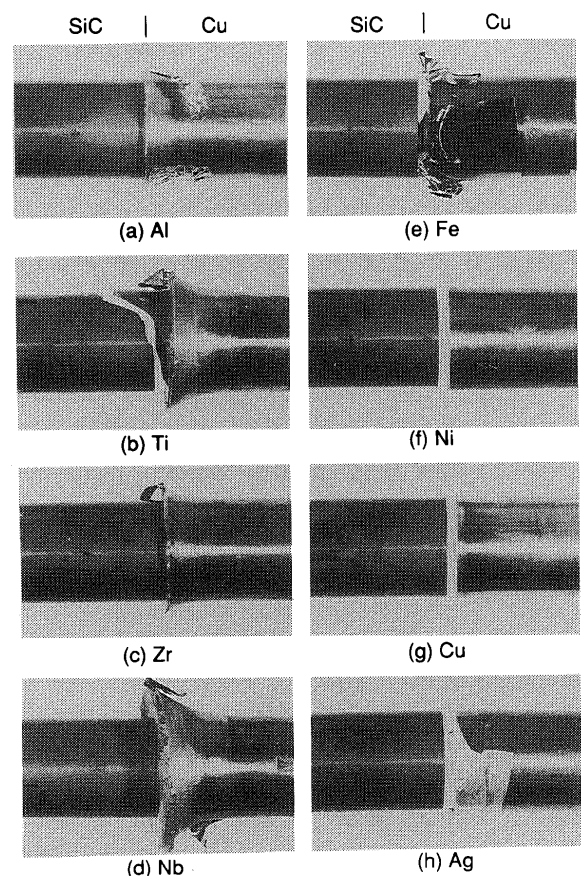

Fig. 1 Appearance of friction-bonded joints of SiC to Cu with the intermediate layers of Al (a), Ti (b), Zr (c), Nb (d), Fe (e), Ni (f), Cu (direct bonding) (g), and Ag (h).
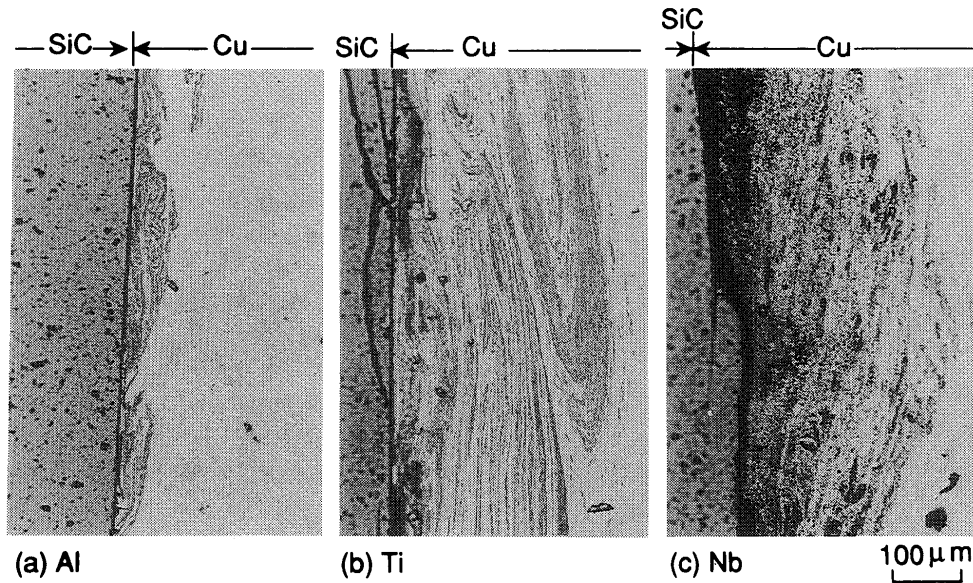
Fig. 2 Microstructures of friction-bonded joints of SiC to Cu with the intermediate layers of Al (a), Ti (b), and Nb (c)
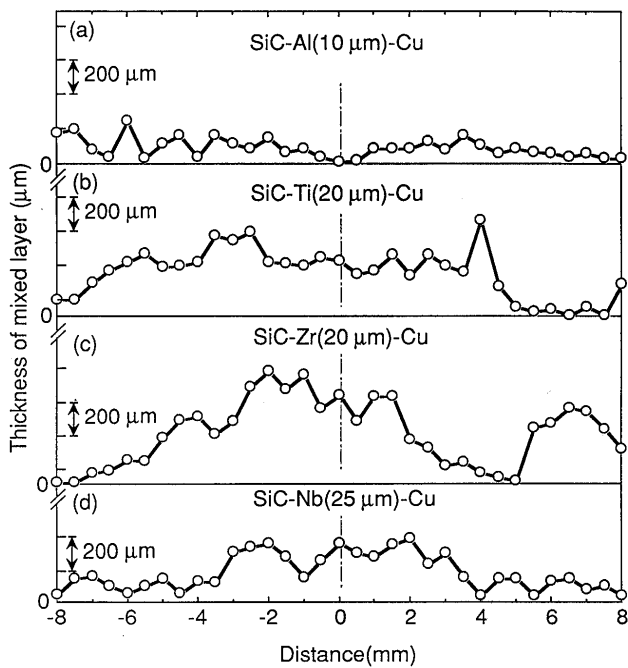

Fig. 3 Width of the mixed region vs. distance from the central axis for SiC-to-Cu joints with the intermediate layers of Al, Ti, Zr, and Nb.

Fig. 4 SEM micrographs of SiC-to-Cu joint and distribution of Cu, Si, and active metals analyzed along the white lines: (a) Ti intermediate layer and (b) Nb intermediate layer.

metals were applied, the deformation of Cu during the bonding process was much increased, and joints were obtained that had strength enough to stand the machining for preparing a specimen of metallographic examination.

As shown in **Fig. 2**, in Cu next to the bond interface of joints with the intermediate layer of the active metals, a region with complicated microstructures was observed in which SiC and the intermediate layer

were mixed with Cu (see Fig. 4). As shown in **Fig. 3**, the width of this mixed region varied remarkably depending on the kind of intermediate layer and the position, and was mostly much greater than the thickness of the intermediate layers (more than 200 μm for the intermediate layers of Ti, Zr, and Nb). In particular, the mixed region in the central part was wide, suggesting that the mixed region in the peripheral part

SiC   Cu

(a)   0.5 μm

(b)   0.1 μm   (c)   0.1 μm
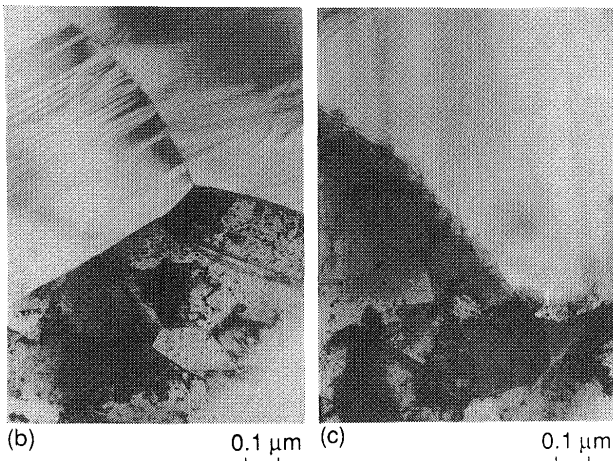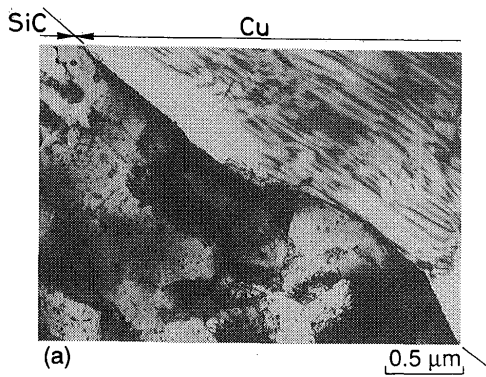
Fig. 5 TEM micrographs of a SiC-to-Cu joint with Nb intermediate layer at a low magnification (a) and at a high magnification (b), (c).

was pushed out of the vicinity of the bond interface with the plastic flow of Cu during the friction and forge processes.

The contents of Cu, Si and the active metals fluctuated remarkably in the mixed region as shown in Figs. 4(a) and 4(b). The peaks of the distribution curve of Si in the mixed region indicate that SiC particles were picked up into the Cu specimen. The SiC particle was observed in the mixed region for all the joints with the intermediate layer of the active metals except for Al. As can be seen from Fig. 4, no reaction layer between SiC and the active metals or between SiC and Cu was observed at the bond interface with SEM. The reaction layer at the bond interface could not be observed even with TEM, as shown in Fig. 5. Thus no reaction layer wider than 10 nm was formed at the bond interface of SiC to Cu with the Nb intermediate layer. The reported width of the interfacial reaction layer observed in the diffusion-bonded and brazed joints of ceramics to metal ranges from a few μm to several 10 μm[1-5]. Therefore, it can be concluded that the reaction layer in the friction-bonded joint is much narrower than those in the diffusion-bonded and brazed joints, even if it is formed.

According to the phase diagram of the Cu-Nb binary system, solid solubility of Nb in Cu is less than 0.1at%[14], though EDX analyses as shown in Fig. 4(a) suggested that the Nb content of the mixed region was

(a)   0.2 μm (c)   0.2 μm
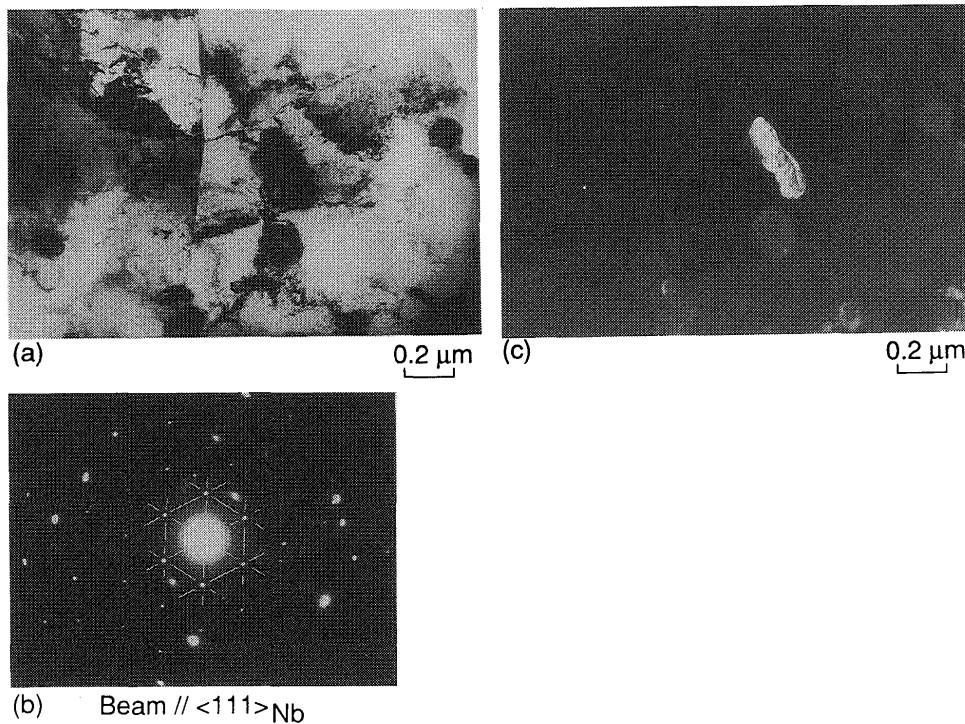

(b)   Beam // <111>Nb

Fig. 6 TEM micrographs of a mixed region of a SiC-to-Cu joint with Nb intermediate layer: (a) bright field image, (b) SAD, and (c) dark field image.

Table 1 Strength and axial displacement of friction-bonded joints of SiC to Cu with the various intermediate layers (A: Fractured during the preparation of a specimen for metallographic or tensile test. N.B.: Not bonded.).

Spec. No.	Intermediate layer Metal	Intermediate layer Thick.(μm)	Weld. param.	Axial displace. (mm)	Tensile test Strength(MPa)	Tensile test Frac. location
S1	Al	10	I	/	/	/
S9	Al	10	I	1.0	A	SiC
S7	Ti	20	I	1.8	/	SiC
S10	Ti	20	I	1.1	4	SiC
S2	Zr	20	I	/	/	/
S11	Zr	20	I	1.1	A	SiC
S8	Nb	25	I	3.7	/	/
S12	Nb	25	I	3.4	41	SiC
S6	Fe	30	I	0.8	N.B.	Interface
S13	Fe	30	II	4.3	A	Interface
S3	Ni	20	I	0.7	N.B.	Interface
S14	Ni	20	II	1.7	N.B.	Interface
S5	Cu(direct bond)		I	0.7	N.B.	Interface
S4	Ag	10	I	0.7	N.B.	Interface

not less than a few %. In order to investigate the morphology of Nb included in the mixed region, TEM observation was carried out. As can be seen in **Fig. 6(a)**, spherical particles with diameters of 10^{-1} μm or less were observed in the mixed region, and these particles were identified as Nb by the electron diffraction pattern (Fig. 6(c)) and the dark field image (Fig. 6(b)). Thus in the mixed region of the friction-bonded joint, an alloying element having only a limited amount of solubility can be dispersed in the form of fine particles with a diameter as small as less than 1 μm.

Table 1 summarizes the result of tensile testing on SiC-to-Cu joints with the intermediate layers. When the intermediate layers of Fe, Ni, Cu (direct bonding), and Ag were applied, the Cu specimen separated from the SiC specimen at the bond interface immediately after the bonding operation without an external load or during preparation of a specimen for metallographic examination. The strength of joints with the intermediate layers of Fe, Ni, Cu, and Ag was hardly increased by increasing the rotation speed and the friction time (condition II), though the longitudinal deformation of Cu during the bonding process was increased significantly. This result suggests that the friction between the SiC surface and a metal surface can only slightly improve the bondability of SiC to the metal. In contrast, joints with the intermediate layers of Al, Ti, Zr, and Nb were fractured in SiC, though the joint strength was much lower than that of the base materials. Cracks on the tensile test of these joints propagated along a path characteristic of the fracture under the strong influence of residual stress caused by the difference in thermal expansion between metal and ceramics[15]. This suggests that the residual thermal stress lowers the

strength of the SiC-to-Cu joint with the intermediate layers of the active metals.

3.2 Friction bonding of partially stabilized zirconia to oxygen free copper

As shown in **Fig. 7**, the friction bondability of ZrO_2 to Cu was also influenced strongly by the intermediate layer of the active metals similarly to that of SiC to Cu, i.e., the use of the intermediate layers of active metals Al, Ti, Zr, and Nb resulted in much greater longitudinal deformation of Cu and much higher joint strength than those of Ni, Cu, and Ag. ZrO_2 could be bonded to Cu by using the Fe intermediate layer, though SiC could not be bonded. This implies that the bondability of ZrO_2 to metal can be improved by an intermediate layer of less active metals than that of SiC. The joint with the Fe intermediate layer, however, failed during preparation of a specimen for metallographic examination, suggesting that the Fe intermediate layer
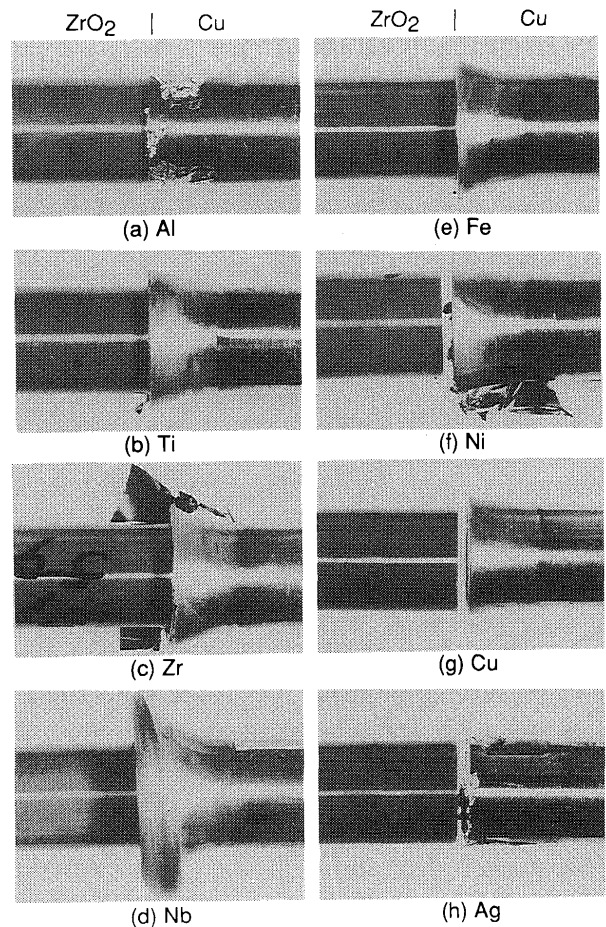

(a) Al
(e) Fe
(b) Ti
(f) Ni
(c) Zr
(g) Cu
(d) Nb
(h) Ag

Fig. 7 Appearance of friction-bonded joints of ZrO_2 to Cu with the intermediate layers of Al (a), Ti (b), Zr (c), Nb (d), Fe (e), Ni (f), Cu (direct bonding) (g), and Ag (h).
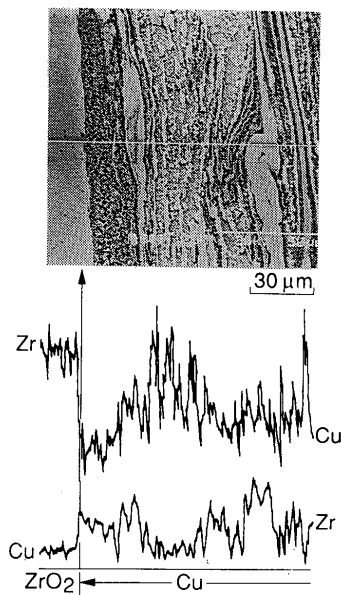
Fig. 8 SEM micrograph of a ZrO_2-to-Cu joint with the Zr intermediate layer and distribution of Zr and Cu analyzed along the white line.

was less effective in improving the bondability than those of the active metals.

In ZrO_2-to-Cu joints with the intermediate layers of the active metals, a mixed region was also observed in which the contents of Cu and the active metals fluctuated widely as shown in **Fig. 8**. In contrast to that of the SiC-to-Cu joint, no ZrO_2 particle was observed in the mixed region.

The width of the mixed region is shown in **Fig. 9** as a function of distance from the central axis of the joint for the intermediate layers of Al, Ti, Zr, Nb, and Fe. When the intermediate layers of Al, Ti, and Zr were applied, the width of the mixed region in the central part was much greater than the thickness of the intermediate layers. The width of the mixed regions of these joints showed a tendency to decrease in the peripheral region, suggesting that the mixed region in the peripheral region was expelled into the collar with the plastic flow of Cu during the bonding process. The mixed regions in the joints with Nb and Fe intermediate layers were considerably narrower than those of Al, Ti, and Zr intermediate layers. Since the longitudinal deformation of Cu of the joint with the Nb intermediate layer was much greater than those of the joints with the intermediate layers of the other active metals, the greater part of the mixed region was probably expelled into the collar during the bonding process for the intermediate layer of Nb. On the other hand, the longitudinal deformation of Cu of the joint with the Fe intermediate layer was much smaller than the intermediate layers of

active metals, suggesting that almost all the mixed region stayed in the vicinity of the bond interface for the intermediate layer of Fe. These results suggest that the mixed region in the joint with the Fe intermediate layer was not developed so much as those in the joints with the intermediate layers of the active metals.

Table 2 summarizes the results of tensile testing on the ZrO_2-to-Cu joint with the intermediate layers. ZrO_2-to-Cu joints with the intermediate layers of active metals Al, Ti, Zr, and Nb were fractured in ZrO_2 in the tensile test, though their strength was much lower than that of the base materials. The crack of these joints propagated along a path characteristic of fracture under the strong influence of the residual stress caused by the difference in the thermal expansion coefficients of the ceramics and metal[15]. This suggests that the residual thermal stress can explain why the ZrO_2-to-Cu joint was fractured in ZrO_2 at stress much lower than that of the base material. Though ZrO_2 can be bonded to Cu with the Fe intermediate layer, the joint obtained was fractured at the bond interface during preparation of a specimen for metallographic examination. Thus the Fe intermediate layer was much less effective in improving the bond strength of the ZrO_2-to-Cu joint than those of the active metals. When the friction time and rotation speed were increased (condition II), ZrO_2 could be bonded to Cu even with the Ni intermediate layer. However, the joint
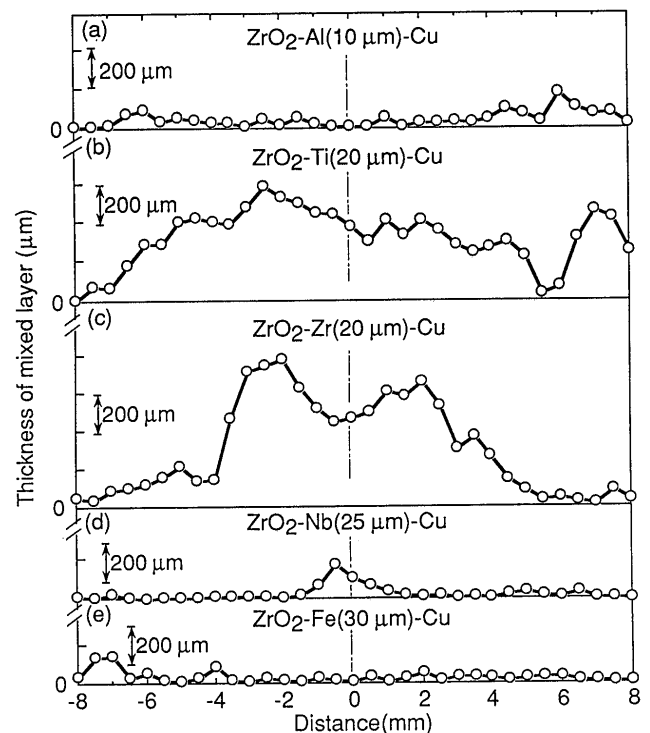

Fig. 9 Width of the mixed region vs. distance from the central axis for ZrO_2-to-Cu joints with the intermediate layers of Al, Ti, Zr, Nb, and Fe.

Table 2 Strength and axial displacement of friction-bonded joints of ZrO_2 to Cu with the various intermediate layers (A: Fractured during the preparation of a specimen for metallographic or tensile test. N.B.: Not bonded.).

Spec. No.	Intermediate layer Metal	Intermediate layer Thick.(μm)	Weld. param.	Axial displace. (mm)	Tensile test Strength(MPa)	Tensile test Frac. location
Z5	Al	10	i	0.5	/	/
Z9	Al	10	I	1.5	16	ZrO2
Z6	Ti	20	I	1.6	/	/
Z10	Ti	20	I	1.9	60	ZrO2
Z1	Zr	20	I	2.0	/	/
Z11	Zr	20	I	1.9	6	ZrO2
Z7	Nb	25	I	5.4	/	/
Z12	Nb	25	I	3.9	80	ZrO2
Z2	Fe	30	I	1.8	/	Interface
Z13	Fe	30	I	1.9	8	Interface
Z8	Ni	20	I	1.0	N.B.	Interface
Z14	Ni	20	II	4.7	A	Interface
Z3	Cu(direct bond)		I	1.0	N.B.	Interface
Z4	Ag	10	I	0.6	N.B.	Interface

with the Ni intermediate layer obtained by increasing the friction time and rotation speed was fractured at the bond interface during preparation of a specimen for metallographic examination. Thus it cannot be expected that the friction of the ZrO_2 surface with a metal surface can significantly improve the bondability of ZrO_2 to the metal.

4. Conclusions

Intermediate layers of metals ranging from the active metal to the noble metal have been applied to the friction bonding of Cu to SiC and to ZrO_2, and their effects on the bondability have been systematically investigated. Results obtained are summarized as follows:

(1) The intermediate layers of active metals Al, Ti, Zr, and Nb improved remarkably the bondability of SiC-to-Cu and ZrO_2-to-Cu joints, while those of Fe, Ni, Cu, and Ag were much less effective.

(2) In Cu adjacent to the bond interface of the joints with the active metals, a mixed region was observed in which the intermediate layer was mechanically mixed with Cu, but no reaction layer between the metal and ceramics specimens could be found even with TEM observation.

(3) The friction of the surfaces of SiC and ZrO_2 with a metal surface was hardly effective in improving their bondability to the metal.

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