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The effect of interlayer metals on the strength of alumina ceramic and 1Cr18Ni9Ti stainless steel bonding

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The effects of interlayers of molybdenum and copper on the strength of alumina ceramic and 1Cr18Ni9Ti stainless steel bonding with $Ag_{57}Cu_{38}Ti_5$ filler metal were investigated. The interfacial morphologies were observed and analysed by scanning electron microscopy and energy dispersive X-ray (EDX) analysis, respectively. The joint strength was examined by shear tests. When using a molybdenum interlayer, the joint strength could be greatly improved because molybdenum not only reduced the interfacial residual stress, but also did not affect the interfacial reaction between the ceramic and the filler metal, and the maximum value was obtained when it was about 0.1 mm thick. When using copper as an interlayer, the joint strength was not increased but decreased, because copper reduced the activity of titanium in the filler metal, resulting in an insufficient interfacial reaction between the ceramic and the filler metal and the formation of poor interfacial adhesion. Therefore, in selecting an interlayer metal to reduce or avoid interfacial residual stress in joining ceramics to metals, in which the interfacial reaction of ceramic and filler metal is important to the joints, the interaction of interlayer metal and filler metal must be considered.

1. Introduction

Active brazing techniques have been widely used in ceramic/metal bonding. Apart from the properties of bonded materials, a strong ceramic/metal joint is dependent on a strong interfacial bonding between ceramic and filler metal. This in turn, is controlled by the interfacial reaction between ceramic and filler metal, and a favourable stress gradient in the ceramic/metal interfacial bond zone which is controlled by an interfacial design. A sufficient interfacial reaction between ceramic and filler metal is dependent on the design of the active brazing filler metal and control of brazing parameters in direct brazing. Much research has been focused on this field [1–5]. On the other hand, in most cases, brazing ceramic to metal is performed at elevated temperature. Owing to the large difference in thermal expansion coefficients of two materials, a significant magnitude of residual stress is produced during the cooling process after joining. Such residual stress created at the bonding zone may cause cracking or reduce the bonding strength. Therefore, it is desirable to minimize the magnitude of the residual stress. One method is the optimum design of the shape of the bonding zone [6]; another method is to introduce an interlayer or a composite interlayer between ceramic and metal [7, 8].

Some theoretical analyses and calculations [9–12], as well as experimental determinations [13, 14] of residual stress have been carried out for ceramic/metal joints, confirming that there is a high residual stress gradient around the ceramic/metal interface, and it is impossible to achieve a strong ceramic/metal joint, or to achieve acceptable joining without an interlayer. There are two kinds of interlayers in physics: a soft interlayer, characterized by copper, and a hard interlayer, characterized by molybdenum or tungsten. Although the chemical compatibility of an interlayer material with an active brazing filler metal and its effect on ceramic/filler metal interfacial bonding are very important, relatively little effort has been directed towards its identification. This investigation aims to improve the joint strength of ceramic to metal by using an interlayer metal and analysing the effect of the reaction between interlayer and filler metal on the joint strength.

2. Experimental procedure

The alumina ceramic (99% purity) and 1Cr18Ni9Ti stainless steel were used as the bonded materials, 15 and 5 mm diameter and 5 mm thick, respectively. The filler metal was $Ag_{57}Cu_{38}Ti_5$ alloy and its thickness was about 50 μm. Copper and molybdenum were selected as interlayers and their thicknesses were 0.05–0.5 mm. Before brazing, the surfaces of all materials to be brazed were polished with abrasive paper and then carefully cleaned in acetone. The assemblage of the sandwich joint, consisting of a ceramic disc and a stainless steel disc, and an inserted interlayer and brazing filler metal between them, was fixed and

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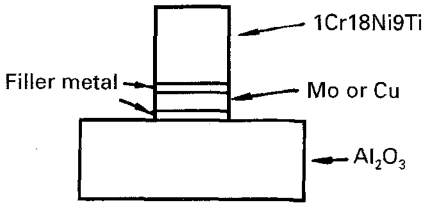


Figure 1 Bonded sample with a molybdenum or copper interlayer.

placed in a vacuum furnace and heated to 850°C at a heating rate of 15 °C min^{-1}, and held there for 30 min, then cooled to 600 °C at a cooling rate of 3–5 °C min^{-1}, and finally cooled to room temperature. A vacuum of about 7×10^{-3} Pa was maintained during brazing. The bonded lap sample is shown in Fig. 1.

The joint strength was measured in air at room temperature by shear tests with a crosshead speed of 0.5 mm min^{-1}. The microstructures and fracture surfaces of joints were observed and analysed by scanning electron microscopy (SEM), and interfacial reaction products were determined by X-ray diffraction (XRD).

3. Results and discussion

3.1. Joint strength

Fig. 2 shows the variation in strength of the ceramic/metal joints as a function of thickness of the interlayer metal (copper). It is clear that the joint strength was lower with a copper interlayer than without it, and the thicker the copper interlayer, the lower was the joint strength.

The copper interlayer has a lower strength and better ductility, but a relatively higher coefficient of thermal expansion ($\alpha \approx 17 \times 10^{-6}$ K^{-1}). An elastic stress analysis [9] has been performed for a ceramic/metal joint with a soft copper interlayer and revealed that a very high residual shear stress existed in the soft interlayer; the relaxation of the residual stress in the joint by a creep or yield mechanism during slow cooling from the joining temperature will be up to about 90% or more. The maximum residual shear stress will be decreased with increasing thickness of the soft interlayer, resulting in an increase in joint strength. Experimental results showed that the joint strength of sailon ceramic to 40Cr steel [8] or zirconia ceramic to 1Cr18Ni9Ti stainless steel [15] could be improved by using a copper interlayer with a suitable thickness. For joining alumina ceramic to 1Cr18Ni9Ti stainless steel with Ag-Cu-Ti active filler metal system, although no analysis nor calculation of residual stress for this system were performed quantitatively, from the standpoint of thermal expansion coefficient, α, matching of alumina ceramic ($\alpha \approx 8 \times 10^{-6}$ K^{-1}), copper ($\alpha \approx 17 \times 10^{-6}$ K^{-1}) and stainless steel ($\alpha \approx 20 \times 10^{-6}$ K^{-1}), as well as lower yield strength for the copper interlayer, copper could definitely decrease the interfacial residual stress and increase the joint strength of alumina ceramic and stainless steel. From Fig. 2, the joint strength is seen to decrease by insertion of a copper interlayer. It can be suggested that there must be other factors introduced by the copper, and the extent of damage to the joints from these factors was much greater than that of improvement to the joint strength by relaxation of residual stress.

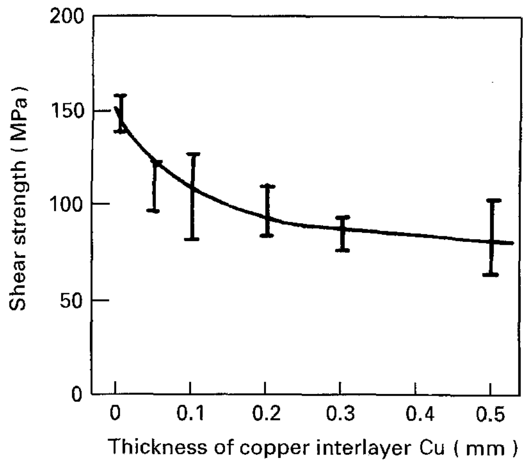


Figure 2 Effect of a copper interlayer on the strength of alumina ceramic and stainless steel bonding at 850 °C for 30 min.

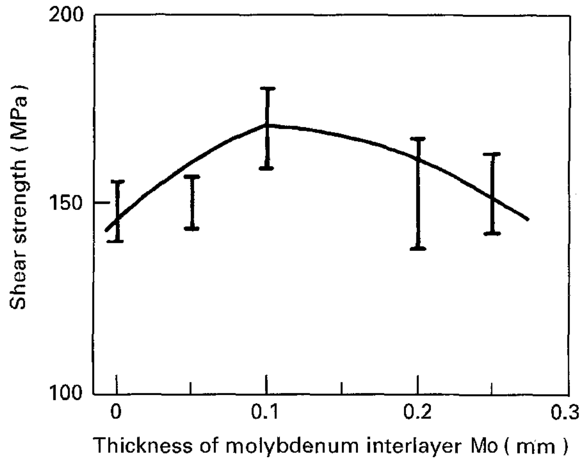


Figure 3 Effect of a molybdenum interlayer on the strength of alumina ceramic and stainless steel bonding at 850 °C for 30 min.

Fig. 3 shows the variation in strength of the alumina ceramic/stainless steel joints as a function of thickness of the interlayer metal, molybdenum. As is seen in the figure, the joint strength of alumina ceramic to stainless steel could be improved by using a molybdenum interlayer with a suitable thickness, and the maximum value was obtained when the thickness of the molybdenum interlayer was about 0.1 mm.

The interlayer metal, molybdenum, has a relatively low thermal expansion coefficient ($\alpha \approx 6.4 \times 10^{-6}$ K^{-1}) which is slightly lower than alumina ceramic, but much lower than 1Cr18Ni9Ti stainless steel. Although the quantitative calculation of residual stress for alumina ceramic and stainless steel bonding with a molybdenum interlayer was not performed, from matching of the thermal expansion for a bonded system, it can be confirmed that the molybdenum interlayer can decrease the residual stress of the joints. By using an interlayer of molybdenum, the maximum residual stress was transferred to the interface between molybdenum and stainless steel, and from the interface between ceramic and molybdenum, as the same that described by Suganuma [11] for Al_2O_3/Nb/Mo/steel bonding. Therefore, from the view point of the

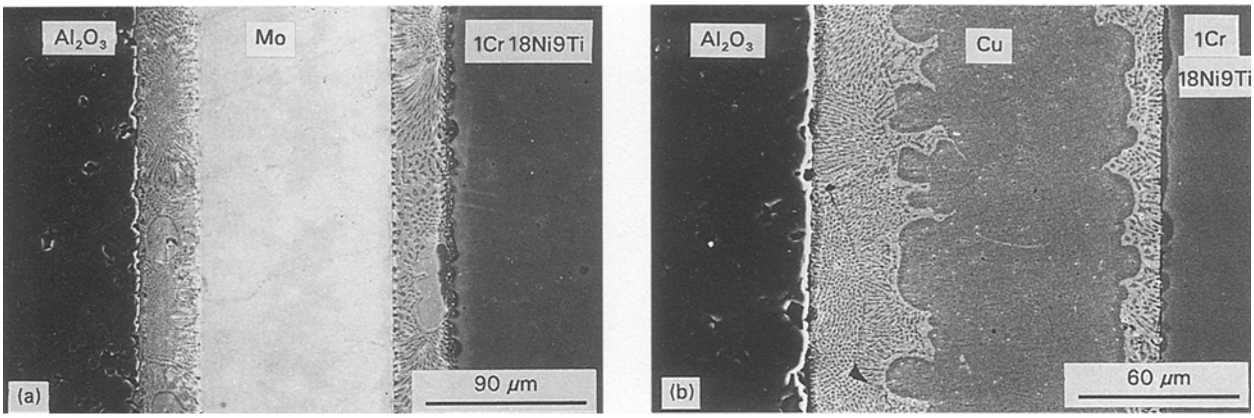
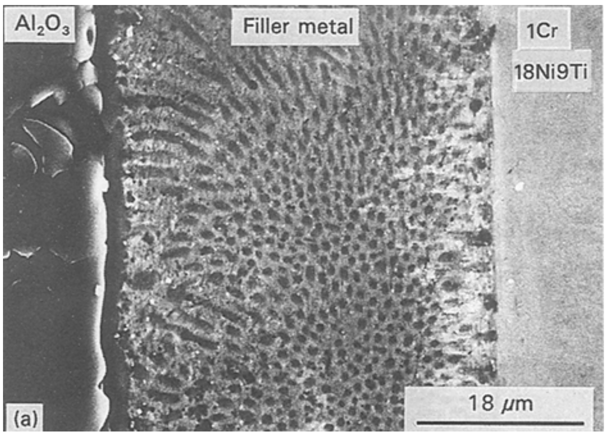
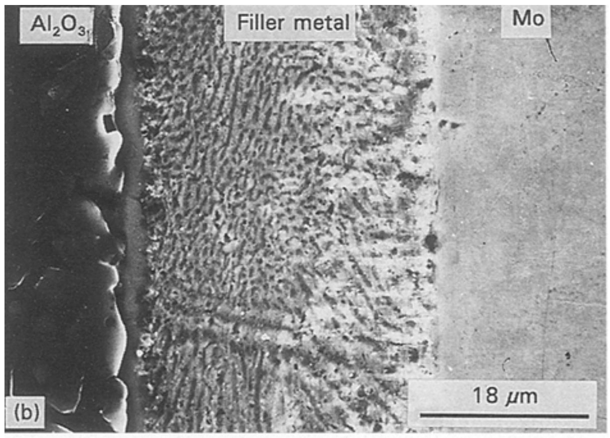


Figure 4 Microstructures of the bonding range of alumina ceramic to stainless steel with a 0.1 mm thick of interlayer of (a) molybdenum, or (b) copper.





interfacial stress, the joint strength can be improved by inserting an interlayer of molybdenum.

3.2. Microstructure of the interface

In order to elucidate the relationship between the joint strength and the interfacial morphologies, microstructures and elemental distribution of the interfacial bond zone were observed and analysed by SEM and (EDX), respectively.

Fig. 4 shows the microstructures of alumina ceramic and stainless steel bonding with a 0.1 mm thick molybdenum or copper interlayer. It can be seen that the interface between the interlayer of molybdenum and the filler metal was smooth, but the interfacial bonding strength was excellent according to the fact that fractures never occurred at the molybdenum filler metal interface during the shear test. The interface between the copper interlayer and the filler metal was not smooth but a sawtooth interface, because silver has a high solubility of copper at the joining temperature and therefore, silver in the filler metal eroded the interlayer copper. Defects, such as cracks or pores, were not found in the bonding range for either kind of joint.

It is also notable that an interfacial reaction layer was found between the ceramic and the filler metal for the three kinds of joints as shown in Fig. 5. The reaction layer thickness when using different thicknesses of molybdenum interlayer was the same and equivalent to that without using an interlayer metal, which

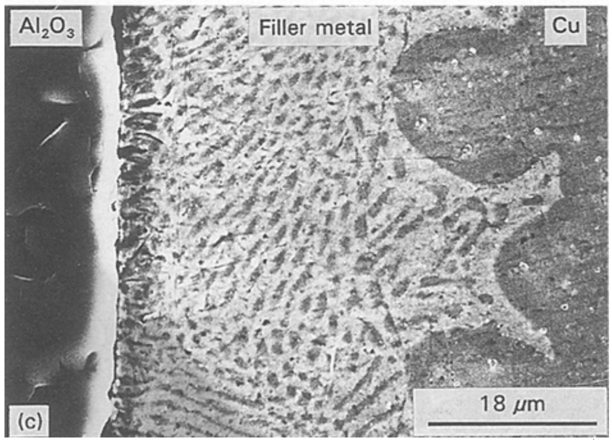


Figure 5 Images of the interfacial reaction layer between ceramic and filler metal bonded (a) without an interlayer and with an interlayer of (b) molybdenum or (c) copper.

indicated that interlayer metal, molybdenum, did not react with the active element, titanium, in the filler metal and thereby was not affected by the interfacial reaction between the ceramic and the filler metal. In the case of inserting interlayer metal, copper, between ceramic and stainless steel, the interfacial reaction layer was much thinner than with or without a molybdenum interlayer, which clearly indicates that the copper interlayer greatly affected the ceramic/filler metal interfacial reaction.

In order to examine the effect of interlayer metal on the ceramic/filler metal interfacial reaction further, elemental distributions were analysed by EDX, as shown in Fig. 6 and Table I.

The results showed that active element, titanium, in the filler metal was mainly concentrated in the interfacial reaction layer for the three kinds of joint, but the content was different and the titanium content at the interface between the ceramic and the filler metal was almost the same for joints with or without a molybdenum interlayer, and it was much higher than that with a copper interlayer. An interlayer of copper decreases the titanium content in the reaction layer possibly because the solution of copper increased the quantity of filler metal and then decreased the concentration of titanium in the filler metal.

In order to analyse the interfacial reaction products, stainless steel and molybdenum as well as copper, 0.1 mm thick, were bonded to ceramics, respectively, and then the metals and filler metals were removed mechanically, and finally the reaction products were examined by XRD, as described in the previous work [16]. XRD analyses showed that with or without a molybdenum interlayer, the reaction products were the same and mainly consisted of Ti_2O, TiO and $CuTi_2$ compounds, and a layer of structures of Al_2O_3/Ti_2O + TiO/Ti_2O + TiO + $CuTi_2$ / $CuTi_2$/ Ag–Cu was formed at the interface, which was consistent with the interfacial reaction results of alumina ceramic and alumina ceramic bonding [16]. In the case of using a copper interlayer, which decreased the content of active elemental titanium at the interface, the reaction products were TiO and $CuTi_2$.

From the above results, it is clear that in joining alumina ceramic to stainless steel with Ag–Cu–Ti active filler metal, inserting a molybdenum interlayer did not affect the interfacial reaction, but copper did.

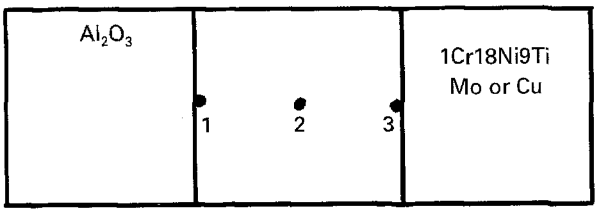


Figure 6 Positions of EDX analyses.

3.3. Fracture analyses

SEM fracture analysis results showed that with or without a molybdenum interlayer, fractures occurred completely in the ceramics, or partially in the ceramic and partially at the interface between the ceramic and the filler metal. However, the joint strength with a molybdenum interlayer was higher than without using interlayer metal, although fractures occurred in the ceramic, which clearly indicated that the ceramics near the interface when bonded without an interlayer must have been severely damaged by the interfacial residual stress arising from the mismatching of thermal expansion coefficients of ceramic and stainless steel. In other words, the interfacial residual stress of alumina ceramic and stainless steel bonding could be decreased effectively by using a molybdenum interlayer.

With a copper interlayer, fractures occurred either completely at the interface between the ceramic and the filler metal or partially in the ceramic and partially at the interface, sometimes at the copper/filler metal interface near the ceramic. The joint strength was lower with a copper interlayer than without it, although the fracture types were the same in the two cases. The joint strength with a copper interlayer was lower when fracture occurred at the interface between the ceramic and the filler metal, than that without copper when fracture occurred in the ceramics, indicating that the interfacial bonding strength was lower with than without copper, and the copper interlayer affected the ceramic/filler metal interfacial reaction, which was consistent with the results of interfacial morphologies and elemental distribution analyses.

3.4. Selection of the interlayer

From the above results and analyses, it can be concluded that because of good matching of thermal expansion coefficients, alumina ceramic and stainless steel bonding with a molybdenum interlayer, the interfacial residual stress can be reduced effectively by molybdenum. On the other hand, the molybdenum interlayer not reacted with titanium in the filler metal at the bonding temperature and therefore unaffected by the interfacial reaction and the interfacial bonding strength between ceramic and filler metal was good, so that the joint strength was increased by inserting

TABLE I EDX analysis results of the joining layer

Interlayer metal	Location	Composition (at %)							
		Ag	Cu	Ti	Al	Mo	Fe	Ni	Cr
Without interlayer	1	0.85	39.47	54.36	5.32				
	2	57.34	42.13	0.53					
	3	36.58	27.35	0.79			26.52	2.51	6.25
Mo	1	1.05	44.10	52.45	2.40				
	2	59.20	40.21	0.59					
	3	38.25	25.28	0.63		36.47			
Cu	1	49.82	40.18	5.82	4.18				
	2	50.62	48.66	0.72					
	3	43.52	55.81	0.67					

a molybdenum interlayer. In this case, the joint strength was mainly controlled by the reduction of interfacial residual stress.

When using a copper interlayer in joining, although it can apparently relax the interfacial residual stress by a creep or yield mechanism, copper affected the interfacial reaction through its solution into the filler metal and reduced the concentration and the activity of titanium in the filler metal, and the joint strength was decreased. In this case, the interfacial reaction was the governing factor for the joint strength but not the relaxation of interfacial residual stress.

Therefore, in selection of an interlayer metal for decreasing or avoiding the interfacial residual stress of ceramics and metals bonding, in which the ceramic/filler metal interfacial reaction plays an important role in the joints, apart from considering the matching of thermal expansion of the bonded materials and interlayer metals, the chemical compatibility of an interlayer metal with an active filler metal should be considered. Firstly, the chemical compatibility of an interlayer metal with a filler metal must be good, in order to form a strong interfacial adhesion of the interlayer with the filler metal. Secondly, the interlayer materials should not react strongly with elements in the filler metal, especially with active elements, or the activity of the active elements in the brazing filler metal would be decreased, which would result in an insufficient interfacial reaction between the ceramic and the filler metal and therefore a low interfacial bonding strength. Clearly, a low joint strength was produced.

4. Conclusion

The effects of interlayer metals on the strength of alumina ceramic and 1Cr18Ni9Ti stainless steel bonding using $Ag_{57}Cu_{38}Ti_5$ filler metal were investigated. From the viewpoint of the interfacial reaction, the influence of interlayers on the joints was also analysed.

The joint strength could be improved considerably by using a molybdenum interlayer and the maximum value was obtained when the molybdenum was 0.1 mm thick. The molybdenum interlayer increased the joint strength, because it not only reduced the interfacial residual stress but also did not react with the filler metal, especially not with the active element titanium, and therefore was not affected by the interfacial reaction of the ceramic and filler metal. While using copper as an interlayer, the joint strength was not increased but decreased, because copper was corroded by silver and its solution into the filler metal reduced the activity of active elemental titanium in the filler metal and then resulted in an insufficient interfacial reaction between the ceramic and the filler metal.

Therefore, in selection of an interlayer metal to reduce or avoid interfacial residual stress in joining ceramics to metals, in which the interfacial reaction between ceramic and filler metal had a strong influence on the joints, the interaction of the interlayer metal with the filler metal, especially with active elements, and its effect to the joints, should be considered.

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