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Residual Stress and Bonding Strength in the Electrical Sialon Ceramics Joint Made by Using the Brazing Metal Layer

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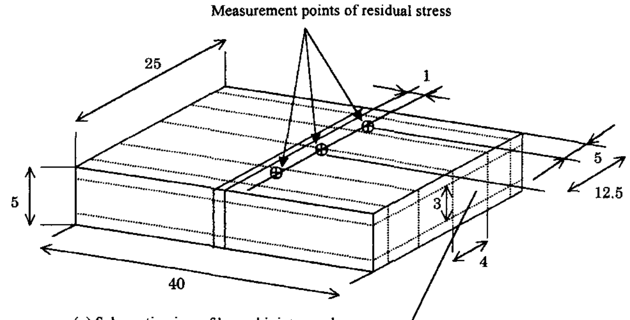
Electrical Sialons which have some TiN contents were joined with Ag-Cu-Ti active brazing metal layer having a thickness from 30 μm to 400 μm at a temperature from 1113 K to 1213 K in a vacuum. Residual stress in the brazed joint specimens was not observed when the thickness of brazing metal layer was 30 μm. However, the residual stress of 80 MPa was detected when the thickness of brazing metal layer increased up to 400 μm. When the brazing temperature was 1113 K, four-point bending strengths of 520 MPa and 310 MPa were obtained for the brazed joint specimens with the thicknesses of brazing metal layer of 30 μm and 400 μm, respectively. While the four-point bending strength increased as the brazing temperature was raised. The maximum value of the four-point bending strength was about 700 MPa which was obtained at a condition of the brazing metal thickness of 30 μm and the brazing temperature above 1163 K. However, the four-point bending strength decreased with increasing the bending test temperature. A remarkable decrease of the bending strength was observed at the test temperature of 873 K, in which the bending strength was 300 MPa.

Key Words : Vacuum Brazing, esidual Stress, Electrical Sialon, Four-point bending strength, X-Ray diffraction

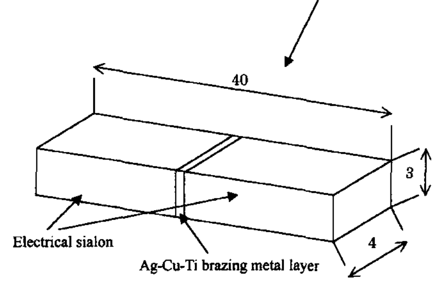
1. Introduction

Many attentions have recently been paid to ceramics as a new material, because they have peculiar properties which are not obtained in metallic materials and polymers. The practical use of ceramics having the peculiar electric and magnetic characteristics is spreading to the fields of electronics and mechatoronics. And also the use of structural ceramics having the excellent mechanical and thermal characteristics[1] is spreading to the fields of mechanical industries such as motor parts and some kinds of industries. The structural ceramics are expected to be further developed in the future. In such a case, bonding technology for joining the ceramics is very important to manufacture the big and complex shaped ceramic structures. The bonding technology for joining between brittle ceramic and tough metal is also indispensable one to conquer the brittleness of ceramics. For the bonding technology, there are a lot of studies in which some of them using active brazing metals[2,3] rarely show high bonding strength to be enough to manufacture the structural ceramic products. However, more systematic studies are desired to develop the bonding technology into practical applications.

The aim of this study is, therefore, to establish the joining technique by using active brazing metals for the joining of electrical sialons in vacuum. The electrical sialons[4,5] are known to be a



(a) Schematic view of brazed joint sample.



(b) Schematic view of brazed joint specimen for four-point bending test.

Figure 1 Schematic view of brazed joint sample and brazed joint specimen for four-point bending test.

special ceramics, because they are easy to make the complicate shaped parts by using an electro-discharge machine.

2. Experimental Procedure

2.1 Specimen used

Samples used in this experiment were electrical sialon specimen with a size of 20×25×5 mm (HCN-40 produced by Hitachi Metal Co. Ltd.). The electrical sialon is containing a slight amount of TiN. The electrical sialon specimens were grinded to the surface roughness of Ry $<2\,\mu$m. On the other hand an active metal composing of 70.5 mass%Ag, 27.5 mass%Cu and 2 mass%Ti (produced by Tanaka Precious Metals Co. Ltd.) was used as a brazing metal material. They were washed in acetone solution for 5 minutes by using an ultrasonic cleaning equipment.

2.2 Joining process and the condition

Using the brazing metal, the electrical sialons were joined on their surface of 25×5 mm in a vacuum of 5.0×10^{-2}Pa to make the brazed joint specimen of 40×25×5 mm as shown in Fig. 1. The thickness of brazing metal layer during joining was controlled by inserting a W wire having different diameter between the electrical sialon specimens. The brazing condition is shown in Table 1.

2.3 Measurement of residual stress

Residual tensile stress in the direction perpendicular to the joint surface was determined from measurement of diffracted X-Ray peak shift to an incident beam angle ψ, which is well known as the $\sin^2\psi$ method. The measurement was conducted on the line separated by the distance of 1.0 mm from the brazed joint interface at the central region and also on the line by the distance of 5.0 mm at the both edge regions in the brazed joint specimen as shown in Fig. 1. To compensate a residual stress associated with grinding process, the measured residual stress was deducted by a value of residual stress at the point separated by the distance of 15.0 mm from the joined interface, which was supposed to be enough distance for removing the grinding effect. The condition for X-Ray stress analysis is shown in Table 2.

2.4 Measurement of bending strength

Four specimens for four-point bending test were cut out from the brazed joint sample using a cutting machine and a grinding machine. They are expected to have almost same strength. The size of specimen for the test was 40×4×3 mm as shown in Fig. 1. The grinding was conducted in the longitudinal direction of the brazed joint specimen to control the effect of grinding marks on the bending strength.7) The joint strength or bending strength was measured using Instron machine-4507, in which the procedures of JIS-R1601 and R1604 were adopted for the test at room temperature and elevated temperatures up to 873 K in air. To increase the accuracy of the measurement, four or eight specimens were adopted for each test. The condition for the four-point bending test is shown in Table 3.

Table 1 Brazing Condition.

Heating rate	0.42(K/s)
Brazing temperature	1113~1213(K)
Brazing time	300(s)
Thickness of active brazing metal layer	30~400(μ m)

Table 2 Condition of X-Ray stress analysis.

Cr-Kα	30(kV), 40(mA)
Diffraction	(311) plate of TiN
Diameter of incident collimator	ϕ 2(mm)
Stress constant, K*	-914.1(MPa/degree)
Detector	PSPC with V filter

*(250GPa, 0.19, 127degree)6)

Table 3 Condition of four-point bending test.

Specimen length	40(mm)
Specimen width	4(mm)
Specimen thickness	3(mm)
Upper span	10(mm)
Lower span	30(mm)
Crosshead speed	0.0083(mm/s)
Testing Temperature	293, 473, 673, 873(K)

3. Results and Discussion

3.1 Thickness of brazing metal layer and residual stress

In the case of bonding hard ceramic and soft metal using the Ag-Cu-Ti brazing metal, it is said that the soft brazed metal deforms plastically and reduces the residual stress which occurred at the joint interface during cooling from brazing temperature to room temperature due to the large mismatch in their thermal expansions. By the way, the value of heat expansion coefficient of the Ag-Cu-Ti brazing metal(20.0×10^{-6}K^{-1}) is about 4 times larger than that of the electrical sialon(5.0×10^{-6}K^{-1}). This fact shows that for joining the two ceramic specimens with the Ag-Cu-Ti brazing metal the brazed metal itself becomes a source of the residual stress which occurs due to the difference in their heat expansion coefficients. The bonding strength of the brazed joint ceramics is known to depend on the amount of residual stress. Therefore, it is suggested that there is an optimum thickness in the brazing metal layer to obtain the highest bonding strength for the ceramics/metal bonding8). In this experiment for joining the electrical sialons, the effect of the thickness of brazing metal layer on bonding strength was investigated because the amount of residual stress depends on the thickness of brazing metal layer.

Figure 2 shows the distribution of the residual tensile stress in the specimens jointed at 1113 K under the condition of Table 1. The residual stresses were averaged for each thickness of brazed metal layer to compare with each other specimen. And Fig. 3 shows the relation between its averaged residual stress and the thickness of brazing metal layer in the specimens jointed at 1113 K. The distinct distribution8) of residual stress was hardly observed in the direction parallel to the joint interface in this experiment, though it is known to be observed a symmetric stress distribution with a peak value at the central region in the case of the ceramic /Ag-Cu-Ti/metal. In general, the residual tensile stress should be symmetrically distributed in an ideal sample, though the stress in this experiment did not symmetrically distributed. This may be due to brazing failure at sample edges which was sometimes observed. Even so, the difference in residual stress between the edge and central regions was relatively large in the case of 400 μ m thickness of the brazing metal layer comparison to the case of the thickness under 100 μm. It is also found from Fig. 3 that when the thickness of brazing metal layer increases the averaged residual tensile stress (plus number) in the joined specimen increases due to the difference in thermal expansion coefficients of the electrical sialon and the brazing metal. On the other hand, the value of residual stress for the thickness of 30 μm indicates a minus value, namely compressive stress, which seems to be due to compensation of grinding effect on the residual stress. In the thickness of 30 μm the residual stress due to joining is considered to be negligibly small.

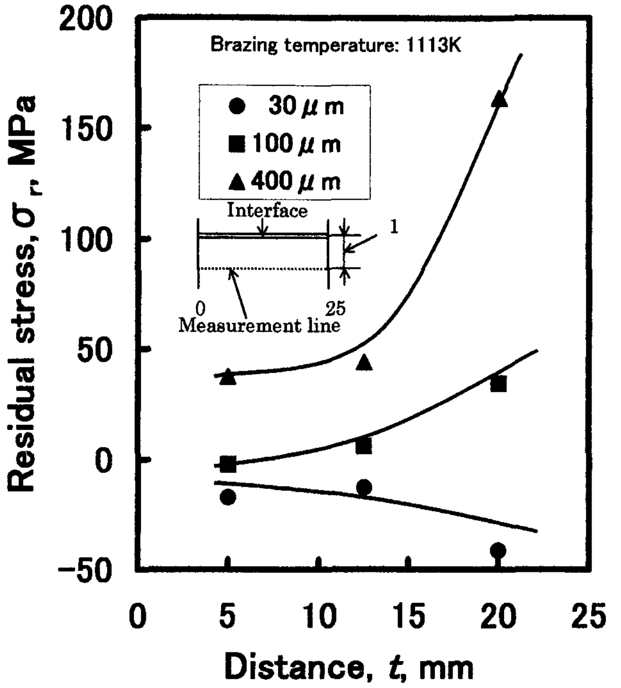


Figure 2 Relation between residual stress and distance from the edge region in the brazed joint interface for various thickness of brazed metal layer.

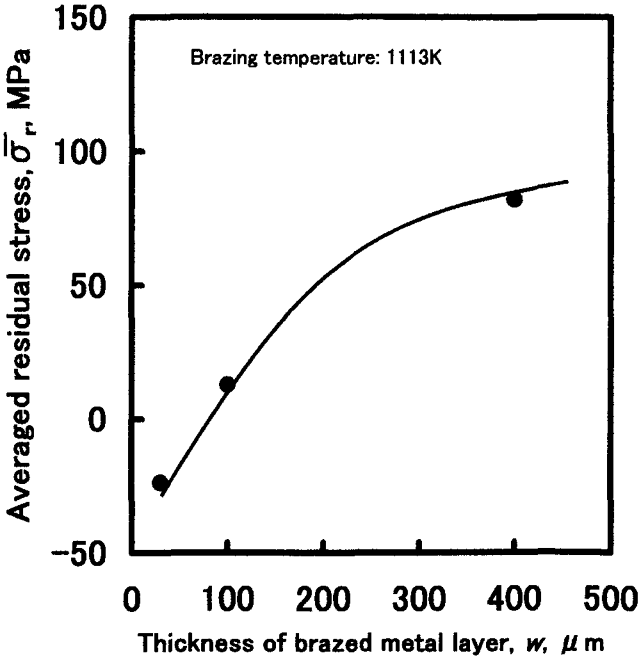


Figure 3 Relation between averaged residual stress and thickness of brazed metal layer.

By the way, Tanaka et al. recommended that the X-Ray collimator having a smaller diameter than ϕ0.3 mm should be used for the measurement of residual stress distribution in metal/ceramic joint specimen.9),10) And also they reported that when the brazed joint sample was cut out, the residual stress redistributes in the specimen though the significant amounts of the stress are remained. By the way, it was hard to obtain an intensity of X-Ray diffraction enough to analysis the residual stress because the electrical sialon is a mixture of TiN and SiAlON : the intensity of specific X-Ray diffraction is weaker in the case of two phase material than that of single phase material. Therefore, the collimator having ϕ2 mm diameter for X-Ray beam was used for this experiment, in which the measured value of residual stress was slightly smaller than that in the case of ϕ0.3 mm because the intensity obtained was averaged in the large size X-Ray spot. However, it was enough to analyze the residual stress.

Figure 4 shows the relation between the four-point bending strength and the averaged residual stress. The bending strength decreases as the residual stress increases. Therefore, it is found that decreasing the thickness of Ag-Cu-Ti brazing metal layer is effective to decrease the influence of thermal expansion mismatch on the residual stress and to increase the bonding strength.

Therefore, it is concluded that in the cases of the ceramic/ceramic bonding and the bonding of the small expansion mismatch materials by using the brazing metal the thinning of brazing metal layer should be sufficiently considered to obtain reasonable joining strength. And also, stress relief effect due to plastic deformation of

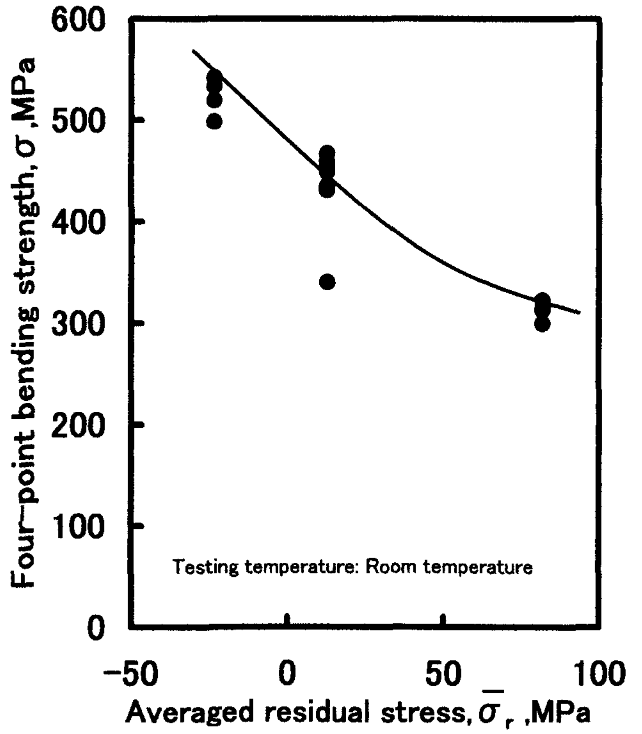


Figure 4 Relation between four-point bending strength and averaged residual stress.

brazing metal layer and thermal stress generation effect due to large thermal expansion of brazed metal should be considered especially in the case of ceramic/metal bonding which has large difference in their thermal expansion coefficients.

3.2 Effect of bonding temperature on the bonding strength

Although the residual stress did not detect in the case of brazing by using the 30 μm thickness of brazing metal layer at 1113K, the bonding strength showed only 520 MPa which was less than the strength of electrical sialon itself. This suggests that there is a critical limit in the adhesive strength at bonding interface in the specimen.

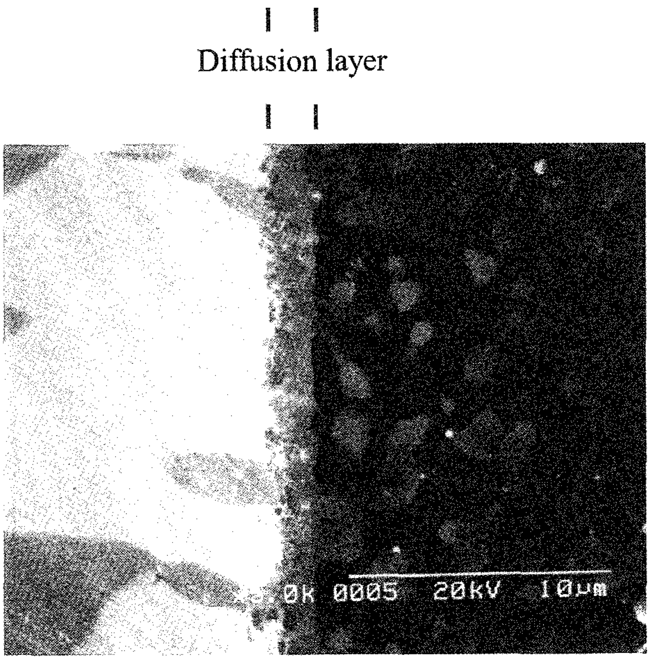


Figure 5 SEM image of Ag-Cu-Ti/electrical sialon interface in the specimen brazed for 300s at 1113 K.

Figure 5 shows a SEM image of the Ag-Cu-Ti brazing metal/electrical sialon interface in the specimen brazed for 300 s at 1113 K. In the interface region, a diffusion layer[8] enriched with Ti element was detected by EDX analysis in analogy with the bonding of electrical sialon to some metals. It has been reported that in the bonding of silicon nitrides with the Ag-Cu-Ti brazed metal a diffusion layer was formed at the interface of Ag-Cu-Ti/silicon nitride and the main reaction products at the interface were TiN and Ti_5Si_3[11,12,13], which improved the wettability[14] of brazing metal and occurred a high bonding strength. The wettability of brazing metal seems to be an important factor to obtain a high bonding strength. Therefore, the wettability of brazing metal was also investigated in this experiment.

Figure 6 shows temperature dependence of contact angle of an Ag-Cu-Ti liquid metal droplet on the plane surfaces of electrical sialon, TiN and Ti, in which the plane film surfaces of TiN and Ti were made by ion plating of TiN and Ti elements on the surface of electrical sialon, respectively. These contact angles were measured after holding each droplet for 300 s at each temperature. The value of contact angle at a lower temperature of 1113 K shows comparatively large values of 33 degree for the electrical sialon surface and 55 degree for Ti film surface, though these angles decrease as the measuring temperature increases. On the other hand, the contact angle in the case of TiN film surface showed lower values than 10 degree immediately after melting of the Ag-Cu-Ti brazing metal, which was observed even at lower measuring temperatures. Based on these results of SEM observation at the interface and the EDX analysis for the Ag-Cu-Ti/electrical sialon system, the diffusion layer formed at the interface was estimated

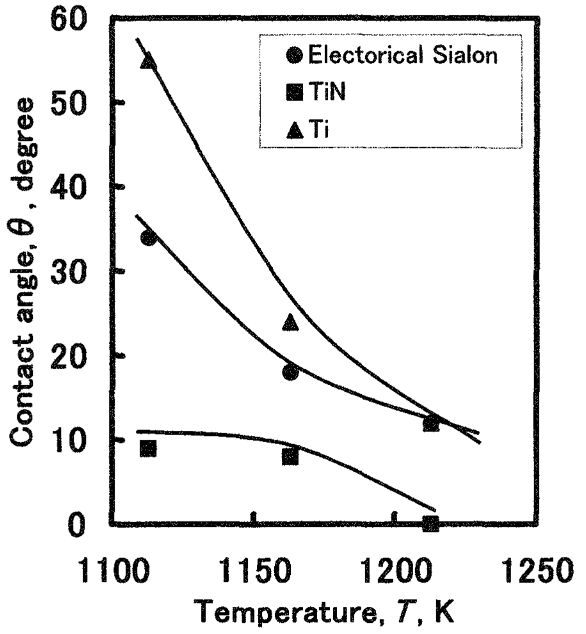


Figure 6 Temperature dependence of contact angle of an Ag-Cu-Ti liquid metal droplet on the plane surfaces of electrical sialon, TiN and Ti.

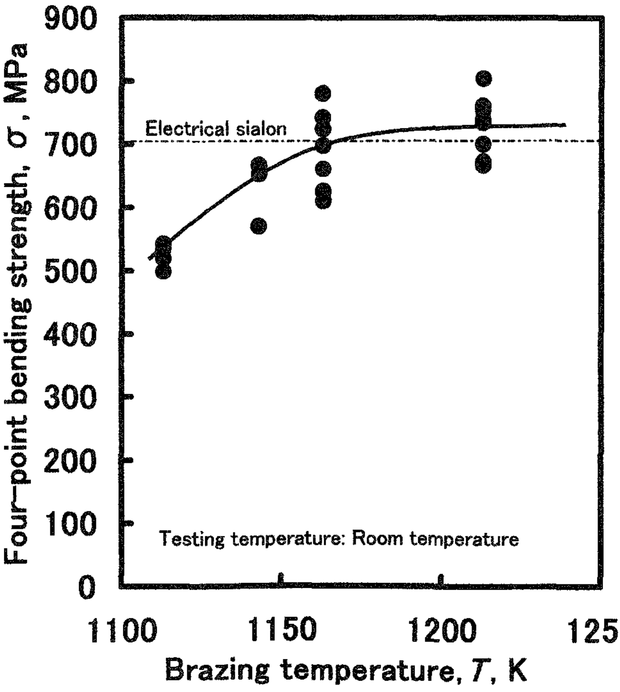


Figure 7 Relation between four-point bending strength and brazing temperature in the specimen with 30 μm thickness of the brazing metal layer.

to have a similar composition to that for the Ag-Cu-Ti/silicon nitride system. Therefore it is presumed that the wettability and the reaction products at the interface are important parameters in the process for bonding the electrical sialon ceramics with brazing metal and that these parameters in the Ag-Cu-Ti/electrical sialon system are favored to obtain a good bonding strength by raising the brazing temperature. Then, the optimum condition for making a titanium rich diffusion layer was investigated to increase the bonding strength by raising the brazing temperature.

Figure 7 shows the result obtained on the relation between the four-point bending strength and the brazing temperature in the specimen with 30 μm thickness of the brazing metal layer. When the brazing temperature is high, the bending strength shows higher values. And the bending strength indicates as the same value of 700 MPa as the strength of electrical sialon itself above at 1163 K.

Figure 8 shows the BSE images of Ag-Cu-Ti brazing metal/ electrical sialon interface in the specimens brazed for 300 s at (a) 1083 K, (b) 1113 K, (c) 1163 K and (d) 1213 K. In the Ag-Cu-Ti brazing metal region, a drastic change in the microstructure is observed depending on the brazing temperature. In the region the white part shows Ag rich phase and the gray part shows Cu and Ti rich phase. The microstructure at 1113, 1163 and 1213 K shows an eutectic structure. On the other hand, the microstructure at 1083 K shows the original structure of the brazing metal before melting, even though the melting temperature of the brazing metal was estimated to be 1063 K. In this experiment at 1113 K, the eutectic structure after melting and the original structure before melting were sometimes observed at the same time. Therefore, the drastic change in the microstructure is due to difference in the melting and brazing temperatures. In the interface region, however, the thickness of the diffusion layer enriched with Ti element is observed to increase with increasing the temperature. The thickness value, however, was less than 3 μm even at 1213 K, which was about 1/4 thickness of the value in the case of the metal/Ag-Cu-Ti/electrical sialon bonding[8]. That is, the thickness of the diffusion layer did not become thicker in the electrical sialon/Ag-Cu-Ti/electrical sialon bonding than in the metal/Ag-Cu-Ti/electrical sialon bonding even at higher brazing temperature. Furthermore, a very thin and continuous reaction layer with high concentration of Ti element was formed between the diffusion layer and the electrical sialon, which will be described in some detail in Fig. 9. This reaction layer is known to be a good one for raising the bonding strength. This fact suggests that the concentration of 2 mass% Ti in the brazing metal is not enough to obtain the good reaction layer sufficiently thicker for the bonding in the ceramic/Ag-Cu-Ti/ceramic bonding than in the metal/Ag-Cu-Ti/ceramic bonding, because the reaction area between the Ag-Cu-Ti brazing metal and the electrical sialon is two times larger in the ceramic/Ag-Cu-Ti/ceramic bonding than in the metal/Ag-Cu-Ti/ceramic bonding. The continuous reaction layer enriched with high concentration of Ti element was also formed on TiN as well as SiAlON at the higher bonding temperature, in which the thickness of the continuous reaction layer was about 0.3 μm at 1213 K. In the continuous reaction layer, the elements of Ti, N and Si were mainly detected by EDX analysis. Based on the brightness of the BSE image, the compound phases of TiN and Ti_5Si_3 (Titanium siliside) were suggested to be formed in the continuous reaction layer.

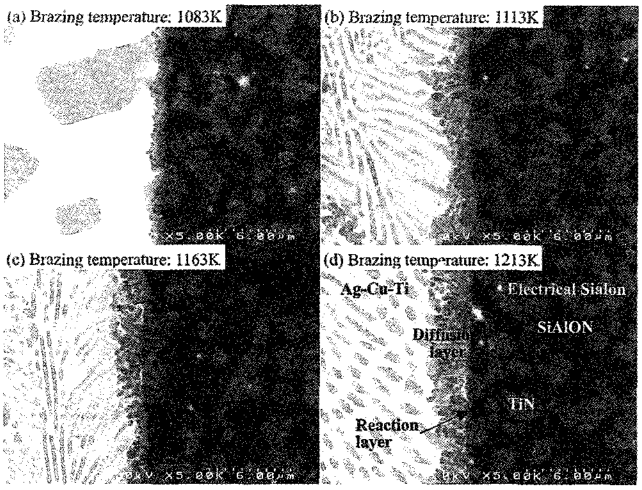


Figure 8 BSE images of the Ag-Cu-Ti/electrical sialon interface in the specimens brazed for 300 s at (a) 1083 K, (b) 1113 K, (c) 1163 K and (d) 1213 K.

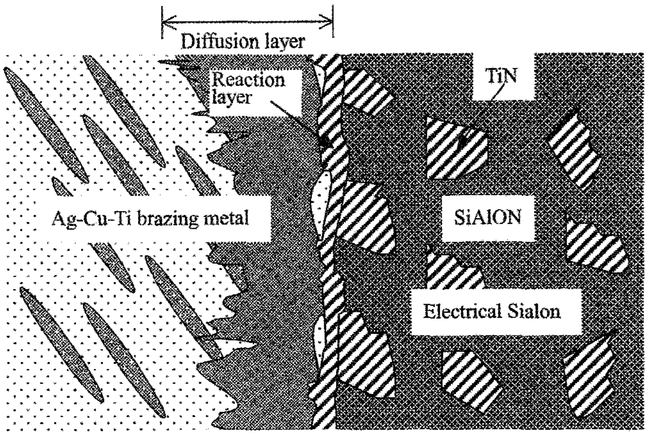


Figure 9 Schematic view of the Ag-Cu-Ti/electrical sialon interface in the specimen brazed for 300 s at 1213 K.

By the way, as shown in Fig. 9 the continuous reaction layer connecting TiN particles in the electrical sialon matrix seems to be a kind of an anchor which makes the bonding between the Ag-Cu-Ti brazing metal and the electrical sialon to be higher one. Therefore, the continuous reaction layer connecting TiN particles seems to play an important role for raising the bonding strength, especially in the case of the brazed specimen having the smaller residual stress and showing the higher bonding strength than the strength of the electrical sialon itself.

3.3 Bonding strength at high testing temperatures

Figure 10 shows the testing temperature dependence of four-point bending strength for the brazed joint electrical sialon and the single electrical sialon (parent material for the joint specimen) specimens. The four-point bending strength of the single electrical sialon specimen shows almost constant values or the slightly decrease with increasing temperature in the whole temperature range. While, the strength of brazed joint electrical sialon specimen shows the notable decrease with increasing the temperature. Especially, the decrease above 873 K is remarkable. In this experiment, fracture after the bending test was observed in the electrical sialon near the brazed interface for all the brazed joint electrical sialon specimens. Authors previously reported[15] that in the case of the brazed joint of electrical sialon to some metals the four-point bending strength showed 85% of the strength of the electrical sialon. The strength was known to also decrease with increasing the temperature. And the fracture occurred in the electrical sialon near the brazed interface up to 473 K, though it occurred at the interface between the brazing metal and the metal in the metal/Ag-Cu-Ti/ceramic bonding above at 673 K, and the four-point bending strength decreased to under 200 MPa. The residual stress in the brazed joint electrical sialon seems to play an important role for the bonding strength in this experiment.

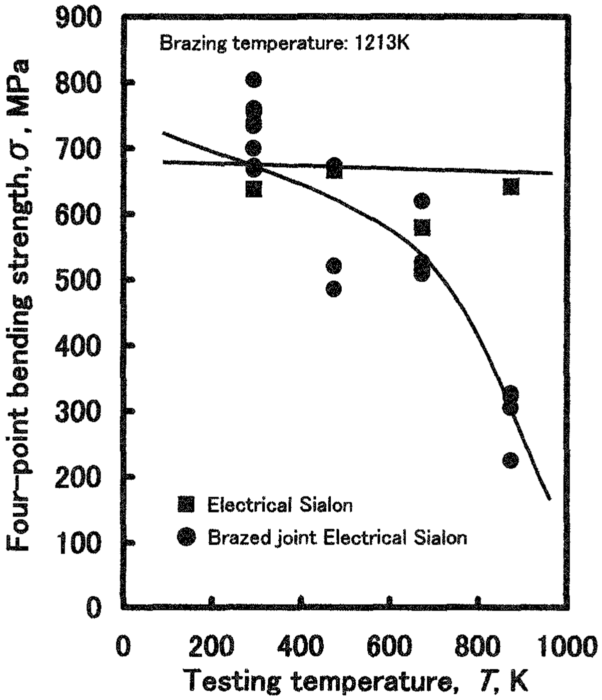


Figure 10 Relation between four-point bending strength and testing temperature in the specimen with 30 μm thickness of the brazing metal layer.

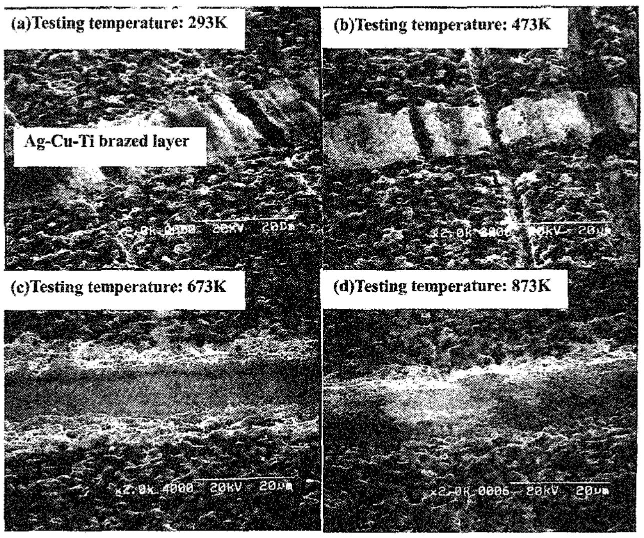


Figure 11 SEM images of the Ag-Cu-Ti brazing metal layer in the tension side joint surface of the specimen after four-point bending test. The test was conducted at (a)293 K, (b)473 K, (c)673 K and (d)873 K.

Figure 11 shows SEM images of the Ag-Cu-Ti brazed metal layer in the tension side joint surface of the specimen after four-point bending test. The test was conducted at 293 K, 473 K, 673 K and 873 K. It is recognized that the SEM image dose not change up to 473K, though grinding marks on the Ag-Cu-Ti brazing metal layer vanish at 673 K and the brazing metal layer looks to heape up about 5 μm at 873 K. In this case, deterioration of the brazed bonding interface was hard to postulate because the fracture did not occur at the interface but occurred in the electrical sialon up to 873 K though morphological change in the brazed metal layer was observed. And also, deterioration in the strength of the electrical sialon itself dose not observed up to 893 K. Therefore, the decrease in the strength of the brazed joint specimen may be due to the thermal stress or residual stress in the electrical sialon, which is caused by the difference in thermal expansion coefficients of the electrical sialon and the Ag-Cu-Ti brazing metal at the testing temperatures. In the brazing joint method mentioned above, the joint strength at higher testing temperatures seems to be eventually controlled only by the melting temperature of the brazing metal. However, it may be also necessary to take account of the thermal stress due to the thermal expansion mismatch between the brazing metal and the ceramics at appreciable temperatures. And it is also important for the joint to get the high bonding strength of the brazing metal interface. Ceramics/brazing metal interface is concluded to have sometimes an advantage over metal/brazing metal interface, because at the ceramics/brazing metal interface the favorable reaction product for the strength is often formed as mentioned above.

4. Conclusions

Electrical sialons were joined with an active brazing metal of Ag-Cu-Ti in a vacuum. Relation between the residual stress in the electrical sialon and the thickness of brazing metal layer at the joint interface was studied. Relations among the residual stress, the bonding strength and the bonding temperature were also examined. The following results were obtained.

(1) When the thickness of the brazing metal layer increases, the bonding strength decreases due to the increase in the residual tensile stress at the direction perpendicular to the joint interface.

(2) The optimum condition for obtaining the highest bonding strength is the brazing with the 30 μm thickness of brazing metal layer for 300 s at above 1163 K, in which the bonding strength is almost same as the strength of electrical sialon itself.

(3) The formation of the continuous reaction layer connecting TiN particles in the electrical sialon matrix seems to play an important role as an anchor for raising the bonding strength.

(4) The value of the bonding strength is almost constant up to 673 K, though the strength at 873 K shows remarkable decrease due to the thermal stress caused by the difference in the thermal expansion coefficients of the electrical sialon and the brazing metal.

(5) The fracture of the interface was hardly observed in the electrical sialon joint up to 873 K, though the fracture is known to be often observed at the interface between brazing metal layer and some metals.

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