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Brazing Si_3N_4 Ceramic to AISI 5140 Steel under Pressure

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Pressures (0 to 40 MPa) were applied to the joints of Si_3N_4 ceramic to 5140 steel during vacuum brazing with Ag-Cu-Ti active filler metal. Pressurization started at various temperatures (873, 973, and 1073 K) and ended at room temperature during cooling. Results show that there is an optimum starting temperature to pressurize, at which the maximum room temperature shear strength of the joint is obtained.

Keywords
brazing, ceramic-metal, pressure, shear strength

1. Introduction

JOINING ceramic to metal is important for applications in which ceramics are bonded to structural components. Preparation of strong, reliable ceramic-to-metal joints is becoming an increasingly important technological problem. For instance, many advanced designs for automotive engines require means for high-strength ceramic-to-ceramic and ceramic-to-metal joints (Ref 1, 2). The strength of the ceramic-to-metal joints is, however, influenced by the residual stresses and the stress concentration caused by the differences between the thermal expansion coefficients and the elastic moduli. The thermal expansion difference between the materials becomes one of the major obstacles to obtaining sound, highly reliable joints (Ref 3).

Many experimental studies were conducted regarding the material combinations and the joining method. Some analytical studies on the thermal and residual stresses also were conducted (Ref 4-7). The active filler metal brazing method is widely used. Commercial Ag-Cu-Ti brazes, which are ductile and have melting temperatures (T_m) of 878 to 1103 K, are finding important applications with a wide range of ceramics, most notable with Si_3N_4 components used to produce high performance automobile turbochargers. Joints produced with these brazes can be strong and reliable; thus M.G. Nicholas and S.D. Peteves (Ref 8) found that Si_3N_4 brazed with Ag-Cu-Ti has a mean strength of 822 MPa and a Weibull modulus of 11.1. Those commercial brazes wet well but do not flow rapidly into wetted capillary gaps between workpieces as does the Ag-Cu eutectic. The liquid flow rate is not determined simply by balance between a surface energy driving force and a viscous impedance. Active metal braze flow involves lateral growth of a wettable reaction product ahead of the liquid front and, hence, could be controlled by slow solid-state diffusion processes (Ref 8). In this study, pressures are applied to the joints to force the liquid brazes to flow along the interfaces and to fill the gaps.

Many experimental studies were conducted to decrease the stress concentration and the residual stresses. Thermoelastic-plastic finite-element analysis showed that the stress concentration can be reduced by changing the shape of the ceramics of the joint (Ref 9). Investigations of cracking in multilayered ceramic-to-metal composites show that the dominant cracking behavior depends on the volume fraction and yield strength of the filler metal (Ref 10). Pressurization can reduce the thermostress and stress concentration caused by the different thermal properties of the two materials. Pressurizing on the liquid filler metal can decrease the volume fraction of the filler metal. Therefore, the strength of the joint should be improved.

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2. Experimental Details

Si_3N_4 ceramics and AISI 5140 steels were joined by the active metal vacuum-brazing method. China National Standard BAg72CuTi (GB 10086-88) was used as brazing filler metal. (This braze is known in the U.S.A. as "Ticusil.") Chemical compositions and properties of test materials are listed in Tables 1 and 2, respectively. Brazing was performed in a vacuum furnace with a pressure bar, as shown in Fig. 1. A vacuum of 7×10^{-3} Pa (5×10^{-5} torr) was maintained during brazing. Brazing was done at a temperature of 1173 K with a 10 min hold. The heating rate was less than 20 K/min, and the cooling rate was less than 15 K/min between 1123 and 923 K and 1.5 K/min between 923 and 293 K.

Table 1 Chemical compositions of test materials

Element	Composition, wt%
5140 steel	
C	0.41
Si	0.29
Mn	0.73
Cr	0.97
P	0.02
S	0.03
Ag-Cu-Ti	
Ag	72
Cu	28
Ti	3

Table 2 Material properties

Property	Si_3N_4	5140	Ag-Cu-Ti
Modulus of elasticity, GPa	304	206	...
Poisson ratio	0.27	0.3	...
Expansion coefficient, $\times 10^{-6}$	3.0	12.0	...
Bending strengh, MPa	790	...	...
Tm, K	2173	...	1051

Pressures were applied to the joints by the loading bar at various temperature ranges (1073 to 293 K, 973 to 293 K, and 873 to 293 K). The stresses were 5, 10, 20, and 40 MPa, respectively. Prior to brazing, the brazing surface of Si_3N_4 was first ground with SiC grains of 0.147 μm; then it was boiled in 10% NaOH solution for 10 min. The steels and the filler metals were polished with 15 μm sandpaper and cleaned with ultrasonic vibrations in acetone.

Shear tests were performed on thermomechanical simulator Gleeble 1500 (Duffers, Duffers, NY 12140) at room temperature. Loading rate was 0.2 mm/min. Dimensions of Si_3N_4 are 4 by 6 by 12 mm, and dimensions of 5140 steel are 5 by 12 by 12 mm. Filler metals are 0.2 mm thick plate. The brazed seam thickness was measured under optical microscope.

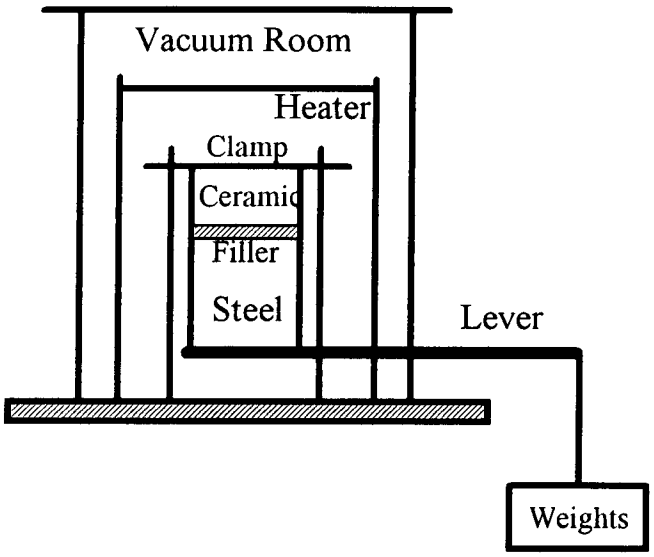

Fig. 1 Schematic brazing furnace

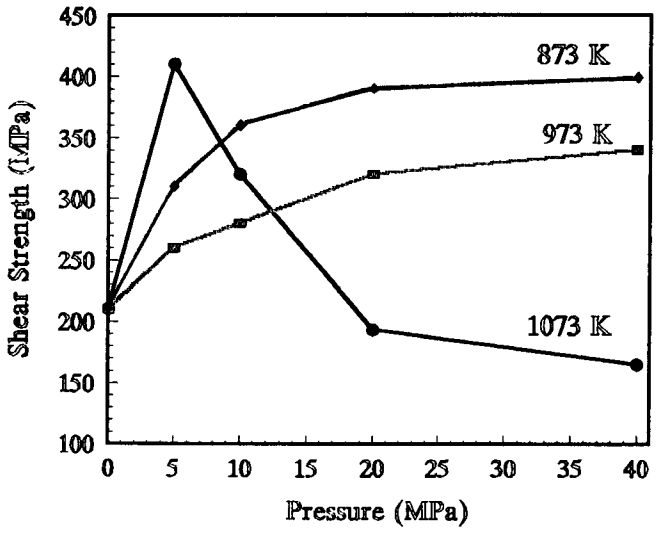

Fig. 2 Room temperature shear strength of the joints brazed under various pressures and pressurized at various starting temperatures

3. Results and Analysis

Pressurized during cooling, the joints have higher shear strength than the unpressurized ones. Results are shown in Fig. 2. When pressures were applied from 1073 K (above the T_m of the filler) to room temperature, 5 MPa pressure had an obvious effect on the joint shear strength. The highest strength was obtained at 10 MPa. Higher pressures than 10 MPa resulted in lower strength. Pressure of 40 MPa caused the lowest strength even lower than that of unpressurized ones. Pressure is not always beneficial to the joint strength when pressurization starts above the T_m of the filler.

When pressures were applied from 973 K (below the T_m of the filler) to room temperature, increase of the joint strength was very rapid at low pressure. At high pressure, this increase was slow. The same result was obtained with the joints pressurized from 873 K to room temperature. There is a transition pressure point, above which the increase of the joint strength with the increase of pressure is not apparent. See Fig. 2. This transition pressure for pressure starting temperature 973 K is smaller than that for 873 K. At the same pressure, the shear strength of the joints pressurized from 973 K is much higher than those pressurized from 873 K. That is, higher pressure does not always give the higher strength when joints are pressurized from the temperature below the T_m of the filler. The effect of the pressure deals with the pressure starting temperature. The higher the starting temperature is, the higher the joint strength will be. At low pressure (below 10 MPa), the joints pressurized from the temperature above the T_m of the filler (1073 K) have higher strength than the ones pressurized from the temperature below the T_m of the filler (973 and 873 K). However, at high pressure, results were different. Joints pressurized at temperatures above the T_m of the filler have lower strength than those pressurized at temperatures below the T_m of the filler. Therefore, the pressurization method must be carefully chosen to obtain the highest shear strength.

Figure 3 shows the thicknesses of the brazed seams. Here the conclusion is that pressurization can reduce seam thickness. When pressurization started at 1073 K during cooling, almost all the braze metal had been squeezed out. These seams are not more than 0.03 mm thick and are thinner than those pressurized starting at a lower temperature. When pressurization started below the T_m of the filler during cooling, the seam thickness increased with increase of the pressure. This result was the same as that of the strength. The relationship between the strength and the seam thickness was found. When the seams are more than 0.03 mm thick, the joint strength increases with the decrease of the seam thickness. However, when the seam is less than 0.03 mm thick, the joint strength decreases with the decrease of the seam thickness. Therefore, the joint shear strength did not always increase with the decrease of the seam thickness, as shown in Fig. 3. Thus, it is not always true that the thinner the brazed seams are, the higher will be the joint strength.

Figure 4 shows the statistic probability of fracturing in ceramic. Results show that the higher the pressure applied on the joint starting at the temperature below the T_m of the filler, the lower will be the probability of fracturing in ceramic. However, with pressurization starting at the temperature above the

T_m, the probability of fracturing in ceramic with the lower pressure is smaller than that of unpressurized ones. That with higher pressure is even higher than that of unpressurized ones, as shown in Fig. 4. This may be the main reason for the lower shear strength of the joint at higher pressure.

4. Discussion

4.1 *Starting Temperature for Pressurization*

The choice of starting temperature for pressurization has a great effect on joint properties, as shown. When pressurizing starts at the temperature above the T_m of the filler during cooling, the pressure squeezes almost all the molten filler. On the other hand, when pressurizing from the temperature below the T_m of the filler during cooling, the pressure just compresses the solid filler metal. The volume fraction of the filler in the former joints is much less than that of the latter ones. Because the joining of Si_3N_4 to 5140 steel with Ag-Cu-Ti is performed by the reaction of Ti with Si and N, the Ti activity is an essential factor for achieving high strength joints (Ref 10). The Ti activity depends on the brazing temperature and the flow of the filler metal. Those Ag-Cu-Ti brazes wet well, but do not flow rapidly into wetted capillary gaps between workpieces as does the Ag-Cu eutectic. When pressurizing above the T_m of the braze, the molten braze was forced to flow along the interfaces and can fill the gap. Thus the joint strength was increased. Therefore, the conclusion is made that pressurization starting at the temperature above the T_m of the filler has an advantage over that below the T_m when the pressure is not more than 10 MPa.

4.2 *Effect on Joints*

Pressure increases the flow rate of the filler metal, both liquid and solid. Flow of the active metal braze involves lateral growth of a wettable reaction product ahead of the liquid front and, hence, could be controlled by slow solid-state diffusion processes (Ref 10); therefore, the reaction between the filler and the ceramics should be improved under pressure. Proper reactions can improve the bond force of ceramic to metal. However, extensive reactions would decrease the bond force because of the brittle reaction products (Ref 8). This may be one of the reasons that low strength is obtained at high pressure when pressurizing started at 1073 K.

Investigations of cracking in multilayered ceramic-to-metal composites show that the dominant cracking behavior depends on the volume fraction and yield strength of the metal (Ref 8). Pressurizing on the liquid filler can decrease the volume fraction of the filler, as shown in Fig. 3. The relationship between the strength and the joint thickness is shown in Fig. 5. With the joint thickness decreasing, the joint strength increases when the joint thickness is more than 0.03 mm. This may be caused by the volume decrease of the low-strength filler metal. How-

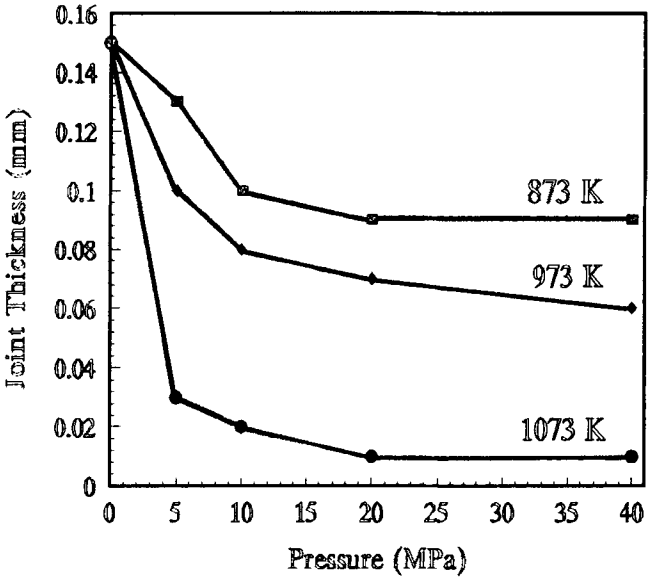

Fig. 3 Effect of pressure on the joint seam thickness

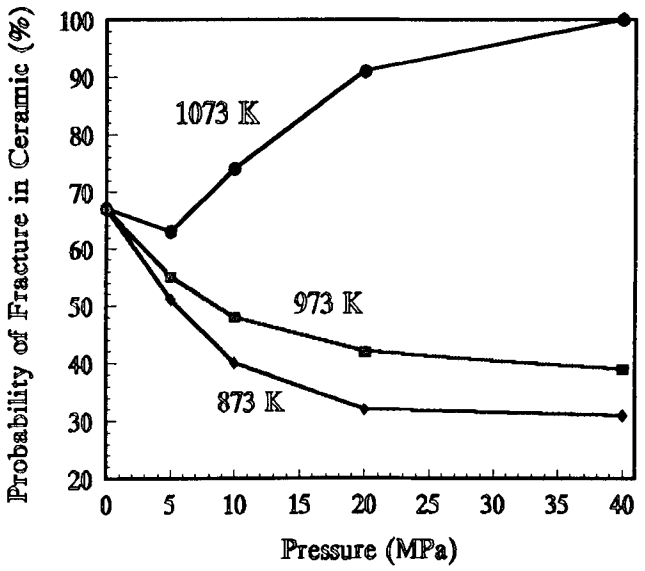

Fig. 4 Probabilities of fracturing in ceramic

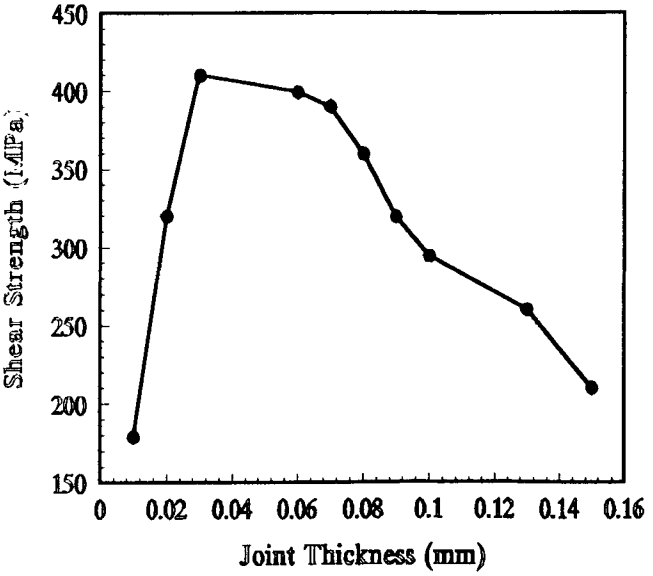

Fig. 5 Strength at various joint thicknesses

ever, excessive decrease of the filler harms the joint strength. This may be caused by the brittle reaction products.

Pressurization can reduce the thermal stresses and stress concentration caused by the different thermal properties of the two materials. First, when pressurizing at the temperature above the T_m, the decrease of the volume fraction of the filler metal reduces the tensile effect of the liquid fillers on the ceramic. Second, pressure applied on the solid joint can restrain the thermal compression and decrease the thermal stresses caused by the compression. Therefore, the residual stresses and stress concentration would be remitted, and the strength of the joint would be increased, as shown in Fig. 2. Since the residual stresses and stress concentration were reduced, not eliminated completely, the probabilities of fracturing in ceramic increase as the joint strength increases.

5. Conclusions

- Pressurizing above the melting temperature (T_m) of the filler metal during cooling can improve the joint strength greatly; pressurizing below the T_m can improve joint strength gradually. The best pressurizing temperature is approximately above the T_m of the filler during cooling. However, high pressures do not always improve joint strength.
- Pressurizing properly can increase joint shear strength, but pressurizing improperly can decrease joint shear strength. Pressurization method must be carefully chosen.
- It is not always true that the thinner the brazed seams are, the higher will be the joint strength. There is a critical thickness point, above which the joint strength increases with the decrease of the brazed seams, but below which the joint strength decreases with the decrease of the brazed seams.

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References

1. H. Mizuhara and E. Huebel, Joining Ceramic to Metal with Ductile Active Filler Metal, *Weld. J.*, Vol 65 (No. 10), p 43-51, 1986
2. J.P. Hammond, S.A. David, and M.L. Santella, Brazing Ceramic Oxides to Metals at Low Temperature, *Weld. Res. Suppl.*, Vol 47 (No. 10), p 227-232, 1988
3. H. Kobayashi, Y. Arai, H. Nakamura, and T. Sato, Strength Evaluation of Ceramic-Metal Joints, *Mater. Sci. Eng. A*, Vol 143 (No. 1-2), p 91-102, 1991
4. B.J. Dalgleish, A.P. Tomsia, K. Nakashima, M.R. Locatelli, and A.M. Glaeser, Low Temperature Routes to Joining Ceramics for High Temperature Applications, *Scr. Metall. Mater.*, Vol 31 (No. 8), p 1043-1048, 1994
5. O. Kimura and T. Kawashima, Effect of Interlayer Thickness on Residual Thermal Stresses in a Ceramic-to-Metal Cylindrical Joint, *J. Am. Ceram. Soc.*, Vol 76 (No. 3), p 757-759, 1993
6. K. Asami and N. Shirski, Influence of Residual Stress on Tensile Fracture Stress in S45C/Si_3N_4/S45C Bonded Joints, *Trans. Jpn. Soc. Mech. Eng.*, Part A, Vol 59 (No. 561), p 1202-1207, 1993 (in Japanese)
7. J. Intrater, Review of Some Processes for Ceramic-to-Metal Joining, *Mater. Manufact. Proc.*, Vol 8 (No. 3), p 353-373, 1993
8. M.G. Nicholas and S.D. Peteves, Reactive Joining, Chemical Effects on the Formation and Properties of Brazed and Diffusion Bonded Interfaces, *Scr. Metall. Mater.*, Vol 8 (No. 1), p 1091-1096, 1994
9. T. Yada and H. Koguchi, Reliability Evaluation of Joints of Ceramics and Metal, *JSME Intl. J.*, Series 1, Vol 34 (No. 2), p 163-170, 1991
10. M.C. Shaw, D.B. Marshall, M.S. Dadkhah, and A.G. Evans, Cracking and Damage Mechanisms in Ceramic-Metal Multilayers, *Acta Metall. Mater.*, Vol 41 (No. 11), p 3311-3322, 1993