Interlayer selection and thermal stresses in brazed Si_3N_4–steel joints

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The influence of Cu, Kovar, Mo, and W interlayers on the magnitude and distribution of thermal stresses and on the tensile strength of brazed Si_3N_4–steel joints is examined using a combination of finite element modelling (FEM) calculations and direct experiment. The FEM takes into account plastic flow and changes of mechanical properties of the interlayer, the steel substrate, and the filler metal as temperature decreases following the brazing operation. Joints made using low yield strength/high ductility interlayers, such as Cu, have lower thermal stresses and higher strengths than those made using low thermal expansivity/high yield strength interlayers, such as Mo or W. A composite interlayer comprising Cu and W will produce the lowest thermal stresses during brazing. Increasing the thickness of the interlayer decreases the thermal stresses produced during brazing, since the rigid restraint effect due to the high yield strength/high elastic modulus steel substrate is reduced. MST/1442

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Introduction

Active filler metals comprising Ag–Cu–Ti alloys have been commonly employed in brazing Si_3N_4 to steel[1] for applications in advanced heat engine construction. However, the marked difference between the thermal expansion coefficients of Si_3N_4 and mild steel implies that the joint interface will be acted on by residual stresses produced as a result of thermal expansion mismatch; this decreases the final tensile strength of dissimilar joints. Although it has been suggested that ductile interlayer materials placed between the ceramic and the metal can decrease the thermal stresses generated during the joining operation, it is not entirely clear how the mechanical properties of the interlayer and substrate material control thermal stress reduction – a general framework is required for selection of an ideal interlayer material.

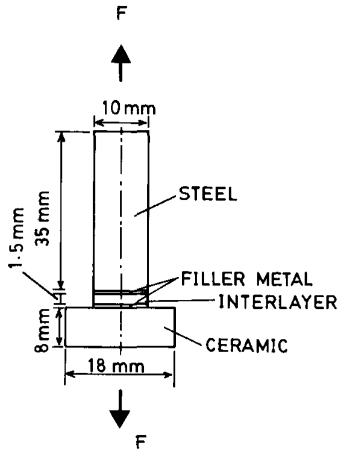
This paper examines the influence of various interlayers on the stresses produced during Si_3N_4–steel joining. The approach taken involves a combination of finite element modelling (FEM) calculation and tensile testing of brazed joints. The results indicate that the selection of interlayer materials for Si_3N_4–steel brazing depends not only on thermal expansion mismatch, but also on the thickness of the interlayer and on the variation of interlayer mechanical properties (elastic modulus, yield strength, and work hardening rate) with temperature.

BACKGROUND

Much research has focused on developing interlayer materials that will decrease the thermal stresses generated during ceramic–metal joining. Naka *et al.*[1] examined the joining of Si_3N_4 to other ceramics (alumina, zirconia, etc.) and to metal substrates (Fe, Cu, Invar, AISI 3014 stainless steel, etc.) and found that joint tensile strength decreased as the difference in thermal expansivities of Si_3N_4 and the ceramic or metal substrates increased. Nicholas and coworkers[2,3] also found a clearcut relation between final joint strength and increasing thermal expansivity mismatch when diffusion bonding alumina to a range of metal substrates. These results were explained on the basis that high thermal mismatch stresses facilitated joint failure at low applied loads. Based on this argument, the use of interlayers which have thermal expansivity values equal to, or close to, that of the ceramic should provide optimal joint mechanical properties. However, among the pure metals, low thermal expansivity is generally associated with high yield strength (e.g. W and Mo) and many investigators have indicated that the use of ductile metal interlayers may overcome thermal stress problems.[4–8] Although the use of Al, Al–AlSi, Fe, Nb, and Cu interlayers has proved successful, it is not entirely clear how the interlayer properties affect the distribution and magnitude of thermal stress produced during ceramic–metal bonding.

Analytical and FEM calculations have been used to evaluate the residual stress distribution produced by thermal expansion mismatch. Naka *et al.*[1] calculated the stress distribution produced when Si_3N_4 was joined to stainless steel and indicated that (i) the regions of maximum principal stress were almost perpendicular to the joint interface and (ii) these stresses were greatest at the outer edges of the joint (at the weld toe region). Similar indications were obtained[4,7] from FEM calculations of the residual stresses produced when alumina was joined to steel using different interlayer materials. Elastic stresses only were considered in these FEM calculations, plastic flow in the metal substrate being ignored, and the mechanical and thermophysical properties of the interlayer material (elastic modulus, shear modulus, Poisson's ratio, and thermal expansivity) were assumed to be constant as the temperature decreased following bonding. As a result, Yamada *et al.*[4] and Saganuma *et al.*[7] pointed out that their calculated values overestimated the stresses present in the actual joint. In this regard, Hamada *et al.*[8,9] used FEM calculations to estimate the residual stresses produced when carbon steel was diffusion bonded to alumina. Although the FEM calculations of Hamada *et al.* allowed for plastic flow in the interlayer material, they did assume that key parameters such as thermal expansivity, yield strength, elastic modulus, etc. were independent of temperature. In spite of this deficiency, Hamada *et al.*[8] did indicate that the principal stresses were lower when the thickness of the Cu interlayer exceeded 1·0 mm and this explained readily the higher joint strength in test welds produced using 2 mm thick Cu interlayers. Yamada *et al.*[4] also noted that the strength of alumina–alumina joints produced using Al–AlSi interlayers was higher when the thickness of the interlayer was increased.

This paper examines the joining of Si_3N_4 to mild steel using a joining method that combines active brazing with the insertion of various metal interlayers. The FEM calculations take into account the changes that occur in



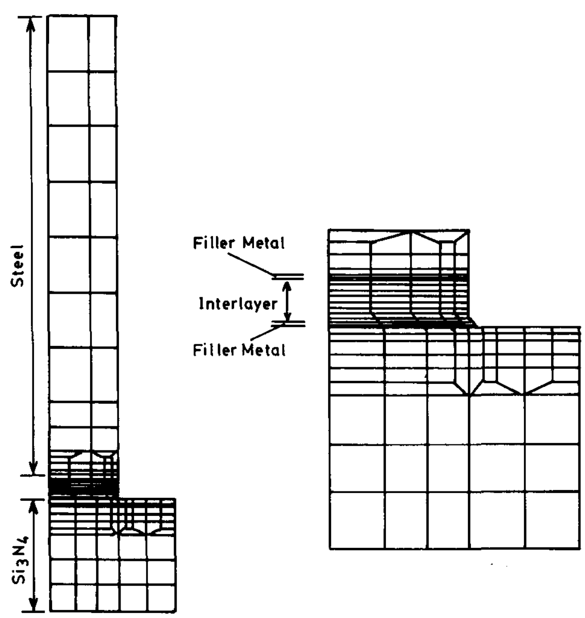
1 Dimensions of tensile test specimens; F denotes force applied during tensile testing

interlayer properties (elastic modulus, yield strength, strain hardening rate, and thermal expansivity) as temperature decreases following brazing. Also, plastic flow in the interlayer, in the steel substrate, and in the Ag–Cu–Ti filler metal is allowed for in the FEM calculations. The test results indicate that, although thermal expansivity mismatch does play a basic role in producing residual stresses during cooling after brazing, a combination of high ductility and low yield strength in the interlayer material has a dominant influence on final joint strength. Also, the thickness of the interlayer has a critical influence on the levels of thermal stress produced during ceramic–metal joining.

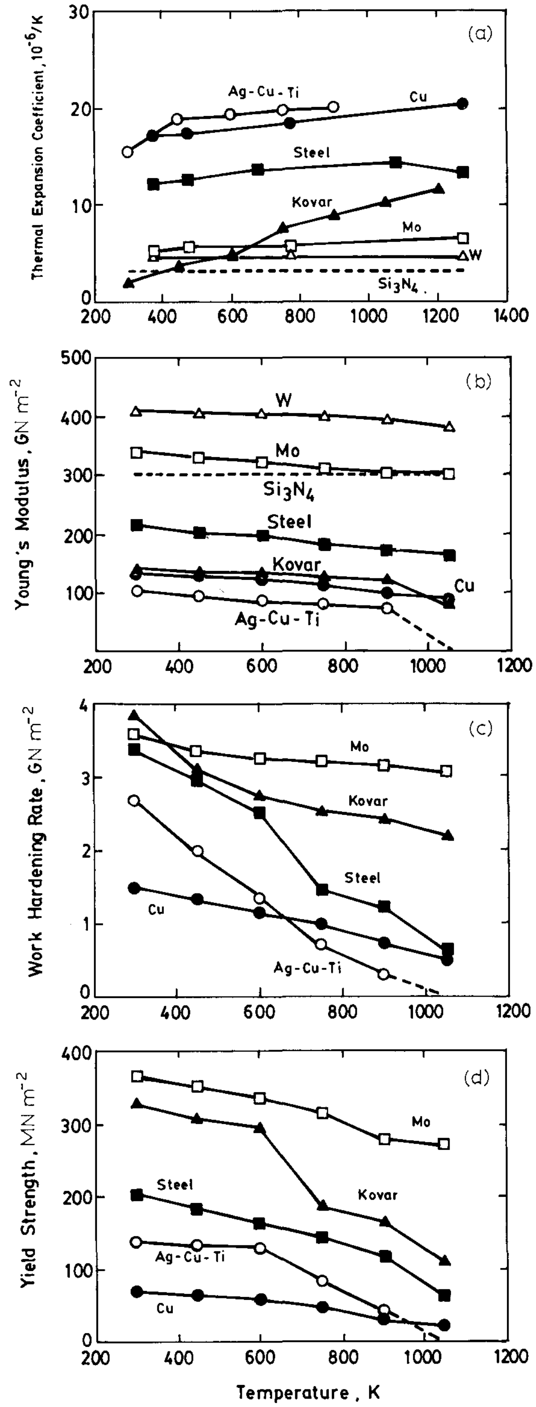
Experimental procedure

TENSILE TESTING

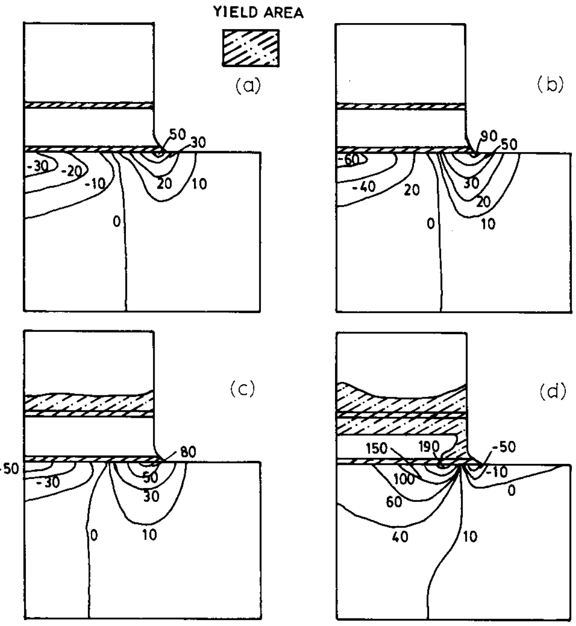
Hot pressed and sintered Si_3N_4 ceramic and mild steel were vacuum brazed using an active Ag–27Cu–3Ti (wt-%) filler metal. The specimen dimensions are shown in Fig. 1. The vacuum pressure during brazing was less than 5×10^{-5} torr, the brazing temperature was 1153 K, and the holding time was 600 s. The cooling rate in the temperature range between 1153 and 923 K was 20 K min^{-1}, and was 1·5 K min^{-1} at temperatures below 923 K. The width of the active Ag–Cu–Ti filler metal was 0·4 mm before brazing and about 0·2 mm after brazing. The mechanical properties of joints produced using 1·5 mm thick Cu, Kovar, Mo, and W interlayer materials were evaluated. The crosshead speed during tensile testing of completed joints was 2·5 mm min^{-1} and the tensile results



2 Configuration used during FEM calculations

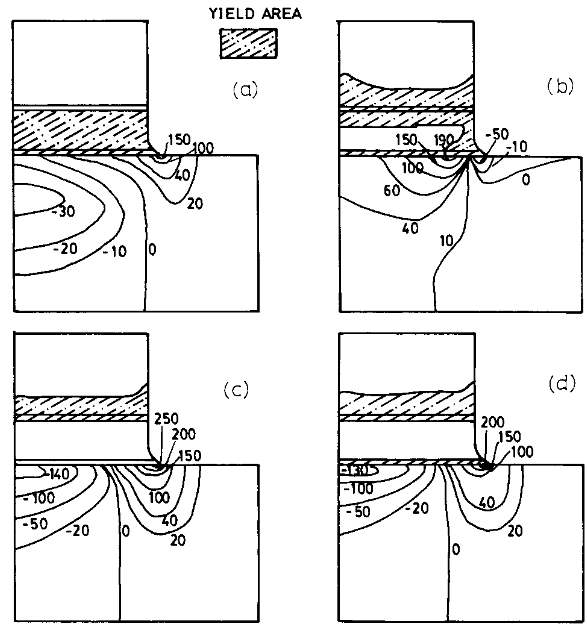


3 Variation of *a* thermal expansion coefficient, *b* elastic modulus, *c* strain hardening rate, and *d* yield strength with temperature for several interlayer materials, steel, and Ag–Cu–Ti filler metal



a 900 K; *b* 700 K; *c* 500 K; *d* 300 K

4 Magnitude and distribution of residual stresses (in MN m^{-2}), at various temperatures, in joints produced using Kovar interlayer material



a Cu; *b* Kovar; *c* Mo; *d* W

5 Magnitude and distribution of residual stresses (in MN m^{-2}) in joints produced using various interlayer materials

presented are the mean of two or three brazing tests for each interlayer material.

FEM CALCULATIONS

The FEM configuration comprised 4–8 node, two dimensional axisymmetric elements (the finite element grid is shown in Fig. 2). Elastic stresses only were assumed in the ceramic material and for the W interlayer (since it had a much higher yield strength than that of the steel substrate or the Ag–Cu–Ti filler metal). An elastic–linear hardening, stress–strain model was applied for the steel substrate, filler metal, Cu, Kovar, and Mo interlayers. The ceramic test specimens were 18 mm in diameter and of 8 mm thickness. The steel sample was 35 mm in length and 10 mm in diameter. The interlayer thickness was 1·5 mm and, in all calculations, the thickness of the Ag–Cu–Ti filler layer following brazing was taken as 0·2 mm.

During the heating cycle, the Si_3N_4 and steel substrates were assumed to be separate. During cooling after brazing, the FEM calculations evaluated the temperature range from the filler metal melting point (1073 K) to room temperature (300 K). The influence of temperature on interlayer, steel substrate, and Ag–Cu–Ti filler metal properties was taken into account during the FEM calculations. Figure 3 shows the change of thermal expansivity, elastic modulus, strain hardening rate, and yield strength as temperature decreased following the brazing operation. The values of yield strength, strain hardening rate, and elastic modulus for each interlayer material and of thermal expansivity for Ag–Cu–Ti filler metal and Kovar were determined by direct experiment. The Poisson's ratio of Ag–Cu–Ti was assumed to be that of Ag (0·367) and the thermal expansivities and Poisson's ratios of the various interlayers were obtained from Ref. 10.

It is considered that fracture will occur in Si_3N_4 when the maximum principal stress equals the fracture strength in this material. As a result, the direction and magnitude of the maximum principal thermal stresses in Si_3N_4 were calculated. The von Mises yield criterion was applied when assessing yielding of the steel substrate, Cu, Kovar, and Mo interlayers and the Ag–Cu–Ti filler metal.

Results

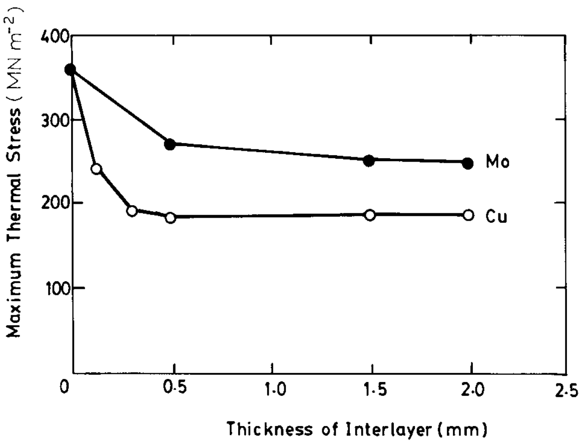
EFFECT OF INTERLAYER MATERIAL ON THERMAL EXPANSIVITY MISMATCH STRESSES

Figure 4 shows the variation of maximum principal stresses produced in a joint made using a Kovar interlayer. The magnitude of the residual stresses and the dimensions of the yielded zone increased as temperature decreased. The stress distribution in the ceramic at 300 K was different from that at higher temperatures (500 and 700 K). This occurred because the thermal expansivity of Kovar became less that that of Si_3N_4 when the temperature decreased below 400 K. It follows from Fig. 4 that the magnitude and distribution of thermal stresses is markedly dependent on changes of material properties as temperature decreases following brazing.

Figure 5 shows the effect of various interlayer materials on the magnitude and distribution of thermal stresses. Joints produced using Mo and W interlayers had similar principal stress distributions but different stress magnitudes (the principal stresses were higher when a Mo interlayer was used). Also, the use of high yield strength W and Mo interlayers promoted yielding of the steel substrate (*see* Fig. 5). When a Kovar interlayer was used, the interlayer region was partially yielded, a compressively stressed region was produced at the weld toe, and the calculated tensile stresses were as high as 196 MN m^{-2} (*see* Table 1). When

Table 1 Calculated maximum principal stresses in Si_3N_4–steel joints when using different interlayer materials

Interlayer	Interlayer thickness, mm	Maximum thermal stress in ceramic, MN m^{-2}
Cu	1·5	178
Kovar	1·5	196
W	1·5	228
Mo	1·5	256
Cu–Kovar	0·5–0·8	132
Cu–W	0·5–0·8	148



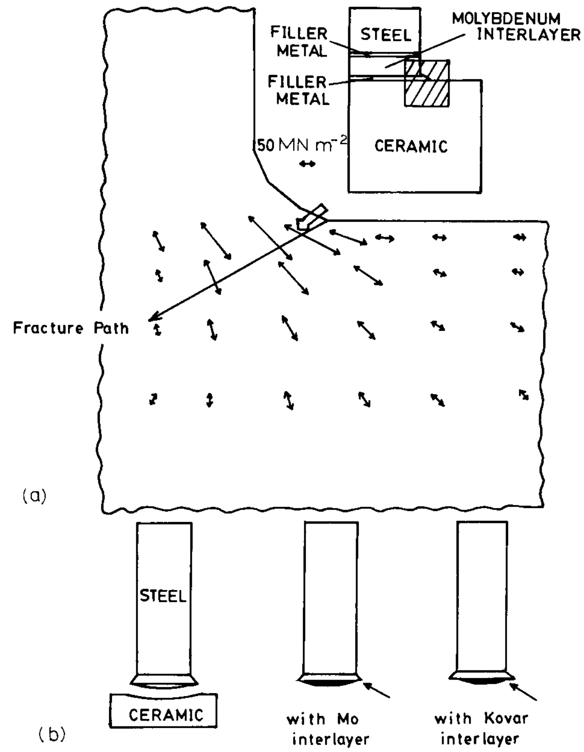
6 Effect of Cu and Mo interlayer thickness on principal stresses produced during joining

a Cu interlayer was used, complete yielding of this material produced joints that had the lowest calculated principal stress values (178 MN m^{-2}; *see* Table 1).

Figure 6 shows the relationship between interlayer thickness and the maximum calculated principal stress values; lower thermal stresses were produced when the interlayer thickness was increased.

TENSILE TEST RESULTS

Figure 7 shows the relationship between joint tensile strength and the calculated maximum principal stresses in joints made using different interlayer materials. The calculated values of thermal stress corresponded with the tensile test results, in that the highest joint strength was associated with the lowest levels of thermal stress. Figures 4 and 5 indicate that the highest stresses are produced at the joint interface, particularly at the weld toe region, and this generally accounted for the mode of failure which occurred during tensile testing (fracture initiation at the weld toe region followed by propagation into the bulk ceramic). However, the thermal stress calculations indicate that a compressively stressed region is produced at the weld toe region when a Kovar interlayer is used (*see* Fig. 5). The presence of this compressively stressed region explained why fracture initiated in a region away from the weld toe (*see* Fig. 8*b*). When a Mo interlayer was used, the weld toe was in tension and the fracture initiated at this region (Fig. 8*a*).

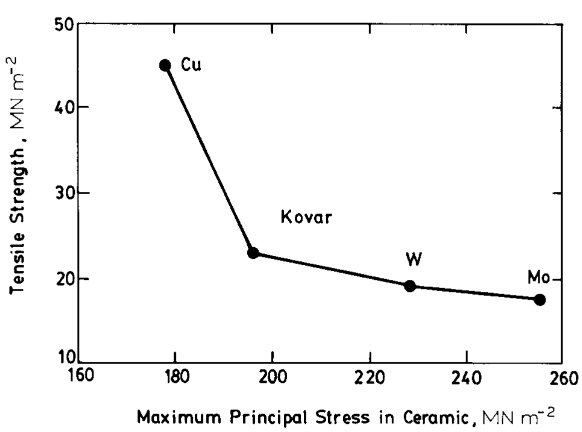


8 *a* Residual stress distribution when steel is joined to Si_3N_4 using Mo interlayer (length and direction of arrows indicate magnitude and direction of principal stresses); *b* fracture paths in joints between steel and Si_3N_4 when Mo and Kovar interlayers are used

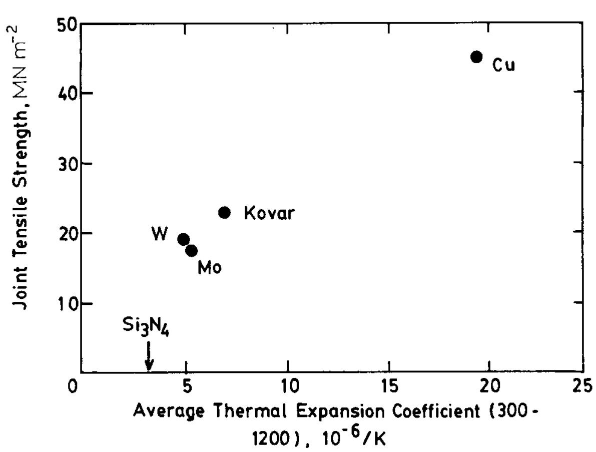
Discussion

SELECTION OF INTERLAYERS

If thermal expansivity mismatch is entirely responsible for the development of residual stresses in dissimilar joints, the lowest values should occur when W and Mo interlayers are used (since their thermal expansivities are the lowest for the interlayer materials examined in this study). However, joints made using W and Mo interlayers had the highest calculated thermal stresses and lowest tensile strengths (*see* Figs. 7 and 9). In fact, the highest tensile



7 Relationship between calculated maximum principal stress in Si_3N_4 and joint strength produced using various interlayers



9 Relationship between joint tensile strength and thermal expansion coefficient of various interlayer materials

strength values were produced when a Cu interlayer was used. This was caused by significant yielding of the Cu interlayer, i.e. the deleterious influence of the large thermal expansivity value for Cu was counteracted by its ability to yield and relax the thermal stresses generated during cooling following brazing. The thermal mismatch stresses acting on the ceramic/metal interface were large, even when W and Mo interlayers were used (*see* Fig. 5). This would be expected, since the lowest thermal expansivity interlayer examined in this study (W) had a value which exceeded that of Si_3N_4 (i.e. $4{\cdot}5 \times 10^{-6}$ compared with $3{\cdot}0 \times 10^{-6}$). The interlayer did not yield when W and Mo were used and, consequently, the tensile strength of the ceramic–metal joint should depend totally on the thermal expansivity difference between the ceramic and the metal interlayer. This explains why the joint tensile strength found when using W interlayers exceeded that found when Mo was used (*see* Fig. 7).

The results in this study indicate that low yield strength/high ductility interlayer materials are more effective in decreasing thermal expansion mismatch stresses when steel is joined to Si_3N_4 material. It is noteworthy that the Cu interlayer produced higher joint strengths than Kovar (*see* Fig. 7). In effect, the formation of a compressively stressed region at the weld toe (when using a Kovar interlayer) shifted the initiation point of failure, but did not produce higher tensile strength. Table 1 gives the calculated thermal stresses produced when composite Cu–W and Cu–Kovar interlayer materials are considered (the Cu is adjacent to the ceramic material in each case). It is apparent that the use of composite interlayers is an effective method of decreasing the residual thermal stresses produced during dissimilar welding.

The interlayer thickness has a marked effect on the level of thermal stresses (and final joint strength) produced during ceramic–metal joining. The results in Fig. 6 indicate that the highest calculated maximum principal stresses occurred when the interlayer thickness was less than 0·5 mm. Hamada *et al.*[8] found a similar relationship between interlayer thickness and calculated thermal stresses in alumina–steel joints, and Yamada *et al.*[4] indicated that the highest joint strengths in alumina–steel joints occurred when the interlayer thickness exceeded 2 mm. The relationship between interlayer thickness, stress–strain characteristics, and calculated thermal stress values has been examined recently by Zhou *et al.*[11] The calculated thermal stresses produced during ceramic–metal bonding depend on whether the resulting thermal stress exceeds the yield point of the interlayer material. For fully elastic conditions, the thermal stress σ is determined by the well known relation

$$\sigma = \Delta\alpha\Delta T \frac{E_{\rm I}E_{\rm II}}{E_{\rm I} + E_{\rm II}} \qquad (1)$$

where $\alpha_{\rm I}$ and $\alpha_{\rm II}$ are the thermal expansivities of the metal and ceramic rods ($\Delta\alpha = \alpha_{\rm I} - \alpha_{\rm II}$), the temperature change is $\Delta T = T_2 - T_1$, and $E_{\rm I}$ and $E_{\rm II}$ are the elastic moduli of the metal and ceramic rods. When the thermal stress in the metal rod exceeds its yield strength, the determining equation for thermal stress is

$$\sigma = \sigma_{\rm Iy} + (E_{\rm Ip}\Delta\alpha\Delta T) \qquad (2)$$

where $E_{\rm Ip}$ is the linear strain hardening coefficient and $\sigma_{\rm Iy}$ is the yield strength of the metal (linear elastic–linear plastic conditions are assumed). In equation (1), the thermal expansivity mismatch term has the dominant effect on the thermal stress level produced during joining. When a low yield strength interlayer such as Cu is used, equation (2) indicates that the yield stress of the interlayer plays a dominant role. In addition, the mechanical properties of the substrate and the thickness of the interlayer material will have a significant effect on the yielding process. When the interlayer is extremely thin, the rigid restraint produced by the higher yield strength/higher elastic modulus steel substrate will induce triaxial stresses in the interlayer and increase its apparent elastic modulus and yield strength. When this occurs, the tensile strength measured during uniaxial testing will be much less than that in the actual joining situation. When the interlayer thickness increases, the restraint effect of the steel substrate decreases and the Cu yields such that lower thermal stresses are produced during ceramic–metal joining. When the thickness of the low yield strength interlayer is considerable, the final mechanical properties of the ceramic–metal joint will depend only on the mechanical properties of the interlayer and the ceramic material.

Conclusions

The influence of various interlayer materials (Cu, Kovar, W, and Mo) on the magnitude and distribution of residual stresses and on the final tensile strength of brazed Si_3N_4–steel joints was examined. Plastic flow and changes of the interlayer, substrate, and filler metal mechanical properties as temperature decreased following brazing were taken into account during FEM calculations. The following conclusions were drawn.

1. During Si_3N_4–steel joining, the use of low yield strength/high thermal expansivity Cu interlayers produced higher joint strengths than low thermal expansivity Mo and W interlayers. These results were accounted for by yielding of the Cu interlayer, i.e. yielding relaxed the stresses produced by the large difference in thermal expansivities of Cu and Si_3N_4. The use of composite Cu–W and Cu–Kovar interlayer materials will produce the lowest thermal stresses in Si_3N_4–steel joints.
2. The residual stresses resulting from thermal expansivity mismatch decreased as the interlayer thickness increased. This occurred because the rigid restraint due to the higher yield strength/higher elastic modulus steel substrate inhibited yielding of thin interlayers. When the thickness of the Cu interlayer increased, the rigid restraint effect decreased and this allowed the interlayer material to yield and relax the thermal stresses generated during ceramic–metal joining.
3. The results in this paper suggest strongly that selection of the optimum interlayer materials for ceramic–metal joining depends not only on thermal expansion mismatch, but also on the thickness of the interlayer and on the change of interlayer mechanical properties (elastic modulus, yield strength, and work hardening rate) with temperature.

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