Influence of shape and size on residual stress in ceramic/metal joining

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The effects of shape and size on the residual stress on the surfaces of silicon nitride/Invar alloy joints have been examined by means of the strain gauge method. The highest residual stress perpendicular to the interface appeared near the corners in the rectangular bond face joint. It was tensile in the silicon nitride and compressive in the Invar alloy. The highest tensile stress in the rectangular bond face joint was larger than that in the circle bond face joint. The larger the diameter of the cylindrical joint used, the larger was the tensile stress induced. The residual stress parallel to the interface was compressive in silicon nitride while that in the Invar alloy was tensile.

1. Introduction

In recent years, great efforts have been focused on joining ceramics to metals to establish processes for a wide range of industrial uses. Several important problems, however, still remain unsolved. Among them, how to produce atomic bonds at ceramic/metal interfaces and how to compensate for residual stress due to thermal expansion mismatch between two constituents are the most critical.

The quality of interfacial bonding depends on chemistry [1, 2] and lattice matching [3–5]. Precise understanding of these is required to establish the understanding of bonding processes. However, it is possible to produce a relatively strong bond empirically at ceramic/metal interfaces by several methods. For example, the active brazing method is one of the effective processes to obtain strong interfaces [6, 7]. The role of the active elements is well understood [1, 7].

On the other hand, the thermal expansion mismatch effect is a serious problem because, even if a strong interface could be achieved, joints with large residual stress are easily broken. Nicholas *et al.* [8] have succeeded in achieving a strong joint between alumina and an austenite stainless steel, which have a large thermal expansion mismatch, by using a soft metal. Some of the present authors have also developed several effective methods [9–12]. In any case, it is very important to know what residual stress is, how large it is and how to reduce it in order to obtain a sound and strong joint.

The aims of the present work are to understand the effects of joint size and shape on the residual stress in the silicon nitride/Invar alloy joints when aluminium is used as a brazing material, and to elucidate the effect of varying the metallic materials to be bonded on the residual stress. These joints can be obtained by a simple joining process, low temperature, low pressure and short-time bonding [11].

2. Experimental procedure

Pressureless-sintered silicon nitride used in this work contained alumina, magnesia and yttria as sintering additives. Four cylinders of 5, 7, 14 and 20 mm diameter and 10 mm high and one 15 mm × 20 mm × 10 mm block, were used. The bond surfaces, one end of the cylinders and a 15 mm × 20 mm face of the block, were ground to average surface roughness 0.2 μm. Super Invar alloy (Fe–32% Ni–5% Co), K-EL50, was supplied by Touhoku Tokushukou Co., Ltd. From this alloy four cylinders and one block with the same dimensions as the silicon nitride specimens were prepared and the faces to be bonded were finally ground to average surface roughness of $\sim 0.2\,\mu$m. Kovar alloy (Fe–29% Ni–15% Co), K-ET51, was also supplied by the same company and one 15 mm × 20 mm × 10 mm block was prepared. A 200 μm thick sheet of aluminium, AA1050, was used as a brazing metal.

The two constituents between which an aluminium sheet was put were tied fast with molybdenum wire as shown in Fig. 1a. They were slightly pressed at the brazing temperature by thermal expansion mismatch between the molybdenum wire and the metals. Bonding was carried out in a vacuum of 5×10^{-5} torr. The bonding temperature was 1073 K, the bonding time was 10 min, and the cooling was controlled at a rate of $20\,\mathrm{K\,min^{-1}}$. Fig. 1b shows examples of the joints. There was little difference in the interfacial microstructures of the joints regardless of the size and shape of the joints. In addition, the interfacial structure of the silicon nitride/Kovar alloy joint was almost the same as that of the silicon nitride/Invar alloy

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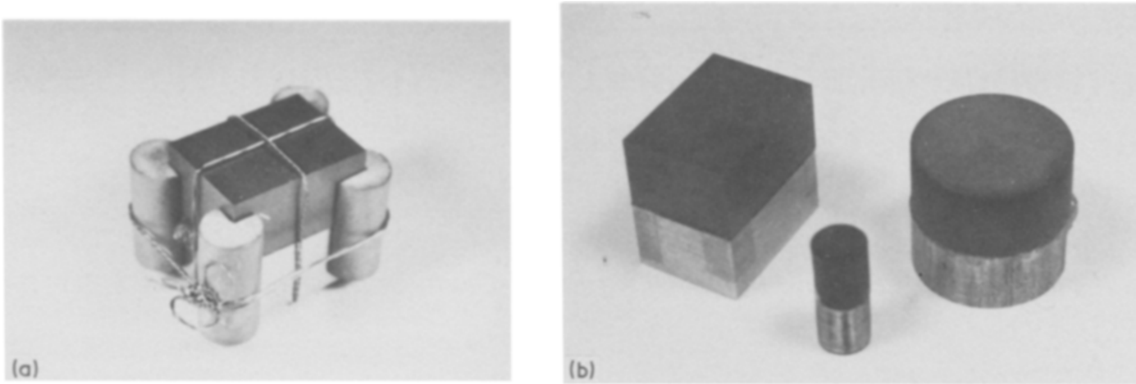

Figure 1 (a) Construction of joints. (b) Three joints, one with a 15 mm × 20 mm rectangular face, and one with a 5 mm and a 20 mm diameter cylindrical face.

joint. This seems to be consistent with the fact that the chemical compositions of the two alloys are very similar. Fig. 2 shows the microstructure of the silicon nitride/Invar interface. A thin aluminium layer remained adjacent to the silicon nitride and a reaction layer was formed between aluminium and the alloys. Fine cracks growing perpendicular to the interface were recognized in the reaction layer. The network of these fine cracks played a role in relieving the residual stresses in the joints.

The thermal expansion of each constituent was measured using a Fizeau-type dilatometer. Residual stress was measured by the strain gauge method [13]. Self-compensating type strain gauges, which had a 1 mm × 1 mm face, were pasted on the surface of the joint. Then the joint was cut along the interface, within the aluminium layer and partly in the reaction layer, with a no-strain cutting machine. The difference in the indications of the strain gauge before and after cutting leads to evaluation of the residual strain in the joint. The residual stress was calculated using the Young's moduli: 300 GPa for the silicon nitride, 140 GPa for the Invar alloy and 136 GPa for the Kovar alloy.

3. Results and discussion

3.1. Thermal expansion of materials

Fig. 3 shows the thermal expansion curves as a function of temperature up to 1073 K, which is the brazing temperature. Both metals show the well-known peculiar expansion curves which show small expansion below 473 and 673 K for the Invar alloy and for the Kovar alloy, respectively. Below these temperatures, the thermal expansion mismatches between the metals and silicon nitride are small. In particular, below 473 K the thermal expansion curve of the Invar alloy fitted very well to that of silicon nitride, but that of the Kovar was slightly larger. However, beyond 473 K, that of the Invar increased drastically and became larger than that of the Kovar. Because the interfacial structures of the silicon nitride/Invar and the silicon nitride/Kovar joints were indistinguishable, the difference in the residual stress which is shown below should be mainly caused by the mismatched expansion characteristics.

3.2. Residual stress in the joint with square bond surface

Fig. 4 shows the residual stress perpendicular to the interface along the lines 1 mm from the interface. Tensile stresses are plotted as positive and compressive stresses as negative values.

On the silicon nitride side, significant tensile stresses are present near the corner. The stresses were lower nearer to the centre, becoming negligible or compressive. On the other hand, the residual stresses were always compressive on the Invar alloy side. The highest compressive stress appeared at the centre and decreased as the corner was approached. Changes of the residual stresses on both sides were very similar.

The most important stress in a ceramic/metal joint is the tensile residual stress which appears near to or at the interface and at the free surface in the ceramic

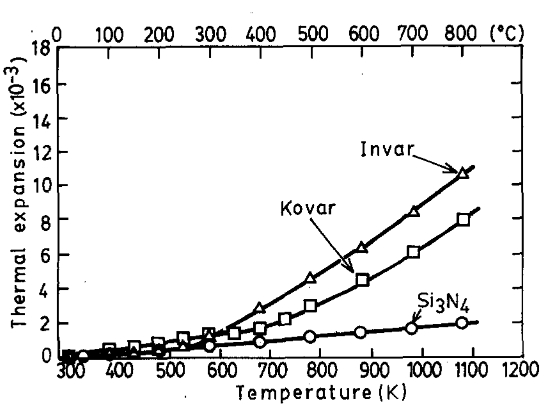

Figure 2 Interfacial microstructure of the silicon nitride/Invar alloy joint(SEM).

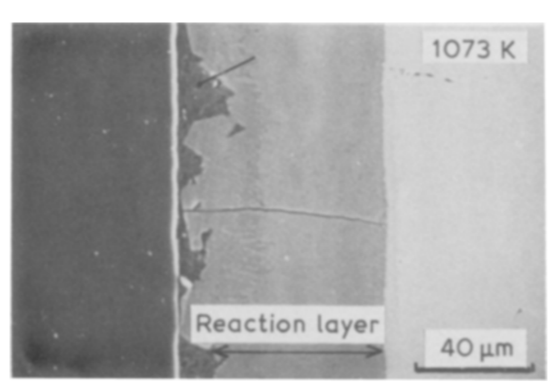

Figure 3 Thermal expansion curves of constituents.

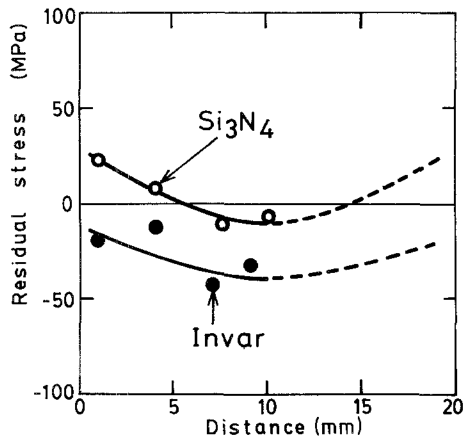

Figure 4 Residual stress acting perpendicular to the interface at points 1 mm from the interface in the rectangular face joint.

side [11, 12]. The corner stress in the rectangular joint is this stress. However, the maximum stress plotted in Fig. 4 might not be the actual maximum because the strain gauge used in this study had an area of 1 mm × 1 mm. Roughly speaking, the stress obtained in Fig. 4 is the average value for the square. However, the highest stress should be precisely at the interface and the corner 1 mm away. However, it is impossible to measure residual stress at such a position and, therefore, the corner stress plotted in Fig. 4 is treated as the highest stress in the present study.

Fig. 5 shows the change of the residual stress across the interface acting perpendicular to the interface along a line 1 mm from the corner. The highest tensile stress appeared nearest to the interface in the silicon nitride and the highest compressive stress at the point nearest to the interface in the Invar alloy. These stresses continued to decrease further away from the interface. The tensile stress in the silicon nitride became almost negligible at a point 7 mm away from the interface. Thus, it is apparent that the severest residual stress appeared near the corner nearest to the interface in the square bond face joint.

3.3. Effect of metallic materials on residual stress

Fig. 6 shows the effect of the metallic materials on the

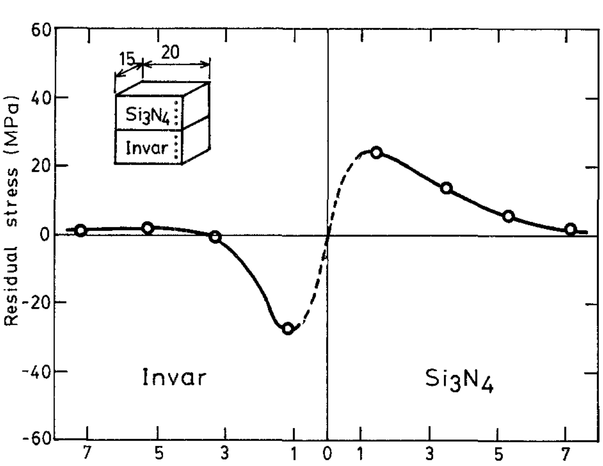

Figure 5 Residual stress acting perpendicular to the interface at the points 1 mm from the corner in the rectangular face joint. Dimensions in millimetres.

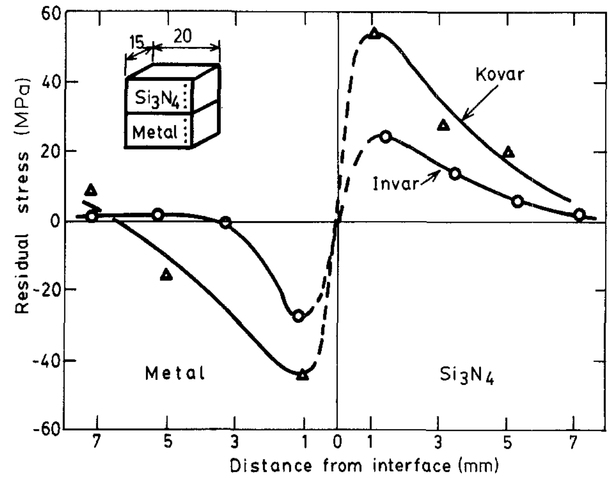

Figure 6 Effect of the kinds of metals on residual stress. Dimensions in millimetres.

residual stress acting perpendicular to the interface. It is apparent that the silicon nitride/Kovar joint has a greater residual stress than the silicon nitride/Invar joint. Considering the thermal expansions of these two metals and the silicon nitride shown in Fig. 3, beyond about 600 K the mismatch between the Kovar and the silicon nitride is smaller than that between the Invar and the silicon nitride. Below about 600 K the tendency is reversed. The fact that the residual stress in the silicon nitride/Invar joint is smaller than that in the joint with the Kovar indicates the importance of the temperature range below 600 K for the occurrence of residual stress in these systems.

In the previous work it was found that the formation of a fine crack network in the reaction layer between the aluminium and the Invar alloy or the Kovar alloy played an important role in relieving the residual stress in the joint [14]. Crack formation began at about 800 K on cooling from the bonding temperature for both systems. However, the bonding strength of the silicon nitride/Invar joint was higher than that of the joint with the Kovar, which corresponds to the difference of the residual stresses measured in both joints.

3.4. Effect of joint shape on residual stress

The comparison of the residual stress between the rectangular bond face joint and the cylindrical one of 20 mm diameter is shown in Fig. 7. Both results were similar but the highest stress measured in the rectangular bond face joint was slightly larger than that with the circular bond face. In addition, as suggested from Fig. 4 the measured highest stress in the former joint is not the actual highest one, which will be at the corner. Hence, the residual stress in the rectangular bond face joint would be higher than that in the circular bond face joint.

3.5. Residual stress parallel to interface and comparison of measurement methods

Fig. 8 shows the residual stress acting parallel to the bond interface at points 1 mm away from the interface in the silicon nitride/Invar rectangular bond face joint. A compressive residual stress was present in the silicon

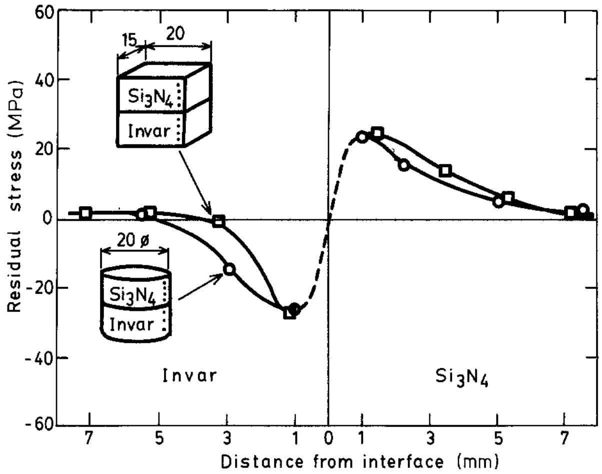

Figure 7 Effect of joint shape on residual stress. Dimensions in millimetres.

nitride while the Invar alloy was in tension. This is just the reverse of the perpendicular residual stress pattern shown in Fig. 4. However, once more the stress was smaller at the centre than at the corner.

Fig. 8 also shows a comparison of measurement methods, the strain gauge method and the X-ray method of the residual stress. The $\sin^2\psi$ method was adopted for the latter method using an incident Cu$K\alpha$ X-ray. The (222) peak and the (411) peak were selected for the Invar and for the silicon nitride, respectively. The data by both the methods in the figure were obtained from the same joint.

The residual stresses measured in both cases changed similarly with distance from the corner but differed greatly in magnitude. The changes in stress values obtained for both materials by the X-ray method were larger than those derived by the strain gauge method. Several reasons for this difference are possible. One of the most important reasons is the difference in the special resolution of stress distribution of the two methods. The areas needed for measurements on both methods are illustrated in Fig. 9. Because the X-ray method used in the present work needed a larger area in order to obtain a measurable intensity of diffracted X-rays, the stresses measured by it are given as average values over a wider area than those by the strain gauge method.

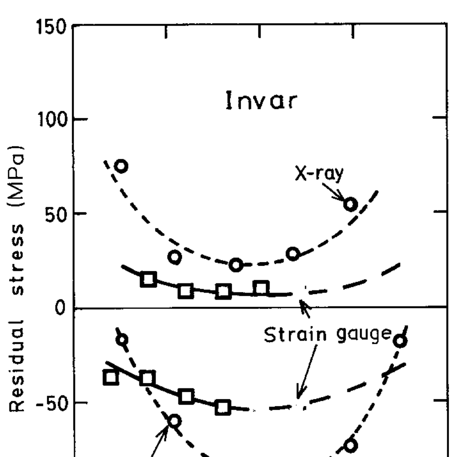

Figure 8 Residual stress parallel to the interface at points 1 mm from the interface and comparison of measurement methods.

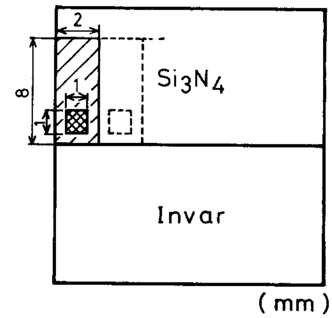

Figure 9 Comparison of measured areas in the strain gauge method and the X-ray method.

The X-ray method is one of the recommended non-destructive methods for measurements of residual stress in materials and has been widely applied. However, it is very difficult to measure residual stress within a narrow area, probably about scores of micrometres, which is required for the study of ceramic/metal joints. Such resolution is very important for investigation of brittle materials and high-intensity X-ray microdiffraction should be one of the powerful techniques for this purpose.

3.6. Effect of joint size

Fig. 10 shows the effect of size, diameter, on the highest tensile residual stress of the cylindrical joint. It is apparent that increased diameter produced larger residual stresses.

The joint has two main ways of relieving the residual stress. One is the formation of a fine crack network in the reaction layer between aluminium and the Invar. The other is the plastic deformation of the aluminium layer. These ways work very well for the smaller joint. For example, the residual stress in the 5 and 7 mm diameter joints was almost zero. On the other hand, the residual stress increased gradually with increasing diameter and a previous report showed that the strength of the silicon nitride/Invar

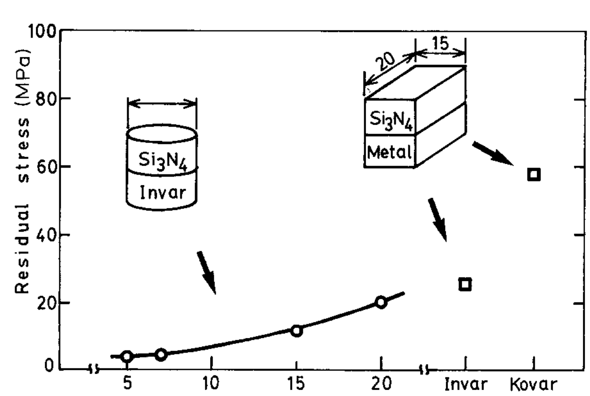

Figure 10 Effect of diameter of bond face on the highest tensile residual stress in cylindrical joints. Dimensions in millimetres.

joint of 5 mm diameter was about 150 MPa on average [14]. Hence, the residual stress of 20 MPa is not a serious problem. Larger joints or joints of a more complicated shape will require additional consideration.

4. Conclusion

The present work was concerned with several factors influencing the residual stress of the silicon nitride/Invar(or Kovar) joints produced using an aluminium filler. The strain was measured by the strain gauge method. Several important results were obtained.

1. The highest residual stress perpendicular to the interface appeared near the interface at the corner of a rectangular bond face joint. It was tensile in silicon nitride and compressive in Invar or Kovar.

2. The joint with Kovar had larger stress than that with Invar.

3. The residual stress parallel to the interface in the silicon nitride was compressive while that in Invar was tensile.

4. The rectangular bond face produced a larger residual stress than the circular bond face.

5. Increasing the diameter of the cylindrical joint produced larger tensile residual stresses.

6. There still remain several problems. One is the difference in the measured values between the strain gauge method that is destructive and the X-ray method that is non-destructive. The latter is of wider application but requires an increased X-ray intensity and resolution. Another problem is how wide a face can be bonded. From the present results, the wider bond face should produce larger residual stress. In practice, face scores of cm^2 in area need to be bonded and further work on residual stress is needed before that can be achieved.

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