Joining Ceramics to Metals using Metallic Foam

A. A. Shirzadi\textsuperscript{1}, Y. Zhu\textsuperscript{2} and H. K. D. H. Bhadeshia\textsuperscript{1}

\textsuperscript{1}Department of Materials Science and Metallurgy
University of Cambridge, Pembroke Street, Cambridge CB2 3QZ, UK

\textsuperscript{2}School of Mechanical Engineering & Automation
Beijing University of Aeronautics and Astronautics, Beijing, China

Keywords: metal-ceramic bonds; metallic foam; brazing; thermal cycling

Abstract

A general method for brazing ceramics to metals using a compliant metallic foam as a buffer layer has been developed. Using stainless steel foams, bonds between alumina and 316 stainless steel with shear strengths up to 33 MPa have been achieved. The resultant ductility enhances the resistance of the joint to thermal cycling; AlN-Inconel 600 bonds exhibited good thermal shock resistance. Alumina-stainless steel bonds withstood more than 60 thermal cycles between 200 to 800°C in air.

Introduction

There are many potential applications where joints between ceramics and metals can usefully be exploited in engineering structures. A common joining methods is brazing using filler metals, containing Cu, Ag and Ti. This can give integral joints but because of the thermal expansion differences between the metal and ceramic, the joints do not perform well during thermal cycling. Indeed, the joining process itself introduces residual stresses in the system during cooling from the brazing temperature. We aim here to alleviate these difficulties by placing a compliant buffer layer separating the metal and ceramic.

Previous work on Mitigation of Ceramic-Metal Interfaces

The thermal expansivity ($\varepsilon$) of engineering ceramics (e.g. alumina and aluminium nitride) are normally much smaller than that of most alloys (e.g. stainless steels and nickel base superalloys). When constrained to cool together, residual stresses resulting from the differences in thermal expansion can compromise the integrity of the joint. Thus, joints between alumina (Al$_2$O$_3$ $\varepsilon=8.1\times10^{-6}$ K$^{-1}$) and a metal are less likely to fail due to thermal expansivity mismatch than the joints between AlN ($\varepsilon=4.5\times10^{-6}$ K$^{-1}$) and the same metal. It is believed that reported large variations in shear strength (11.8 to 67.5 MPa) of Fecralloy-SiC diffusion brazed joints is mostly due to the residual stresses arising from thermal expansivity mismatch. Most bonds failed in the first thermal cycle and none could withstand more than three cycles \cite{1}. Similar variations in the shear strength of brazed alumina-stainless steel samples have been reported \cite{2}. The problems can be minimised by optimising the brazing conditions but the expansion mismatch is an intrinsic feature of the joint and cannot be avoided. As a result, buffer layers are sometimes used to make a gradual transition between the metal and ceramic.
The compensation interlayer approach relies on inserting one or more interlayers with an expansivity which is between that of the metal and ceramic being joined (Fig. 1). The use of more than one interlayer gives a smoother transition in thermal properties. A triple interlayer of Kovar /tungsten/ nickel and a proprietary braze were used to join SiC to a nickel-based superalloy [3]. Despite variable strengths, an average four-point-bend strength of 63 MPa was reported. The samples brazed without any interlayer had no or very low strengths.

A generalisation of the multiple interlayer approach involves the use of a functionally graded interlayer in which the material properties vary continuously across the thickness of the layer (Fig. 1). The fabrication of functionally graded materials with a linearly or non-linearly varying expansivities can be difficult. For instance, it is more practical to replicate a functionally graded interlayer by sintering a sandwich of three alumina-Cr composites when joining alumina to steel. A finite element analysis of a bond between alumina and steel, consisting of three alumina-Cr composite with 75, 50 and 25% alumina, showed that the residual stress at the alumina/composite interface could be half the value in the absence of the composite interlayers [4]. The FE analysis was based on the assumption that "crack-free perfect bonds" formed at all five interfaces (between the alumina, 3 composites and steel). However, the formation of "perfect bonds" are unlikely especially when joining alumina to a composite with 75% alumina at a relatively low temperature of 900°C for only 30 min.

The flexible interlayer approach uses a thin, dimpled or corrugated sheet of metal as an interlayer which is then vacuum brazed to the metal and ceramic parts with a filler metal- see Fig. 1. The effects of the shape of interlayer (i.e. dimpled and corrugated), dimple size, loading direction and mechanical properties of the interlayer on the shear strength of the bonds have been analysed using finite element analysis and in most cases the calculations were found to be consistent with experimental data [5]. A maximum shear strength of 25 MPa was achieved when using the dimpled steel interlayer. The flexibility of the interlayer should help reduce residual stresses but the bond strength must clearly depend to that of the interlayers and the adhesion of the brazing filler metal to the ceramic and metal.

Application of a metallic foam

The objective of the present work was to develop a joining method which meets the following criteria:
- produces ceramic-metal joints capable of resisting severe thermal cycles;
- is a generic solution applicable to various combinations of ceramics and metals;
- is simple, hence, does not require fabricating complicated interlayers;
- is economical and utilises non-precious metal available commercially.

Most of the early work on metal foams was predominantly on the processing and manufacture of aluminium foams [6,7]. Foams of various steels and nickel-based superalloys, have also been developed [8]. Published work on joining stainless steel foam to conventional 316 stainless steel, showed that bonds with mechanical strength higher than those of the parent foam can be produced by transient liquid phase bonding [9].

Given stainless steel foams can survive high temperatures and have shown promise in the context of bonding, it was decided to use 316 stainless steel foam as the flexible interlayer as illustrated in Fig. 2.
Experimental procedure

The base metals and ceramics used were Inconel 600, 316 stainless steel, aluminium nitride and alumina. The open-cell foam with 75% porosity was of 316 stainless steel. The compositions and thicknesses of the metal foils and brazing filler metals are given in Table 1.

The base metals and ceramics were cut into 10 mm diameter discs of 5 mm thickness. The foam was originally in the form of a disc of 60 mm diameter and 10 mm thick. Wire-cut electrical discharge machining was used to cut smaller discs of 10 mm diameter. Thin slices with thicknesses of 0.2, 0.4 and 0.6 mm were made using a high precision cutting machine.

A K-type thermocouple was spot welded to the base metal in order to monitor and control the temperature during bonding. Prior to bonding, all samples were ground with emery paper of 1200 grit, then washed thoroughly in ethanol and finally rinsed in acetone. The foam was cleaned ultrasonically in a bath of acetone. The specimen was placed in a vacuum bonding rig with an induction heating system. The direct measurement of the temperature at the joint vicinity, and precise control of actual heating and cooling rates are the advantages of the induction heating system. No significant temperature gradient was expected after holding the samples at the bonding temperature for a few minutes. The same induction heating system was used for the thermal cycle tests with a heating rate up to 200°C min\(^{-1}\).

The vacuum chamber was evacuated down to a pressure of 10\(^{-4}\) to 10\(^{-5}\) mbar. Details are shown in Figure 3.

The combinations studied, including the bonding conditions and outcome of each bonding attempt, are presented in Fig. 4.

Joint assessment

Bond line microstructures and fracture surfaces were examined by optical and scanning electron microscopy. X-ray energy dispersive spectroscopy (EDS) was used to determine the compositions of the new phases forming at the bond.

The bond strengths were evaluated by shear tests (Fig. 5) which were conducted only on the samples which survived being dropped onto a hard floor from about one meter height. To assess the role of the intervening metal foam in reducing the residual stress, some of the bonded samples were exposed to heating and cooling cycles in air, between 200 to 800°C with the dwell times of 3 and 5 minutes, respectively (for alumina-stainless steel samples brazed with Cusil-ABA). 800°C was considered the highest service temperature of the brazing metal and the stainless steel foam. Maintaining a fast and constant cooling rate below 200°C proved difficult since it required complicated cooling system. Therefore the thermal cycle tests were carried out between 800 and 200°C. The bonds between aluminium nitride and Inconel 600 were subjected to thermal cycles between 500 to 800°C in air, with a heating and cooling rate of 200°C min\(^{-1}\) and virtually zero dwell times. The number of cycles to failure was estimated by frequent unloading and visual examination of the samples. The most likely reason for interfacial cracking or failure was the residual stress build up due to the expansivity mismatch.
Results and Discussion

A summary of the experimental results is given in Fig. 4; use of the foam as a buffer clearly has advantages. Fig. 6 illustrates a substantial plasticity of about 1.5 mm across 10 mm diameter sample during the shear testing. The ductile failure of the samples, made using a metal foam, was attributed to the flexibility of the foam and probably the reduction in residual stresses resulting from bonding.

The stainless steel foam with 75% porosity was weak with a maximum shear strength of about 13.3 MPa. Infiltration of molten Ag-Cu-Ti eutectic filler metal into the foam gave the composite structure illustrated in Fig. 7. Note that the porosity of the foam was reduced due to the infiltration of molten filler metal as well as the deformation of the foam under the bonding load.

Optimisation of foam thickness

The integrity of joints proved to be dependent mostly on the thickness of the foam when other factors are kept constant. This is expected since thicker foams are better able to accommodate the thermal expansion differences between the adjacent ceramic and metal. However, the flexibility that makes a foam useful results in a reduction in the shear strength of the joint as the foam thickness is increased. Thicker foams are less likely to be fully penetrated by the fixed quantity of braze, so that the properties of the foam (containing voids) then dominate the overall resistance to shear.

The microstructures of bonds made using 0.2, 0.4 and 0.6 mm thick stainless steel foams are shown in Fig. 8; all samples were brazed under identical conditions using the same filler metal. The porosity at the joint clearly is exacerbated as the foam thickness becomes larger. The shear properties and thermal cycling data of these joints are illustrated in Fig. 9; because the joints were unloaded and assessed every 5 cycles, the actual cycles to failure could be at most 5 cycles less than those indicated in Fig. 9.

A few samples were directly brazed without using intervening metal foam. Some of these samples cracked within the ceramic during the joining process and none withstood the post-bonding thermal cycle tests. The random failure of the directly brazed samples is consistent with the previous work which showed a wide variation in the bond strength of such joints between ceramics and metals [1].

Fig. 10 shows the fracture surfaces of the samples brazed without and with foam after thermal cycling (200 to 800°C in air). The former showed a “cup & cone” fracture within the ceramic after the first cycle whereas the latter failed from the ceramic-foam interface after more than 60 cycles. To summarise, mechanical and thermal tests indicate that the acceptable thickness of the foam is 0.2 to 0.4 mm for the specific materials and joint configurations investigated here.

These preliminary results apply to the particular joint configurations and material combinations investigated here. It would be interesting in future work to develop a model for the process, one which includes a consideration of the optimum foam thickness, of the penetration of the braze into the pores within the foil and which gives insight into the shear and tensile strength of the assembly.
Conclusions

It is found that the use of a metallic foam as a buffer layer between ceramic and metal could be an effective way of avoiding thermal expansivity mismatch between the two materials when bonded together by brazing. It has been demonstrated that the joints are tolerant to severe thermal cycling tests.

The number of thermal cycles (200 to 800°C in air) to failure was 67±3 during the thermal cycling test. During shear testing, the failure mode was ductile due to the plasticity in the foam-based region. The maximum shear strength of the alumina-316 stainless steel bonds was of 33 MPa.

References


### TABLE CAPTIONS

Table 1: Compositions of brazing foils.

<table>
<thead>
<tr>
<th>Foil / Shim</th>
<th>Thickness / mm</th>
<th>Nominal composition / wt%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ti</td>
<td>0.01</td>
<td>99.9 Ti</td>
</tr>
<tr>
<td>Cu</td>
<td>0.07</td>
<td>99.9 Cu</td>
</tr>
<tr>
<td>Al-6082</td>
<td>0.250</td>
<td>Al-1Si-0.9Mg-0.7Mn-0.5Fe</td>
</tr>
<tr>
<td></td>
<td></td>
<td>-0.25Cr-0.2Zn-0.1Ti-0.1Cu</td>
</tr>
<tr>
<td>Cusil-ABA brazing foil</td>
<td>0.50</td>
<td>63Ag-35.25 Cu-1.75Ti</td>
</tr>
</tbody>
</table>
Fig. 1: Schematic presentation of previous approaches to join two materials with a large mismatch in coefficient of thermal expansion, such as ceramics and metals.
Fig. 2: Joint configuration when using a stainless steel foam as a flexible interlayer (not to scale).
Fig. 3. Bonding set-up used for joining metals to ceramics with intervening brazing foils and foam. This arrangement was also used for thermal cycling experiments conducted in air.
Fig. 4: Summary of joining experiments. Only the final approach on bonding alumina to Inconel 600 is discussed in details in this work. Bonding conditions in brackets.

- **Aluminium nitride | Inconel 600**
  - Solid-state bonding with no interlayer
  - 1000-1200°C, 30 mins, 6-11 MPa
  - Very weak bonds or no bonding at all.

- **Aluminium nitride | Ti | Inconel 600**
  - Brazing using Ti foil as filler metal
  - 1 min at 1200°C and 30 mins at 1000°C, 30-11 MPa
  - Liquidation at interface and formation of strong bonds but AlN cracked during post-bond thermal cycle between 500 to 800°C

- **Aluminium nitride | Ti | Foam | Ti | Inconel 600**
  - Implementing stainless steel foam (0.3 mm thick) as a buffer interlayer
  - 1 min at 1200°C and 30 mins at 1000°C, < 1 MPa
  - Strong bonds between foam and Inconel but no bond formation at AlN-foam interface

- **Aluminium nitride | CU | Ti | Foam | Ti | CU | Inconel 600**
  - TLP bonding and implementing stainless steel foam as a buffer interlayer
  - 5 mins at 900°C, < 1 MPa
  - Still no bond formation at AlN-foam interface probably due to having a much lower temperature than Inconel. It must be noted that AlN has a very low electrical conductivity hence irresponsive to induction heating

- **Aluminium nitride | Al-6082 | Foam | Ti | Inconel 600**
  - Implementing stainless steel foam (0.3 mm thick) as a buffer interlayer
  - 4 mins at 1200°C and 4 mins at 1100°C, < 1 MPa
  - High strength bonds withstanding rapid thermal cycles between 200 to 800°C in air without interface failure or cracking of AlN
  - Work continued using a commercial brazing filler metal (Cusil)

- **Alumina | Cusil | Foam | Cusil | Inconel 600**
  - Optimising foam thickness based on mechanical test and microstructure
  - 20 min at 850°C, 1.2 MPa
  - Bond with shear strength up to 33 MPa withstanding up to 71 thermal cycles between 200 to 800°C in air
Fig 5: Shear test arrangement used to assess ceramic-metal bond strength.
Fig. 6: Alumina-stainless steel sample bonded using a stainless steel foam and two layers of commercial brazing filler metal as shown schematically in Fig. 2. Arrows show the orientation of the applied shear stresses, resulting in considerable plasticity before fracture in any part of the joint.
Fig. 7: Stainless steel foam (A) after infiltration of Ag-Cu-Ti eutectic filler with a large silver concentration (B). Copper rich phases appear as light grey areas (C). Black regions represent empty pores.
Fig. 8: Cross sections of the joints between alumina and conventional 316 stainless steel made using stainless steel foams with various thicknesses and Cusil filler metal.
Fig. 9: Shear and thermal cycle data of alumina-stainless steel bonds with foam and Ag-Cu-Ti braze. Thermal cycling was in air and between 200 to 800°C with a 5 minute dwell time at high temperature.
Fig. 10: Fracture surfaces of joints without and with metallic foam following thermal cycling between 200 to 800°C in air. Number of cycles to failure were <1 and 60±4 for samples without and with a foam interlayer, respectively.