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The elevated temperature strengths of alumina–aluminium and magnesium–aluminium samples

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Pairs of alumina cones were soldered with aluminium at 1000° C and tested in tension at 20 to 500° C. The fracture strengths of the samples fell between the ultimate tensile strength of aluminium and the fracture strength of alumina, reaching a maximum at a temperature that depended on the thickness of the aluminium solder layer. The sample fracture surfaces produced by room temperature strength tests were entirely ceramic but became increasingly metallic at higher test temperatures. In contrast, the fracture strengths of magnesia cones soldered with aluminium did not peak between 20 and 500° C, and the location of the fracture surfaces could not be related to the testing temperature or sample strengths. It is argued that the effects observed with alumina–aluminium samples are due to the conflicting influence of the testing temperature on relaxation of residual stresses within the ceramic and the ability of the metal solder layer to deform. In the case of the reactive magnesia–aluminium system, strengths seemed to be largely determined by the formation of a $MgO.Al_2O_3$ spinel layer at the ceramic–metal interface during soldering and by the fragility of the porous ceramic.

1. Introduction

Ceramic-to-metal joining can play an important part in the manufacture of electrical, electronic and vacuum components. Many applications of such equipment call for the seals to be of high integrity even when exposed to elevated temperatures during or prior to operation. However, most assessments of the vacuum tightness or strength of ceramic–metal joints have been conducted at room temperature, and the objective of the work described in this paper was to obtain interfacial strength data over a range of temperatures and hence to assess the relevance of room temperature observations.

High temperatures could affect the strengths of ceramic–metal interfaces by changing the mechanical properties of the bonded materials, by relaxing stresses due to mismatches in thermal expansion coefficients, and by promoting interdiffusion or chemical interaction of the components. In an attempt to distinguish between some of these effects, a study was made of the elevated temperature strength characteristics of samples of a relatively inert ceramic–solder combination, alumina–aluminium, and of one that was notably reactive, magnesia–aluminium. These particular combinations were selected not only for their representativeness but also because of the availability of some relevant bonding, wetting and reactivity data. Thus the room temperature strengths of alumina–aluminium interfaces formed by pressure bonding have been measured by Dawihl and Klinger [1], Klomp [2, 3] and Heidt and Heinke [4]. Similarly, Nicholas [5] showed the room temperature strengths of interfaces formed at 670 to 1018° C to be 82 $MN\,m^{-2}$ while

Carnahan *et al.* [6] and Champion *et al.* [7] characterized the high temperature wetting behaviour of the system and associated it with the formation of volatile Al_2O. Bond strength data are not available for the magnesia–aluminium system but Al-Taie [8] and McEvoy *et al.* [9] studied its wetting and reactivity behaviour at 730 to 1020° C.

2. Experimental materials and techniques

The ceramics used to form the bonded samples were 10 mm high truncated cones with a base diameter of 10 mm and a top, bonding, surface diameter of 5 mm. The alumina was WADE UL 995, 99.5% Al_2O_3 with traces of CaO, MgO and SiO_2, obtained from English Glass Company Ltd. It had a density of $3.85\,g\,cm^{-3}$, a closed porosity of 3.2% and a grain size of about $30\,\mu m$ as illustrated in Fig. 1. The alumina had a rough surface with an average asperity height of $3.2\,\mu m$. The magnesia cones obtained from the Kyoto Ceramic Company had average asperity heights of $7.8\,\mu m$, and were 98.5% pure with 0.3% Al_2O_3, 0.2% CaO, 0.2% Fe_2O_3 and 0.4% SiO_3. The magnesia density was $3.2\,g\,cm^{-3}$ and it had 8% closed porosity, 5% open porosity and a grain size of about $16\,\mu m$.

The aluminium used in the study, 99.999% pure material made by Cominio Ltd, was rolled into 1 and 0.1 mm thick sheets from which 5.5 mm diameter discs were punched.

Before being soldered, the ceramic cones were ultrasonically cleansed in trichloro-ethane and acetone and fired in air at 1000° C for 1 h, while the aluminium discs were ground with no. 600 SiC abrasive paper and ultrasonically cleansed in acetone. An aluminium disc was placed between two ceramic cones and the sample inserted in a graphite die prior to soldering at 1000° C in an induction furnace evacuated to 0.05 Torr. After the sample had been at temperature for 10 min, power was switched off and it was allowed to cool.

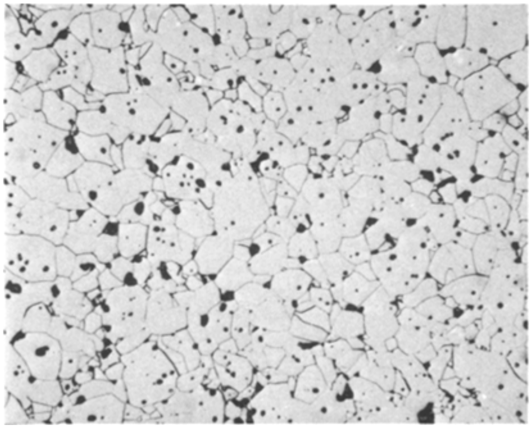

Figure 1 UL 995 alumina etched with H_3PO_4, × 100.

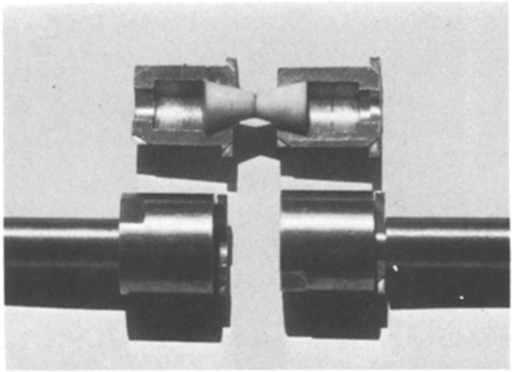

Figure 2 Tensile testing jig used to hold soldered ceramic samples.

The sample bond strengths were measured at temperatures ranging from 20 to 500° C using a special chamber which was attached to an Instron machine and evacuated to 10^{-4} Torr. The sample was heated to the desired temperature and equilibrated for 30 min before being pulled at a crosshead speed of $0.5\,mm\,min^{-1}$ using the holding jig shown in Fig. 2. To minimize the chance of ceramic fracture at the jig edge, a $20\,\mu m$ aluminium foil, which is not shown in the figure, was placed between the sample and the jig. The temperature was measured by a thermocouple placed near the sample which had been calibrated previously with reference to a second thermocouple located in a dummy sample.

Tests to provide comparative ceramic and metal strength data were conducted using bobbin-shaped samples with the same dimensions as a pair of bonded truncated cones. Before being placed in the Instron attachment, the ceramic bobbins were fired in air for 1 h at 1000° C and the aluminium bobbins were annealed for 1 h at 350° C.

3. Results

3.1. Alumina–aluminium

The thicknesses of the aluminium interlayers decreased during soldering from an initial 1 mm to 0.75 ± 0.05 mm or 0.1 mm to 0.04 ± 0.02 mm. Samples produced using both thick and thin interlayers had appreciable bond strengths at room temperature, 61 to $78\,MN\,m^{-2}$ for the thick inter-

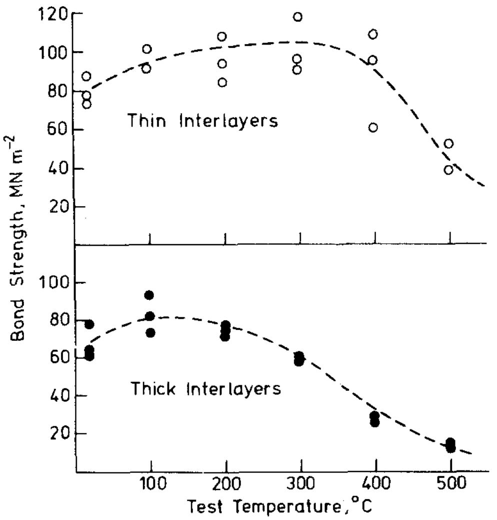

Figure 3 The effect of testing temperature on the fracture strength of alumina samples soldered with (a) thin and (b) thick aluminium interlayers.

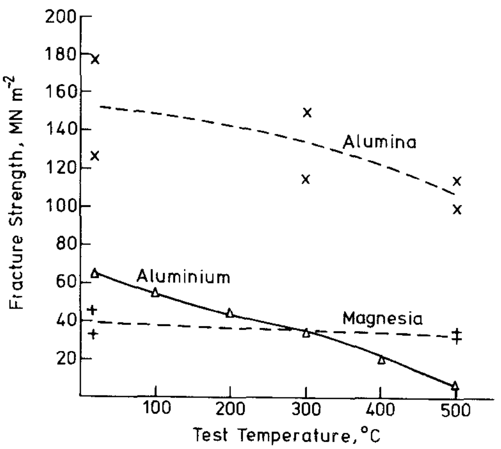

Figure 4 The effect of testing temperature on the ultimate tensile strength of aluminium bobbins and the fracture strengths of alumina and magnesia bobbins.

layer samples and 73 to 89 MN m^{-2} for the thin interlayer samples. Increasing the testing temperature initially caused the interfaces to become somewhat stronger as shown in Fig. 3. Although strength data are rather scattered, particularly for samples soldered with the thinner interlayers, Fig. 3 demonstrates that the interfaces remained strongly bonded at temperatures as high as 300° C for samples with thick, and 400° C for samples with thin interlayers. In contrast, the strengths of alumina and aluminium bobbins decreased steadily with temperature (Fig. 4).

Optical and scanning electron microscopy showed the fracture surfaces of soldered samples tested at low temperatures to contain many ceramic cleavage facets while those of samples tested at high temperatures displayed metal dimples (Fig. 5). Near-complete failure within the aluminium occurred at temperatures of 200 to 300° C for samples with thick interlayers and at 400 to 500° C for samples with thin interlayers (Fig. 6).

3.2. Magnesia–aluminium

The strengths of samples produced using 0.1 mm interlayers, which decreased in thickness to about 0.04 mm during the soldering process, were rather scattered but tended to decrease with the testing temperature (Fig. 7). The strengths of magnesia bobbin samples tested at 20 and 500° C were only slightly larger than magnesia–aluminium soldered samples (Fig. 4).

Molten aluminium penetrated porosity in the magnesia during soldering (Fig. 8), and reacted with the ceramic to form a product identified by X-ray diffraction as $MgO.Al_2O_3$ spinel.

Electron probe microanalysis of cross-sectioned

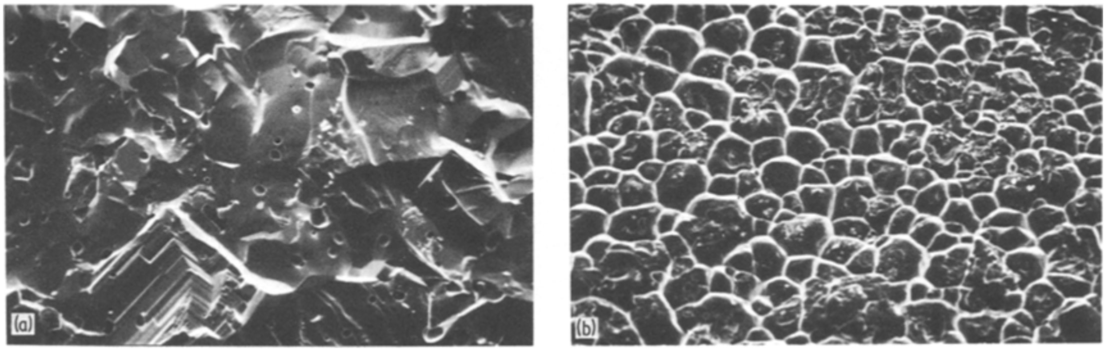

Figure 5 Scanning electron micrographs of the fracture surfaces produced at (a) room temperature and (b) 400° C of alumina samples soldered with thin aluminium interlayers, × 250.

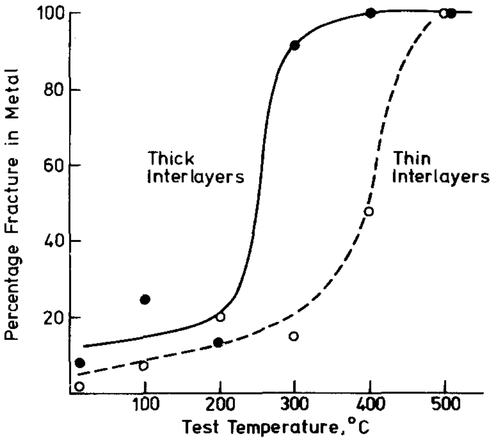

Figure 6 The effect of temperature on the location of the fracture surfaces of alumina samples soldered with thin and thick aluminium interlayers.

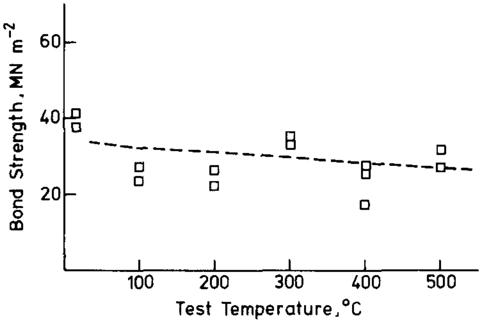

Figure 7 The effect of temperature on the fracture strengths of magnesia samples soldered with aluminium.

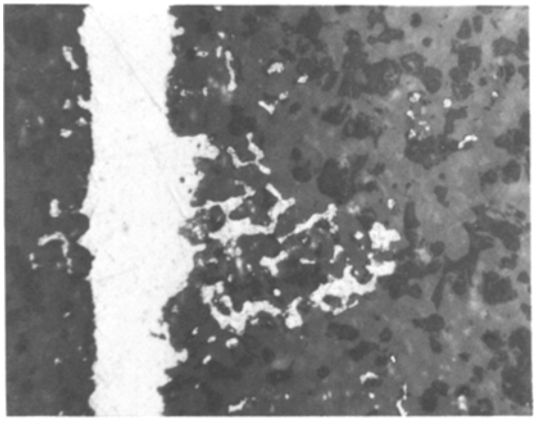

Figure 8 Cross-section of a soldered magnesia sample showing penetration of the white aluminium into interconnected porosity, × 270.

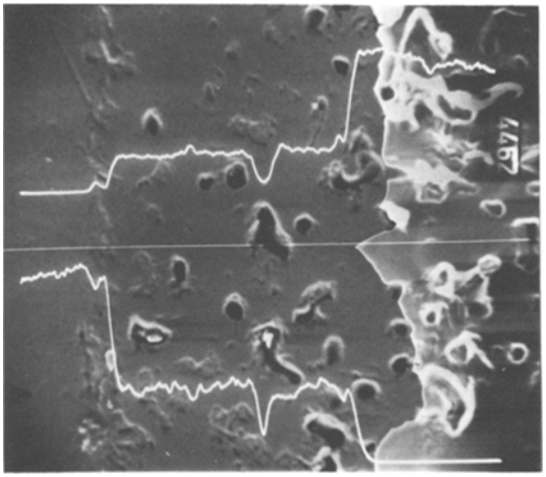

Figure 9 Electron probe microanalyser composition trace across the interface of a soldered magnesia sample. Upper trace, Mg; lower trace, Al.

samples confirmed the presence of a zone containing aluminium and magnesium, extending to a depth of 60 μm in some areas, as illustrated in Fig. 9, but many fracture surfaces displayed in magnesia and metallic features as well as the spinel. The metallic features tended to be more evident on the fracture surfaces of samples tested at high temperatures but there was no clear correlation between sample strengths and the proportion of their fracture surfaces occupied by the spinel phase.

4. Discussion

The ultimate tensile strength of aluminium decreases rapidly with temperature (Fig. 4) and, therefore, the most striking observation presented in this paper is that aluminium soldered alumina samples at first increased in strength with rising temperature and retained at least room temperature strengths until 300 to 400° C, 60 to 70% of the solder melting point expressed in degrees Kelvin. The strengths of the magnesia samples fell with increases in the testing temperature, but their strength at 500° C was only 25% lower than that at room temperature, in contrast to the 90% decrease in the ultimate tensile strength of aluminium.

The strength data presented in this paper are thought to be unique in that we know of no other information of this type for these systems. The only other high temperature interfacial strength data available relate to more complex systems, but they display some features similar to those observed in this work. Thus Thornton [10] found strength maxima during the elevated temperature testing of samples of silicate glass joined to a 17% chromium–7% nickel steel, but not AISI 303 which contains a small amount of manganese and silicon. Similarly, Witt [11] reported strength maxima for samples of ceramic glazes joined to various steels. More relevantly to the present work, Kogan and Markowskii [12] found that elevated temperature strength tests of samples of metallized technical grade alumina soldered with copper or silver not only revealed strength maxima but also retention of near room-temperature strengths, 50 MN m^{-2}, up to 600° C, about 60 to 70% of the solder melting points expressed in degrees Kelvin.

By definition, a body will fracture at the part least able to resist the applied stress. However, comparison of the strength data in Fig. 3, 4 and 7 with the fracture location data in Fig. 6 and the scanning electron micrographs in Fig. 5 suggests that failure did not always occur within the material that had the lowest bulk strength. This discrepancy, however, may be an artefact of the testing technique since the aluminium data plotted in Fig. 4 are ultimate tensile strengths, that is, the failure load of a bobbin divided by the original cross-sectional area of its waist. The aluminium in the bobbin samples was free to deform but that present in the solder layers between the alumina or magnesia truncated cones was not. Thus the data for aluminium in Fig. 4 are not as relevant as true failure strengths, the load divided by the actual bobbin waist area of the moment of failure. This can be twice as large as the ultimate tensile strength for aluminium and hence comparable to the failure strengths of soldered samples breaking within the aluminium interlayer or at the ceramic–solder interface.

Although the strengths of the soldered alumina samples were less than the true failure strengths of the alumina bobbins, fracture occurred within the ceramic during tests at or near room temperature. The fragility of the soldered ceramic at these temperatures could be due to the presence of stress concentrators such as pores or second-phase inclusions or to the effects of residual stresses. The first possibility is considered unlikely because it would also have influenced the strengths of the alumina bobbins. On the other hand, stresses would be generated within the solder and the ceramic and at their interface as the samples cooled after fabrication due to the gross mismatch in the thermal expansion coefficients of alumina and aluminium, about 8 and $24 \times 10^{-6}\,°C^{-1}$, respectively. Such residual stresses within the ceramic will be compressive and parallel to the interface immediately adjacent to the solder but will be tensile in more remote locations. During strength testing, the stress pattern will be made even more complex by the imposition of an external force normal to the interface. However, regardless of their nature and distribution, the significant increase in the strengths of samples produced by testing at 100° C rather than room temperature suggests that they are easily relieved. Thus the strength peaks observed with alumina–aluminium samples can be attributed to a combination of stress relaxation in the ceramic and softening of the metal. The relaxation of residual stresses is achieved at low temperature and the effect of further heating will depend upon the ability of the softening aluminium to deform. The constraints exerted by attachment of the aluminium to relatively rigid ceramic bodies will be less with thick than thin interlayers. Hence, it is to be expected that the temperature at which the soldered samples weakened and failure occurred predominantly within the aluminium was lower for those with the thicker interlayers.

This relatively simple dependence of sample strengths on physical interactions suggested by the alumina–aluminium data provides an inadequate description of the results obtained for magnesia–aluminium samples. Magnesia was chosen as a second ceramic to provide a material with a different, more marked, chemical reactivity with aluminium. Such reactions did occur on a fairly extensive scale with Fig. 9 suggesting an interdiffusion coefficient of up to $6 \times 10^{-12}\,m^2\,sec^{-1}$ at 1000° C in reasonable accord with the $3 \times 10^{-12}\,m^2\,sec^{-1}$ at 1027° C that can be derived from the work of McEvoy *et al.* [9]. It can be argued that the production of the $MgO.Al_2O_3$ spinel at the interface should decrease the strength of the soldered magnesia sample since it has a lower crushing strength than comparable magnesia [13] and it reduces the coefficient of thermal expansion of the ceramic in immediate contact with the aluminium from about 13.5 to $9 \times 10^{-6}\,°C^{-1}$. However, it is probable that the most significant difference between the alumina and magnesia cones used in this work was not their reactivity, but their porosity.

The effect of porosity on the behaviour of alumina–niobium samples has been evaluated by

Elssner and Pabst [14] who found the location of fracture planes to depend on the ceramic quality; samples produced using fine-grained high-density alumina fractured at the ceramic–metal interface while others produced using porous alumina fractured within the ceramic near the interface. Similarly, in the present work with porous magnesia, fracture occured always at least partially within the ceramic. This is thought to be due to the low strength of the magnesia, only 40 $\mathrm{MN\,m^{-2}}$ as compared to about 130 $\mathrm{MN\,m^{-2}}$ for fully dense material [15], and the complex stresses created by thermal contraction of the aluminium that had penetrated into pores within the magnesia (Fig. 8).

The present work, therefore, suggests that the elevated temperature strengths of soldered ceramic samples can be greatly affected by constraint and structure factors. It is not possible to predict component fracture characteristics only on the basis of interface behaviour studies, the behaviour of carefully designed components potentially being better than would be suggested by such studies. The same factors influence both room and elevated temperature bond strengths and this work has shown, at least for alumina–aluminium and magnesia–aluminium samples, that room-temperature strengths provide a reasonable guide to behaviour at higher temperatures. For example, stressed alumina vacuum components strongly soldered with thin aluminium interlayers should be able to ensure UHV baking temperatures, 250 to 400° C, without weakening significantly.

Acknowledgements

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