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An investigation into microwave bonding mechanisms via a study of silicon carbide and zirconia

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It is known that the glassy grain-boundary phase present in low-purity aluminas has two primary functions during direct microwave bonding. Firstly, it increases the dielectric loss of the host ceramic, allowing heating to occur; secondly, the bonding mechanism itself has been found to be based on viscous flow of the glassy grain-boundary phase. However, some evidence has also been found for the bonding of individual grains where they come into direct contact across the join line. To investigate the role of grain-boundary phases further, the microwave bonding of two different grades of silicon carbide and one grain of zirconia has been studied. A single-mode resonant cavity operating at 2450 MHz was used for both studies. The temperature and axial pressure were varied and the bonding time was kept to a minimum. Analysis of the resultant bonds indicated that both reaction-bonded silicon carbide and partially stabilized zirconia could be successfully joined using microwave energy with bonding times typically 10 min or less. For reaction-bonded silicon carbide ceramics, the silicon grain-boundary phase softened at the bonding temperature, allowing the butting faces to be "glued" together. Unlike the glassy grain-boundary phase for alumina ceramics, the silicon phase did not allow grain motion but always formed a discrete and continuous layer at the interface, even under optimum joining conditions. The work with zirconia confirmed that it is possible to join ceramics without the presence of a substantial grain-boundary phase. The mechanism is thought to be either solid-state diffusion and/or grain-boundary sliding. *© 1998 Kluwer Academic Publishers*

1. Introduction

A previous study [1] into the effect of composition on the direct microwave bonding of alumina ceramics revealed that, the lower the purity of the alumina, the more successful is the microwave bonding process. This was attributed to the two roles played by the glassy grain-boundary phase present in such ceramics. Firstly, a major function was to increase the dielectric loss of the host ceramic, allowing heating to occur. Secondly, the bonding mechanism itself was found to be based on viscous flow of the glassy grain-boundary phase. By softening, the glassy phase allowed the alumina grains to move under the imposed load. Provided that sufficient migration across the bond line of both glassy phase and grains occurred, then a fully homogeneous microstructure could be obtained in which no obvious glassy line existed at the boundary, making it indistinguishable from the bulk material. This led to mechanical bond strengths as high or higher than that of the parent material with the bonded samples never breaking at the bond line during four-point bend tests when optimum conditions were used. In addition, some evidence was also found for the bonding of individual grains where they came into direct contact across the bond line. This was believed to imply that direct joining of ceramics without a substantial grain-boundary phase should be possible if sufficient energy could be deposited to allow the ceramic to be heated. Additional work was clearly required if the joining mechanisms involved during microwave bonding were to be illuminated further. To this end, two grade of silicon carbide, namely, reaction-bonded silicon carbide (RBSC) and hot-pressed silicon carbide (HPSC), and one grade of zirconia, namely, zirconia partially stabilized with 8 wt % Y_2O_3 (Y-PSZ), have been investigated.

Rapid high-quality bonds have been obtained between silicon carbide ceramics by a number of

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researchers [2–6]. When RBSC samples were directly microwave joined [2, 5] a Si-rich band was always observed at the bond line. This is in contrast with results obtained for low-purity alumina ceramics where grain movement allowed a homogeneous bond line to be obtained [1]. When aluminium was used as an interlayer bonds as strong as the base ceramic itself resulted [4]. RBSC has also been bonded to sintered SiC (SSC) [2, 3]. The results suggested that, although substantial cracking had occurred in the interfacial region, Si from the RBSC had wetted and bonded to the SSC. Although high-strength direct microwave bonds (400 MPa) have been produced between two SSC samples by utilizing conditions of 2050 °C and 8 MPa [5], indirect approaches using interlayers have resulted in bonds being produced under substantially less demanding conditions. Dense and homogeneous interfacial regions could be achieved when Si interlayers were used under conditions of 1450 °C and 2–5 MPa with 5–10 min at temperature [6]. The width of the bond was shown to be dependent on the method employed for applying the interfacial material. A plasma-sprayed Si layer on one of the SiC specimens produced a bond which was less than 5 μm thick. Alternative approaches using loose Si powder or Si slurries in watch oil resulted in bond widths of 50 μm and 10 μm, respectively. Attempts at using *in-situ* combustion synthesis of SiC/TiC interlayers were less successful owing to oxidation of the metals prior to reaction and the formation of a porous reaction product [6].

No reports of previous attempts to microwave bond zirconia ceramics have been found in the literature. This is believed to be due to the ceramics high susceptibility to thermal shock, making control of heating rates critical.

2. Experimental procedure

Two grades of silicon carbide and one grade of zirconia were investigated; details of their physical characteristics are listed in Table I. The HPSC samples were square-sectioned rods whilst the RBSC samples were small discs and the Y-PSZ samples existed in two forms, as rods of circular cross-section and circular thin-walled tubes. The heating system, which consisted of a TE_{102} single-mode microwave applicator powered by a 300 W, 2.45 GHz magnetron, has been described elsewhere [1] together with the general experimental arrangements. Because of the size of the RBSC samples, high-purity (99.5%) alumina push rods were used to hold the samples in place in the centre of the microwave applicator as shown in Fig. 1. In all cases the butting faces of the samples were diamond ground prior to the bonding experiments. A matrix of experiments was performed for each ceramic in which the bonding temperature and axial pressure were varied. For the SiC, the conditions were in the ranges 1190–1375 °C and 0.5–2.0 MPa whilst for the Y-PSZ they were in the ranges 1230–1560 °C and 2.7–3.2 MPa. Heating and cooling rates were maintained at the fastest levels found to be acceptable. Details of the experimental conditions may be found in Table II and Table III.

After joining, the rods were sectioned and polished using normal preparation procedures. A scanning electron microscope fitted with an energy-dispersive X-ray detector was used to obtain secondary-electron (SE), back-scattered electron (BSE) and elemental images. For the zirconia samples, when very successful

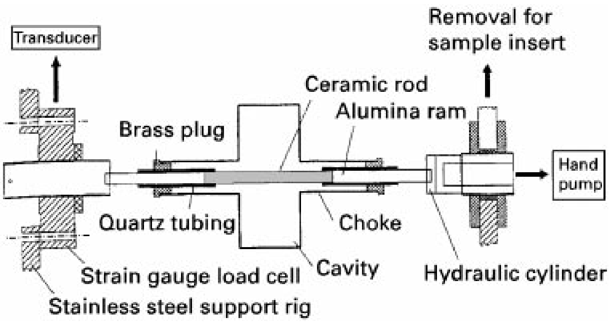

Figure 1 Schematic diagram of the sample support and loading system.

TABLE II Microwave joining parameters for silicon carbide

Parameter (units)	Value for the following materials	
	HPSC–HPSC	RBSC–RBSC
Joining or maximum temperature (°C)	1310	1190–1375
Maximum power used (W)	600	300
Axial pressure (MPa)	0.5	0.5–2.0
Typical heating rate (°C s^{-1})	0.6	2.0
Typical cooling rate (°C s^{-1})	4.6	3.0
Holding time (min)	–	Up to 2
Typical total processing time (min)	30	15

TABLE I Physical properties of materials investigated

Material	Density (g cm^{-3})	Grain size (μm)	Sample dimensions	Supplier and grade
HPSC	3.15	4	Square-sectioned rods: 5 mm × 5 mm × 50 mm	Cercom, USA; Type N
RBSC	3.10	10	Circular discs: diameter, 15 mm; length, 9 mm	Tenmat, UK; Refel F
Y-PSZ	5.98	0.4	Circular-sectioned rods: diameter, 7 mm; length, 40 mm Tube of 1.8 mm wall thickness: diameter, 6.4 mm; length, 45 mm	Viking Chemicals, Denmark

TABLE III Microwave joining parameters for Y-PSZ

Parameter (units)	Value for the following materials	
	Y-PSZ–Y-PSZ rods	Y-PSZ–Y-PSZ tubes
Joining or maximum temperature (°C)	1400–1565	1230
Maximum power used (W)	370	300
Axial pressure (MPa)	2.7 ± 2.0	3.2 ± 2.0
Typical heating rate (°C s^{-1})	0.5 at < 900 °C; 3.7 at > 1000 °C	2.8 at < 800 °C; 0.5 at > 1000 °C
Typical cooling rate (°C s^{-1})	3.5	4.0
Holding time (min)	10	10
Typical total processing time (min)	30	30

bonds were produced, the only method of determining the location of the bond was via an approximately 100 μm misalignment of the rods and tubes which showed up as steps at either end of the bond line. It was then possible to track across the bond line with the scanning electron microscope. In addition to electron microscopy, microhardness measurements were made along a series of lines running perpendicular to the join and spanning a distance of 3 mm either side of the join. Eight randomly distributed indentations were also measured across the section of each untreated ceramic. The dimensions of each sample were also measured both before and after bonding to allow the extent of deformation caused by the thermomechanical cycle to be determined. The measurements were then converted into percentages of the area and longitudinal strain.

3. Results and discussion

3.1. Silicon carbide

The processing parameters achieved are outlined in Table II. Despite all attempts, including the introduction of an adjustable iris design, the HPSC could not be heated to a temperature in excess of about 1300 °C. This did not permit the ceramics to be bonded; temperatures in the range 1950–2100 °C are typically required for conventional diffusion bonding [7]. The applicator did become very hot, however. On the basis of these experimental observations and the results of dielectric measurements [8], it is believed that, as the temperature of the HPSC was raised, the electrical conductivity of the material increased, resulting in a decrease in the penetration depth of the microwaves. Thus the ceramic began to reflect the microwaves rather than to absorb them. This resulted in a steady-state condition being reached, at 1310 °C, when increases in power level did not result in an increased temperature. This problem was overcome in other studies by using either interfacial materials which could be microwave heated to the necessary temperature [3, 6] or a hybrid heating system [5]. Since this work was intended to be a study of microwave bonding mechanisms, these approaches were not adopted.

Owing to the presence of the silicon in its structure, the RBSC ceramic could be joined under all conditions examined, however, the quality of bond varied significantly. At low temperatures and pressures, e.g. 1190 °C and 0.5 MPa, a general lack of contact across the width of the sample was found. The butting surfaces were separated by a 10 –20 μm gap which was only partially filled by free silicon (Fig. 2). As the temperature and pressure were increased, the width of the interfacial region reduced and became increasingly filled with silicon. The gap was reduced to 5–10 μm under conditions of 1235 °C and 1 MPa, whilst by 1350 °C and 1.5 MPa it had become 3–5 μm and was completely filled with silicon (Fig. 3). Increasing the processing conditions to 1375 °C and 2.0 MPa produced a bond which suggested that they were excessive. A region of enhanced porosity spanning the width of the sample was observed and free silicon was found on the surfaces of the rods, indicating that it had flowed out of the bond region. An examination of the one sample which fractured along the bond line during machining clearly demonstrated the formation of an inverse temperature profile during the microwave heating process, as shown in Fig. 4. The highest concentration of free silicon at the centre of the sample indicated that this was the hottest region. The amount

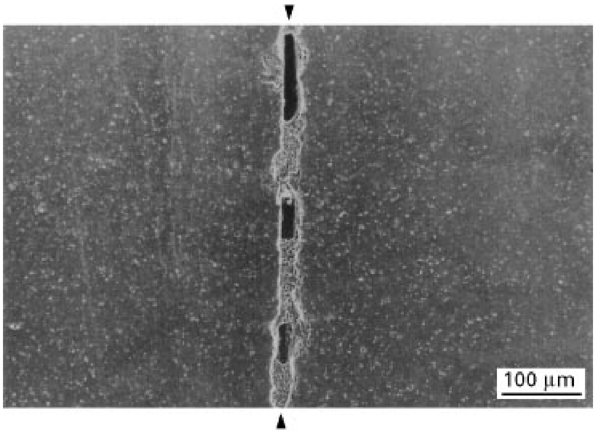

Figure 2 Scanning electron micrograph of RBSC joined at 1190 °C and 0.5 MPa. A sparse distribution of free Si fills the gap which separates the butting surfaces.

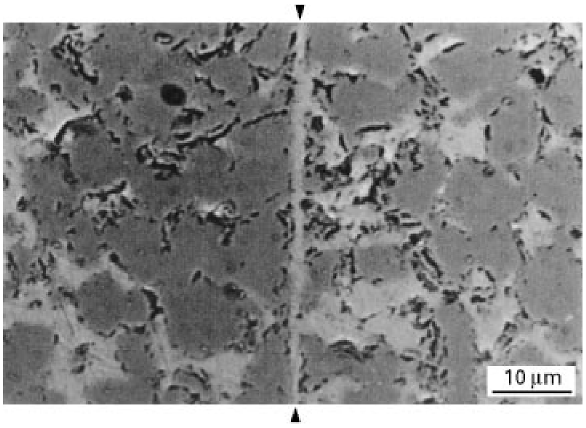

Figure 3 Scanning electron micrograph of RBSC joined at 1350 °C and 1.5 MPa. A layer of free Si completely fills the gap separating the butting faces.

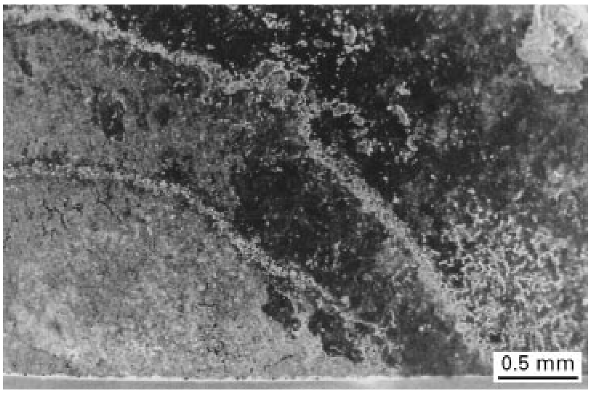

Figure 4 The fracture surface of an RBSC-RBSC sample joined at 1250 °C and 1 MPa in 2 min. The distribution of free Si on the fracture surface indicates the presence of an inverse temperature profile.

of silicon transported to the butting surfaces decreased on moving towards the edge of the sample, indicating a lower temperature.

The BSE imaging mode on the scanning electron microscope demonstrated the lack of movement of the α-SiC grains under all conditions. The butting surfaces remained defined by the straightness of the SiC–Si–SiC interface. The amount of Si present in the form of a layer at the bond was comparable with that observed elsewhere within the material and was not therefore considered to be especially deleterious to the bond. This argument was also proposed by Ahmad *et al.* [3]. In addition, the grain size did not appear to have been affected by the microwave heating process under any conditions, nor did the samples suffer from any deformation.

These observations indicate that the silicon grain-boundary phase is softening and providing a "glueing" effect. The optimum temperature is that which causes the silicon phase to soften sufficiently to fill the void between the butting faces, but not so extensively as to flow out of the bond region. A schematic illustration of the movement of free silicon is shown in Fig. 5. Within reason, the higher the pressure, the narrower is the interfacial region. This is expected to lead to higher strengths in the resulting bond. No evidence for local rearrangement of the SiC grains across the bond interface was found in any of the samples examined and hence it was not possible to produce a join in which a homogeneous bond line was achieved.

From a practical standpoint, the controlled microwave heating of the RBSC was not easy. The material displayed a rapid increase in complex permittivity at about 1150 °C which resulted in a large shift in the tuning of the cavity which had to be manually brought back under control. A different design of applicator might have alleviated this problem [9]. Nevertheless, the necessary degree of control could be achieved allowing RBSC samples to be directly joined with just 2 min at temperature and a total processing time of about 30 min. This compares very favourably with the time of 1 h required by Iseki *et al.* [10], using a conventional bonding technique. Ahmad and Black [2] have suggested that enhanced diffusion of the silicon at the grain boundaries of the silicon carbide, and the enhanced mass transport into the bond region, accounted for the faster bonding of the pieces to be joined. No evidence to refute this suggestion has been found in the present work. However, the results of a comparison of the processing times needed to produce similar bonds using microwave and conventional heating in a previous study using alumina [1], suggested that the efficiency of heat transfer into the sample played a major role. Mircowave energy causes very rapid volumetric heating of the bulk material, whereas for conventional processing the heat needs to be conducted into the sample. As the size of sample increases, this effect will become more noticeable, provided that the depth of penetration for microwaves into the ceramic is not exceeded.

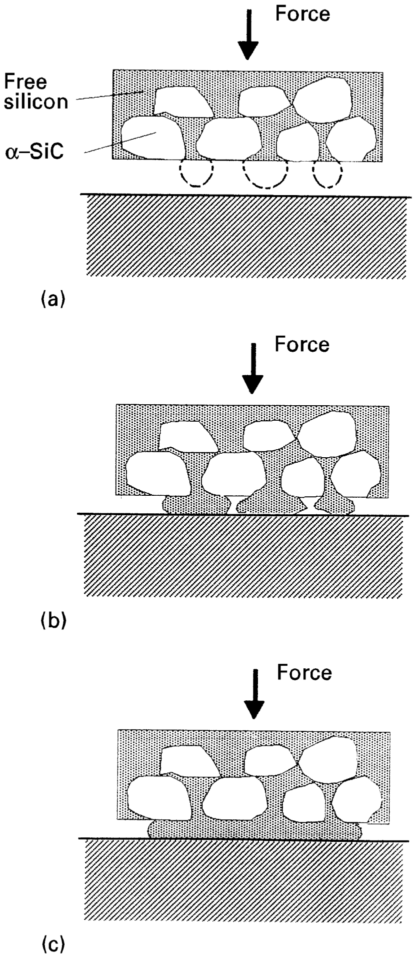

Figure 5 A schematic illustration of the movement of free silicon onto the butting surfaces. (a) Free silicon bleeds out onto the surface of the RBSC. (b) Silicon spreads across the surface of the RBSC, assisted by an applied axial force. (c) The individual globules coalesce to form a continuous interlayer.

3.2. Zirconia

The processing parameters achieved are outlined in Table III. Zirconia proved to be a difficult material to join owing to the large thermal expansion associated with zirconia ceramics ($\alpha = 10.6 \times 10^{-6}\,{}^{\circ}\mathrm{C}^{-1}$). This made control of the applied pressure difficult and it should be noted that the values quoted had a large

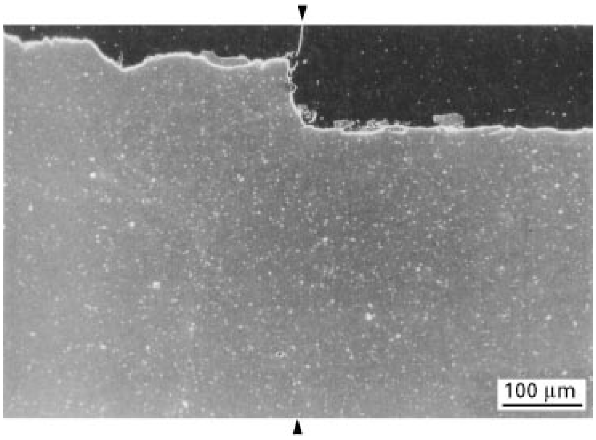

Figure 6 Y-PSZ rods joined at 1400 °C and 2.7 MPa in 10 min. The misalignment at the edges is the only method of identifying the location of the bond using the SE imaging mode.

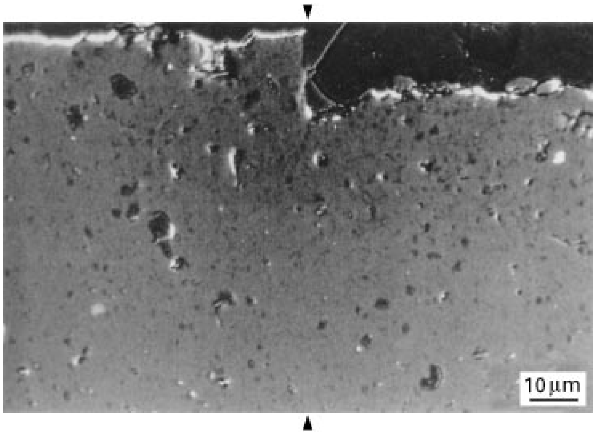

Figure 8 BSE image of Y-PSZ rods joined at 1565 °C and 2.7 MPa in 10 min. The bond cannot be detected except via the misalignment at the edges of the sample.

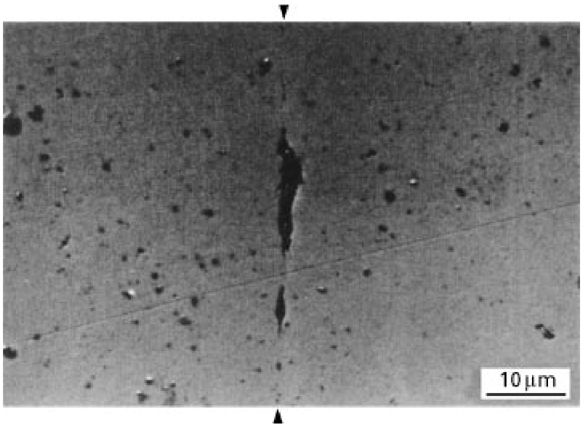

Figure 7 BSE image of the sample shown in Fig. 6. Note the region of elongated porosity at the bond line.

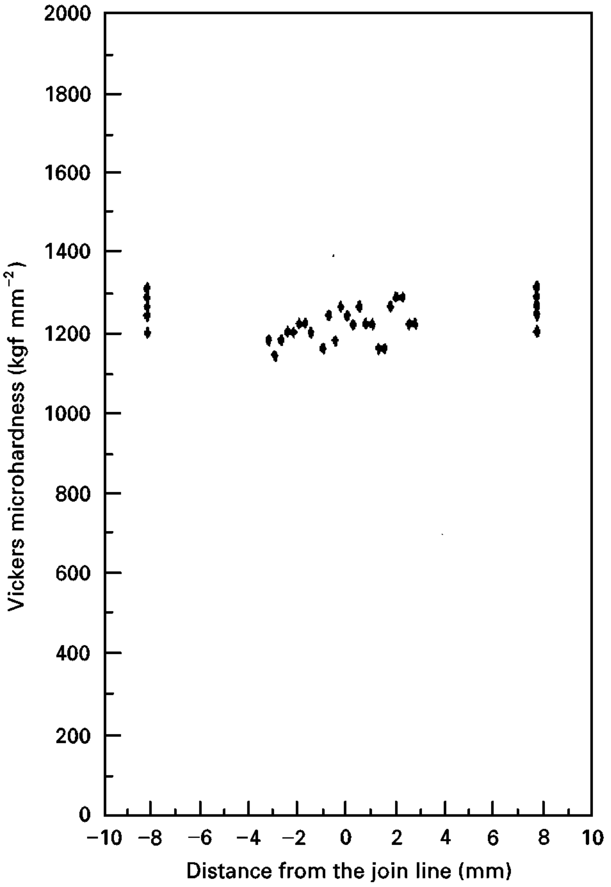

Figure 9 Microhardness measurements taken across the bond line of the zirconia rods joined at 1565 °C and 2.7 MPa in 10 min. The values shown at ± 8 mm represent the bulk untreated ceramic.

degree of scatter when the temperature was below 1000 °C. Above this temperature the load applied began to fall, reaching zero by 1500 °C. The thermal expansion also yielded severe problems with thermal shock, limiting heating rates if cracking of the samples was to be avoided. Nevertheless, it proved possible to produce bonds which were undetectable by scanning electron microscopy, except via the sample misalignment mentioned earlier.

Conditions of 1400 °C and 2.7 MPa axial pressure resulted in a bond line which was undetectable in SE imaging mode for the zirconia rods (Fig. 6). However, use of BSE imaging revealed the presence of isolated porosity across the width of the sample (Fig. 7). Between the regions of porosity a fully homogeneous microstructure had replaced the two contacting surfaces. Increasing the bonding temperature to 1565 °C produced a bond which was homogeneous across the whole width of the sample under both SE and BSE imaging modes (Fig. 8). The microstructure appeared unaffected by the microwave heating process and the microhardness measurements taken across the bond line were comparable with those made on the bulk untreated material (Fig. 9). However, the very fine grain size made it difficult to determine whether grain rearrangement across the bond line had occurred (Fig. 10). Since there is no substantial grain-boundary phase present which could have softened at the bonding temperatures, joining is thought to have occurred as a result of solid-state diffusion and/or, owing to the very fine grain size, a degree of grain-boundary sliding. No evidence has been found in the literature to suggest which mechanism is most likely.

Following the success of joining the zirconia rods, joining tubes was attempted. Although it was

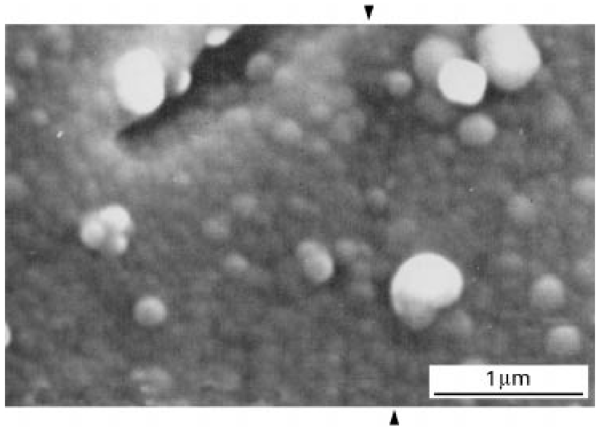

Figure 10 SE image of the zirconia rods joined at 1400 °C and 2.7 MPa in 10 min. The sample has been etched to show the microstructural continuity at the join.

possible to bond them, they were very susceptible to thermal shock and the resultant samples were very fragile.

An interesting feature observed during the microwave heating of the zirconia rods was a noticeable increase in waveguide wavelength on reaching 900–1000 °C. This was ascribed to the rapid change in dielectric properties of zirconia at about this temperature [11] which will have caused a substantial change in the electric field distribution. The consequences of this effect was a need for a rapid change in the tuning conditions at a time when thermal expansion was necessitating great care in the control of the axial pressure. No such phenomenon was observed during heating of the zirconia tubes, presumably owing to the much lower quantity of ceramic present which will have had a smaller effect on the *E* field.

4. Conclusions

Both RBSC and Y-PSZ could be joined, without the use of interlayers, using 2.45 GHz microwave energy and a single-mode applicator with bonding times of 10 min or less and total processing times of about 30 min. HPSC could not be heated to temperatures of 1310 °C or above using the current system and this did not permit bonding to occur. It is believed that at high temperatures the ceramic became highly reflective to microwave radiation as a result of increases in its electrical conductivity.

Because of the presence of the silicon in the structure, the RBSC could be successfully bonded under conditions of about 1350 °C and 1.5 MPa applied axial pressure. At lower temperatures and pressures a general lack of contact across the width of the sample was found, whilst under excessive conditions a region of enhanced porosity spanning the width of the sample was observed and free silicon was found on the surfaces of the rods, indicating that it had flowed out of the bond region. The operational bonding mechanism for the RBSC thus involved the softening of the silicon grain-boundary phase, allowing the butting faces to be "glued" together. However, unlike the glassy grain-boundary phase for alumina ceramics [2], the silicon phase did not allow movement of the α-SiC grains across the bond line but always formed a discrete and continuous layer at the interface even under optimum joining conditions. This prevented the bond from being homogeneous. The reasons for these two types of behaviour are not certain, but are probably due to the different viscosities and wetting characteristics of the two phases. The size of the grains is not thought to be relevant since both the alumina [2] and the RBSC ceramics had similar grain sizes. Under the optimum conditions, the amount of silicon present in the form of a layer at the bond line was comparable with that observed at the grain boundaries and was not therefore considered to be especially deleterious to the bond.

The work with Y-PSZ, a ceramic which absorbs microwaves readily but which does not possess a substantial grain-boundary phase which will soften at the bonding temperatures, has confirmed for the first time that it is possible to bond such ceramics directly using microwave energy. Samples were successfully joined at 1565 °C using an axial pressure of 2.7 MPa. Solid-state diffusion and/or grain-boundary sliding are believed to have been the operational mechanisms. However, the large thermal expansion coefficient of this ceramic, and consequent low thermal shock resistance, made this ceramic difficult to join.

Mircowave bonding is thus most easily affected when (i) the ceramic is sufficiently absorbent of microwave energy to heat to the required temperatures and (ii) grain-boundary phases are present in the base material which will soften at the joining temperature, allowing the butting faces to be "glued" together. For materials without one or other characteristic, appropriate interlayers can act as a suitable substitute. However, the work with zirconia has shown for the first time that it is possible to obtain direct joins without such grain-boundary phases or interlayers.

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