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Joining of sialon ceramics by a stainless steel interlayer

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Abstract

In direct diffusion bonding of sialon to stainless steel, thermal residual stresses arise due to the difference in coefficient of thermal expansion of the two materials. These stresses frequently lead to failure of the bond. This behaviour is further influenced by the formation of interfacial reaction layers between ceramic and metal and the problem is essentially one of asymmetry of stresses in the interface between dissimilar materials. The present study demonstrates that a thin layer of austenitic stainless steel can be used as an interlayer to join two sialon components. In such a case the distribution of residual stresses is symmetrical across the composite join and provided that the thickness of the steel layer is less than a critical value, then fracture on cooling from joining temperature does not occur. The development of this process is described and a finite-element model has been used to predict the properties of the interfacial reaction layer between steel and ceramic which are consistent with the experimental observations. © 2001 Elsevier Science Ltd. All rights reserved.

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1. Introduction

Problems associated with joining of silicon nitride ceramics have been well documented and reports have been published on techniques involving direct diffusion bonding,[1,2] brazing[3] and the use of ductile metallic interlayers[4,5] among others. The latter method is particularly attractive where high-temperature applications are envisaged provided that the metallic interlayer is thermally stable and does not form brittle intermetallic phases which may result in failure of the joint either during fabrication or in service. In all such applications however, there are thermal stresses which arise due to differences of coefficient of thermal expansion between the materials and which must be accommodated if successful bonding is to be achieved. It is the magnitude and orientation of such stresses in fabrication and use which dictate the constitution of the joining materials. The present work was the result of such a specification where a sialon-sialon bond was required for use at temperatures above those at which a brazed joint loses mechanical strength and to operate in an air environment. Previous work[2] had shown that stainless steel will bond effectively to sialon by direct diffusion bonding and so it was decided to study the behaviour of an austenitic stainless steel as a ductile interlayer in joining two sialon components. The particular interest of the study was the distribution of thermal stresses which arise after cooling from bonding temperature during fabrication of the joint and the influence of interfacial reaction layers on such stresses.

There has been relatively little experimental work reported on direct diffusion bonding of silicon nitride or sialons to stainless steel. The underlying problems as indicated above relate to reaction between the constituents of the steel and ceramic to form brittle interlayers and to the residual thermal stresses which arise due to the difference in coefficient of thermal expansion between the ceramic and metal. Krugers and Ouden[6] studied the bonding of AISI 316 to silicon nitride (reaction bonded and hot pressed) and showed that good bonds could be produced although iron silicides were formed in the interface. Suganuma et al.[4] followed the approach of earlier workers[7] in using soft compatible interlayers of aluminium metal to avoid the interfacial reaction problems and to accommodate the thermal

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stresses and Brito et al.[8] used a similar approach with nickel as an interlayer in joining silicon nitride to other metals. The approach to assessing residual thermal stresses in ceramic-metal joints, including those with interlayers has been studied by Iancu and Muntz[9] who give a simple analytical model which provides a useful comparison with finite element analysis methods as used in this study.

2. Experimental

The sialon used in these experiments is Cookson Syalon 201 and is a yttria-densified sialon with a z-value of 0.75. The microstructure is one of fine, acicular β' grains with an amorphous grain-boundary phase rich in yttrium. The stainless steel is of AISI 316L grade (17.19% Cr, 11.54% Ni, 2.06% Mo, 0.02% C). Both materials were used in the form of 10 mm diameter rods cut to appropriate thickness for the joining experiments. The surfaces to be joined were prepared by polishing to a 6 μm diamond finish and rinsed in an ultrasonic bath immediately before the experiment. The joining process was carried out in an induction-heated die in which the samples were embedded in boron nitride powder to avoid reaction with the graphite components of the die; the gas environment consists of carbon monoxide and nitrogen from autogenous reaction in the die. The joining temperature was 1250°C and was measured by a disappearing-filament pyrometer sighted on the external surface of the die and calibrated against the melting points of pure metals. During the experiment the samples were held in a small compressive stress of about 20 MPa and the heating and cooling rates were 20°C/min. The samples after joining were sectioned and examined by standard optical and electron metallographic techniques.

3. Results and discussion

Previous work[2,10] has shown the nature of the reactions which take place in the interface between sialon and AISI 316 stainless steel and so only a brief description of the essential features is given here. There is an exchange reaction between the sialon (which may be considered as an impure silicon nitride) and the transition metal components of the stainless steel[1,2,6,10] which results in the formation of metal silicides and dissolution of nitrogen in the steel. The reactions to form silicides are the result of dissociation of the ceramic and cause the formation of a eutectic liquid phase in the metal close to the interface which enhances the bonding process and results in penetration of the metallic phase into the ceramic structure adjacent to the interface. An example is shown in Fig. 1. The interface is irregular but coherent and the bond is strong. The second (dark) phase in the interface which is evident in the figure is found by EDX analysis in the SEM to be rich in yttrium, silicon and aluminium from the sialon and its intergranular phase and which have segregated to the metallic liquid during the bonding process. The observed phase has then precipitated on cooling. There is however a greater extent of diffusion of silicon into the metallic layer than is evident from the figure and is detected by EDX analysis but also by change in the hardness of the steel layer. This is seen in Fig. 2 where there is a gradient of microhardness from the interface into the steel layer. It is clear also that the steel is strengthened at the interface relative to the bulk steel and therefore has a gradation of properties from the ceramic through the steel layer. In principle, this is a desirable situation as the hardness (strength) gradient helps accommodate the stresses generated on cooling the bond. The situation in bonded samples, where a thin layer of steel is used as a ductile interlayer, is that the effect is symmetrical with silicon diffused into the steel interlayer from the ceramic components on both sides [Fig. 2(a)].

In the ceramic side of the couple there has been considerable penetration of metal while the steel shows extensive precipitation of prismatic crystals adjacent to the interface and which were not present in the original steel (Fig. 1). These two effects are due to the chemical reactions which take place at the interface between steel and sialon. The effect is that silicon nitride (sialon) decomposes to give silicon dissolved in the steel and releases nitrogen, some of which also dissolves in the steel. The solidus temperature of the diffusion-modified steel is below the joining temperature and so a liquid metal phase penetrates the ceramic as the nitride is attacked. Similarly, on the steel side of the interface and immediately adjacent to it, there is dissolution of silicon and nitrogen in the metal which on cooling reprecipitate as silicon nitride in the metal matrix. These are in the form of the prismatic crystals observed in the metal microstructure (Fig. 1) and which have been identified by EDX analysis in the SEM and by X-ray diffraction as α-silicon nitride.

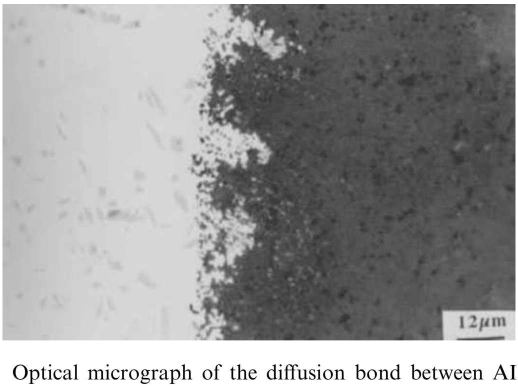

Fig. 1. Optical micrograph of the diffusion bond between AISI 316 stainless steel and sialon ceramic showing the penetration of metal into the ceramic.

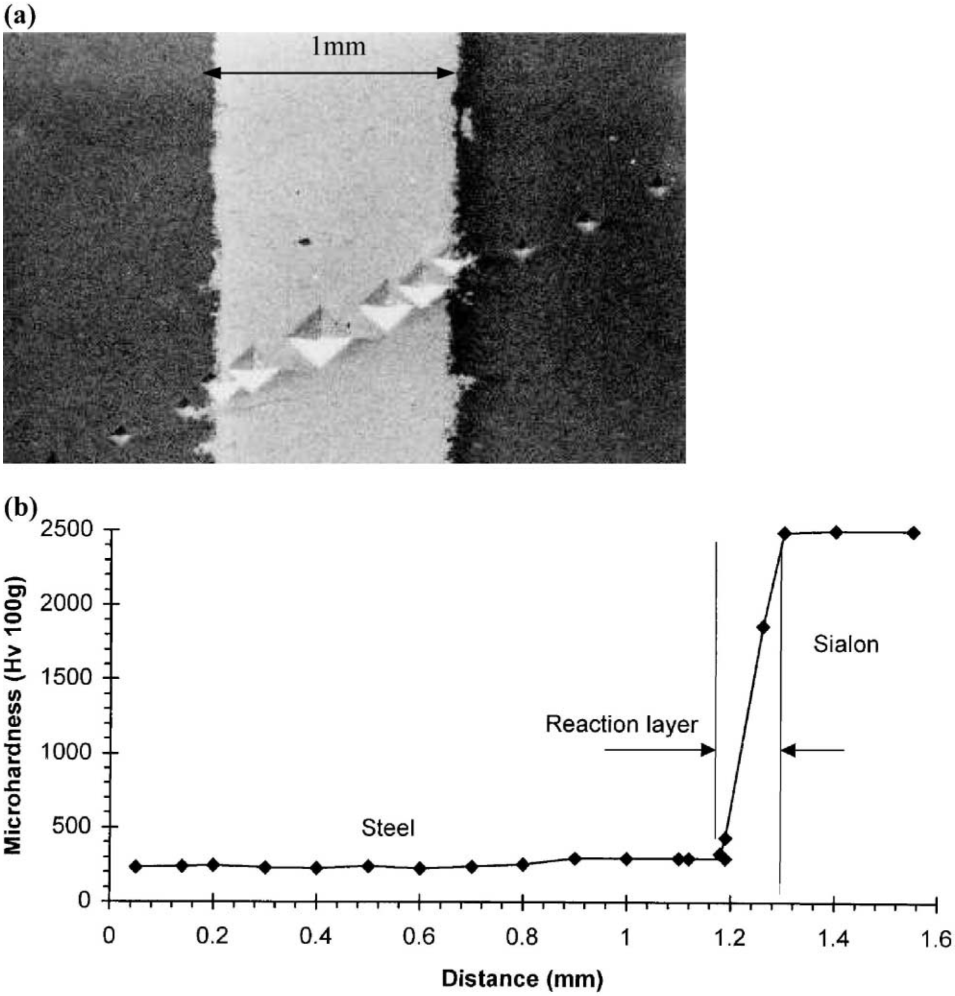

Fig. 2. (a) Optical micrograph of hardness indents in an AISI 316 interlayer between sialon components. (b) Hardness profile across a sialon/AISI 316 interface as in (a).

Previous work[2] has shown that in the case of joining sialon to steel in this way the interfaces formed are coherent and strong but in the asymmetric case of a sialon bar joined to a steel bar the residual stresses generated by cooling from joining temperature and caused by the thermal expansion mismatch between the ceramic and metal are such as to cause fracture. However it was shown[11,12] that when the join is symmetrical, that is a layer of steel sandwiched between two sialon bars, then the distribution of stresses is such that the joint does not fail on cooling. This observation is however dependent on the thickness of the steel interlayer and on the extent of the interfacial reactions described above.

Finite element modelling (FEM) has been used to study the predicted stresses caused by thermal expansion mismatch during cooling from bonding temperature. ANSYS 5.3 general purpose finite element software was used as the FEM package for calculation of thermally induced stresses on the basis of linear elastic behaviour. The axisymmetric nature of the geometry of the samples allowed a 2-D representation to be used to calculate the plane stresses σ_x and σ_y (x perpendicular to and y parallel to the ceramic–metal interface). Certain assumptions are also inherent in the model and these are:

1. The sample shrinks freely in the x- and y-directions.
2. No external mechanical load is applied and hence a residual stress field is created by the mismatch of thermal expansion coefficients alone.
3. Failure occurs within the elastic range of the materials.
4. There are perfect interfaces between the materials.
5. The metal and ceramic are taken as two plates in contact initially without any reaction between them and in the case where reaction is considered the reaction layers are taken as single parallel plates between the sialon and steel.

Clearly, assumptions (4) and (5) are contrary to the real situation described above but the significance of this is discussed below.

The parameters used for the FEM calculations[12] are given in Table 1 and are taken from manufacturer's data provided by the suppliers of the sialon and stainless steel (values of coefficient of expansion and stresses have also been verified in the author's laboratory). While it would be desirable to incorporate the temperature dependence of the parameters in Table 1 in the calculations such data is not available for the grade of sialon used and so room temperature data are used as an approximation.

The calculations were first performed for the case of perfect contact between the sialon and steel with no interfacial reaction. From the discussion presented above this is clearly an unrealistic scenario but it is important to establish the basis for comparison and to identify, if any, the effects of the interfacial reactions. The results for a steel layer thickness of 1.6 mm show that there is a variation in stresses perpendicular and parallel to the interface and a corresponding distortion of the joint. An example of the calculation is shown in Fig. 3. In the *x*-direction (perpendicular to the interface) there is a tensile stress of almost 500 MPa in the centre of the section but the free ends of the steel are under compression. Stresses parallel to the interface are induced by the shear stresses in the interface and have high values (of the order of 1060 MPa) which would be considerably in excess of the fracture strength quoted by the manufacturers for this grade of sialon (825 MPa). The conclusion would follow that this bond would fail and this was in fact the case in experimental work. Fig. 4 shows an example of cracks in the sialon following bonding with this steel thickness and geometry and these are seen to follow the predicted stress profile which is compressive at the free ends and rising tensile toward the centre of the section.

The results presented graphically in Fig. 5 show the variation of the stresses calculated from the ANSYS software as a function of the steel interlayer thickness. These show an increasing (but non-linear) dependence of σ_x on steel thickness. A calculation[11] based on the simple analytical model of Iancu and Munz,[9] which gives a linear dependence on steel thickness, shows a value of 330 MPa at 1 mm steel thickness compared to 380 MPa in Fig. 5(a). There is a constant, high level of interfacial stress σ_y at steel thicknesses in excess of approximately 1 mm which decreases with smaller thicknesses but is still considerably above the value of 825 MPa for fracture of the ceramic. These calculations of the value of σ_y are contrary to the experimental observations which show that at thicknesses of steel of 1 mm or less a good bond could be obtained with no evidence of fracture or cracking.

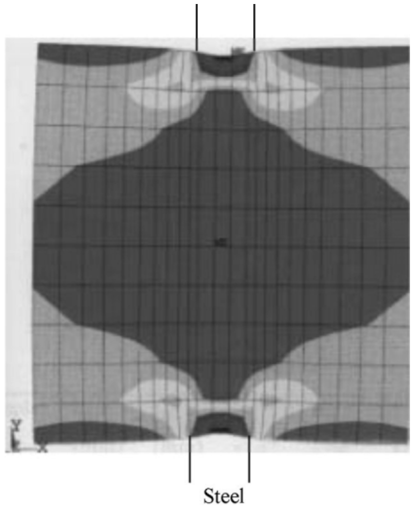

Fig. 3. Example of the ANSYS output for calculation of the thermal stresses for bonding of sialon with an AISI 316 stainless steel interlayer using the parameters given in Table 1.

Table 1
Parameters used in the FEM calculations

Material	Modulus (E, GPa)	Poisson's ratio (ν)	Coefficient of thermal expansion (α, $K^{-1}\times10^6$)	Stress (σ, MPa)
Sialon	300	0.22	3.1	825 (fracture)
AISI 316	200	0.25	22.56	286 (yield)

The difference between the observed and predicted behaviour clearly lies in the effect of the interfacial reaction on the properties of the join. It was therefore decided to model the effect of the reaction interlayers by inserting estimated values of the physical parameters of the interlayer into a multi-layer model of the interface. Clearly, the interlayer is not a single entity as Fig. 1 shows that there is penetration of metal (irregularly) into the ceramic and diffusion of silicon and nitrogen into the steel (as shown by the hardness profile, Fig. 2). However, a simple model of the interface layer was devised as described below.

Given the experimental observations that a thickness of less than 1 mm led to an integral stainless steel bond between sialon pieces, an experiment was conducted with an initial steel layer thickness of 0.8 mm in order to characterise the dimensions and nature of the interfacial reaction layers. This is the experiment illustrated in

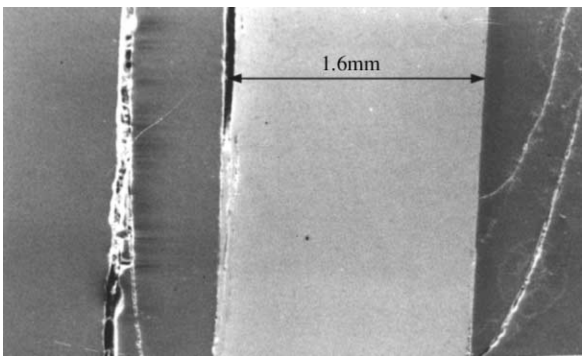

Fig. 4. Optical micrograph of cracking in steel-bonded sialon parts where the steel thickness (1.6 mm) is greater than the critical value.

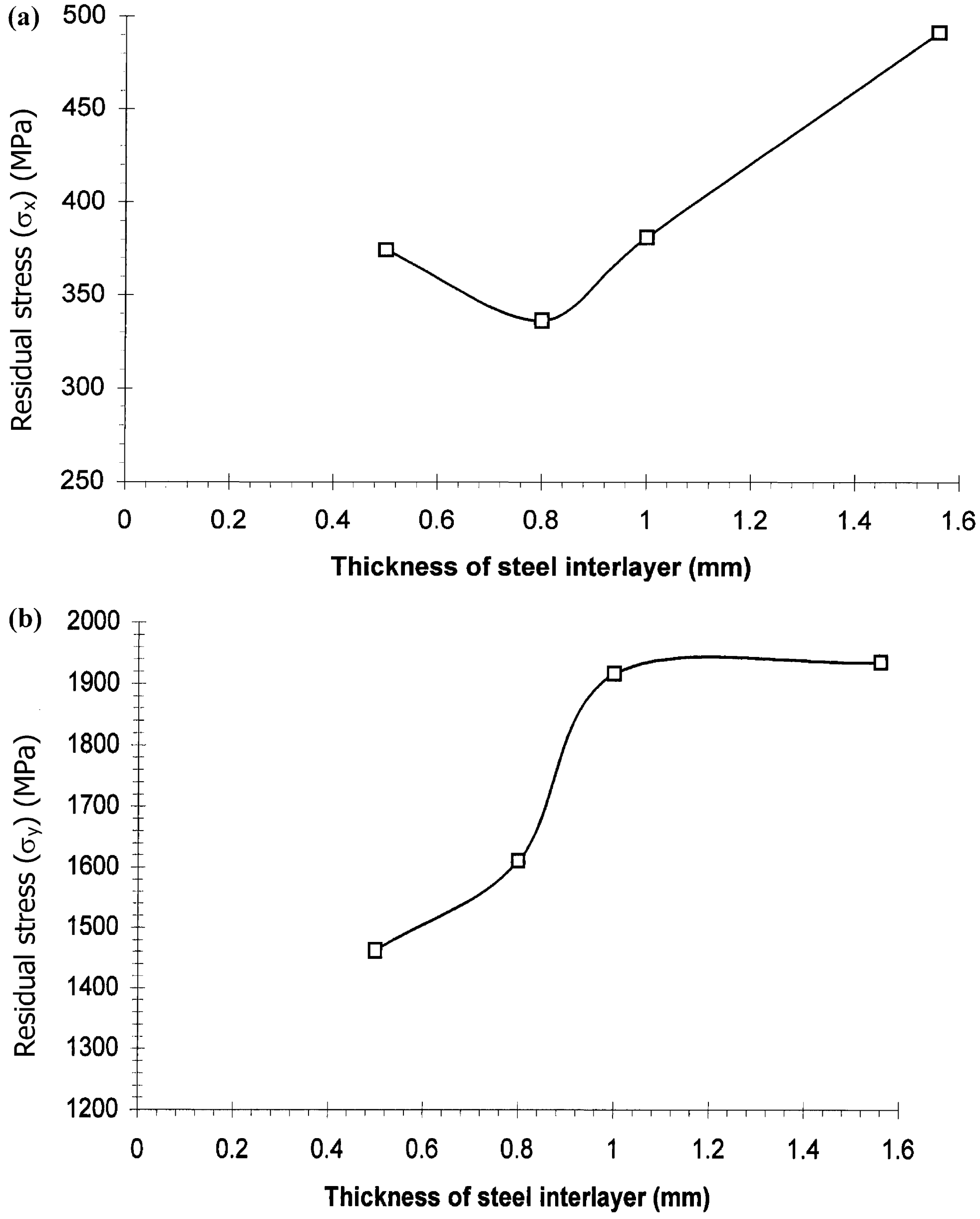

Fig. 5. Plots of stresses σ_x and σ_y (from the ANSYS analysis) against thickness of the steel interlayer.

Fig. 6 and the microstructures and phases have been described above and in detail elsewhere.[2,11,12] From measurements of the reaction layers it is estimated that the interfacial reaction layer thickness is 0.26 mm which comprises of 0.06 mm penetration into the sialon and 0.2 mm of diffusion depth into the stainless steel [on each side of the steel layer, as shown in the schematic of Fig.6(b)]. These dimensions have then been introduced to the ANSYS data and estimates made of the physical parameters for the layer. In the model, the data for the sialon and steel are fixed and variations are made to the Young's Modulus and the coefficient of thermal expansion of the interfacial layer. Given the hardness of the reaction layer in the steel, which is intermediate between that of sialon and steel, the modulus was varied from the value for the stainless steel (200 GPa) to that of the sialon (300 GPa). The coefficient of thermal expansion is assumed to be lower than that of the metal (higher hardness and the presence of large precipitates) and is varied from $3.1–10\times10^{-6}$ °C^{-1}. The results of iterative calculations show that for the stress in the sialon to be less than 825 MPa, the fracture stress of the sialon, then a modulus of 250 GPa and a thermal expansion coefficient of less than 6×10^{-6} °C^{-1} are required for steel thickness of 0.8 mm. The modulus value is reasonable for such a material and the stress is relatively insensitive to changes in that value. However, the stress is extremely sensitive to changes in the expansion coefficient and increasing the value above 6×10^{-6} °C^{-1} leads to a rapid increase in the calculated stress. This value of the expansion coefficient

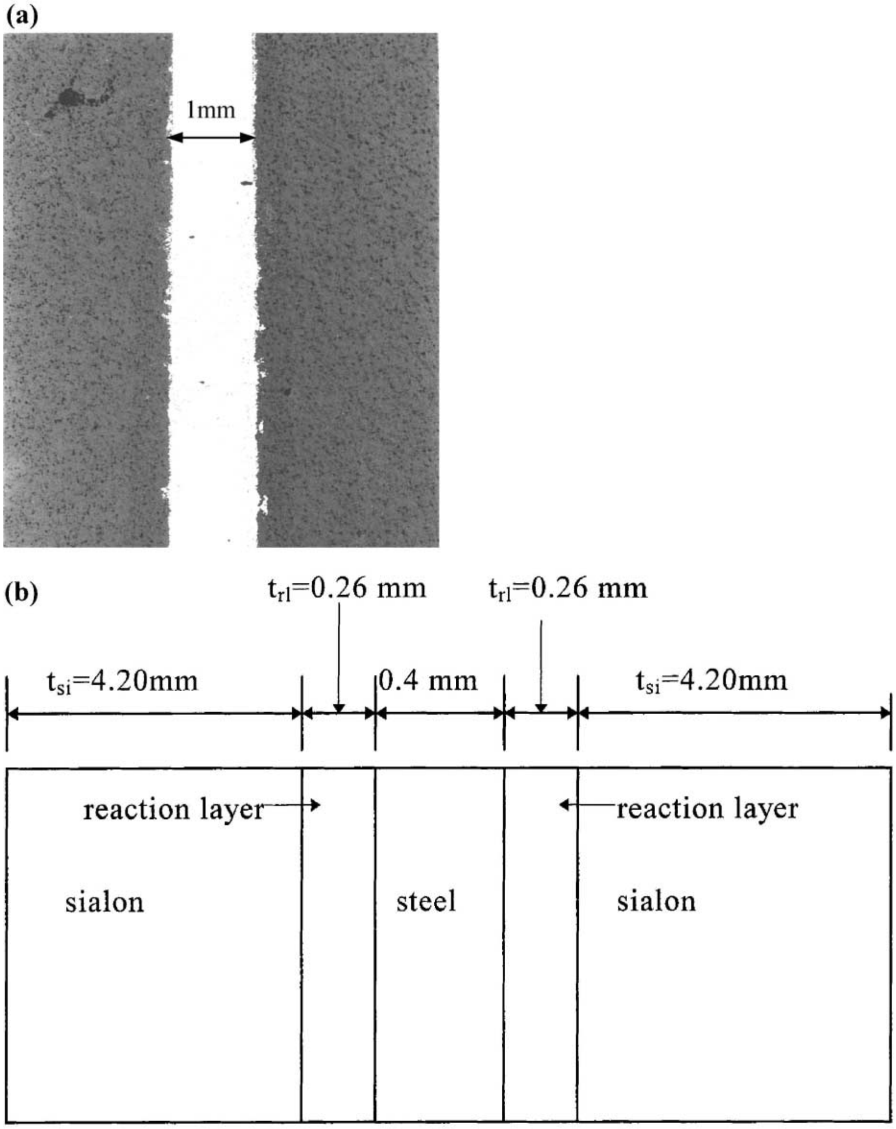

Fig. 6. (a) Optical micrograph of a coherent sialon–sialon bond with a steel interlayer. (b) Schematic diagram of the relative thicknesses of the component parts of the bond in (a).

is, however, reasonably in line with values in the range of 6–8.5×10^{-6} °C^{-1} which have been measured in the authors' laboratory for sintered sialon-stainless steel particulate composites containing 10–45% steel. The reaction layers shown in Fig. 1 are clearly more "metallic" than these composites but the coefficient of expansion value required from the stress calculation are reasonably in line with these observations.

4. Conclusions

The nature of the interfacial reactions and the residual thermal stresses which occur on joining sialon to stainless steel have been characterised. The data and experimental observations have then been applied to the use of an austenitic stainless steel as a ductile interlayer to join two sialon components. It is shown that below a critical thickness of steel the residual stresses are less than the fracture stress of sialon and a sound join is obtained. Finite element modelling has been used to derive values of the Young's modulus and coefficient of thermal expansion of the interfacial reaction layer which are consistent with the observed behaviour. The derived values (Young's modulus 250 GPa and coefficient of thermal expansion 6×10^{-6} °C^{-1}) are also consistent with the microstructural nature of the interlayer.

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