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Interface reactions and bonding strength in aluminium alloy (AA-7075)–alumina diffusion bonds

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Diffusion bonds between alumina and high-strength aluminium alloy (AA-7075) have been produced and studied in the present work. Direct diffusion bonding in the solid state was tested as a possible joining method for both materials. The nature of the AA7075–Al_2O_3 interface was investigated paying special attention to the chemical interaction processes between the alloying elements and the ceramic material, as well as their influence on the joint strength. SEM images and energy-dispersive microanalysis were used to determine the formation of reaction layer between both parent materials. Shear strength was used as an optimum method to evaluate the bond strength and the influence of the bonding parameters on it. A maximum shear strength of 60 MPa was achieved using bonding temperatures and pressures of 360 °C and 6 MPa, respectively, during very prolonged bonding times (100 h). Fractographic studies of the failure surface gave additional information on those aspects.

1. Introduction

The development of ceramic science and technology during recent years has resulted in the design of a new generation of ceramics which can be applied as structural materials. This has been possible because of their high wear resistance, high hardness and elevated resistance to high temperature, in addition to a lower density than most of the metals [1]. One of these applications which required the previous properties is the field of propulsion engines, where an increasing power with an important saving in combustible consumption can be obtained if ceramic valves are used [2].

The employment of these kinds of valve or other components in an engine requires the production of metal–ceramic joints with a minimum mechanical strength. One of these dissimilar joints is the union between structural aluminium alloys and ceramic materials as alumina.

The present paper summarizes the results obtained in the study of one of those dissimilar joints. It is the union of AA7075 with alumina using direct diffusion bonding.

Both microstructural studies (SEM and energy dispersive spectroscopy, EDS) and a mechanical test (shear test) were carried out on diffusion-bonded specimens to determine the nature of the bond interface, the interaction between both parent materials and its influence on the joint strength.

2. Experimental procedure

The parent alloy used in this investigation was a high-strength aluminium alloy (AA-7075) with a nominal composition (wt %) of Al–5.6 Zn–2.5 Mg–1.6 Cu–0.23 Cr. It was received in sheet form with a thickness of 3 mm in a "T4" condition.

The parent ceramic was a pure alumina fabricated in a sintering process with variable contents of sodium ($< 0.005\%$) and magnesium ($< 0.1\%$), and low porosity ($< 3\%$).

The bonding surfaces of both parent materials were carefully prepared by grinding with SiC emery paper, finishing the operation with 600 grade for the aluminium alloy, and with 6 μm diamond past for the ceramic material. Both surfaces were cleaned in an ultrasonic bath with acetone just before bonding.

Diffusion joints were carried out in a special vacuum furnace with a level system to apply compressive load. All bonding tests were made in a lightly oxidizing atmosphere (5×10^{-3} torr; 1 torr = 133.322 Pa) obtained in the furnace chamber with a rotatory pump.

Preliminary diffusion bonding tests were made to determine the range of bonding conditions for which an excessive plastic deformation of the metallic sheet could be avoided (Fig. 1). These preliminary trials were produced using bonding temperatures in the range 360–400 °C for times between 300 and 3600 min (Table I). The bonding pressure for all these tests was fixed at 6 MPa in an attempt to obtain intimate contact between both faying surfaces at the same time, in order to control the final overall thickness deformation in the aluminium sheet to values lower than 10%.

After these preliminary trials, a second sequence of bonding tests was carried out to determine the strength of the metal–ceramic joints produced. They were made using low temperatures (360 and 380 °C) and prolonged bonding times (3200–6000 min). Bonding pressure was again fixed at 6 MPa (Table II).

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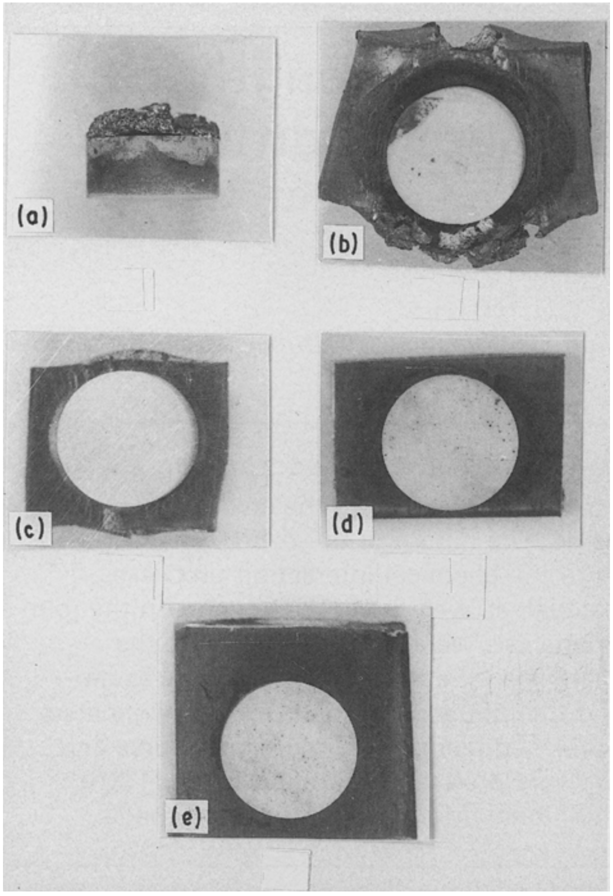

Figure 1 Preliminary bonding trials. For conditions, see Table I.

TABLE I Preliminary diffusion-bonding tests

Trials	P (MPa)	T (°C)	t (min)	Observation
a	6	440	300	Partial melting
b	6	425	3600	High deformation
c	6	400	3600	High deformation
d	6	380	3600	Deformation < 10%
e	6	360	3600	No deformation
f	6	360	3000	No deformation
g	6	330	3600	No bonding

TABLE II Diffusion-bonding conditions and shear strength in AA7075–alumina joints

Trials	P (MPa)	T (°C)	t (min)	σ (MPa)
C1	6	360	3200	13.6
C2	6	360	3600	18.0
C3	6	360	4000	43.8
C4	6	360	6000	60.2
C5	6	380	3200	14.0
C6	6	380	3600	33.5
C7	6	380	4000	–

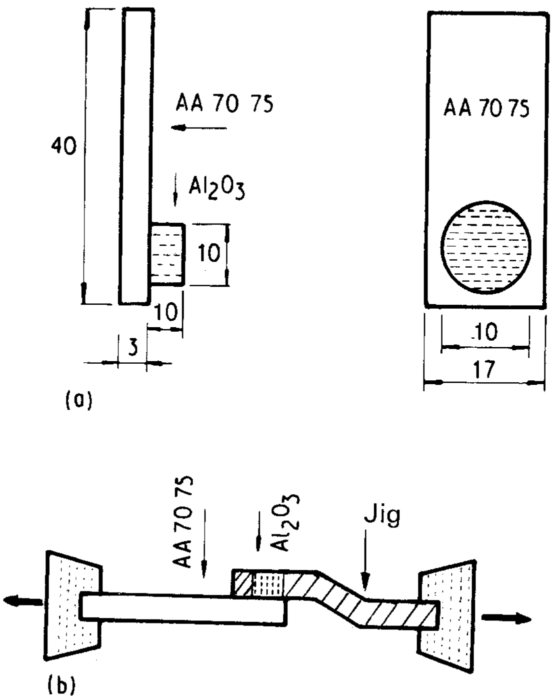

Figure 2 (a) Shear test piece used for testing AA7075–alumina diffusion bonds; dimensions in mm. (b) Jig used for shear testing.

During the diffusion bonding of all the specimens tested, and once the bonding time at the fixed temperature was over, the joints were subjected to a slow cooling process ($\approx 0.8\,°\mathrm{C\,s^{-1}}$) down to 200 °C. Subsequently, bonded specimens were held at that temperature for 3 h before removing them from the chamber furnace. Both the slow cooling and the postbonding annealing process were applied to relieve the residual stresses generated during cooling, because the mismatch effect originated through the bond interface due to the different expansion coefficients present in both parent materials.

The strength of the bonded specimens was evaluated by a shear test carried out at room temperature with a crosshead speed of $0.01\ \mathrm{mm\,s^{-1}}$. The shear test piece used is illustrated in Fig. 2a. It was made by bonding a ceramic cylinder, 10 mm diameter and 10 mm high, on one edge of a rectangular blank (40 mm × 12 mm) of the alloy. The shear test was carried out using a special jig which minimizes the bending components and makes sure that failure occurs by shearing (Fig. 2b).

3. Results

3.1. Microstructure of diffusion-bonded joints

The microstructure of the bond interface in the alumina–AA7075 diffusion bonds was characterized using scanning electron microscopy (SEM) and EDX microanalysis. It was proved that high continuity that can be obtained between both parent materials (Fig. 3) where the formation of a diffusion compound by reaction between the alloying elements and alumina, was observed (Fig. 4). This compound forms an almost continuous layer that is spread through the bond interface, occasionally penetrating through the alumina grain boundaries connected with the original bond interphase.

Semiquantitative analyses realized on different zones of alumina–AA7075 diffusion joints are shown in Table III. These results show an important enrichment in zinc and specially in copper in the compound formed in the bond interface, in relation to the contents of both elements measured in the metallic

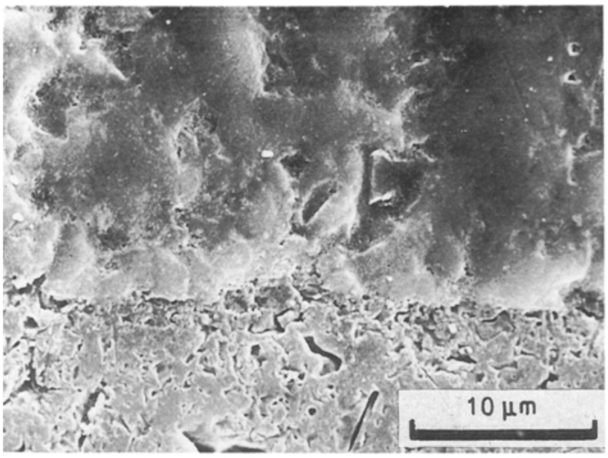

Figure 3 AA7075–alumina diffusion-bond interface.

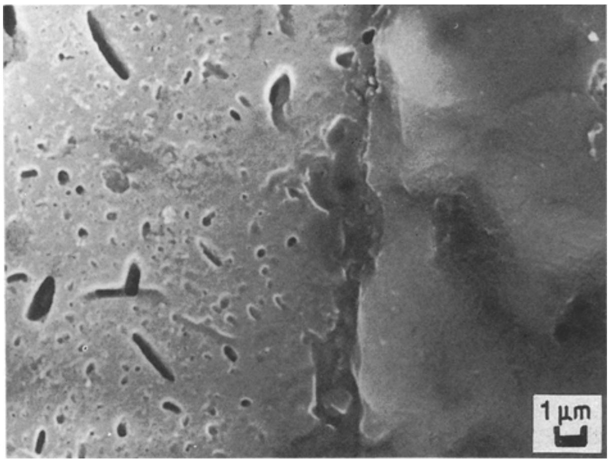

Figure 4 Detail of diffusion layer formed during bonding of AA7075 to alumina.

TABLE III Semi-quantitative EDS microanalysis in different zones in an AA7075–alumina diffusion bond. I is the emmisivity (intensity K_α)

Zone	KI			$II_0(\times 10^3)$	
	Al	Cu	Zn	(Cu/Al)	(Zn/Al)
AA7075 matrix	643.5	2.4	9.2	3.7	14.3
Precipitate	582.7	9.6	12.9	16.4	22.2
Parent alumina	309.5	–	–	–	–
Interface	268.0	5.8	4.8	21.6	17.7
Alumina (5 m)[a]	298.6	0.9	1.0	2.9	3.3

[a] Alumina matrix at 5 m to the bonding interface.

matrix. The relative intensity of the X-ray K_α for copper and zinc measured in the interface is similar and even higher than those obtained in precipitates rich in both elements. However, the intensity of the K_α line for aluminium, measured under the same conditions, is lower than that obtained for the aluminium matrix and even for most of the precipitates, the amount being similar to that measured in the alumina. This proved that the diffusion compound was probably an oxidizing phase.

Although the presence of magnesium was also detected in the diffusion layer, it was very difficult to measure the real intensity of its K_α line.

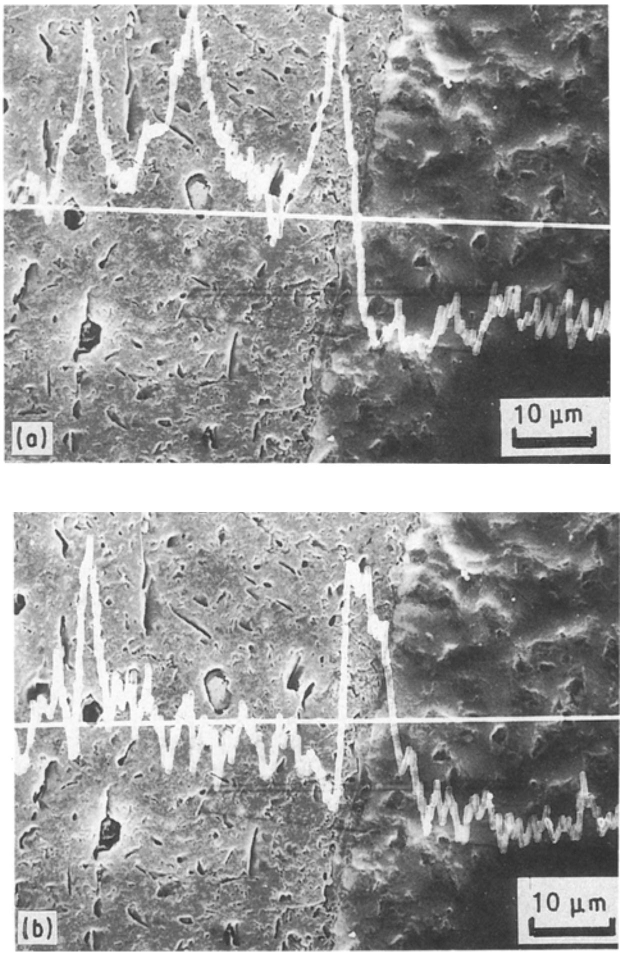

Figure 5 X-ray line profiles for alloying elements of AA7075: (a) ZnK_α and (b) CuK_α.

In the same way, the presence of a very low concentration of copper and zinc diffused into the alumina matrix close (~ 5 µm) to the bond interface was observed. Fig. 5a and b represent the line profiles of zinc and copper characteristic X-radiation (K_α line), respectively. They have been measured along the white straight lines marked in the figure and, in both cases, show an increase of the intensity just in the bond interface.

3.2. Shear strength of diffusion-bonded joints

The realization of the preliminary bonding test (Table I), made it possible to determine the range of bonding conditions which provide a thickness reduction in the metallic sheet lower than 10%. Fig. 1 shows some of these trials bonded at temperatures from 360–440 °C and bonding times shorter than 3600 min. In joints bonded at 440 °C (Fig. 1a), the aluminium alloy underwent incipient grain melting. Although this phenomenon did not occur at temperatures in the range 400–425 °C (Fig. 1b and c), the high plasticity of the alloy under this condition causes excessive deformation in the alloy (> 10%) even when short bonding times were used. Only these joints bonded at 360–380 °C presented a thickness deformation in the range considered (Fig. 1d and e). For this reason, all the diffusion joints for shear testing were made in this temperature range.

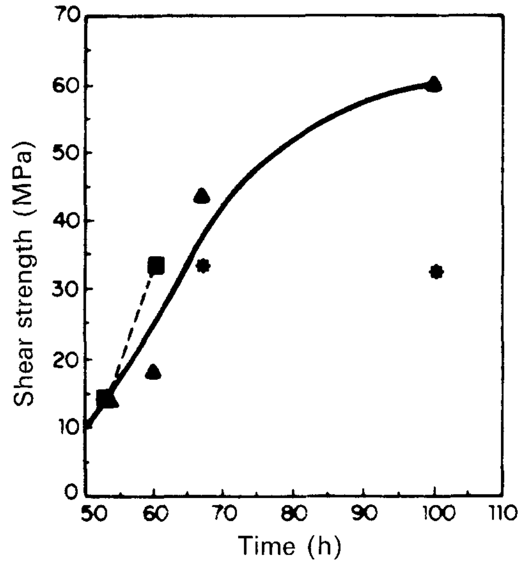

Figure 6 Shear strength versus bonding times in diffusion joints of AA7075–alumina bonded at (▲) 360 and (■) 380 °C. (★) > 10% deformation.

The measured shear strengths in AA7075–alumina diffusion joints are given in Table III, as well as their bonding parameters. The maximum shear strength obtained was approximately 60 MPa. These values were determined as a relation between the failure load and the initial contact area between both parent materials.

Fig. 6 represents the dependence of the bonding time on the average shear strength for two different temperatures. Temperatures higher than 360 °C and bonding times longer than 70 h produced a deformation higher than the established limiting value (10%) and the measurement of the strength under these conditions is not possible.

The higher integrity of the diffusion joints bonded at 360 °C over long periods is seen from the increase in the plastic strain introduced into the metallic sheet during the shear test before failure occurs. Fig. 7 shows some of the load versus elongation curves obtained in the shear testing of three bonded specimens. The failure in joints bonded with times shorter than 60 h was always located in the bond interface, and it was produced with a very limited strain (Fig. 7a). On increasing the bonding time to the range 60–70 h, the shear strength reached values close to 40 MPa; and, although the failure occurred again through the alloy–alumina interface, some points of rupture in the ceramic were observed. This occurred with increasing elongation (Fig. 7b). Bonding times of 100 h and temperatures of 360 °C allowed the maximum strength to rise to values higher than 60 MPa, with an important elongation in the parent alloy (Fig. 7c). Under these bonding conditions, failure occurred principally through the bulk ceramic.

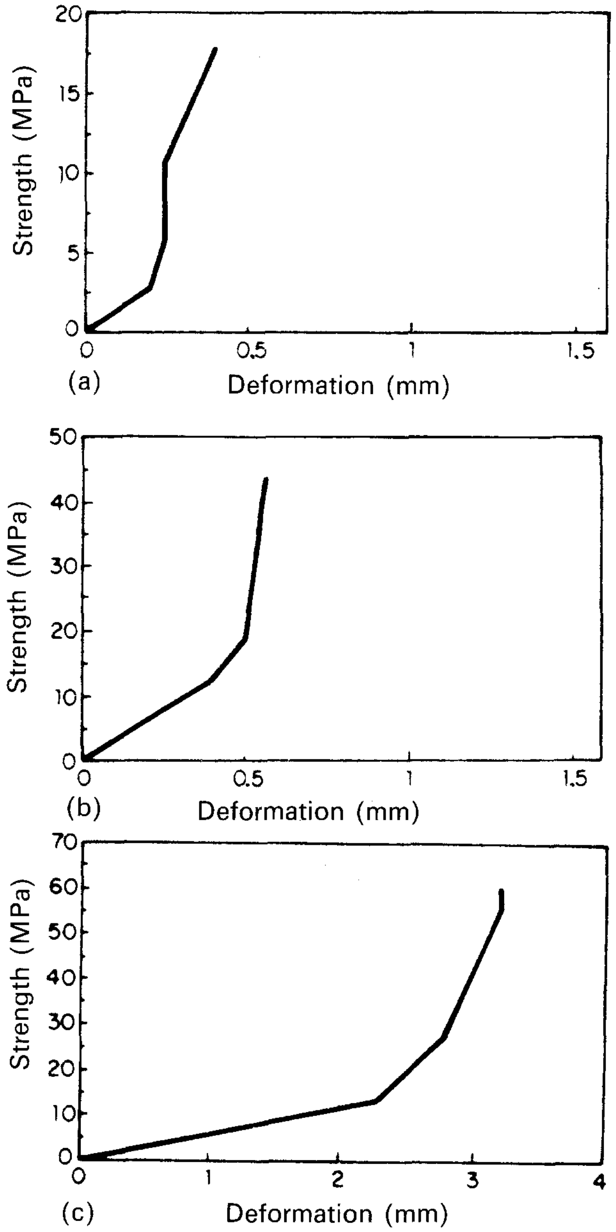

Figure 7 Strength–deformation curvés obtained during shear testing of AA7075–alumina joint: (a) 380 °C, 3200 min; (b) 380 °C, 3600 min; (c) 360 °C, 6000 min.

3.3. Fractographic study of diffusion joints tested

The fractographic study carried out on specimens tested to shear provided additional information on the chemical interaction between the parent materials and its influence on the mechanical strength of the joint.

Failure in joints bonded with the shortest times (< 60 h) which presented a very limited strength (< 15 MPa), occurred through the bond interface. No evidence of the formation of oxidized products on the metallic surface was observed and the joint was preferentialy produced by penetration of the aluminium alloy by creep in the alumina voids, producing a mechanical union.

On increasing bonding time, the zones where the failure occurred through the bulk alumina grew. These zones presented the typical concoidea fracture produced by residual stresses generated during cooling, which could not be completely eliminated by post-bonding annealing (Fig. 8a and b).

The failure in a joint bonded during 100 h occurred principally in the bulk ceramic, although an edge effect was observed where the bond integrity was very low.

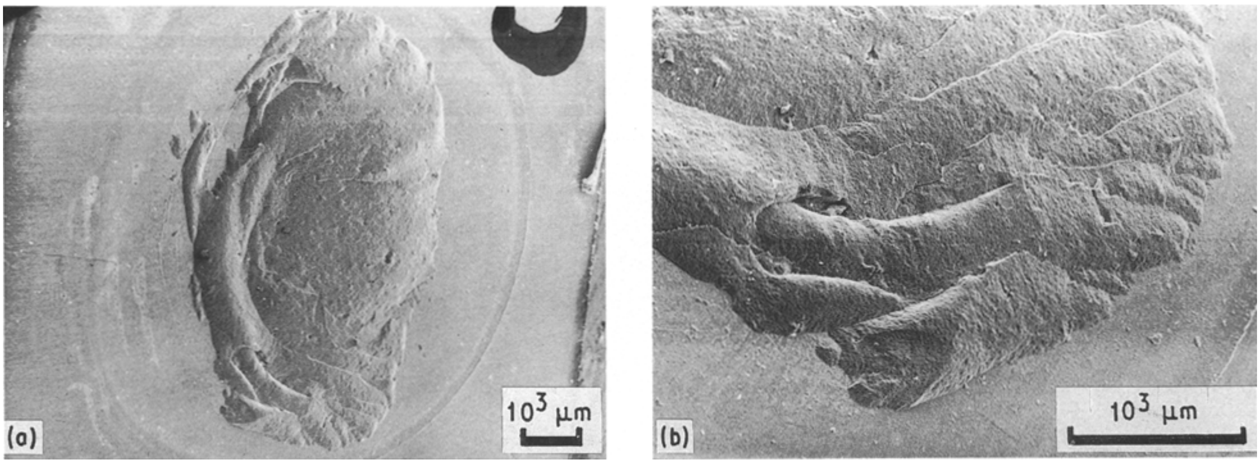

Figure 8 Fractography of a AA7075–alumina diffusion bond tested to shear. (a) Concoideal fracture, (b) Detail at higher magnification.

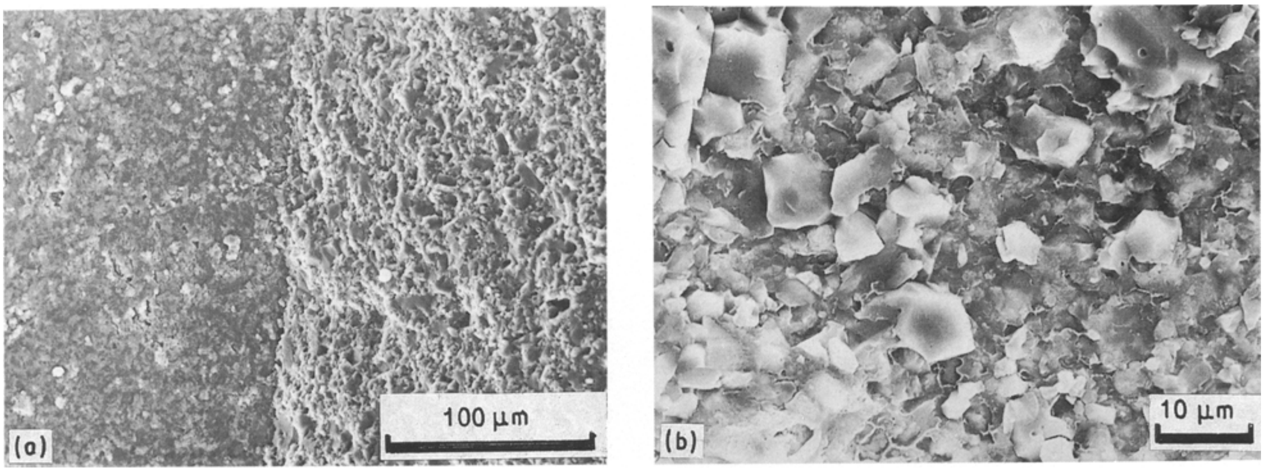

Figure 9 (a) Fractography of an AA7075–alumina bond. (b) Detail of Al_2O_3 grains adherent to the aluminium matrix by reaction layer formation.

The study, at higher magnification, of the fracture zones close to the ceramic failure areas shows the presence of alumina grains adhering to the metal surface; the presence of a layer of complex oxide analysed in the polished surface was also detected (Fig. 9a and b). The proportion of alumina grains decreases toward the joint edges, where the union is only achieved by a mechanical effect and no sign of chemical interaction was detected. However, zones of failure by shearing of the aluminium alloys could also be observed (Fig. 10).

4. Discussion

Microstructural study by SEM, both on polished and fracture surfaces, combined with EDX microanalysis, provided sufficient information to explain the method of formation of the alumina–aluminium alloy joint.

These studies have shown the important role that the presence of the alloying elements in the aluminium alloy (zinc, copper and magnesium) play on the formation of a resistant joint with the alumina. These elements favour the formation of complex oxides in the metal–ceramic interface by reaction with the alumina in the oxidizing environment of the diffusion-bonding chamber. It promotes the generation of a chemical bond, the effects of which are added to the mechanical bond typical of ceramic–aluminium joints.

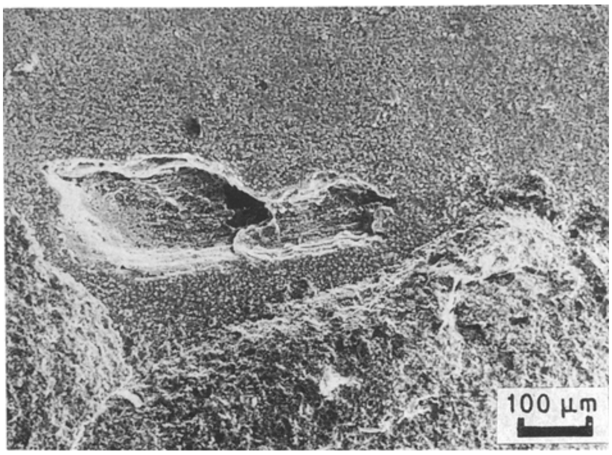

Figure 10 Detail of shearing failure through the metallic material.

With the experimental data obtained here on this kind of joint, a model is proposed to explain its formation. This model is based on that proposed previously for the solid state bonding of alumina to steel via a copper interlayer [3], and, in the same way, that the model consists of three stages dominated by different physical and chemical mechanisms.

The first stage is initiated when both parent materials are brought into contact up to bonding temperature, with the application of a compressive load. This produces an increase of the initial contact area by the participation of plastic deformation and creep mechanisms on the metallic material. Both mechanisms favour the establishment of a mechanical joint as the only type of bond.

For longer bonding times (more than 60 h for the bonding temperature used), in a second stage, a further two mechanisms take part in the bond formation. Firstly, the alloying elements, which are dissolved in the aluminium matrix at bonding temperature, diffuse to the bond interface. At this point, they react with the free oxygen of the chamber atmosphere ($P_{02} \approx 10^{-3}$ torr), inducing the formation of ZnO, CuO and MgO. Table IV gives the formation free energy, ΔG, for these three oxides at 360 °C and P_{02} of 10^{-3} torr [4].

TABLE IV Oxidation reactions of the alloying elements diffused to the bond interface. Formation free energy at 360 °C and $P_{O_2} = 0.001$ torr

Reaction	ΔG (kJ/mol O_2)
$2Cu + O_2 = 2CuO$	– 75.4
$4Cu + O_2 = 2Cu_2O$	– 174.2
$2Zn + O_2 = 2ZnO$	– 498.3
$2Mg + O_2 = 2MgO$	– 997.9

Once alloying element oxides are formed, in a third stage, which takes place only after very prolonged times (≈ 100 h), they react with the alumina by topotactic reactions giving the formation of complex oxides. These oxides are probably of the spinel type ($Al_2O_3 \cdot XO$), and its formation has been already detected in other types of alumina–metal joints [5, 6]. These oxides formed a reaction layer, with variable thickness (< 2 μm), which never forms a continuous film. The limited amount of oxide formed in the specimens tested and its complexity makes correct identification very difficult. The layer formed is probably constituted by a mixing, in variable proportion, of three types of spinels: AlO_2Cu, Al_2O_4Zn and Al_2O_4Mg. The slow kinetics of these reactions at bonding temperature explain the scant amount of mixed oxides.

The layer growth will be controlled by the metallic cation mobility in the ceramic (Cu^+, Mg^{2+} and Zn^{2+}) which present a movement in the opposite direction to the Al^{3+}. Traces of copper and zinc have been detected inside the Al_2O_3 matrix close to the reaction layer (< 5 μm).

The formation of the reaction layer explains the higher strength obtained for the joint studied, in relation to those reached in alumina–pure aluminium joints where the only mechanism to produce a resistant union is the mechanical linking by plastic and creep flow of the metal inside the alumina voids [7, 8].

5. Conclusions

1. Resistant joints between alumina and an aluminium alloy (AA-7075) can be obtained by bonding in the solid state at 360 °C with pressures of 6 MPa and prolonged bonding times (100 h).
2. Maximum shear strength of 60 MPa has been obtained with the previous bonding conditions; the application of a post-bond annealing is necessary to relieve residual stresses generated during cooling.
3. The presence, in the aluminium alloy, of active elements such as zinc, copper and magnesium favours the formation of a reaction layer in the bonding interface.
4. The reaction compound detected in the bond interface may be constituted by complex oxides of the type $Al_2O_3 \cdot XO$ (X = Zn, Cu and Mg), formed by the reaction of Al_2O_3 with the alloying element oxides.

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