Welding of dense alumina and aluminium by plastic deformation and diffusion

M. A. MARTÍNEZ, J. RODRÍGUEZ, C. NAVARRO, R. CORTÉS, V. SÁNCHEZ-GÁLVEZ

Department of Materials Science, ETS Ingenieros de Caminos, Canales y Puertos, Polytechnic University of Madrid, Ciudad Universitaria s/n 28040 Madrid, Spain.

The development of a welding system which makes it possible to obtain a close joining of two very different materials is described: high density (low porosity) alumina and aluminium alloy. To determine the characteristics of the contact area, optical microscopy was used, whereas energy dispersive spectroscopic (EDX) microanalysis was employed in the study of the atom diffusion that may take place through the metal-ceramics interface. The Hopkinson bar was used to test the joint resistance in an impact test.

1. Introduction

In the field of new materials, the line that is developing most quickly is that of the joint use of materials such as metals and ceramics, either as a compound material or as welded structural pieces.

The best results have been obtained when a metallic bearing has been used to integrate the ceramic pieces in the structure. In spite of all the problems that remain to be solved, there is an ever increasing use of these compound materials in the automobile, aerospace, nuclear and defence industries [1, 2].

For adequate joining of the metal and the ceramics, close contact between the pieces is required. This can be achieved with or without the use of an intermediate metal or alloy. Once the pieces have been welded, the interface should be capable of resisting the residual stresses generated during the cooling process, which are due to the difference in the thermal expansion coefficients of the materials.

Many researchers have dealt with the binding of ceramics such as silicon carbide, silicon nitride and aluminas of different degrees of purity and density, and metals such as steel [3–11], copper [8, 12, 13], chromium [14, 15], nickel and its alloys [14, 16, 17], lead [18], noble metals [19], etc. These bindings were carried out directly or else by using ductile materials as intermediate components.

There is little reference bibliography on the subject of aluminium (and its alloys)-alumina welding, which is surprising if we consider the importance of these materials in the field of industry. Publications by various researchers are listed in Table I: in some cases aluminium has been used as the ductile material interband to facilitate the welding of another metal and a ceramic material [9, 20, 21, 25] or that of two ceramics [22-25]. However, information is incomplete in all cases, and the processes carried out hardly practicable either from the economic or from the technological point of view.

Our research was aimed at establishing welding

conditions that could be reproduced at relatively low costs at industrial level; we worked with inert argon atmosphere instead of in a vacuum, and at lower pressures and for shorter periods of time than those employed in diffusion welding, which is harder to achieve in the large chambers used for certain pieces. Thus, a special furnace was designed, which makes it possible to exert pressures of 0.1-0.5 MPa on the specimens, which are heated to temperatures of 600-700 °C. This process takes place in an inert argon atmosphere.

Once welding has been obtained, the resulting structures are studied by optical and electronic microscopy and microanalysis [9, 21]. The resistance of the welded junction undergoes an impact test with the Hopkinson Bar, and the results obtained are compared to those of similar specimens made of alumina, aluminium alloy and alumina-aluminium bound with a rigid adhesive.

2. Metal-ceramics binding process

Alumina and aluminium can be joined in two different ways: they can be either bound or welded. Bounding may be the right solution in some cases, but with the disadvantage of the instability of adhesives at temperatures over $150 \,^{\circ}$ C.

The first problem that has to be faced when welding aluminium and its alloys is its high absorption of oxygen. These materials are characterized by their tendency to form, in a very short time, films of very hard, adherent and dense alumina, which protect the rest of the material from oxidation but also prevent the migration of atoms from the metal to the ceramics, making for a difficult diffusion welding process. These oxide films can be broken in two ways: by exerting high pressures at medium temperatures (diffusion welding) or by low pressures at temperatures close to the melting point of aluminium (welding by plastic deformation).

TABLE

Comp 1	Comp 2	Inter	T (°C)	P (MPa)	T (min)	Reference
Si ₃ N ₄	Si ₃ N ₄	Al	860	3.3	15	[20]
Invar	Al_2O_3	Al	500	20-60	1800	[21]
Invar	Si ₃ N ₄	Al	800	0.15	7	[9]
Kovar	Si_3N_4	Al	800	0.15	7	[9]
Al	Al_2O_3	-	580-620	14-75	-	[22]
Al	SiO ₂	_	600-640	8-30	40	[23]
Al-6Mg	PSZ	_	535	9	30-40	[24]
Al-6Mg	ZrO_2		500	0.5-7		[25]
Al-6Mg	MgO		500	1	-	[25]
Steel	Al_2O_3	Al	625	50	30	[25]
	(47.5%)					
Steel	Si ₃ N ₄	Al	800	0.15	7	[9]

Another important problem that must be taken into account in these welding processes is the appearance of stress in the contact area, due to the difference in the thermal expansion coefficients of aluminium alloys and alumina (which is of at least one order of magnitude). After the welding process, a quick cooling of the material generates a certain degree of residual stress, especially at the edges of the pieces and creates an "edge effect" [26]. When the metal does not rearrange its crystalline structure by means of plastic or inelastic deformation, tensions in the contact area appear; the ceramics crack close to the welding, which is the area of maximum stress (Figs 1 and 2). In order to prevent this, an interband of ductile material should be placed between the metal and the ceramics. This material should be thick enough to support elastic, plastic and even slip deformation, as well as extremely slow cooling processes followed by long annealing treatments at low temperatures, making it possible for the intermediate metal to recrystallize so that stress is relieved.

Solid state diffusion welding is a binding process in which the surfaces of the materials to be welded are exposed to medium temperature and high pressures for a long period of time. Research on the diffusion welding of aluminium and alumina suggests the use of temperatures ranging from 400 to $600 \,^{\circ}\text{C}$ (between 50 and 80% of the melting temperature of the aluminium alloy used), pressures between 10 and 80 MPa, and periods which range from 15 to 1800 min. in order to break the aluminium oxide film that protects the metal [21, 22, 24, 25]. This is a slow process, and it is carried out in critical conditions of pressure, temperature and atmosphere.

Welding by plasticization is achieved by means of the plastic deformation of the metal at high temperatures close to its melting point. This technique has been used to join two pieces of a high melting point, putting aluminium sheets of different thickness between them [28, 29]. Under low pressures the plasticized metal penetrates the surface defects of the ceramics. The higher the porosity of the ceramics and the plasticity of the metal, the more resistant the fastening [3, 4, 30].

In both cases, however, the material surfaces are never perfectly polished nor exactly parallel: the real contact starts on the ridges of the surface roughness.



Figure 1 Area of maximum stress caused in a welded $Al-Al_2O_3$ specimen by the difference in contraction during a high speed cooling process.



Figure 2 Breakage of an $Al-Al_2O_3$ weld caused by high speed cooling of the specimen.

This contact area increases with the exertion of pressures at adequate temperatures (Fig. 3). This coalescence is similar to that which takes place in sintering processes. Temperature makes possible the plasticization and the subsequent diffusion, since it causes the migration of atoms through the weld surface to the empty spaces in the ceramics [31].

The metal-ceramics joining may originate chemical reactions of mass transfer which produces transition areas between the metal and the ceramics [32]. These may appear even in solid state, and are facilitated by a



Figure 3 Microscope view of one of the surfaces after blooming.

greater diffusion when working at high temperatures. In some cases, the resulting oxides or metallides may make for greater fragility or for additional strength of the weld, depending on their proportion and their mechanical features.

3. Experimental procedure

There are certain factors that must be taken into account when considering the quality of these welds.

(a) Surface condition: since metal surfaces usually contain oxides, grease, etc, they require appropriate machine work, although polishing is not necessary. Oxides and pollutants are eliminated by descaling, whereas grease is removed with solvents. The ceramic surface requires previous forming, and it should be as porous as possible but not to the point of being rough.

(b) Temperature conditions: temperature control should be extremely precise since the aluminium melting point must not be reached in the process. An ideal temperature is $20 \,^{\circ}$ C below that point, which is enough to make plasticization easier. This temperature has to be reached as quickly as possible, to avoid the formation of superficial oxide films which might prevent an adequate contact between the metal parts. Later, to facilitate the diffusion process, it should be kept between 50 and 80% of the melting temperature of the metal to be welded.

(c) Pressure conditions: the pressure to be exerted on the materials should be high enough to enable the plasticized metal to flow through the pores and intergranular spaces of the ceramics. The lower the pressure, the higher the temperature that has to be reached for the plasticization to take place.

(d) Time: plasticization takes place rapidly, although the subsequent solid state diffusion process is comparatively slow. Finally, a very slow cooling in the furnace is required so that the stress produced by the difference in the thermal expansion coefficients of the materials is relieved gradually.

(e) Atmosphere: it should be non-oxidant, either a vacuum or an inert atmosphere obtained by means of an argon sweep, to avoid the formation of oxide films on the contact surfaces. In some cases, special coatings have been used to prevent the access of oxygen to those surfaces.

Taking all these factors into account, we have designed and built a furnace inside a hermetic chamber with an external cooling coil, which enabled us to operate in a vacuum or a controlled atmosphere. Pressure is exerted on the specimens by means of a pressure system of two pistons located above and below the drum (Fig. 4).

The following materials were used:

2014 aluminium alloy: (3.32% Cu-0.91% Mn-0.79% Si). The contact surface was etched with a solution of hydrofluoric acid (20%) in water for 1 min, which increased surface porosity by dissolving part of the crystalline phases.

A ceramic made of alumina (96%), magnesium and calcium oxides. The contact surface was etched with a solution of hydrofluoric acid (20%) in water for 1 min, which increases surface porosity by dissolving part of the crystalline phases.

The specimens had two different contact areas $(3.135 \text{ and } 16.000 \text{ cm}^2)$ so as to analyse the influence of the weld size in the process. This change in scale made it necessary to alter the heating and load conditions to ensure the equivalence of the thermal cycles undergone by the specimens and the pressures exerted on them. The only difference observed was that when the



Figure 4 Experimental device used in the welding process.



Figure 5 Al-Cu diagram. The vertical arrow indicates the alloy employed.



Figure 6 Temperature-time diagram used in the specimens.



contact area is larger, a longer stress relieving treatment was required after welding.

As mentioned earlier, it is essential to achieve good contact between the surfaces to be welded. To this end, the material was heated to $640 \,^{\circ}$ C at 0.15 MPa. An incipient fusion took place (Fig. 5) as well as the plasticization of the alloy, which flowed through the pores and cracks of the ceramics, producing a mechanical fastening. This whole process occurred in an argon atmosphere which prevented the oxidation of the aluminium. The mechanical fastening obtained was made easier by the metal creep at high pressures and temperatures and the high porosity of ceramics.

Once we had determined the minimum deformation of the aluminium alloy required to produce a sound binding, a steel mould was used (coated in graphite for easier demoulding). This mould made it possible to carry out hot-forming and, at the same time, it limited

Figure 7 (a) Al-Al₂O₃ welding. (b) Welding of two pieces of Al₂O₃ with an Al sheet.

the stroke of the pistons by interrupting the pressure exerted on the specimens (Fig. 4). After welding, a cooling process in inert atmosphere ends the thermal cycle (Fig. 6). This method has enabled us to obtain sound welds of alumina and aluminium and between pieces of alumina (Fig. 7).

4. Results

4.1. Microanalysis and microscopy

Stress relieving and cooling times were long enough to allow the migration of aluminium atoms, producing substitution reactions among them. The resulting aluminium oxide is not chemically different from the original alumina; this explains the impossibility of detecting it by EDX microanalysis. No significant amount of copper was found in the interface or in the ceramics, which proves that no metallides were formed during the tests.

Optical microscopy shows how the metal penetrated the pores and cracks of the ceramic surface, establishing a sound mechanical fastening (Fig. 8). When plasticization was incomplete, some pores in the interface remained open (Fig. 9). A too rapid cooling of the contact area caused cracks in the ceramics (Fig. 10), making for a more fragile joint.

Closer observation with a transmission electron microscope (TEM) shows that the metal had adapted to the ceramic surface, and also revealed some areas in which the aluminium alloy has reached its melting point and slowly solidified later (Fig. 11).

4.2. Impact tests

Several impact tests were carried out on welded aluminium alloy-alumina specimens bound with a rigid adhesive, and on annealed aluminium alloy-alumina specimens.

In these impact tests the Hopkinson bar was used. This device consists of two steel bars of a high elastic limit. The specimen was put between these two steel bars; a projectile was aimed at the free end of the first



Figure 8 Penetration of Al through the cracks and pores of the Al_2O_3 .



Figure 9 Defective contact surface (too porous).



Figure 10 Cracking of a small area of the ceramic welded to Al, caused by the differences in contraction during a medium speed cooling process.



Figure 11 Al penetration into the ceramic surface defects.



Figure 12 Hopkinson Bar impact test results for a welded $Al-Al_2O_3$ specimen.

bar and, on impact, an elastic compression wave was generated. When this wave reached the surface of the specimen, it was partially transmitted and partially reflected. A record of these waves was kept in each test (Fig. 12) and this enabled us to estimate the energy loss.

It is obvious that this loss cannot be put down solely to the deformation or breakage of the material;

TABLE II Energy loss								
Material	Test without specimen	Al-2014 annealed	Al ₂ O ₃	Al–Al ₂ O ₃ rigid bounding	Al-Al ₂ O ₃ welding			
Energy %	7	30	27(*)	27	40			

(*) In Al_2O_3 energy loss is 16% for no broken specimens.



Figure 13 (a) Specimens used in the impact tests. (b) Specimens after the impact tests, which show that there are no cracks in the welded area

other factors must be taken into account, such as friction in the contact areas between the specimen and the steel bars. Consequently, the results of these tests can only be considered as qualitative guidelines for the performance of the different materials tested. A series of tests without specimens were also made, to determine the energy loss of the system itself.

The specimens used were 20 mm diameter and 10 mm height cylinders (Fig. 13).

Projectile speeds were $16-17 \text{ m s}^{-1}$.

Table II shows a summary of the results obtained. These suggest, if only qualitatively, that the welded specimens absorb more energy than the others. In fact their performance is more satisfactory than that of the parts bound with rigid adhesive, which shows that the weld strengthens the material. The fact that the cracks in the ceramics due to the impact did not appear in the welded interface leads to the same conclusion (Fig. 13).

5. Conclusions

A sound and resistant weld was obtained between two materials of highly different expansion coefficient, without using any type of interband.

The welding process carried out is simple and easy to reproduce; it employs an argon atmosphere, not very high temperatures and low pressures, which makes it possel to use it at industrial level.

The quality of the binding was assessed by means of impact tests. As expected, the ceramic material breaks on impact, but the cracks do not appear in the weld interface, as in the case with specimens bound with rigid adhesive. Therefore, the welding process has strengthened the material.

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