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Short communication

A new method for bonding alumina to the alloy titanium-zirconium-molybdenum

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ABSTRACT

Assemblies of TZM alloy with alumina were obtained with a two-step process, much shorter than moly-manganese brazing methods. This new process involves, first, the preoxidation of the alloy in air at 795 °C, followed by hot-pressing in flowing argon, at 1700 °C under 22 MPa. The adherence of the joints, higher than 45 MPa, was explained by two complementary reasons: i) a mechanical contribution, of “tenon and mortise” type and ii) the penetrations of secondary phases (MoO₂ and minor phases coming from alumina additives) into both materials that play the role of nails joining the alloy and the ceramics.

1. Introduction

Many applications need strong bonding between ceramics and metals. Brazing is the most widely spread method; it was well known for long, and particularly the moly-manganese process [1–3]. This process consists in covering ceramics by several coatings in order to create physical and chemical continuities when going from ceramics to metals. Rather strong assemblies were obtained with this process, which presents also several serious drawbacks:

- i) The great number of successive steps, including the reduction of oxide layers, makes this process expensive;
- ii) Industrialization is difficult (the junctions are achieved manually, at least partially);
- iii) The presence of molybdenum trioxide with a low melting point (795 °C) limits the use of such assemblies to relatively low temperatures.

Alternative solutions for bonding ceramics to metals were sought for a long time, especially the diffusion bonding, well-known for ceramics / ceramics or metal / metal junctions [4]. For ceramic-to-metal junctions, the main problem is that there is no reaction thermodynamically possible between the usual ceramics (alumina, zirconia,) and metals, except with the alkaline and alkaline earth metals that have no industrial applications. Indeed, insofar as the formation of interdiffusion layers between both materials is required to obtain a strong diffusion bonding, the two materials must react together: unfortunately the corresponding Gibbs functions are always positive for the reactions of ceramics with the usual metals. From this, the proposed solutions involve at least one

extra interlayer, to be placed at the metal / ceramic interface, and many tests have been attempted: ductile metals such as copper [5] giving a mechanical coupling, in particular because of the surface roughness; additives inside the materials giving a secondary phase [6]; glass interlayers [7,8], titanium and/or zirconium fillers for active brazing [3].

Now, for high temperatures applications, which are particularly sought in military devices, no satisfactory process has been proposed. The specifications for such applications are: resistance at high temperature, good adherence and air tightness. In these conditions, the metals to be considered are not numerous, because they must have high melting points (while, in this respect, most ceramics are good candidates). In the present work, molybdenum was chosen because it is one of the most used among the high melting points metals. In fact, the alloy called TZM, which contains 99.3 at.% of molybdenum, with about the same melting point as molybdenum ($T_F = 2620$ °C, [9]), was preferred because it contains small amounts of titanium and zirconium carbides that gather at the grain boundaries: they block the microstructure and increase the recrystallization temperature, which is interesting for high temperature applications [10]. For the ceramic part, alumina was chosen because of its widespread use.

This couple of materials was also chosen for thermal expansion reasons: the thermal expansion coefficients (TEC) of TZM and alumina are close, in the range $5.5 \times 10^{-6} \text{ K}^{-1}$ [11]– $8.0 \times 10^{-6} \text{ K}^{-1}$ [12], according to temperature. This characteristic is of great importance because, when cooling after performing the bonding at high temperature, the TEC differences induce shear stresses at the ceramic / metal interface, which must be minimized in order to avoid the fracture of the joints.

Assemblies such as TZM / alumina have never been studied before.

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The only published works that associate Mo and Al₂O₃ are articles devoted to molybdenum – aluminium composites [13], or to thin molybdenum films deposited on alumina [14]; they never brought out any reactivity between these materials. The lack of studies on this kind of assemblies probably lies in the reactivity of molybdenum with oxygen, from 500 °C [15], and, likely, it seemed useless to study junctions intended for high temperature uses, when one of the materials (Mo) rapidly corrodes in air. Now, for some new applications, with short durations of use, such assemblies could be of great interest.

About the assembly method selected for the present study, despite the fact that moly-manganese processes could probably be enforced for Mo / Al₂O₃, the chosen route was that of the alloy's preoxidation, followed by hot-pressing (HP). This process that associates preoxidation and HP has never been tested before, neither in this system nor in another one. Preoxidation aims at forming directly an oxide layer susceptible to favour the bonding; this method was already successfully used for other ceramic-to-metal junctions (e.g. for joining 304L steel with alumina [16]), and HP has been known for a long time to give strong ceramic-to-metal bonding [6,17,18].

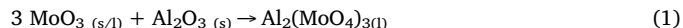
2. Experimental

The TZM and alumina pieces were discs, 10 mm in diameter, 3 mm in thickness cut with a diamond saw from bars provided by Goodfellow Inc. The impurities content was low for both the materials (< 1 at.% for alumina, with a secondary phase mainly composed of silicon, calcium and sodium mixed oxides), while the Mo-based TZM contained Ti (5000 ppm), zirconium (700 ppm), carbon (500 ppm) and tungsten (200 ppm), with < 190 ppm for the other impurities content. The discs were carefully manually polished up to Ra = 0.06 µm, with silicon carbide paper (4000 mesh).

The first step of the process consisted in the preoxidation of the alloy in a furnace, in air, during 30 min at 795 °C, with the heating and cooling rates of 0.5 °C s⁻¹. The choice of working in air was guided by searching for a process as simple as possible in order to make easier the possible future industrialization. During this step, the pieces were superimposed, with TZM at the top and without pressing. The conditions of preoxidation were empirically adjusted in order to obtain pieces already bonded after this treatment: for shorter or longer times, or for lower or higher temperatures, no bonding was achieved. The so-obtained assemblies, shown in Fig. 1, exhibited a crystalline interlayer, more than 100 µm thick, which joins the ceramic and alloy discs. The two pieces could be easily separated by hand, but the fact that they were bonded, even weakly, favoured their further introduction in the HP furnace.

When separating the two discs, the fracture occurred inside the

interfacial zone and the two half were analyzed by X-ray diffraction (XRD, Bruker D8 Advance, Germany) with a copper anticathode and a backward monochromator. The corresponding XRD patterns, as the one given in Fig. 1b, identified mainly molybdenum trioxide MoO₃, but also small amounts of aluminium molybdate Al₂(MoO₄)₃, on the alumina side. MoO₃ was the expected oxide for the treatment temperature chosen (795 °C) that is precisely the melting point of MoO₃ [15]. The presence of molybdate indicates that MoO₃ attacked alumina, following the reaction:



One may notice that the molybdate formation could result either from a solid state reaction or from a solid-liquid one, since MoO₃ was solid or began to melt. The molybdate Al₂(MoO₄)₃, was liquid at 795 °C, since its melting point is 705 °C [19].

The major points were that, after the first step of the process, the discs of alumina and of TZM were weakly bonded, and the surfaces of both the materials became rough because of the oxidation (for the alloy) and because of the reaction with the molybdenum trioxide (for the ceramics), as illustrated in the Fig. 2, which shows also the big crystals of the interfacial zone.

The second step of the process was the hot-pressing of the assembly in a graphite furnace equipped of a single-acting press (LPA Goliath, France). The samples were placed in the graphite die schematized in Fig. 3. The experimental conditions were the following: pressure: 22 MPa; temperature: 1700 °C; heating and cooling rates: 0.25 °C s⁻¹; duration: 3 h; atmosphere: flowing argon (0.032 L s⁻¹).

During the treatment, a liquid phase was driven out the interfacial zone and reacted with the graphite die. For avoiding this harmful reaction, the samples were first wrapped in a papyex[®] sheet (flexible graphite, Mersen, France), before introducing in the die.

After hot-pressing, the samples were strongly bonded. The interface was observed in cross section (Fig. 4) with a scanning electron microscope (SEM, JEOL IT 300, Japan) using the backscattered electrons (BSE) mode.

Fig. 4a presents a general view showing that the interlayer between TZM and alumina, initially more than 100 µm thick, almost disappeared except for a thin grey layer that remained, by places, and which is well visible in the magnification of Fig. 4b. This was the remains of the interlayer, most of which was expelled from the interface under the action of the pressure, as described above.

On the TZM side, many dark grey inclusions appeared, close to the interface, and some of them penetrated deep inside the alloy, up to more than 250 µm. The dark colour indicates that this phase contained light elements, which was confirmed by the microanalyses by energy

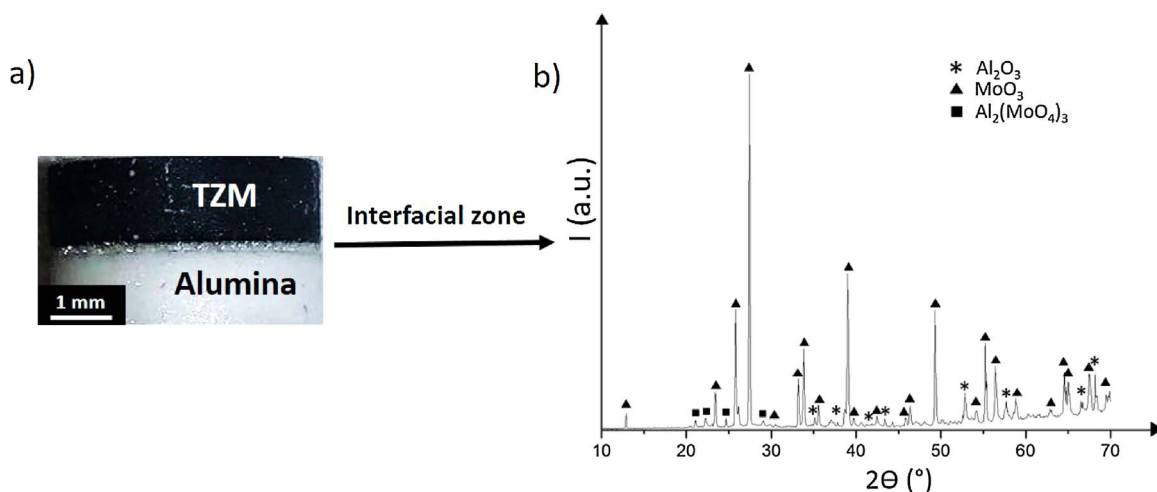


Fig. 1. The assembly TZM / alumina as obtained after preoxidation and before HP:(a) general view and (b) XRD pattern of the interfacial zone after fracture (alumina side).

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