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SHEAR STRENGTH OF AL_2O_3-NB JOINTS PRODUCED BY BRAZING USING A CU-ZN ALLOY AS JOINING ELEMENT

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Abstract: Metal brazing is a widely used technique to join advanced ceramics and metal. In this study, we have investigated the microstructure and mechanical properties of Al_2O_3/Nb joint brazed using a 70Cu-30Zn alloy as joining element. Cylindrical Al_2O_3 disks were produced by sintering of powder at 1550°C and the Al_2O_3/Cu-Zn/Nb/Cu-Zn/Al_2O_3 arrangements were bonded at 920°C to 980°C in argon atmosphere. Effective bonding of Al_2O_3 to Nb was observed at 950°C and 980°C, however joining was not possible at 920°C. Scanning electron micrographs of the reaction layer show the growth of the interface zone increasing with both bonding temperature and time. Evidence of Nb diffusion towards the joining element forming spherical Nb-rich precipitates near to the Al_2O_3 bonding line was evident from the SEM observations. Mechanical characterization of the joints at 980°C showed shear stress results varying from 57 to 127 MPa in samples joined for different times.
Keywords: Alumina ceramic; Brazing; Niobium; Cu-Zn, Shear test.

INTRODUCTION

Alumina (Al_2O_3) is an advanced engineering material used in a wide range of applications, including refractories, structural materials, electronic packaging, catalysts, and sensors (RUYS, 2018) With the development of new ceramic materials the demand to join ceramics to metals has increased. There are three general categories or types of joining process: *mechanical, direct, and indirect joining* (JANCZAK-RUSCH et al, 2014; LAN et al, 2014). At present, there are several mature technologies for the bonding technologies of ceramics and metals, some need an intermediate liquid phase, brazing, and others are produced by diffusion bonding [FIRMATO ET AL, 2011), that are well known in industial manufacturing. Among them, brazing has received far-reaching attention due to its convenience and cost-effectiveness (ZHAO et al, 2015; ZHANG et al, 2002). The liquid state bonding or brazing method, is used to joint ceramic/metal systems; the filler material of brazing is a metal alloy, or, metal glass which melts at high temperature forming a joint interface; moreover, in brazing is necessary a good wettability of the filler metal on the ceramic surface (NASCIMENTO et al, 2005).

In most cases, ceramics must be joined to metals with sufficient bond strength because the poor workability and brittleness of ceramics can be compensated by the metal. Ceja-Cárdenas et al. (2013) recognized the development of diverse Nb-silicide´s in the course of bonding Si_3N_4 to Nb-foil interlayer and using a Cu-Zn foil as joining element by brazing. This may cause the development of tensions at the interface that could result in a premature failure of the joint. On the other side. Mick et al. (2015) have studied a technique to produce Al_2O_3/Ti implant components using glass solder bonding. Two main problems normally occur when joining ceramics to metals: the first is the poor wettability of ceramics by most metals and metallic alloys, which can be overcome by using an active filler alloy. The active element in the filler alloy, such as Ti, Zr, has sufficienlty large driving force to destabilize the ionic or covalent bonding in the ceramics and chemically reacting with one or more elements in the ceramic. The second problem is the significant differences in the physical and mechanical properties betwwen the ceramic, the metal and the brazing alloy, such as coefficients of thermal expansion (CTE) and Young's modulus. These differences can lead to high residual stresses when cooling from the brazing temperature, leading to the component fracture under a low load (BLUGAN et al, 2007; XU, 1994; PARK, 2004;

PARK et al, 2002)]. On the other side, Lemus et al. (2015) study brazing of Al_2O_3 to Ti using a Au-foil as joining element. Previous to joining process Al_2O_3 samples were coated with 2 and 4 μm thick of Mo layer and then stacked with Ti. Al_2O_3–Mo/Au/Ti combinations were joined at 1100°C in vacuum. They observed that the interface shows the formation of a homogeneous diffusion zone, and Mo diffused inside Au forming a concentration line.

Therefore, it is important to study the microstructure and mechanisms of interface formation between metal and ceramic and its relationship to the mechanical strength of the interface. For that reason, the main objective of this work is to study various aspects of the liquid state diffusion bonding of cylindrical samples of Al_2O_3 and commercial pure niobium (99.7%) by brazing using a 25-μm-thick 70Cu-30Zn (wt%) alloy as the joining element.

EXPERIMENTAL METHODOLOGY

Cylindrical monolithic Al_2O_3 disk shape samples, with a 7 mm diameter and 3 mm thickness were initially produced by sintering of α-Al_2O_3 commercial powders using a Carbolite furnace at 1550°C for 120 minutes. Additionaly for joining, commercial pure Nb-foil (99.98%) of 0.25 mm thickness (*Aldrich Chemical Company, Inc. USA)* and a 70Cu-30Zn (%wt) foil of 0.025 mm (25 μm) thickness *(Johnson Matthey Company, USA)* were used. The achievement of the bonding process depends of several factors; an important one is the surface roughness of the joining materials as it controls the initial contact area between the metal/ceramic pair. Therefore, joining experiments were initiitiated with the surface preparation of the materials to be joined. To ensure reproducibility a polishing procedure was followed where all the samples were ground using silicon carbide paper (320 grit) to promote a similar surface roughness finish. Sandwich joint configuration consisted of two disks of Al_2O_3 mounted uniaxially with a disk of Nb and 70Cu-30Zn foil between the metal and the ceramic components with the polished surfaces in contact. The sandwich specimens were positioned in a graphite die immersed in a boron nitride (99.5% pure) powder bed; the purpose of the powder bed was to avoid contact between the sample and the internal walls of the graphite die (CASTRO-SÁNCHEZ, 2017). The joining experiments set-up consisted of a resistance furnace with an millite tube chamber of 80 cm in length by 10 cm in diameter with one side closed. The graphite die, with the joining sandwich, was placed in the furnace, closed and filled with argon, and heated to the preset joining temperature, see Figure 1.

Al_2O_3/Cu-Zn/Nb/Cu-Zn/Al_2O_3 joining experiments were carried out at temperatures of 920°C, 950°C, and 980°C for different holding times. Interface characterization was done on cross-sections of polished joints. Images and analyses of the interfaces were achieved with a JEOL JSM-6400 scanning electron microscope. Mechanical evaluation of the joints was obtained at room temperature using an universal testing equipment with a 25 kN load cell. The load was applied at a constant vertical speed of 0.1 mm/min until fracture. The schematic illustration of the shear test is exposed in Figure 2. Three samples obtained at the specified temperature and time conditions were evaluated and the average the joint strength was calculated.

RESULTS AND DISCUSSION

SINTERING OF AL_2O_3 POWDERS

Disk shape samples measuring 7 mm in diameter and 3 mm in thickness were formed by sintering highly homogeneous commercial Al_2O_3 spherical powders with a size distribution below 100 nm at 1550°C

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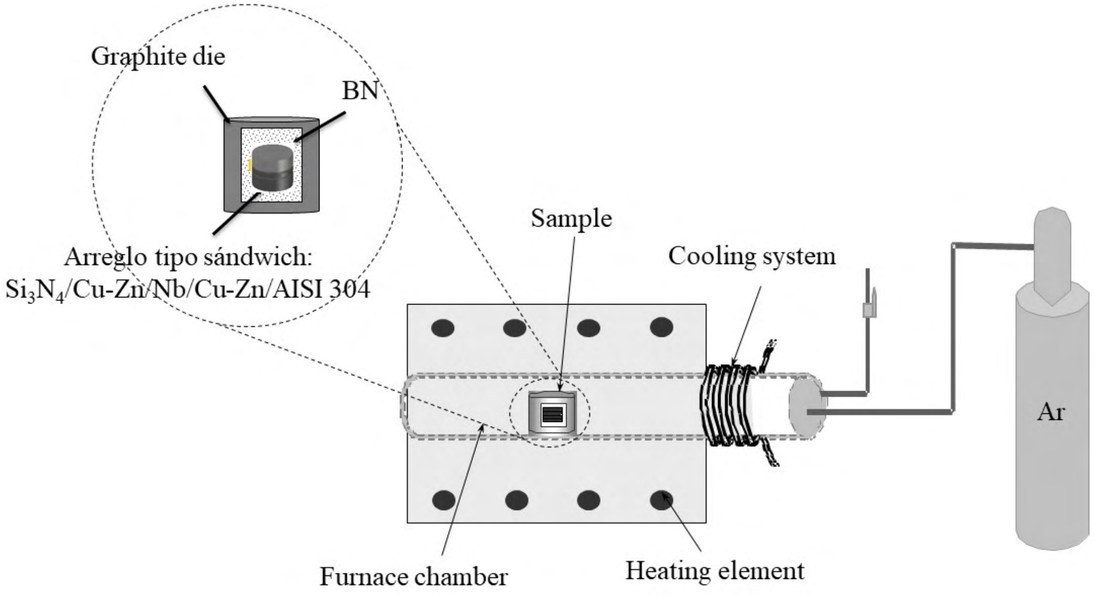

Figure 1- Schematic representation of the samples assembly inside the graphite die

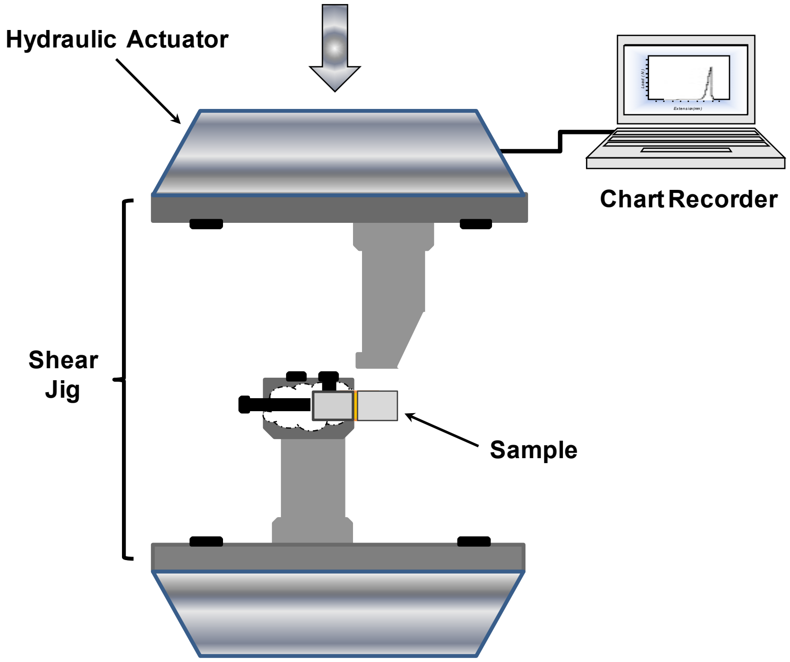

Figure 2- Representation of the mechanical setup used to evaluate the interface shear strength of the joints

for 120 minutes. X-ray diffraction analyses confirmed the formation of the α-Al_2O_3 phase structure as shown in Figure 3a. A relative density higher than 90%.was achieved under these particular conditions. Figure 3b shows a fractured Al_2O_3 sample displaying the nanometric grain structure.

JOINT´S INTERFACE BEHAVIOUR

Bonding of alumina was investigated by joining sample combinations of Al_2O_3/Cu-Zn/Nb/Cu-Zn/Al_2O_3 at temperatures of 920°C, 950°C, and 980°C under argon atmosphere and holding times varying from 5 to 35 min. In thermall processes, a small temperature variation will result in significant deviations in the process kinetics compared to other less dominant parameters. In this case, plastic deformation and diffusion mechanisms are both highly sensitive to temperature variations. In this study results showed that joining of Al_2O_3/Cu-Zn/Nb/Cu-Zn/Al_2O_3 combinations was achieved successfully at all temperatures and holding times, with evidence of Al_2O_3 and metals interaction. However, for samples bonded at 920°C and 950°C detachment occurred during the metallographic preparation. This is related to a low joining strength of the samples produced at solid-state process since the liquid growth at temperatures exceeding 950°C. However, to examine the interfacial contact during the joining process, interface inspection was done on a cross-section of the samples joined at 980°C.

Bonds of Al_2O_3/Cu-Zn/Nb/Cu-Zn/Al_2O_3 were formed through the creation of a reaction zone by the conduction and contact of the Cu-Zn foil with Nb. Cross-sections of the Al_2O_3/Cu-Zn/Nb/Cu-Zn/Al_2O_3 interfaces for samples joined at 980°C for a) 5 and b) 25 min are shown in Figure 4. The interfaces formed by a homogeneous reaction zone created by a chemical interaction arising from the high attraction of Cu, Zn, and Nb.

At the bonding interface of combinations produced at 980°C for 5 min, a crack can be detected along the bonding line with Al_2O_3 as shown in Figure 4a, which was perhaps formed throughout the grounding of the samples. Contrary to this, joints produced at 980°C but longer times, 15 and 25 min, show comparable interface with out porosity and free of cracking. On the other side, increasing time from 5 to 25 min did not increase the interface thickness; however, the creation of a spherical Nb-rich precipitates intermixed inside the reaction zone near the joining line with the Al_2O_3 is present, and the amount of these precipitates increases with time. Figure 5a and 5b show a backscattering image of the zones marked in the cross-section interfaces of Figure 4 for the joints produced at 980°C for 15 and 25 min, respectively, where spherical Nb-rich precipitates can be clearly observed. The EPMA-EDS shows that the highest element in the precipitates phase is Nb, which increases in amount when time rise from 15 to 25 min; nevertheless, the thickness remains practically constant.

An overview of the different components across the Al_2O_3/Cu-Zn/Nb interface was obtained using element line scan analyses of the joints produced at 980°C for 25 min, the results are showed in Figure 6. The components analyzed were Nb, Cu, Zn, O and Al. Analysis was performed starting on the Nb sample over the reaction interface passing through the reaction phases up to the pure Al_2O_3. In the Nb composition profile, it was possible to observed its diffusion inside the interface and obtain its concentration in the spherical Nb-rich precipitates. It was not observed diffusion of Cu or Zn into the ceramic or Nb, resulting in the drop of the Nb concentration profile with a concentration reduction for the Cu and Zn profiles inside the spherical precipitates. Diffusion of Cu, Zn or Nb into the ceramic is

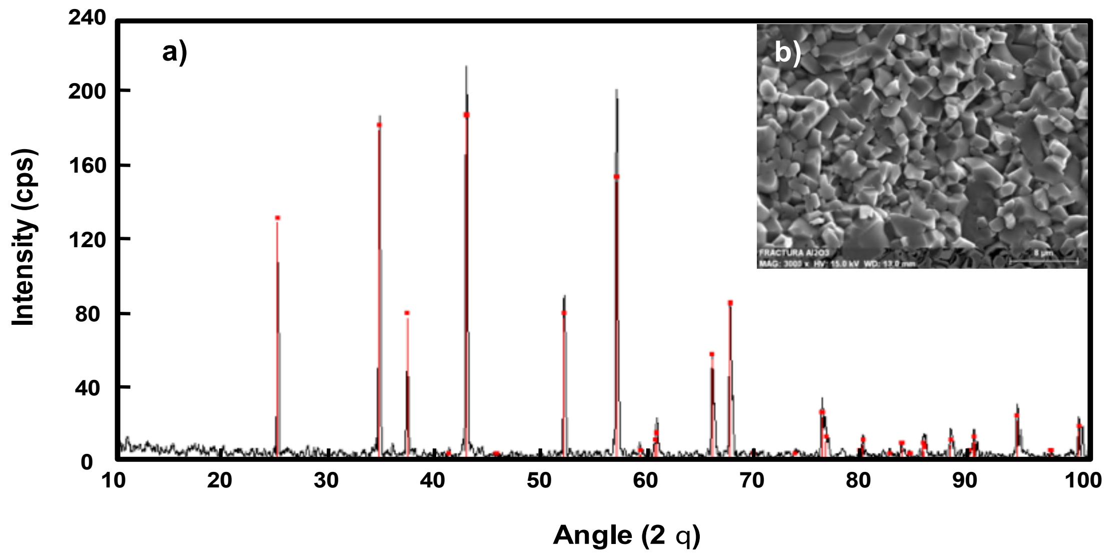

Figure 3- a) X-ray diffraction pattern and b) fracture surface of Al_2O_3 samples sintered at 1550°C for 120 minutes, showing the α-Al_2O_3 phase structure

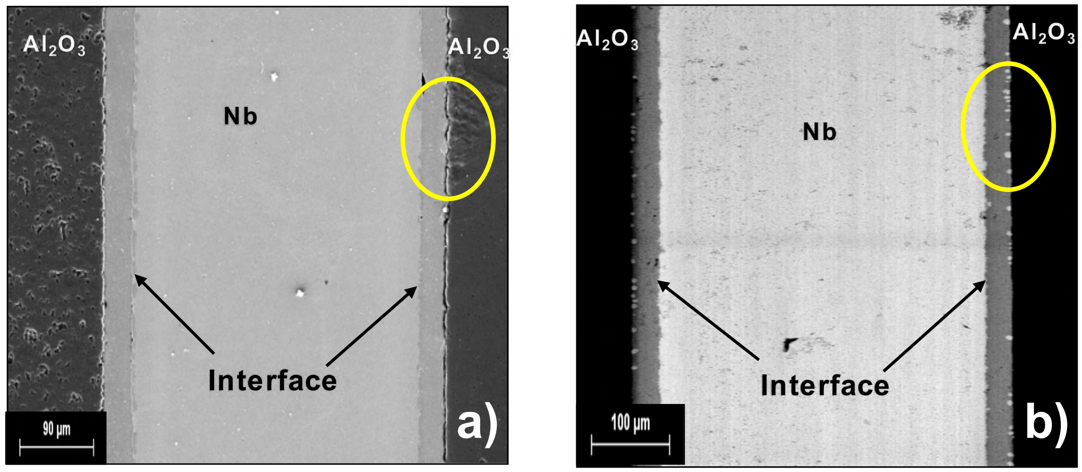

Figure 4- Cross-section of Al_2O_3/Cu-Zn/Nb/Cu-Zn/Al_2O_3 interfaces for samples joined at 980°C for a) 5 and b) 25 minutes in argon

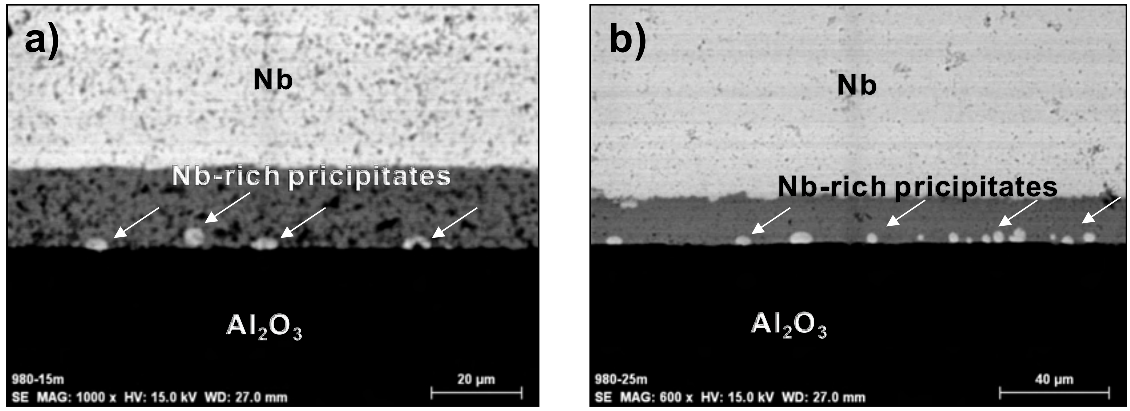

Figure 5- Cross-section of Al_2O_3/Cu-Zn/Nb/Cu-Zn/Al_2O_3 interfaces for samples joined at 980°C for a) 15 and b) 25 minutes in argon showing the spherical Nb-rich precipitates

limited. At the same time liquid creation plays an main part during joining as it increases the speed of interface creation, improving the interaction zone. Consequently, the chemical contact is more pronounced, promoting fast distribution, since liquid is much faster than solid state diffusion. These results are in good agreement with previous experimental findings by Lemus and Drew (2003).

In general, when a ceramic is in contact with a metal, an interaction is estimated to happen. Virtually all diffusion bonding and sintering mechanisms as well as plastic deformation, are sensitive to heat. However, the main point to produce a positive assembly technology lies in the capability to change the interface to accommodate the unlike forms of chemical attachment, i.e. from metallic bonding for the metal to the ionic or covalent bond for the ceramic, to decrease the electronic discontinuity at the combined surfaces. This interface is also indispensable in decreasing the unfavourable things due to two aspects: thermal mismatch and chemical changes between the materials, metals and ceramics (ELREFAEY AND TILLMANN, 2009).

Joints of the Al_2O_3/Cu-Zn/Nb combinations were possible through the creation of a reactive zone as a effect of diffusion and reaction of Cu and Zn with Nb and Al_2O_3, which can be explained by the fact that diffusion takes place faster in metals than in ceramics (LEASAGE, 1994). The EPMA analyses revealed that Nb was partially dissolved and migrated to the Cu-Zn zone forming the spherical Nb-rich precipitates; these analyses also demonstrated that Zn partially evaporated from the Cu-Zn joining element. According to the Cu–Zn phase diagram, when Zn evaporation occurs the melting point of the system increases; rendering the liquid phase more stable for longer periods and ultimately following a complete depletion due to the presence of diffusion elements, which leads to a solid phase diffusion bonding mechanism. Similar behavior was observed by Zhang et al. (2003) who reported the evaporation of Zn at 950°C during self-joining of silicon nitride for various heating times, using (CuZn)85Ti15 as the filler alloy. They concluded that by increasing the holding time on heating, the thickness of the reaction layer increases, whereas the reaction products containing Zn can be hardly observed due to the evaporation of zinc. Reaction products formed inside the interface lead to a different type of joint between the metal and ceramic, and the relative efficiency of these products on the strength of Nb/Al_2O_3 interfaces is not fully understood to this date. However, most reaction layers are brittle and thus potentially detrimental to the interface mechanical properties. In addition, the thickness of the reaction zone in joints is a function of the joining temperature and may dominate the final strength.

MECHANICAL JOINT BEHAVIOUR

The strength of diffusion-bonded joints depends on the nature and microstructure of the interface between the joining materials. To determine the mechanical properties of the joints, samples of Al_2O_3/Cu-Zn/Nb/Cu-Zn/Al_2O_3 were subjected to shear testing. The resistence to fracture was determined for samples bonded at 980°C for 10 to 35 min. The results are shown in Table 1 and represent the average joint strength of at least three samples for each set of experimental conditions. It can be observed that the shear strength of the joints increased from 56 MPa to 127 MPa as a function of bonding time. This can be associated with the formation of spherical Nb-rich precipitates intermixed inside the reaction layer and near the bonding interface with Al_2O_3; the amount of this phase increases with increasing holding time.

The effect of a reaction layer on the interface

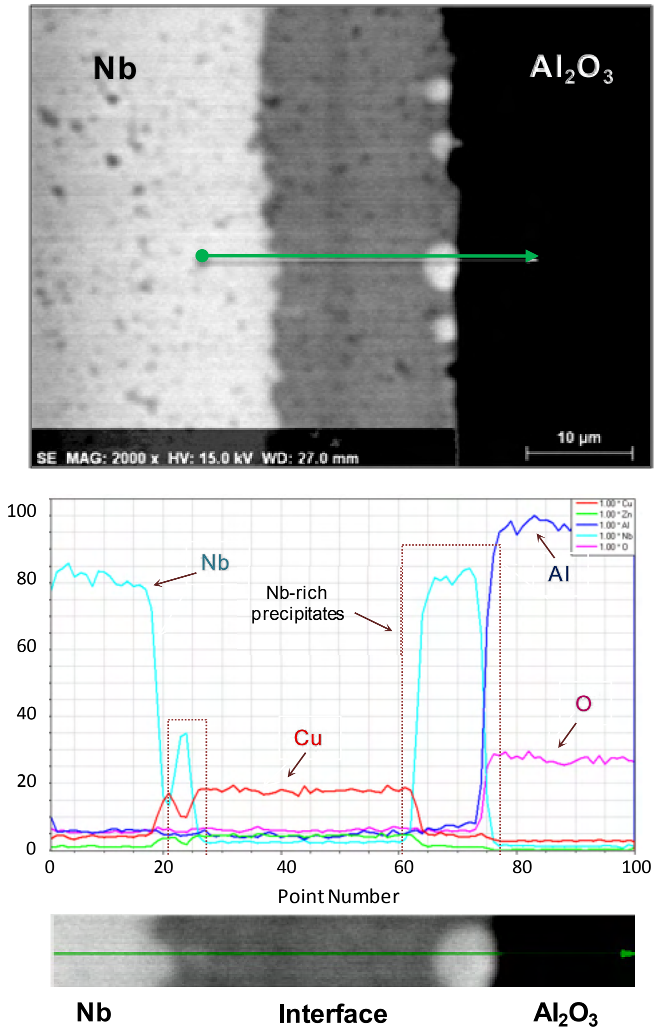

Figure 6- Line chemical analysis across the Al_2O_3/Cu-Zn/Nb interface obtained at 980°C for 25 minutes in argon

Joining of Al_2O_3/Cu-Zn/Nb/Cu-Zn/Al_2O_3 at 980C				
Joining time (minutes)	10	15	25	35
Shear Strength (MPa)	56±15	100±11	116±13	127±08

Table 1- Shear strength as a function of time for samples joined at 980°C

strength depends on a number of factors such as the mechanical properties of the reaction layer and its thickness and morphology, in addition most reaction layers are brittle and thus potentially detrimental to the interface properties. It is clear however that the amount of interfacial reaction played a major role in determining the final mechanical properties of the joints. Furthermore, the nature of the reaction products may also have influenced the mechanical properties of the joints. The maximum shear strength values of the joints (127 MPa at 980°C for 35 min) are comparable to those reported by Chen et al. (2012) of 154 MPa for WC-Co joined to 3Cr13 stainless steel at 1100°C for 10 min, employing Ni electroplated on Cu-Zn alloy as the filler element. They attributed this behaviour to the formation of an interdiffusion zone promoted by the Ni, which enhanced the bond strength. All joint fracture samples showed the same type of fracture mode as shown in Figure 7. The fracture originated and mainly propagated along the Al_2O_3/Nb reaction zone interface with fracture occurring at the interlayer.

Fractography studies show a very homogeneous and predominantly defect-free Cu-Zn brazing surface. This finding indicates good wettability of the joining element. Figure 7a shows the Al_2O_3 areas of the fracture surface of the alumina phase. The surface contained alumina and only a small amount of the joining elements. However, Figure 7b shows that the fracture surface of the niobium compound was nearly entirely covered with the Cu-Zn material of joining element, which was confirmed by EDS analysis. However, Al_2O_3 regions are observed in the niobium side, indicating a strong relationship between the microstructure of the interface and the mechanical strength of the joints. Lemus and Aguilar (2004) observed that as the thickness of these interfaces increases, the joint strength increases at first owing to the creation of a strong bond integrity, reaching a maximum at a certain thickness and then descreasing as the interface continues growing.

Therefore, the reaction layer thickness must be controlled to ensure good joint strength. It is clear that the amount of interfacial reaction played a major role in determining the final mechanical properties of the joints. Furthermore, the nature of the reaction products may also have influenced the mechanical properties of the joints. In summary, the choice of suitable conditions to prepare ceramic/metal joints requires knowledge about the mechanism of the reaction between the materials and the evolution of the interface.

CONCLUSIONS

Based on the results presented in this work, we have demonstrated that:

- It is possible to join Al_2O_3 to Nb using a Cu-Zn foil as joining element.
- Effective bonding was observed at 950°C and 980°C, however bonding was not possible at 920°C.
- Bonding of Al_2O_3 to Nb occurred through the formation of a homogenous diffusion zone on the metal side of the joint.
- Diffusion of Nb inside the reaction interface occurred, forming a spherical Nb-rich precipitates near the bonding line with Al_2O_3. However, the diffusion of Cu, Zn or Nb into the ceramic material is unclear.
- Mechanical evaluation carried out by shear testing resulted in a maximum average value of 127 MPa in joints produced at 980°C for 35 minutes.
- Fracture surface of the Al_2O_3 shows a small amount of the joining elements, however fracture surface of the Nb was nearly entirely covered with the Cu-Zn

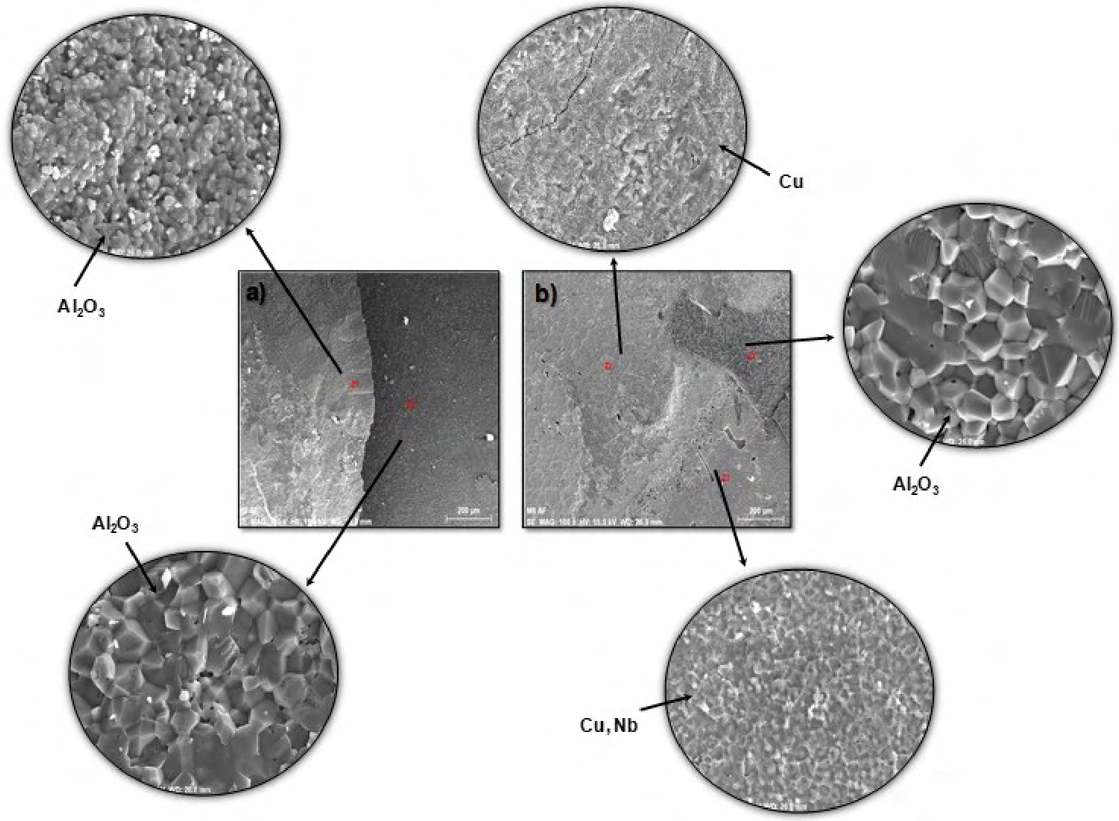

Figure 7- Fracture of Al_2O_3/Cu-Zn/Nb samples after shear testing, a) Al_2O_3 and b) Nb side, showing a residual alumina material remaining on Nb

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material of joining element.

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