INTERNATIONAL JOURNAL OF AUTOMOTIVE AND MECHANICAL ENGINEERING
ISSN: 2229-8649 e-ISSN: 2180-1606
VOL. 21, ISSUE 1, 2024, 11010 – 11017
DOI: https://doi.org/10.15282/ijame.21.1.2024.05.0851

RESEARCH ARTICLE

Solid State Diffusion Bonding of Alumina with Aluminum Alloy

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ABSTRACT - The objective of this work is to study the effect of time during the bonding process of alumina with an industrial aluminum 6060 alloy by the technique of solid-state welding at 600°C. Optical microscopy, scanning electron microscopy with energy dispersive spectroscopy, corrosion tests, and microhardness measurements were used as characterization techniques. The extended time during the bonding process had an effect on the microstructure of the interface and its mechanical properties. Oxygen diffusion was detected across the aluminum alloy/alumina interface. A recrystallization reaction developed on the aluminum side during the bonding process, which affected the aluminum alloy hardness values. By bonding metal to ceramic, it allows the use of ceramics in automotive engines to reduce weight, increase thrust-to-weight ratio and operating temperature, and improve working efficiency.

ARTICLE HISTORY
Received : 08th July 2023
Revised : 16th Nov. 2023
Accepted : 15th Dec. 2023
Published : 20th Mar. 2024

KEYWORDS
Solid-state diffusion
Bonding
Alumina
Aluminum
Mechanical properties

1.0 INTRODUCTION

In materials science, materials are classified into four families: metallic materials, ceramic materials, organic materials and composite materials [1]. Metallic materials and ceramic materials are widely used in industry and each material has its advantages and disadvantages in terms of physical, chemical, and mechanical properties, i.e., each material has its own specific weak and strong properties [2]. Metals are strong, hard and excellent conductors of electricity but in general, their corrosion resistance and high temperature properties are rather poor. Ceramics can be used in an aggressive environment or under severe circumstances such as high temperatures [3]. A disadvantage is that they are fragile materials [4]. Establishing a bond between ceramic and metal can take full advantage of the complementary advantages of both materials in terms of economy and performance [2]. Several techniques have been used to join ceramics to metals, such as brazing and soldering, with or without ultrasonic waves, and solid-phase diffusion bonding [5]. Solid phase diffusion welding is a suitable method for joining ceramics to metal [1], unlike conventional fusion welding techniques such as arc, electron laser and beam welding which are not suitable for effectively bonding ceramics and metals [2]. Diffusion bonding is widely used in the joining of high strength and refractory materials in the aerospace, medical and nuclear industries[6].

Solid-state diffusion bonding is a joining method that forms strong bonds without causing melting of any phase for bonding the two base materials [7]. The diffusion bonding technique is defined by Derby [8] as: *"a solid-state joining process by which two nominally flat surfaces are held together at an elevated temperature using an applied interfacial pressure."* This technique makes it possible to assemble similar materials such as the welding of two metals or to assemble dissimilar materials such as the assembly of a metal with a ceramic [9]. It is known that the parameters that control the joining operation are time temperature, pressure, and atmosphere [10]. It has been found that the requirements of the simple reaction bonding technique are as follows [11]:

- The bonding temperature is below the melting point of the lowest melting material in the system, which is around 90% of the melting point of the material.
- Bonding time usually ranges from minutes to hours (2-3 hours).
- Clamping pressure of 0.5 to 1.5 MPa is necessary to have adequate contact between the surfaces of the materials to be joined.
- The contact surfaces of the two materials to be welded must be well-polished

Solid-state diffusion bonding can be used to bond a wide range of ceramic oxides and metals. For example, zirconia, alumina, beryllium oxide and alumina silicate ceramics can be bonded to valuable industrial metals such as gold, nickel, platinum, and copper[10]. It is important to mention that the structures containing ceramic/metal joints obtained by diffusion bonding can be used in industrial applications where these joints must have high mechanical strength, and also corrosion resistance, and optimal properties at high temperatures [12]. Metelkin et al. [13] investigated the diffusion bonding of four ceramic-metal joints. They joined copper with a variety of ceramic materials such as alumina, sapphire, forsterite and soapstone. Bondings were carried out under a vacuum as well as under a hydrogen atmosphere. The strongest joints were obtained under a hydrogen atmosphere. However, joints have generally failed in ceramics [14]. The

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bonding of alumina to copper has also been studied by Borbidge et al. [15] who found that the bonding process is sensitive to levels of impurities in the copper, especially oxygen, and, therefore, sensitive to the bonding atmosphere, relative to the oxygen content. In addition, some research works have been carried out on other ceramic/metal couples such as the study of Pt/A1203 [16] and Pd/MgO [17]. All couples mentioned above have been successfully bonded by the solid state diffusion technique.

However, limited research has been conducted on the solid-state diffusion welding between metals and alumina. For example, some joints were made between alumina and the following metals: nickel, stainless steel, titanium, Kovar, Nichrome, low-alloy steel, iron, and palladium. Satisfactory results have been obtained on all these ceramic-metal couples. Thamir et al. [18] investigated the solid diffusion bonding of alumina to some metals of high purity by diffusion (copper, silver, zinc, aluminum, tin, and lead). an observed that the bond strength decreases with atomic number, signifying the difficulty of the solid-state diffusion process with metals that have higher atomic numbers. Janghorbhan [19] has successfully assembled solid state diffusion ceramic-metal multilayers using alumina and different aluminum alloys; a spinel structure was observed for one of the couples. Allen et al. [11] studied three pairs of platinum-bonded alumina samples under the same conditions. However, despite using electron probe microanalysis and scanning electron microscopy, they were unable to detect any evidence of metal diffusion into the ceramic. According to Elssner and Petzow [20], the formation of bonds at the interfaces of different metal/ceramic material systems is not well understood and further research is needed to understand and optimize assembly processes for specific applications. The study of welding aluminum alloys on ceramic materials has been of particular interest to researchers. For example, Li et al. [21]studied the brazing of Al_2O_3 ceramic on an aluminum alloy 1A95 with the use of a ternary Al-Si-Mg alloy. They noted the effect of welding time on the properties of the welded joint. Among the aluminum alloys that need to be welded to ceramic materials, is aluminum alloy 6060 which contains, in addition to aluminum, a percentage of magnesium and silicon. This alloy has very good corrosion resistance, excellent formability and weldability [22].

It is in this context that this research work relates to the study of the bond at 600°C of alumina with an Al 6060 alloy through solid state diffusion process. Microstructural observations, chemical composition measurements, X-ray diffraction analyses, corrosion tests and microhardness measurements were carried out on the bonded dissimilar materials.

2.0 EXPERIMENTAL PROCEDURE

In this study, aluminum 6060 alloy and ceramic (alumina) were used as dissimilar base materials. The chemical compositions of the two materials are given in Tables 1 and 2. The ceramic used in this study is alumina Al_2O_3. It is made from a nanopowder and sintered at very high temperatures (1600 to 1700°C). This ceramic is marketed by HITECH CERAMICS (Chennai – India), in the form of white rods 12 mm in diameter.

Table 1. Chemical composition of aluminum 6060 alloy (Wt.%)

Al	Si	P	Cu	Mg	Zn	Fe
98.6	0.57	0.04	0.01	0.53	0.02	0.23

Table 2. Chemical composition of the alumina (Wt.%)

Compounds	Na_2O	CaO	MgO	SiO_2	Al_2O_3
Alumine	0.14	0.15	0.24	0.52	98.95

Before proceeding with the solid state diffusion bonding process, surface samples of aluminum 6060 alloy and alumina were polished and thoroughly ultrasonically cleaned in acetone. Then, the two samples were put in contact and clamped using a specific device under pressure of 15 MPa, as illustrated in Figure 1. The samples were heated to 600°C at a rate of 4°C/min and kept at this temperature for different times (2, 3, 4, and 5 min). To prevent sample oxidation, the bonding was performed under an atmosphere of hydrogen and argon. After each bonding time, the sample was cooled in air to room temperature. For the different characterization techniques, the four assembled samples were cut into two parts perpendicular to the bond line (Figure 2).

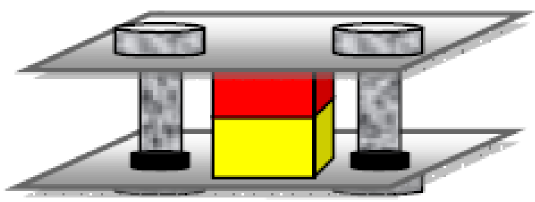

Figure 1. Solid-state diffusion bonding configuration applied for joining alumina to Al 6060 alloy

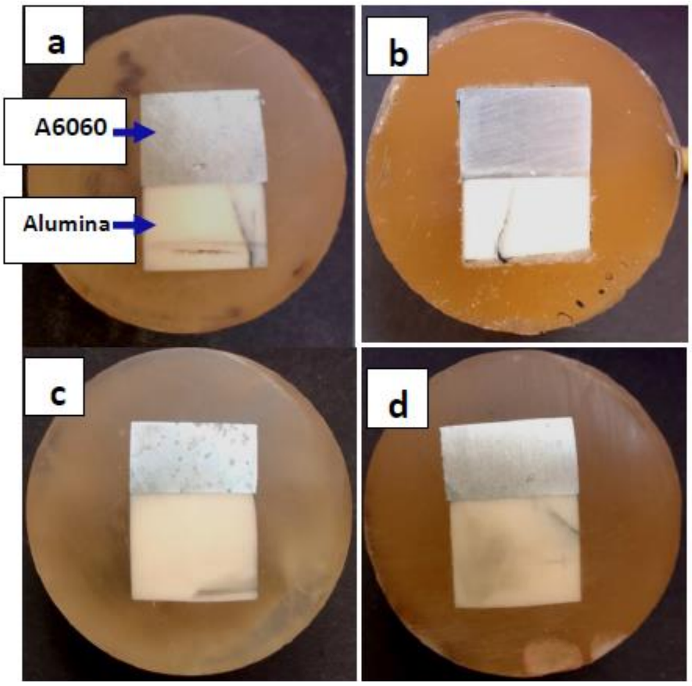

Figure 2. Samples obtained by solid state diffusion bonding at 600°C of alumina with aluminum 6060 alloy by solid state diffusion method for different times(a): 2 min, (b): 3 min, (c): 4 min, and (d): 5 min

For microstructural observations, samples were prepared following the standard metallographic procedure. The aluminum side was etched with HF reagent (10%). The microstructure was revealed using an optical microscope (NIKON) and a type of scanning electron microscope (HITACHI SU8020) equipped with an energy dispersive spectroscopy (EDS) detector. The X-ray diffraction (XRD) study was performed using a diffractometer (EMPYREAN), using the CuKa source in the range of 28° to 90° with voltage U = 45 kV and current I = 40 mA. X'pert high score plus software was used to analyze the results. The hardness across the weld joint was measured by a Vickers tester (SHIMADZV) using 50 gf for the Al 6060 alloy side.

The corrosion tests were performed in an AUTOLAB galvanostat potentiostat with Z view integrated software coupled to an HP microcomputer (Figure 3). The electrode in saturated DHW calomel was used as a reference electrode. It allows to measure or control the potential of the work electrode. It is placed near the work electrode (≈ 2 mm) in order to minimize the ohmic fall caused by electrolyte. Auxiliary electrodes (the electrodes) allow the passage of the current, which passes through the electrochemical cell. They are unassailable platinum electrodes, arranged in parallel with the work electrode in order to obtain a homogeneous current distribution. The work electrode consists of the metal studied, an alloy of 6060 serial aluminum bonded with alumina, coated by an epoxy resin except for the active surface. The electric current is provided by electric wires welded to the surface of the metal not exposed to the electrolyte. Table 3 presents the methods and parameters used during the corrosion test. It should be noted that the surface of the sample subject to electrochemical tests is about 0.80 cm^2. The electrolyte used in which the samples were immersed is a 3.5% by-weight NaCl solution.

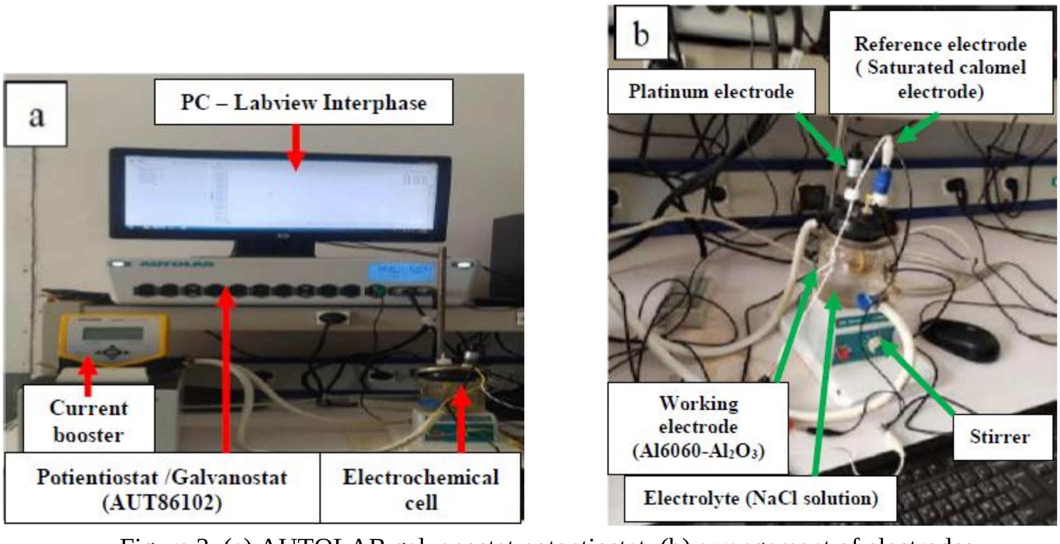

Figure 3. (a) AUTOLAB galvanostat potentiostat, (b) arrangement of electrodes

Table 3. Corrosion test methods and parameters

Method used	Fixed parameters	Values
OCP open circuit potential measurement	Immersion time	3600 (s)
Electrochemical impedance spectroscopy	Frequency range	100 kHz - 0.1Hz
	Signal amplitude	10 mV
	Work potential	Ecorr
Tafel	Initial potential	300 V/Ocp
	Final potential	+300 V/OCP
	Scanning speed	1 mv /s

3.0 RESULTS AND DISCUSSION

3.1 Macrostructural observations

Figure 4 presents the macrostructures of the bonded joint alumina/Al 6060 alloy realized at 600°C for different bonding times. This figure shows a bond with no apparent defect at the interface for the four welded samples. Moreover, the only observation is that the macrostructural evolution is localized in the aluminum alloy part where the grains are lengthened in a direction perpendicular to the interface and these same grains will undergo a recrystallization after a holding time of 4 and 5 minutes at 600°C. This recrystallization may be due to the load applied during diffusion bonding and the holding time being a little prolonged.

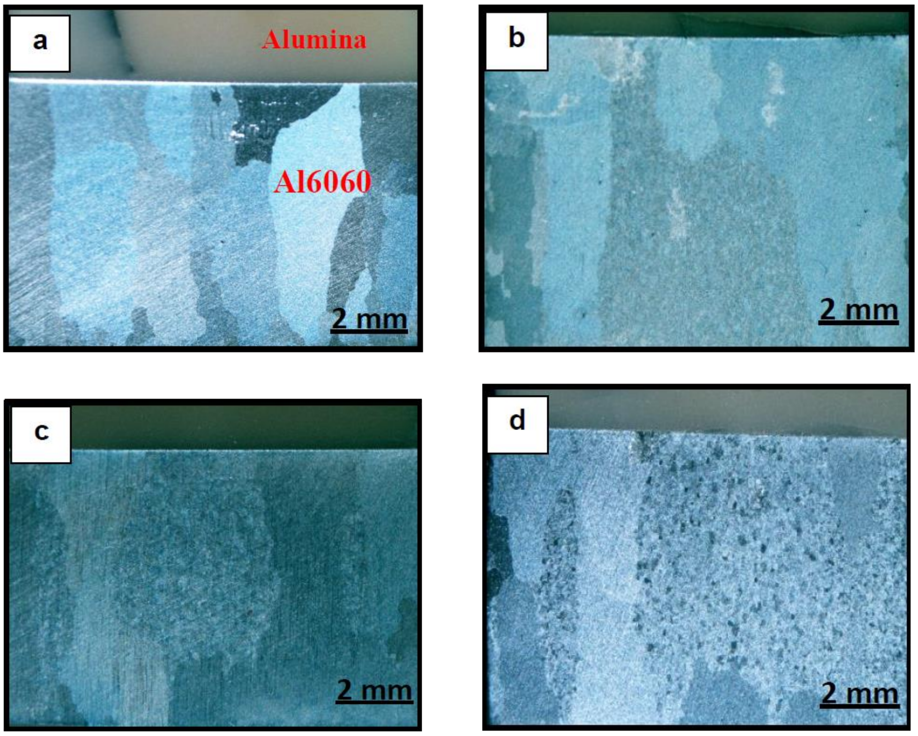

Figure 4. Macrostructures of bonded alumina/Al 6060 alloy at 600°C for different bonding times (a) 2 min, (b) 3 min, (c) 4 min, and (d) 5 min

3.2 Microstructural observation

Figure 5 presents the microstructures of the bonded joint alumina/Al 6060 alloy realized at 600°C for different bonding times. This heat treatment regenerates a thin interface between bonded alumina to Al 6060 alloy. Figure 6 shows more details in the interface during holding time at 600°C. For 2 min of holding time (Figure 6(a)), a thin layer was formed in the interface (indicated by an arrow). By extending the holding time to 3 and 4 min, this interface becomes thicker. We consider the holding time from 2 to 4 min (Figure 6(b) and (c)), as holding time for the diffusion process. However, prolonging the time to 5 min induced plastic deformation in this interface (Figure 6 (d)). Bedjaoui et al. [23] found that, during the diffusion welding process of copper with an aluminum alloy, intermetallic phases form at the interface and more particularly on the side of the aluminum.

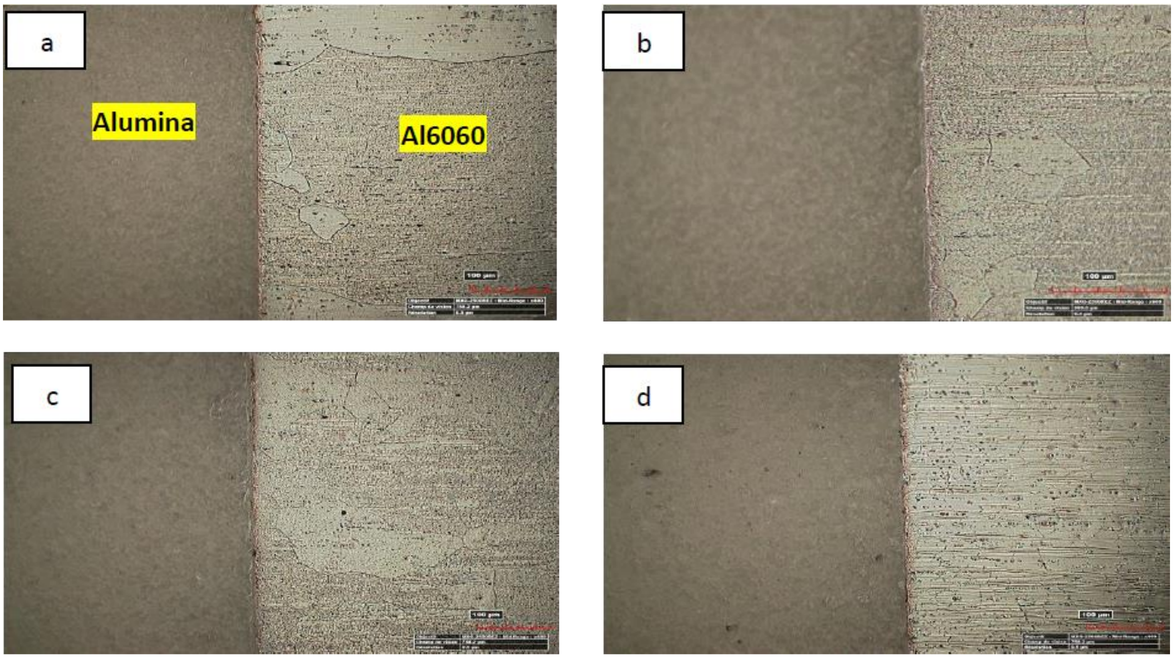

Figure 5. Microstructures of bonded alumina/Al 6060 alloy at 600°C for different bonding times (a) 2 min, (b) 3 min, (c) 4 min, and (d) 5 min

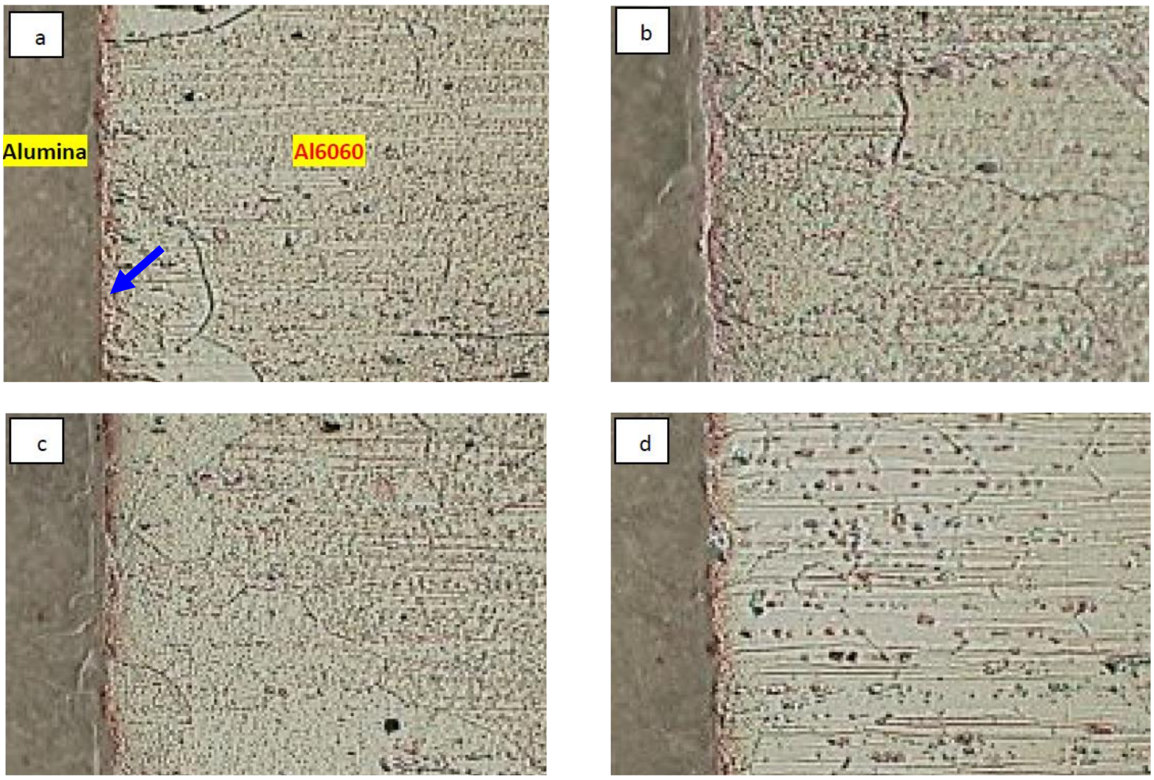

Figure 6. Microstructure evolution of the bonded interface alumina/Al 6060 alloy at 600°C for different bonding times(a) 2 min, (b) 3 min, (c) 4 min, and (d) 5 min

3.3 EDS analysis

Figure 7 presents the EDS analysis along the bonded alumina/Al 6060 alloy. The diffusion of atomic aluminum from the base metal to the ceramic side is confirmed (Green curve) and also the diffusion of atomic oxygen from the ceramic to the aluminum side is confirmed (Red curve). This kind of diffusion created an interdiffusion zone between the two dissimilar materials. As mentioned in Figure 7, the interdiffusion area increases with the extension of the dwell time from 3.5 um to 16 um. This result confirms the effect of extending the holding time on the bonding process, which strengthens the bond between the two dissimilar materials. Interdiffusion of elements across the interface during solid state diffusion bonding was also observed in our previous study of the two bonded dissimilar steels [24].

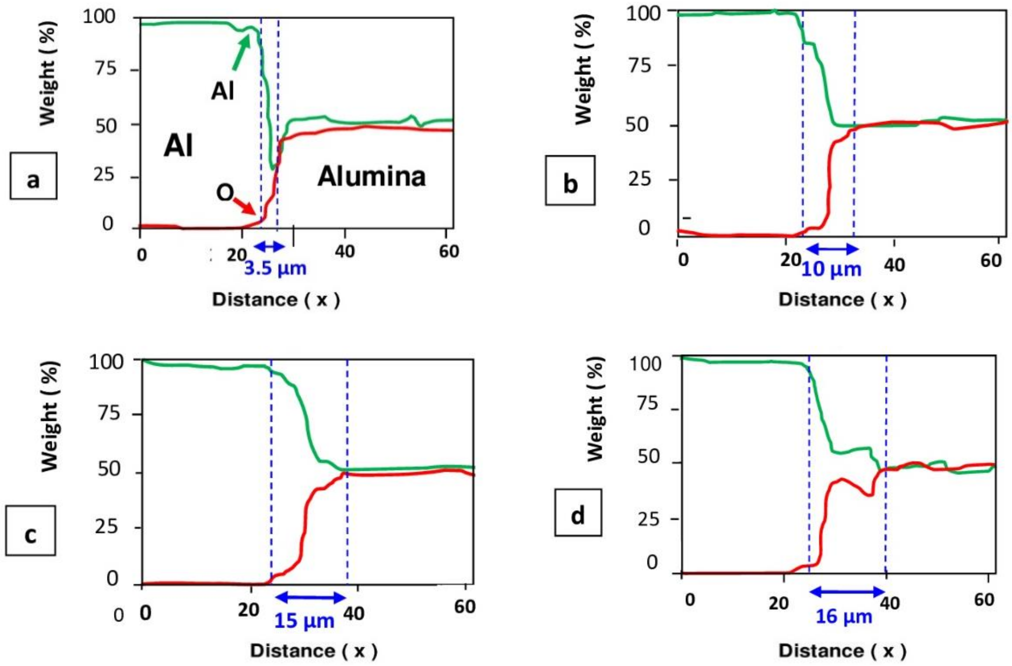

Figure 7. EDS analysis along welded alumina/Al 6060 alloy at 600°C for different bonding times (a) 2 min, (b) 3 min, (c) 4 min, and (d) 5 min

3.4 XRD analysis

In this part of the study, the X-ray diffraction spot is applied to the connecting joint which contains the two dissimilar materials. Figure 8 presents the X-ray diffractograms of the samples bonded at a temperature of 600°C for different holding times. For the first hold time of 2 min (Figure 8(a)), the XRD diffractogram is composed of high intensity peaks of Al, Al_2O_3, and Mg_2Si with small neglected peaks of MgO. The Mg_2Si phases were formed in the aluminum-based matrix. On the other hand, the MgO phase is a new one that was formed at the interface during the short bonding time at 600°C. It was also observed that the Mg_2Si and MgO peaks increase in number and intensity as the hold time increases. (Figure 8(b)-(d)). The increase in the amount of these two compounds is the result of the interaction between the elements of the two dissimilar materials during the interdiffusion process.

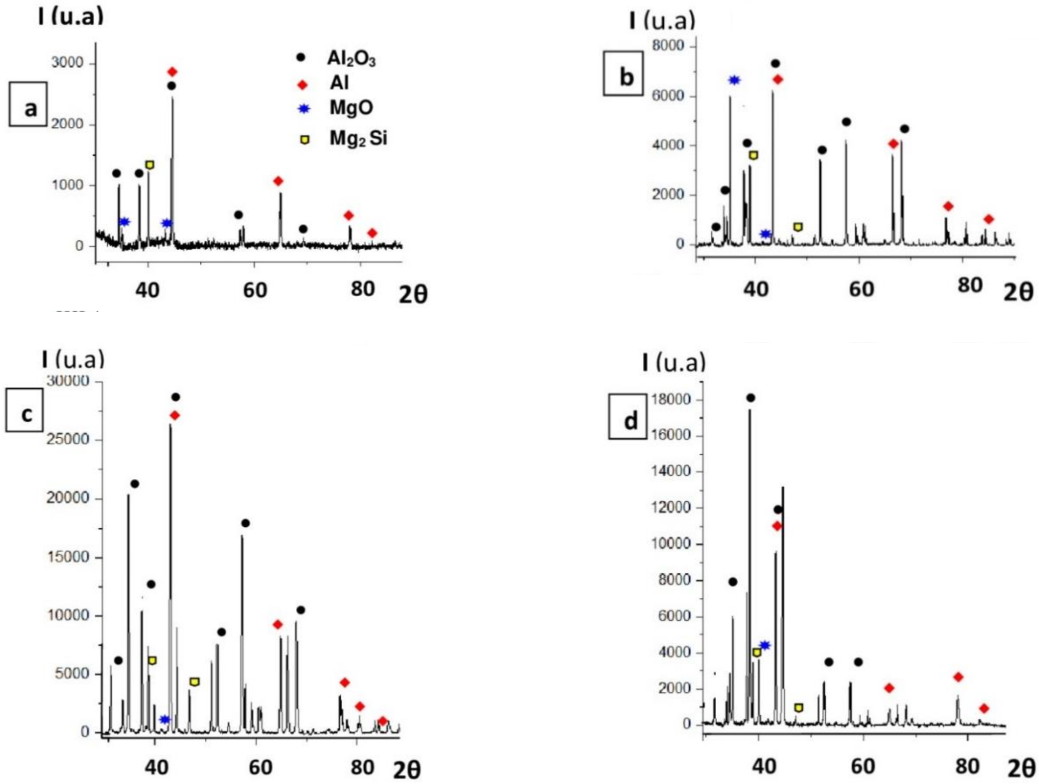

Figure 8. X-ray diffractograms of the bonded alumina/Al 6060 alloy at 600°C for different bonding times (a) 2 min, (b) 3 min, (c) 4 min, and (d) 5 min

3.5 Microhardness measurements

Figure 9 shows the variation of the microhardness of the aluminum side of the bonded joint at 600°C for different welding times (3, 4, and 5 minutes). The main common behavior is the highest hardness values near the bond joint. This mechanical behavior reflects the presence of the interdiffusion zone at the alumina/aluminum 6060 alloy bonding joint. It can also be noted that the hardness curve of the sample welded for 5 min shows the highest hardness at the interface on the aluminum side; this may be due to the formation of hardening compounds such as the MgO compound. Thus, increasing the hardness of the bonded joint increases its mechanical resistance. Mellik et al. [25, 26] found that, when extending the diffusion welding time of an aluminum composite material, the hardness on the welded joint increases, which enhances the weldability of the material.

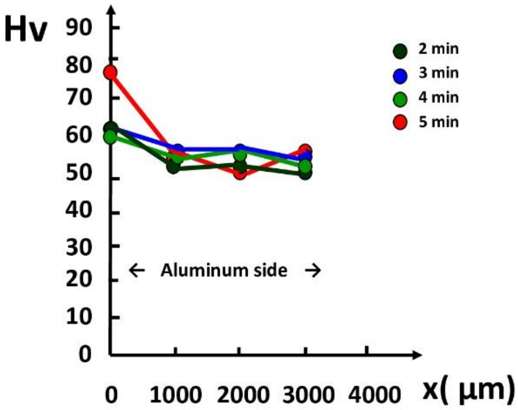

Figure 9. Hardness curves of bonded alumina/Al 6060 alloy at 600°C for different bonding times (a) 2 min, (b) 3 min, (c) 4 min, and (d) 5 min

3.6 Corrosion test

It is important to note that the corrosion testing was performed on the surface containing the two dissimilar bonded materials. The polarization curves of the samples bonded at a temperature of 600°C for three different holding times (3, 4 and 5 min) are presented in Figure 4. The curve of the welded joint at 2 min did not give a significant result, and for this reason, it has not been presented. It can be concluded from Figure 10 that, by increasing the bonding time, the polarization curves show a significant shift in the corrosion potential toward the most negative values; this means that the bonded joint becomes more and more resistant to corrosion.

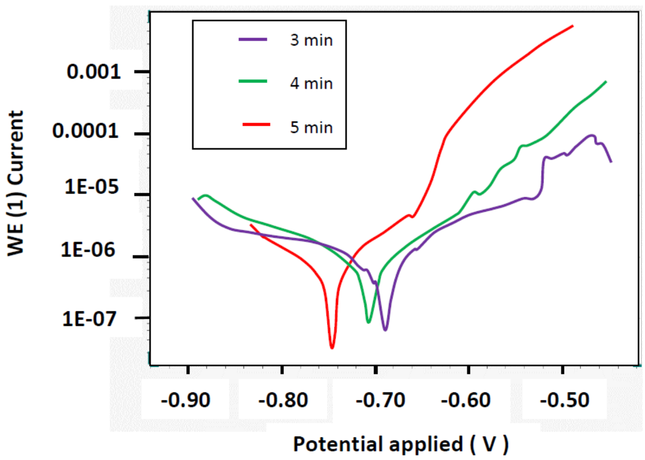

Figure 10. The polarization curves of the bonded alumina/Al 6060 alloy at 600°C for different bonding times (a) 3 min; (b) 4 min, and (c) 5 min

4.0 CONCLUSION

Our investigation represents a contribution to the study of the bonded alumina/Al 6060 alloy. The applied joining process was solid-state bonding at 600°C. The solid-state welding technique was successfully applied by injoining ceramic to aluminum alloy. The inter-diffusion zone in the welded joint has been formed between ceramic and aluminum alloy. This inter-diffusion process is located at the interface and it has been confirmed by EDS analysis The hardness values on the aluminum side showed the highest hardness values are close to the interface. The increase in the bonding time increases the corrosion resistance of the bonded dissimilar materials.

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