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Investigation of the Relaibility of Diffusion Bond Strength Between Ceramic-Metals in Solid State

A THESIS

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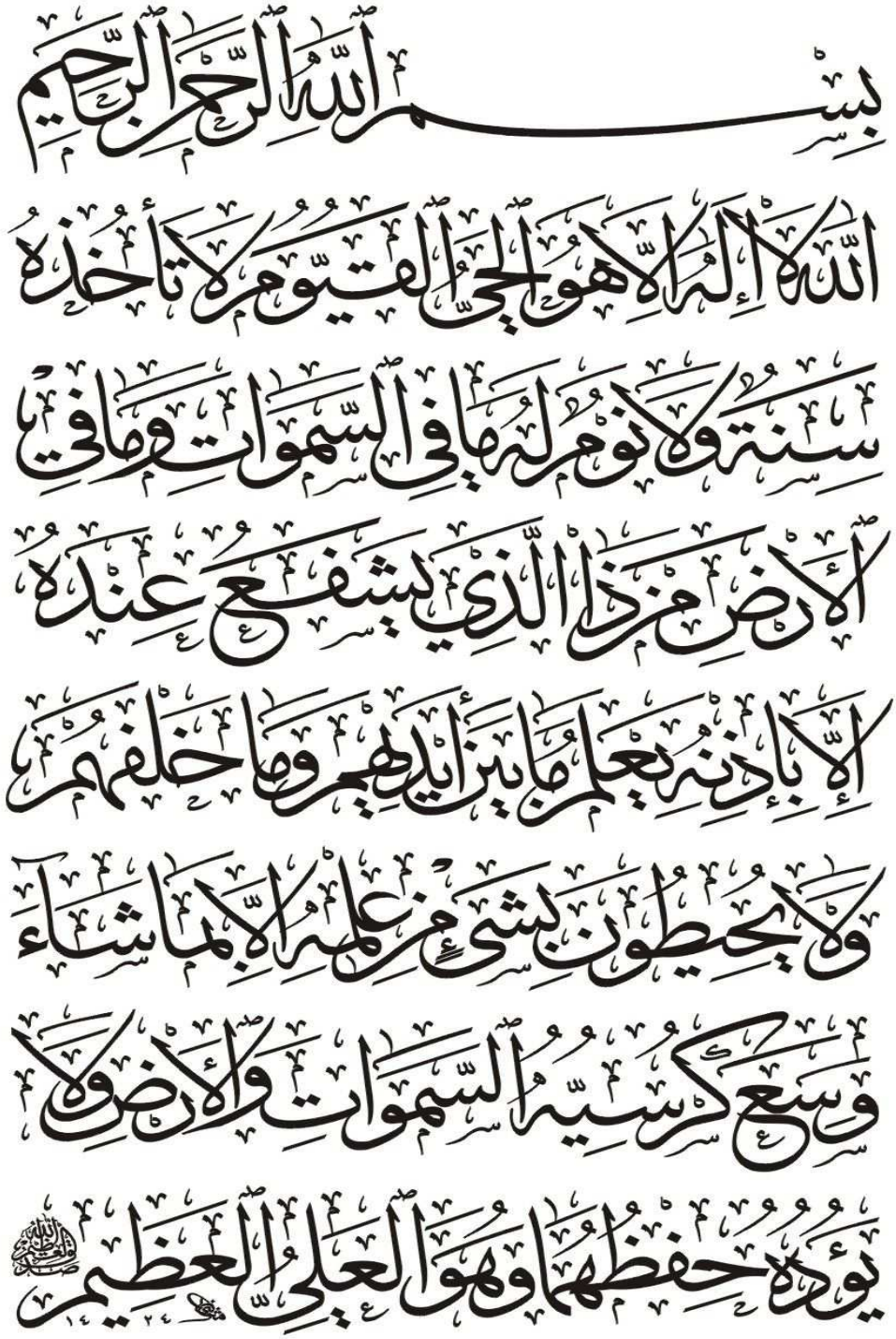

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List of Symbols

Symbol	*Definition*
J_{net}	**The flux for one place to another place**
D	**Coefficient of diffusion**
dC/dX	**Concentration gradient**
C	**Concentration**
X	**Displacement**
d	**Distance between planes**
β	**Geometric factor**
q_d	**Activation energy**
T	**Temperature**
K	**Boltzmann constant**
I_i	**Intensity of characteristic line from element i**
$I_{100\ i}$	**Intensity of that line from pure element i**
W_i	**The weight (W)of the element i**
λ	**Wave length**
OM	**Optical microscope**
XRD	**X-Ray diffraction**
XRF	**X-ray fluorescence**
t	**Time**

Chapter One

Introduction

1.1 Bonding of Dissimilar Materials

1.1.1 Bonding Interaction Types

Many applications in industry, depends on dissimilar material joints. Due to the difference in chemical, mechanical and thermal behaviors of materials, the joining of dissimilar materials presents a challenges, significantly different than similar materials joining [1]. Bonding as a subdivision of both solid-state and liquid-phase welding, is a joining process wherein the principal mechanism is interdiffusion of atoms across the interface. The bonds is a result of chemical and/or physical interaction for the two faced materials prepared for joining.

a. Physical interaction*:* Van der Waals forces include attractions between atoms, molecules, and surfaces. They differ from covalent and ionic bonding in that they are caused by correlations in the fluctuating polarizations of nearby particles (a consequence of quantum dynamics)[2]. Intermolecular forces have four major contributions:

1.A repulsive component resulting from the Pauli exclusion principle that prevents the collapse of molecules.

2.Attractive or repulsive electrostatic interactions between permanent charges (in the case of molecular ions), dipoles (in the case of molecules without inversion center), quadrupoles (all molecules with symmetry lower than cubic), and in general between permanent multipoles.

3.Induction (also known as polarization), which is the attractive interaction between a permanent multipole on one molecule with an induced multipole on another.

4.Dispersion , which is the attractive interaction between any pair of molecules, including non-polar atoms, arising from the interactions of instantaneous multipoles[2].

b. C***hemical interaction****:*** A chemical bond is an attraction between atoms or molecules and allows the formation of chemical compounds, which contain two or more atoms. A chemical bond is the attraction caused by the electromagnetic force between opposing charges, either between electrons and nuclei, or as the result of a dipole attraction. The strength of bonds varies considerably; there are "strong bonds" such as covalent and ionic bonds[3].

per force***c. Mechanical interaction:*** Stress is a measure of the average on which internal forces deformable body of a surface within a areaunit acting between forcesacts. It is a measure of the intensity of the internal particles of a deformable body across imaginary internal surfaces. These internal forces are produced between the particles in the body as a reaction to external forces applied on the body. External forces are either . Because the loaded deformable body is body forces or surface forces , these internal forces are distributed continuumassumed as a continuously within the volume of the material body, i.e., the stress distribution in the body is expressed as a piecewise continuous function (symbol pascal unit for stress is SIof space coordinates and time. The Pa), which is equivalent to one Newton (force) per square meter (unit area)[4].

1.1.2 Bonding Techniques

a. Indirect bonding

1. Diffusion bonding via interlayer

The use of ductile metal interlayer to assist the diffusion bonding of ceramic (and glasses) to metal can offer several advantages. If the interlayer is soft, deformation of the metal work pieces is minimal so they can be bonded in their final forms, and the interlayer is also able to accommodate some mismatched contraction as the component cool after being bonded. Further, if the interlayer metal has a modest melting temperature, it may be possible to bond without degrading the mechanical properties of heat treatable work piece alloys[5].

2. Soldering

, ceramics….etc) are metal Soldering is a process in which two items (joined together by melting and flowing a filler metal into the joint, and . Soft soldering is melting pointthe filler metal having a relatively low characterized by the melting point of the filler metal, which is below [6,7]. solder400 °C (752 °F). The filler metal used in the process is called

3. Brazing

Brazing is a joining process whereby a filler metal is heated above 400^0C and distributed between two or more close-fitting parts by capillary action. The filler metal is brought slightly above its melting (liquids) temperature, while protected by a suitable atmosphere (usually a flux). The filler will then flows over the base material in a process (known as wetting), and then cooled to join the workpieces together. The structures of metal interlayer diffusion bonding joints and brazed joints are similar, but the processes involved in their creation are quite different. Brazing depends on the molten interlayer metal or alloy being able to wet

the work pieces. Although metal work pieces are usually readily wetted by metals, many technologically important ceramics are not special materials (active to metal brazes) or processes (metallization followed by brazing using conventional alloys) have to be adopted. Unlike diffusion bonding, active metal brazing depends on chemical reactions occurring at metal-ceramic interfaces to promote wettability. Ceramics generally lack the delocalized electrons that bind together metal lattices[7].

4. Adhesive bonding

or semi-liquid state that liquid in a mixture An adhesive, or glue, is a or bonds items together. Adhesives may come from either natural adheres sources. The types of materials that can be bonded are vast syntheticor curebut they are especially useful for bonding thin materials. Adhesives (harden) by either evaporating a solvent or by chemical reactions that occur between two or more constituents[8].

Adhesives are an advantageous for joining thin or dissimilar materials,, and when a vibration dampening joint is needed. A disadvantage to adhesives is that they do not form an instantaneous joint, unlike most other joining processes, because the adhesive *needs time to* cure[9].

The adhesive bonding of metals is promoted by careful preoxidation of the metal work piece surface and the presence of components in the adhesive or primer that can form chemical bridges (such as Fe-O-Si sequences between steel and polysiloxanes). Thus although polymeric adhesives have radically different binding characteristics from brazes, there are some phenomenological similarities in their bonding behavior. The principle practical attraction of adhesive bonding to the fabricator is the low temperature at which it can be used, unlike brazing[10].

5. Mechanical bonding

The high volume production of joints by crimping, clamping, shrink fitting, bolting, screwing and other variants makes mechanical attachments the most widely used joining category for dissimilar materials combinations. The processes can be cheap and simple to perform, but the joints lack continuity and in fact demountability is often a crucial requirement. Mechanical bonding is employed in a wide range of domestic applications industry[5].

b. Direct bonding

1. Fusion bonding

The ceramic – metal combination could be fusion welded to produce prototype insulated electrical connectors. The range of ceramic- metal combination that joined in this way is very small since close matching of the melting temperature and thermal contraction characteristics is necessary, if failure by cracking of the brittle ceramic is to be avoided . These problems could be overcome to some extent by melting only the bonding face of the metal workpiece, but this is generally impractical due to the poor ability of liquid metals to wet ceramics[5].

2. Solid state Diffusion bonding

Solid-state diffusion bonding is a process by which two normally flat interfaces can be joined at an elevate Temperature (about 50%-90% of its melting point of the parent material) using an applied pressure for a suitable time.

Diffusion bonding of materials in the solid state is a process for making a monolithic joint through the formation of bonds at atomic level, as a result of closure of the mating surfaces due to the local plastic deformation at elevated temperature which aids inter diffusion at the

surface layers of the materials being joined[11]. The aim of diffusion bonding is to bring the surfaces of the two pieces being joined sufficiently close that interdiffusion can result in bond formation. However, there are two major obstacles that need to be overcome in order to achieve satisfactory diffusion bonds. Firstly, even highly polished surfaces come into contact only at their asperities and hence the ratio of contacting area to facing area is very low. Secondly in certain materials, the presence of oxide layers at the faying surfaces will affect the ease of diffusion bonding. For some metallic alloys, their oxide films either dissolve in the bulk of the metal or decompose at the bonding temperature (e.g. those of many steels, copper, titanium, tantalum, columbium and zirconium). In practice, because of inevitable surface roughness and also the presence of oxide layers on most faying surfaces, it is neither feasible to bring together the surfaces of two pieces within interatomic distances nor to establish complete metal-to-ceramic contact by simply putting two pieces together.

For example of diffusion is the motion of vacancy and the doping of semiconductors, which are used as electronic components. Diffusion bonding of most metals is conducted in vacuum or in an inert atmosphere (normally dry nitrogen, argon or helium) in order to reduce detrimental oxidation of the faying surfaces. Bonding of a few metals which have oxide films that are thermodynamically unstable at the bonding temperature (e.g. silver) may be achieved in air[1].

a. Diffusion mechanism

Fig.(1.1) used to illustrate activation energy schematically. A carbon atom is small (r =0.07 nm) where r is radius of carbon atom and can sit interstitially among a number of face center cubic (fcc) atoms (such as

iron atom). If it has enough energy it can squeeze between the iron atoms, to the next interstice when it vibrates in that direction, at 20^0 C. There is only a small probability that it will have that much energy. At higher temperature the probability increases. The other diffusion mechanism (interstitial) is sketched in fig(1.2). When all the atoms are the same size, or nearly so the vacancy mechanism becomes predominant. The vacancy may be present either as part of defect structure or because of extensive thermal agitation[12].

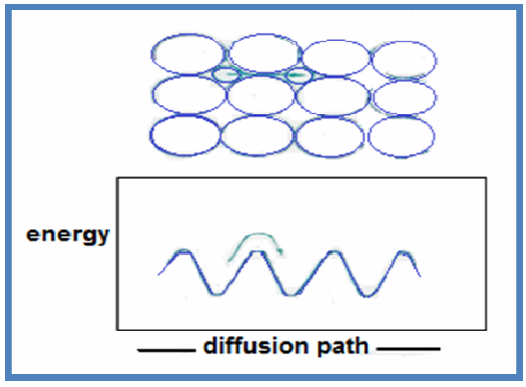


Figure (1.1): Atom movement[12]

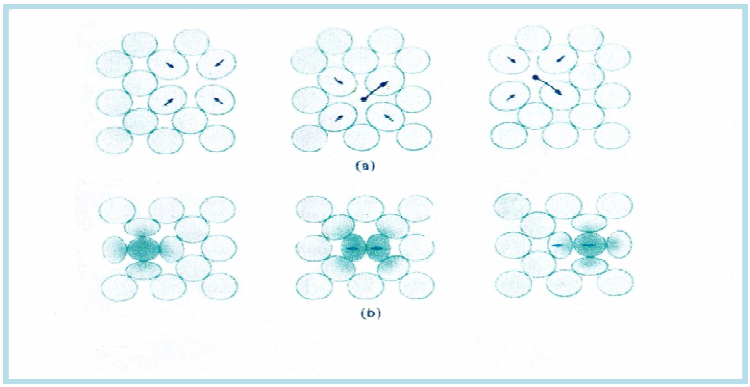


Figure (1.2): Diffusion mechanism. (a) By vacancies. (b) By Interstitialcies[12]

Solid state diffusion can be mathematically described by two differential equations which are called Fick's first and second laws.

Fick's first law can be express from this relation:

$$J_{net} = -D\frac{dC}{dx} \quad \text{.....(1.1)}$$

The units are:

$\left(\frac{atoms/m3}{m}\right)\left(\frac{m2}{sec}\right) = \frac{atoms}{(m2)(sec)}$. Where J_{net} is the flux of atoms diffuse from one place to another, dC/dx is the concentration gradient, and D is the diffusivity (or coefficient of diffusion). The negative sign in the equation means that the flux of diffusion species is from higher to lower concentration [1].

Fick's first law allows to calculate the instantaneous mass flow rate or flux past any plane in a solid, but it gives no information about the time dependence of the concentrations. The time dependence is contained in Fick's second law, which can be derived by using Fick's first law and the concentration of mass. The Fick's second law is:

$$\left(\frac{dC}{dt}\right) = D\left(\frac{d}{dx}\right)_x\left(\frac{dC}{dx}\right) \quad \text{..... (1.2)}$$

The notation $(dC/dt)_x$ means that dC/dt is the derivation of C with respect to time (t) while direction(x) is held constant , and similarly for (dC /dx). Equation(1.2) provides a relationship between C varied with t, x and D (recall that D depends on the temperature and structure).

The diffusion coefficient is defined by:

$$D = d^2\beta\nu \exp(-q_d/KT) \quad \text{..... (1.3)}$$

Where d is the interplaner spacing planes in the crystal, this distance measured in the direction of diffusion, β is the geometric factor, ν is for diffusion, T is vibration frequency, q_d is the activation energy temperature & K is the Boltzmann constant [13].

Depending on the equation (1.2) the diffusivity varies with many important factors:

1. The nature of the solute atoms.

2. Nature of solid structure.

3. Higher temperatures provide higher diffusivities, because the atoms has higher thermal energies and therefor greater probabilities of being activated over the energy barrier between atoms.

4. The grain size of metal effect on the diffusivity for example Carbon atoms have a higher diffusivity in ceramic than do nickel atoms in ceramic because the carbon atom is small one.

5. Copper atoms diffuse more readily in aluminum than in copper because the cu-cu bonds are stronger than the Al-Al bonds (as evidenced by their melting temperature).

6. Atom has higher diffusivity in bcc iron than in fcc iron because the former has a lower atomic packing factor (0.68 versus 0.74) and the fcc structure has larger interstitial holes; however, the passageways between the holes are smaller in the fcc than in the bcc structure [13].

b. Advantages of solid-state diffusion bonding

The advantages of solid-state diffusion bonding can be summarized as follows[5]:-

1. The process has the ability to produce high quality joints so that neither metallurgical discontinuities nor porosity exist across the interface. The optical micrograph of diffusion bonding of cobalt-base super alloy was shown in fig.(1.3).

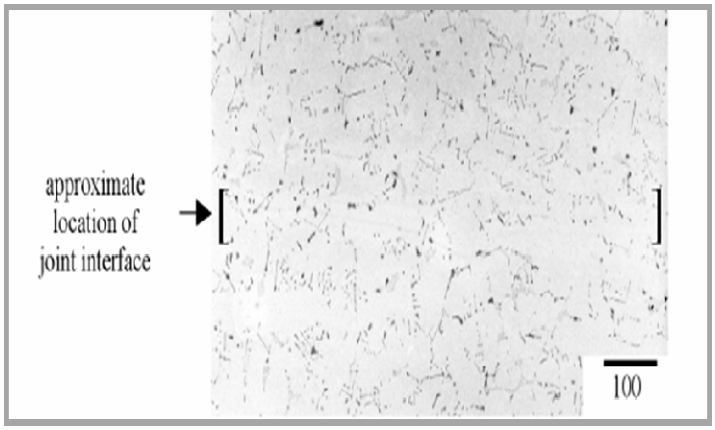


Fig (1.3) Optical micrograph of the diffusion bond in a cobalt-base super alloy, free from flaws, voids and loss of alloying elements[14]

2. Joining of dissimilar materials with different thermo-physical characteristics, which is not possible by other processes, may be achieved by diffusion bonding. Metals, alloys, ceramics and powder metallurgy products have been joined by diffusion bonding. Fig.(1.4) shows dissimilar bonds in different Al alloy and Ti alloy tested by bending and torsion tests. No preferential failure at the joint interfaces occurred.

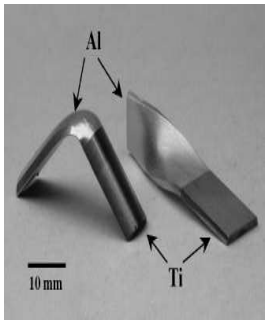


Fig. (1.4) Bonding of different alloys of Al and Ti[14]

3. High precision components with complicated shapes or cross sections can be manufactured without subsequent machining. This means that good dimensional tolerances for the products can be attained.

4. Apart from the initial investment, the consumable costs of diffusion bonding are relatively low as no expensive solder, electrodes, or flux are

required (although the capital costs and the costs associated with heating for relatively long times may be high).

5. Diffusion bonding is free from ultraviolet radiation and gas emission so there is no direct bad effect on the environment health and safety standards are maintained[14].

c. Limitations of diffusion bonding

Diffusion bonding has many limitations, and among them are the following[11,5]:-

1. Great care is required in the surface preparation stage. Excessive oxidation or contamination of the faying surfaces would decrease the joint strength drastically. Diffusion bonding of materials with stable oxide layers is very difficult. Production of thoroughly flat surfaces and also precise fitting-up of the mating parts takes a longer time than with conventional welding processes.

2. The initial investment is fairly high, and production of large components is limited by the size of the bonding equipment used.

3. The suitability of this process for mass production is questionable, particularly because of the long bonding times involved[5].

d. Problems with solid-state diffusion bonding

The aim in diffusion bonding is to bring the surfaces of the two pieces being joined sufficiently close that inter diffusion can result in bond formation. There are two major obstacles that need to be overcome in order to achieve satisfactory diffusion bonds[11]:-

1. Even highly polished surfaces come into contact only at their asperities and hence the ratio of contacting area to faying area is very low.

2. In most metals, the presence of oxide layers at the faying surfaces will affect the ease of diffusion bonding [11].

1.2 General properties of ceramic and metal

Ceramics and metals used in joined samples specified with many important mechanical and thermal properties.

1.2.1 Ceramic:

A ceramic material is often understood as restricted to inorganic *crystalline* oxide material. It is solid and inert. Ceramic materials are brittle, hard, strong in compression, weak in shearing and tension. They withstand chemical erosion that occurs in other materials subjected to acidic or caustic environment. Ceramics(Al_2O_3) generally can withstand very high temperatures such as temperatures that range from 1,000 °C to 1,600 °C. The specific properties of ceramic material are high mechanical strength, wide range of thermal expansion, stability of geometric shapes, accuracy of their dimensions, no special annealing required and low vapor pressure at high temperatures. One of common types of ceramics is alumina which represents about 25% of the earth crust and used in this research. Pure alumina (α-alumina) is poly – crystalline material, has a hexagonal structure with two Al_2O_3 molecules per unit cell[15,16].

1.2.2 Metal:

Metal in joined samples can be as a structure or as a joining means. In any case the important physical properties for joining are:-

a. Melting points:

Metals tend to have high melting points because of the strength of the metallic bond. The strength of the metallic bond varies from metal to

metal and depends on the number of electrons in each atom delocalises into the sea of electrons, and on the packing[1].

1. Metals like sodium and potassium have relatively low melting points mainly because each atom only has one electron to contribute to the bond.
2. Elements are also inefficiently packed (8-co-ordinated), so that they aren't forming as many bonds as most metals.

They have relatively large atoms (meaning that the nuclei are some distance from the delocalised electrons) which also weakens the bond[12].

b. Thermal conductivity:

Metals are good conductors of heat. Heat energy is picked up by the electrons as additional kinetic energy (it makes them move faster). The energy is transferred throughout the rest of the metal by the moving electrons[13].

c. Malleability and ductility:

Metals are described as ***malleable*** (can be beaten into sheets) and ***ductile*** (can be pulled out into wires). This is because of the ability of the atoms to roll over each other into new positions without breaking the metallic bond. If a small stress is put onto the metal, the layers of atoms will start to roll over each other. If the stress is released again, they will fall back to their original positions, as shown in fig.(1.5). Under these circumstances, the metal is said to be elastic[13].

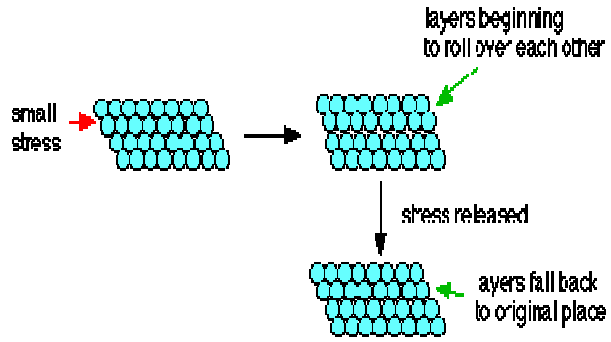


Fig.(1.5) Small stress put on the metal[13]

If a larger stress is put on, the atoms roll over each other into a new position, and the metal is permanently changed, as shown in fig.(1.6).

Fig.(1.6)Large stress put on the metal[13]

d. The hardness of metals:

The rolling of layers of atoms over each other is hindered by grain boundaries because the rows of atoms don't line up properly. It follows that the more grain boundaries there are (the smaller the individual crystal grains), the harder the metal becomes.

Offsetting this, because the grain boundaries are areas where the atoms aren't in such good contact with each other, metals tend to fracture at grain boundaries. Increasing the number of grain boundaries not only makes the metal harder, but also makes it more brittle[1].

e. Controlling the size of the crystal grains

If a pure piece of metal is present then one can control the size of the grains by *heat treatment* or by *working the metal*. Heating a metal tends to shake the atoms into a more regular arrangement, decreasing the

number of grain boundaries, and so making the metal softer. Banging the metal around when it is cold tends to produce lots of small grains. Cold working therefore makes a metal harder. To restore its workability, reheating were needed. The regular arrangement of the atoms can be breakup by inserting atoms of a slightly different size into the structure. Alloys such as brass (a mixture of copper and zinc) are harder than the original metals because the irregularity in the structure helps to stop rows of atoms from slipping over each other[6]. Table (1.1) shows some properties of metals and alumina.

Table (1.1) Material properties[1,10,13]

Sample	Atomic number (Z)	Mass number (A)	Density (g/cm^3)	Melting point (^0C)	Thermal expansion $x10^{-6}$ (K^{-1})	Atomic radius (Pm)
Cu	29	٦٤	8.9	1084	16.5	128
Ag	47	١٠٨	10.49	962	18.9	144
Al	13	٢٧	2.7	660	23.1	143
Zn	30	٦٥	7.14	420	30.2	134
Pb	82	٢٠٧	11.4	327	28.9	175
Sn	50	119	7.3	٢٣٢	22	140
Al_2O_3	-	-	3.98	٢٠٤٠	8.2	-

1.3 Joint Strength Measurement

The joint strength, as the most important property of the joint can be characterized by the use of fracture mechanics. The strength of the joint

system can be define as the amount of energy necessary to separate the ceramic from metal at the interface in the form of an applied load . The application of the load is either a tensile or a shear stress.

In tensile stress, the applied force will be normal to the interface, while in shear stress the applied force is parallel to the interface. Figure (1.7) shows the basic principles of this stress type[17].

There are three major requirements for a testing method, shear or tensile stress to evaluate the mechanical properties of ceramic –metal joint. First a testing method should lead to accurate and consistent results. Second a testing method to be able to evaluate effects of processing variables on the joint properties. Third, the result of the test should provide meaningful engineering parameters that can be utilized for designing the ceramic-metal joints[18].

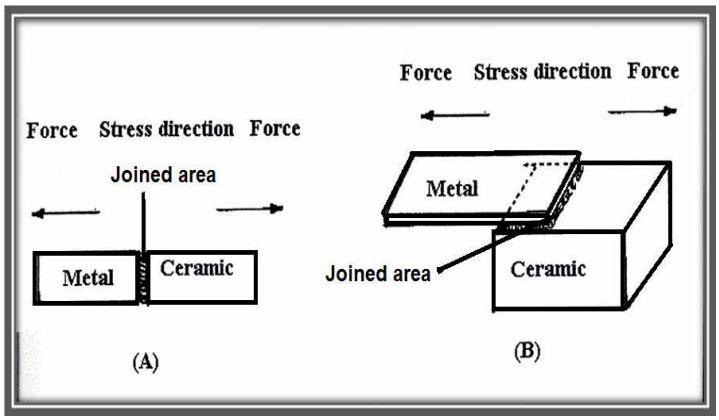


Fig (1.7) Types of stresses in ceramic to metal joint A-Tensile stress B- Shear stress[3]

1.4 Fracture surface characterization

The fractured surfaces can be examined by X-ray methods [elemental analysis by using X-ray fluorescence (XRF), phase identifications by using X-ray diffraction (XRD)], and microscopic methods using optical microscope (OM).

1.4.1 Elemental Analysis:

The X-Ray fluorescence spectrometry is the best technique to give rapid information about qualitative and quantitative elemental analysis. Having made the qualitative survey on material, quantitative evaluation of concentrations can be made by reducing the net intensity of specific X-ray lines (K-lines) characteristic of the element to mass concentrations. After intensity correction for background, the ratio of intensities for a given line from the sample and pure standard element is directly proportional to the weight per unit area of the element in the spacemen being analyzed[19]. If I_i be the intensity of characteristic line from element i in the sample and I_{100i} be the intensity of the line from pure element i. Then (W_i/a) which is the weight (W) per unit area (a) of the element i in the sample will be given by[20]:-

$$I/I_{100i}=W_i/a \quad \text{..... (1.4)}$$

This equation shows that the analytic line intensity I_i from material layer is linearly related to the weight of the element in the sample (W_i/a). If we are dealing with a three-component mixture, (for example), then:

$$W_i + W_j + W_k = 100\% = 1 \quad \text{..... (1.5)}$$

And a relative intensity R_i of an element in the sample is given by:

$$R_i = I_i / I_{100i} \quad \text{..... (1.6)}$$

Where i, j and k are the elements in the sample. For multi-elements, the interfering effects are present; and fluorescent intensity can depart widely from proportionality due to the amount present of other elements. These effects are either absorption effects or enhancement effects [21,22,23], and will have a special mathematical treatments and experimental calibrations[24].

1.4.2 Phase Identification:

X-ray diffraction technique is a powerful tool for investigating the solid state phases [20]. Suppose the monochromatic X-ray are directed towords a crystal and a parallel beam which interacts with all atoms in the region of the crystal to which it can penetrate. The additional path traversed is 2d Sinθ for a ray which suffers specular reflection from the second plane rather than the first. All specular reflected components will be able to combine constructively in phase if this distance is a multiple of). Thus the condition for efficient specular reflection λwavelength ((Bragg' s law) is:

$$n\lambda = 2d \sin\theta \quad \text{..... (1.7)}$$

Where n is an integer. Useful information is provided by the peak' s characteristic [21,22]:-

1. Position: The position of the peak, measured as the angle θ.

2. Intensity: the relative intensity of the peaks, measured either as the peak height or more correctly, as the area under its profile.

3. Shape: The shape of the peak, of which its breadth is a useful guide, provides information regarding crystallite size and lattice imperfections, including strain [23].

1.4.3 Microscopic methods:

The optical microscopic technique provides a direct and easy method for examining fractured surfaces and to determine its morphology providing a good method for dimension measuring method down to the wavelength of visible light [24].

1.5 Literature Review

Due to industrial requirements for joining of the dissimilar materials such as ceramic to metal or alloy, in 1920 many researcher had been investigate the capability of joining[16,25]. The Diffusion Hypothesis was the most commonly accepted hypothesis, considers the contribution of interatomic diffusion during bond formation [26] .The difference in the energy level of surface atoms and of bulk atoms is the basis of this hypothesis. All attempts at modelling diffusion bonding have two main aims. The first is to optimize the process variables e.g. surface finish, bonding temperature, pressure and time so that the proper bonding conditions for a particular material can be identified. Secondly, a model attempts to obtain a reasonable and profound understanding of the mechanisms involved and their relative contributions not only for different bonding conditions but also for different materials being joined[27].

The realization of joining ceramic to metal date back to 1934 when Siemens, et.al. at Telefunken, and Allgemenine in Germany [16,28] independently began to develop ceramic sealing techniques. In 1953 King, et.al., were expected that the migration of interface away from the voids was assumed to occur during the second stage and remaining isolated voids were removed by volume diffusion in the third stage [29] .

In 1966 Cline[9] was reach to the same results of King, he found two stag models assuming localized plastic deformation followed by the diffusion controlled process using the recrystalization model of parks.

In 1975 Garmong , et.al.[30], study the effects of corresponding wavelength and roughness of the joining surface removable of the voids was modulated by using the center equations derived by Coble in1970.

In 1982, Derby et.al.[8], an intensive sintering equations assuming six different diffusion mechanisms, and makes a modification on the existing model in order to reduce the discontinuity in bonding rate, especially between the second and final stage.

In 1986 Borbidge , et.al. [31], were discussed the bonding between wide range of materials , metals (such as steel, copper, Titanium, Platinum and Gold), and ceramics Al_2O_3 and silicon nitride. They concluded that the reaction bonding of solid state process can be used to join a wide variety of ceramics to metals, and the interlayer of suitable metals can be used to facilitate bonding in those cases.

In 1989 various techniques were developed in order to obtained the best bond in ceramic to metal interface, and have been discussed by M. G. Nicholas, et.al.[32], and Mamora N. et.al. [33].

In 1990 Nakamura, et.al. [34], were studied the solid state bonding of silicon nitride ceramic with nickel chromium alloy interlayer. Joining was performed by hot pressing between 1000^0C and 1350^0C (in argon), under uniaxial pressure in the range of 50 to 100 MPa. They found the formation of the interfacial poress (which not distributed uniformly at the bond interface), the scatter in strength is relatively large.

In 1992 C.D. Qin, et.al. [35], were studied a diffusion bonds of Ni/ZrO_2 and Ni /NiO_2 /ZrO_2 . They found that subsequent annealing in air after bonding improves bond strength and annealing in vacuum reduced strength. This attributed to the formation of thin oxide layer during annealing in air which enhances adhesion to the ceramic, where as annealing in vacuum creates de bonding voids at the spaceman edges.

In the same year Crank J.[30] and Bernard Y. [36], were studied joining of ceramic to metal based on three main features compatibility of thermal expansion coefficient for the parts to be joint.

In 1999 Irfan, et.al.[37], were studied the joint of copper and aluminum materials coupled by diffusion and friction welding methods, which are able to weld in a solid state without melting. This study was showed that the intermetallic phases (Al_2Cu, AlCu, Al_4Cu_9) occurred during the welding processes which have very important effects on the joint mechanical properties. As a result of diffusion welding process which takes long time and outer appearance of welding place is more uniform, the strength of the bonding zone is lower than of that pure aluminum. In 2000 Sarkis, et.al.[38], were studied solid state diffusion bonding of metals to ceramic and they concluded that the bonding due to chemical interaction between the metal and the ceramic is found to be unlikely and the braking strength of the metal-ceramic joint showed a linear relation with the melting point of metals involved.

In 2001 A. A Shirzadi, et.al[39], worked to overcome problems with surface oxides, when joining alumina with different alloys and compared for both solid state diffusion bonding and conventional transient liquid phase(TLP) diffusion bonding.

In the same year Jasenka , et.al.[37], were determined whether the rate of cooling induced structural changes in a high gold alloy and/or whether these changes modified the bond strength between the gold alloy and hydrothermal ceramic. Analysis of variance joints reveled no (statistically) significant difference in bond strength among the three types of specimens, suggesting that the mode of alloy cooling had no substantial effect either on the microstructure of high gold alloy or on the

quality of bond between gold alloy and hydrothermal ceramic. In 2008[40] a diffusion bonding method has been developed that enable large transfer of single crystal lithium niobate thin film to silicon substrates. Transmission electron microscopy confirms the interface evaluation via diffusion bonding which combines interfacial diffusion.

Many authors[41,42] have studied the fracture surface characterization and microcraking behavior at elevated temperatures. Advanced technologies were used to evaluate properties and analyze the joint strength. Different methods of test provides important information about the mechanical properties of the joint which were well established in many references[40,43,44,].Fracture surfaces were analyzed using optical microscope (OM), for studying microstructures and examining the interface layers [35,17].

1.6 Aim of this work

The purpose of this study was to evaluate the diffusion bonding in solid state at a contact surfaces between alumina and submitted pure metals Cu, Ag, Al, Zn , Pb, and Sn. For achieving this process, tensile fracture test was carried out for conducting the reliable of joint strength. Fracture surfaces were attempted by X-ray methods listed below:

a. Elemental analysis using X-ray fluorescence (XRF).

b. Phase identification using X-ray diffraction (XRD).

The fractured surfaces were examined microscopically by using optical microscope (OM).

Chapter Two

Experimental Part

2.1 Introduction

This chapter deals with procedure of samples preparation (cutting, polishing and decontamination processes), joining processes and hence investigation of bond strength. Fracture surface identifications were achieved by using a non- destructive methods such as X-Ray (XRF,XRD) and microscope techniques.

2.2 Materials

The materials used in this investigation were; metals such as Sn, Pb, Zn, Al, Ag, and Cu, while the ceramic was alumina (Al_2O_3).

High purity metals were used as a thin foils (99.9 wt% purity). Polycrystalline α-alumina (99.8 wt% purity and 25µm of average particle size) were used in preparing the joined samples.

2.3 Sample Preparation

2.3.1 Ceramic Preparation

All ceramic samples were prepared from alumina (Al_2O_3), and cutted by diamond cutter machine into rectangular shape (dimensions of (1x1x0.5) cm^3). To get normally flat surface and free of obstacles, wet grinding and polishing of all samples were achieved(to obtain the proper surface). The grinding and polishing processes were carried out on alumina parts as follows:

a. Grinding stage: The alumina was wet grind by silicon carbide metallographic papers with different grads of 360, 800, and 1000 respectively. Distilled water was used as a cooler to avoid any possibilities of cracking in alumina samples.

b. Polishing stage: The alumina samples were polished by using special cloth with diamond paste. These two stages were done using Strues machine of Denmark made.

In order to obtain clean alumina surface, it was necessary to remove any contaminated materials (such as greasing), chemical cleaning were conducted by immersing the alumina pieces in acetone for 20 minutes followed by immersing in dilute nitric acid for five minutes and subsequent rinsing in distilled water. Finally the alumina pieces were fired in air at temperature 1000 ^{0}C for about one hour, the samples were kept in a desiccator ready to use, as it is recommended by some researcher [3,18,45].

2.3.2 Ceramic Density and Porosity Measurement

A preliminary tests for measuring the density and porosity for the prepared alumina samples have been performed according to ASTM-C373-88. The bulk density (ρ) is defined as the following equation[46]:

$$\rho = D_r/(W-S) \quad \text{(2.1)}$$

Where:

D_r = dry weight in (gm)

S = suspended weight in (gm)

W = saturated weight in (gm)

On the other hand the apparent porosity as percentage (p) is defined as in the following equation[47]:

$P = [(W-D_r)/V] \times 100$ ……..……..….(2.2)

Where V is the exterior volume in cm^3. The accuracy of measuring density and hence the porosity was within ± 2%.

2.3.3 Metal Foils

The high purity metals (99.9 wt%) of 1mm thick rolled foils were prepared into square dimensions similar to that alumina(1x1x0.5) cm^3 of dimension (1x1x0.1) cm^3. The metals surfaces were prepared to joining by immersing it for about 30 minutes in acetone followed by polishing until it is visually seen to be free of oxide. The sheets were then kept in a desiccator ready to use.

The selection of metals were mainly according to their melting points. A wide range were chosen from Sn up to Cu (see table (1.1).

2.4 Joining Procedure

Metal foils were inserted between two pieces of alumina and fixed by a fixture of stainless-steel. Figure(2.1) represents the fixture set up.

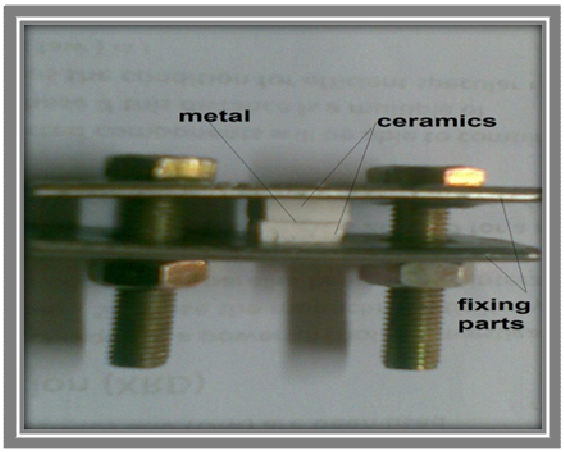
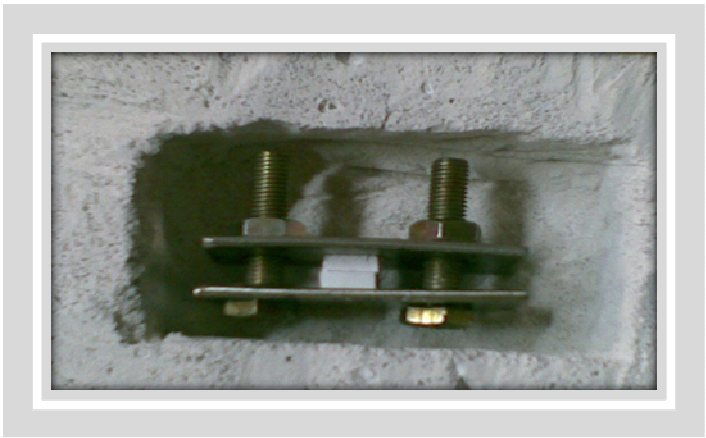


Fig. (2.1) Joining Fixture Set up

Pressure was then applied by torque-spanner with value ($3x10^5$ Pa). The pressure is equally distributed on each side of the fixture. To reduce the metal oxidation process, the assembly was embedded in alumina powder in order to isolate the joining set up from the ambient conditions as it is illustrated in figs. 2.2,a and b.



(a) Before isolation

(b) After isolation

Fig.(2.2) Joining Set up preparations

The system to be joined was fixed in the center of the furnace chamber and then heated to about $0.9T_m$ (where T_m is the melting point of each metals) under atmospheric condition. For this purpose Qallenhamp furnace(made in England) was used. The soaking time were two hours. The joining assembly was allowed to cool

down in the same way of heating schedule as shown in fig.(2.3).The final step was the carefully pressure released from the sample. Fig.(2.4) shows a typical joined sample.

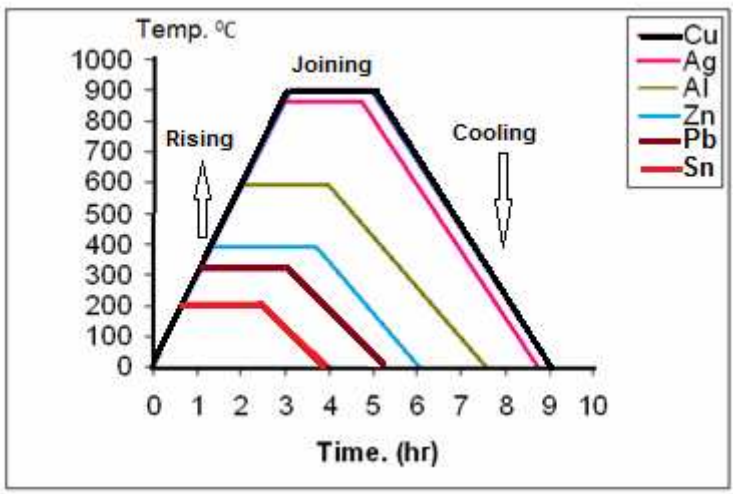


Fig. (2.3) Time-Temperature joining schedule

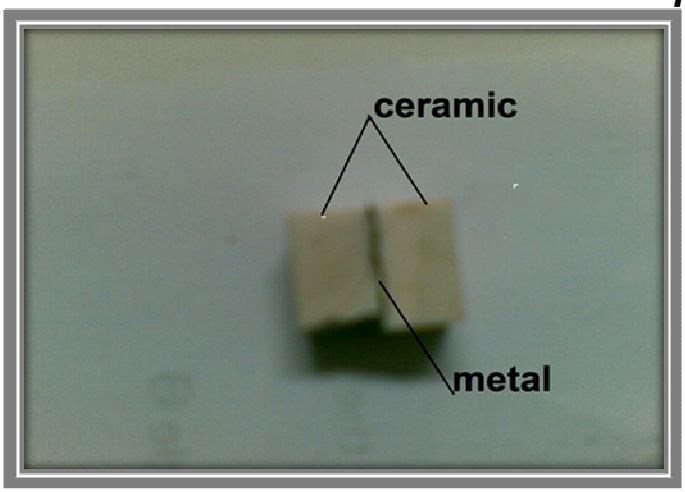


Fig.(2.4) Joined sample

Some tried experiments were performed to select the best soaking time starting 0.5 hr up to 2.5 hr. Joining temperature (starting from 0.5 T_m up to 0.9 T_m). Experiments revels that 2 hr for 0.9 T_m were convenient, since a suitable bond were achived.

2.5 Joining tests

2.5.1 Tensile Test

Measurement of the joint strength was carried out by using a tensile machine ***TINIUSOLSEN*** , with XP computer, made in England. The tensile force was applied at the crosshead speed of 3 mm/min. A suitable designed fixture (as shown in fig.(2.5)) was used to hold the alumina- metal assembly on the tester machine. The fracture stress in (N/m^2=Pa) was calculated from the force at fracture point of this assembly measured in Newton (N) , divided by fracture surface area (m^2).

Fig. (2.5) Joining sample under test.

2.5.2 Microscopic Test

The fracture surfaces were examined using optical microscope at 50-1000 magnification. For this purpose Nikon optical microscope fitted by camera digital Nikon with XP computer, made in Japan, was used to achieve these tests. Fig.(2.6) show the optical microscope instrument used to carry out these testes.

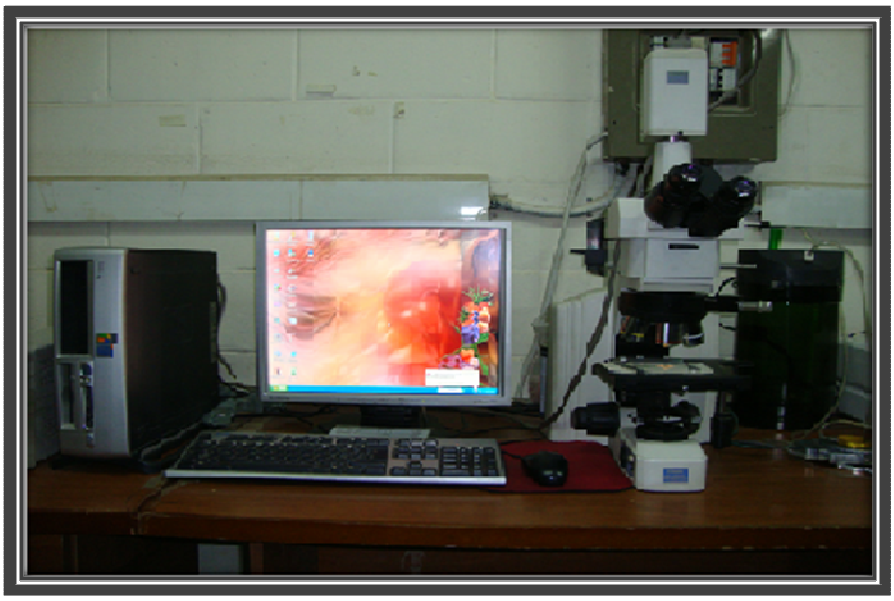


Fig.(2.6) Optical microscope used in this investigation

2.5.3 Elemental Analysis

X-ray Fluorescence (XRF) instrument type wave length dispersive X-ray Fluorescence(WLDXRF) work by exposing a sample to a beam of X-rays from tungsten target. The qualitative analysis of materials have been carried out by using this spectrometry of Twin-X-simple, Oxford instrument as shown in fig.(2.7), with the following operation conditions:

1.X-ray tube target = Tungsten

2.Power =50 kV, 1mA, 250 W

3.Cone angle =25^{0}

4.Vacuum =10^{-3} mbar

The atoms of the sample absorb the X- ray energy and become temporarily excited and then emits secondary X-rays. Each element emits x-rays at a unique energy, known as characteristic energy of the emitted element. An XRF analyzer can provide qualitative and quantitative analysis regarding the composition of the material being tested. Elemental composition can be determined by making use of calibration curves between intensity in count per second (CPS) and the composition.

Fig (2.7) WDXRF instrument (Twin-X-simple)

2.5.4 Phase Identification

A typical X-ray diffraction tests were conducted for phase identification of all materials used for joining process and fractured surfaces to detect any changes in the joined samples. The phase identification were carried out by using Lab. XRD-6000, SHIMADZU, (made in Japan) machine as shown in fig.(2.8). Copper is the X-ray tube target with monochrometer ($K\alpha_1$ radiation) operate at 40kV and 30 mA. The 2□ rang was taken from 20^0 up to 60^0. The rang limit was taken according to the basic informations for each element in this study.

Fig.(2.8) X-ray Diffractrometer type Lab XRD-6000

Chapter Three

Results and Discussion

3.1 Alumina Density and Porosity

The density and porosity of alumina which is used in this investigation were calculated according to equations (2.1)and (2.2) respectively. The results were listed in table (3.1).

Table (3.1) Alumina density and porosity results

Ceramic	Sintering temp.(^{o}C)	Density g/cm^{3}	porosity %
Al_2O_3	1650	3.82	4.03

The benefit of the porosity of alumina is may be for introduce the interlocking bonds and enhances the diffusivity of metal atoms through alumina.

3.2 Joining Strength

Typical tensile tests results were illustrated in fig.(3.1) which shows the maximum joining strength (applied force per unit area necessary to separate the joint faces) with the extension in (mm).

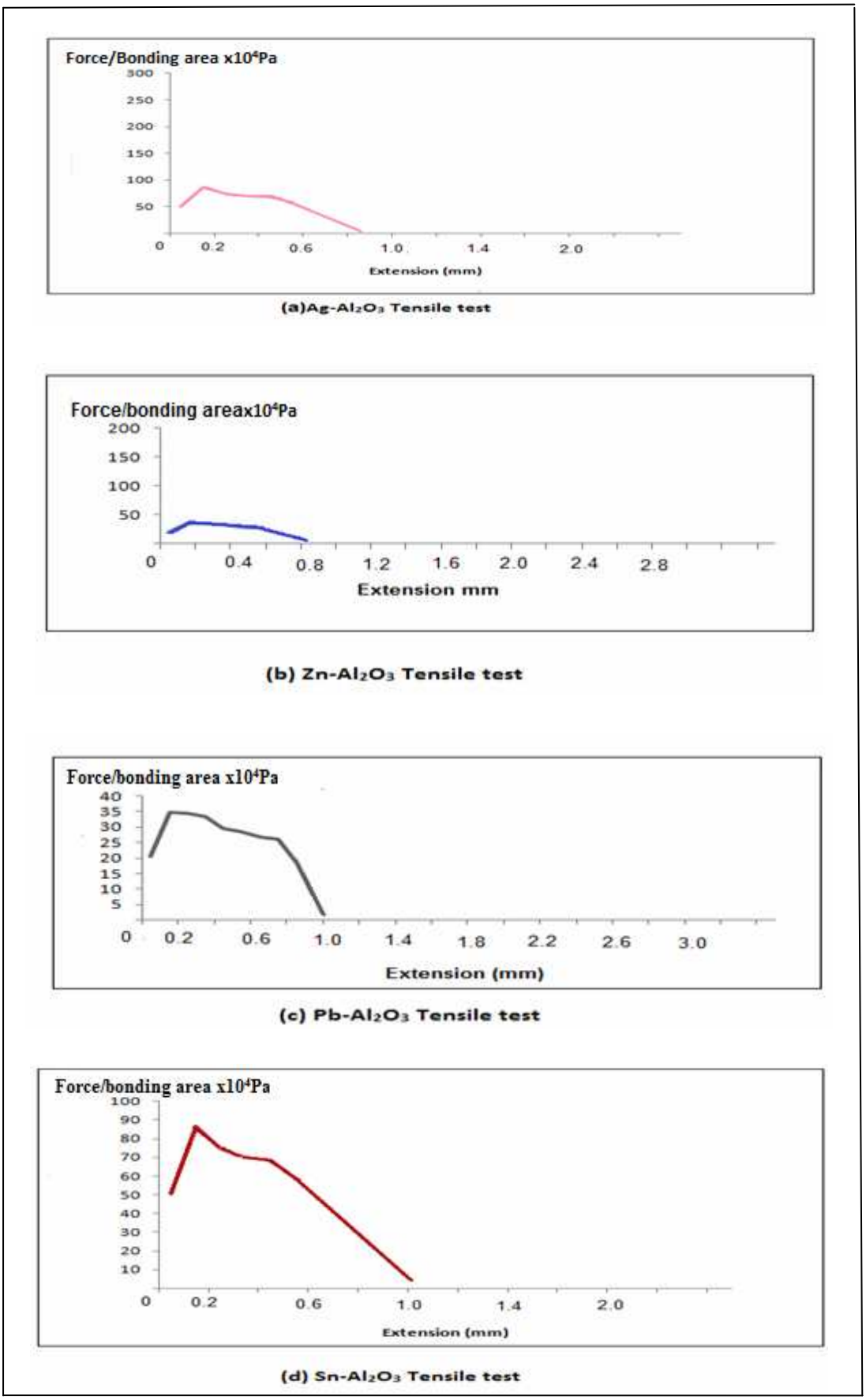


Fig. (3.1)Typical Tensile tests for metals bonded to alumina.

This separation needs a different forces for the different metals, even the whole samples were carried out at the same initial conditions as mentioned in chapter two. Table (3.2) represents fracture strength for all ceramic metals joined samples at 0.9 of its melting temperature.

Table (3.2)Fracture strength for ceramic-metals joined samples at bonding pressure of 0.3 MPa and 2hr socking time

Metal	Bonding Temp. (≈0.9 $T_m{}^0C$) $10^{\mp}$	Bonding Strength $x10^4 Pa(N/m^2)$	Bonding Catoagoria
Cu	900	Nill	weak
Ag	864	86.5	Strong
Al	594	Nill	weak
Zn	378	35	medium
Pb	295	35	medium
Sn	208	86.5	Strong

Bonding strength were identified as weak (Cu, Al), medium (Zn, Pb), and strong bonding (Ag, Sn). The weakness of the bond were due to the affinity of the metals (Cu, Al) to the oxygen, and 2hr socking time was too high create a satisfactory bond. Actually it is enough to enhance the oxidation process and weakening the bond. However, it seems to be with less effect for the other metals (Ag, Zn, Pb) which were illustrated in fig.(3.2).

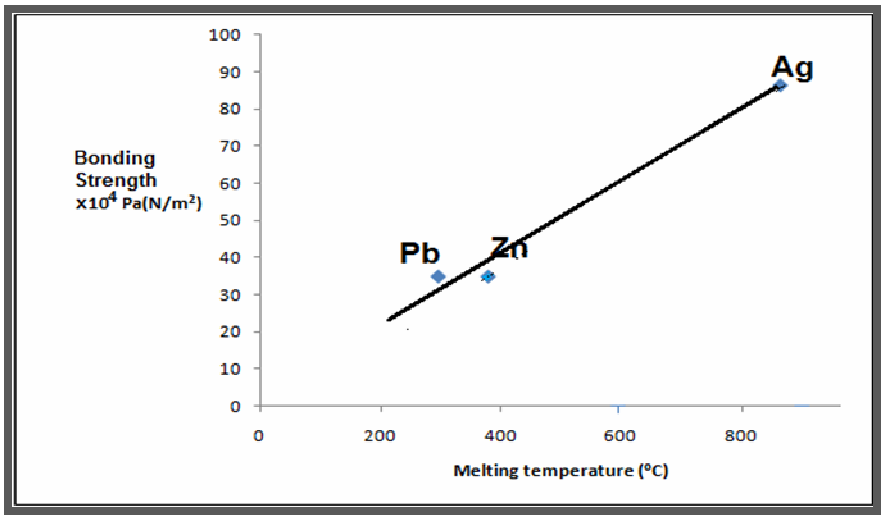


Fig (3.2) Fracture strength of ceramic-metal bonds as a function of bonding temperature

It is clear the linear relationships for the bonding strength with melting temperature of the mentioned metals. This behavior was well studied by others [38], but for the lower socking time (2 min.). In other hand, the effect of 2hr socking time may be more clear for increasing the joint strength for (Sn). Meanwhile, to sketch these results as it is shown in fig.(3.3), the results reveals well the over whole effect.

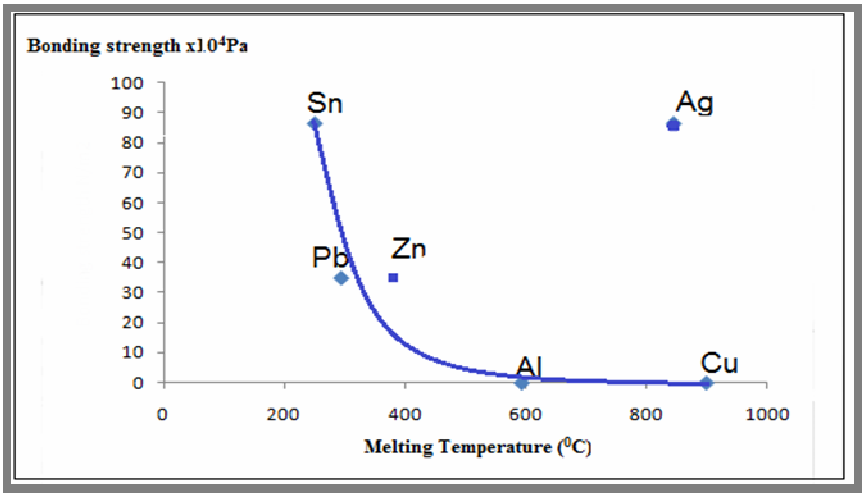


Fig (3.3) Fracture strength of ceramic-metal bonds as a function of bonding temperature

From the results of three categories of the joints, one can explain the effect of metals properties on the bonding strength, and the reasons of why some joint samples are high while the others are low or even weakly failed. The effect of metals oxidation was the most effective parameter. If the metals process shows a brittle layer growth of metal oxide in the interface regain and then this brittle layer will break the joint. This behavior was studied also by Amir, et.al.[39].

In copper-alumina joint samples the surface of metal oxidized and became black, brittle layer at the interface. Fig.(3.4)shows the fracture Cu-Al_2O_3 joint sample break at the center of the metal, which became as a brittle metal.

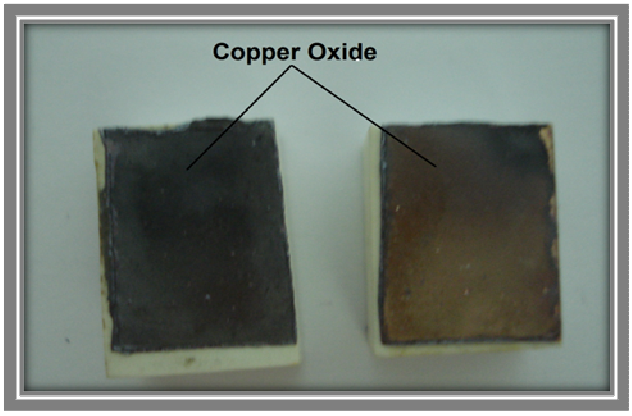


Fig.(3.4) Photographic illustration of copper- alumina joint sample, shows the diffusion and joining of copper oxides to alumina and fracture in metal

Tin (Sn) metal was benefit by increasing the socking time (joining time) and became with more joint strength with minimum level of oxidation. Some metals such as Ag, Zn and Pb shows a very thin oxidation layers, but without actual effect on joint strength.

Another reasons were effects the joints results such as the compatibilities of thermal expansions between metals and ceramic as

we have already discussed in chapter one, and its effects on the joint strength. Fig.(3.5) represents the relationships between bonding strength with the compatibility of thermal expansion of the joint partners.

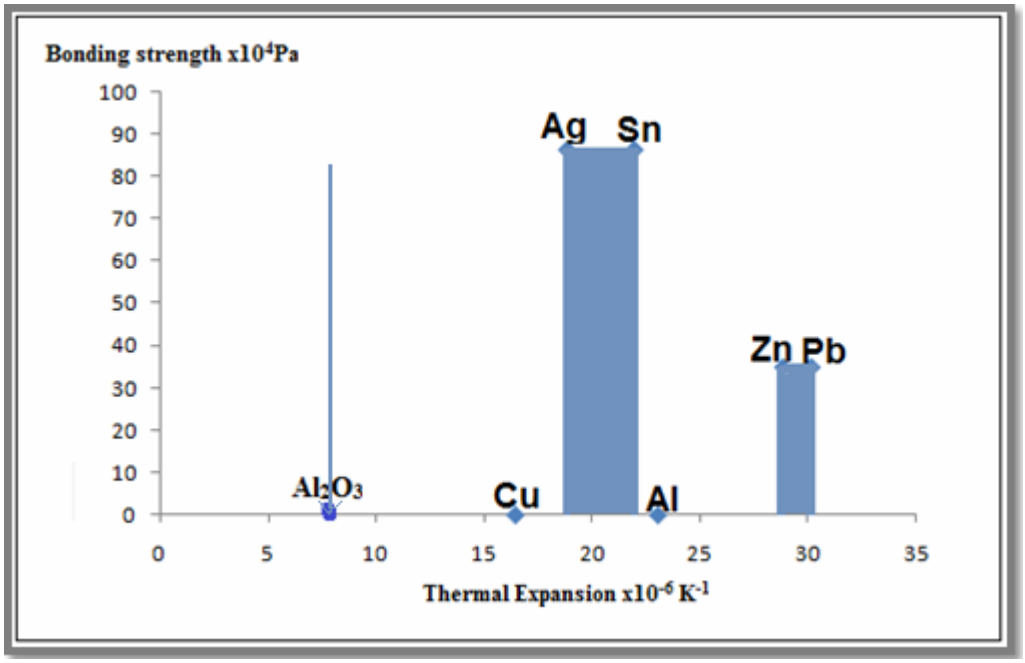


Fig.(3.5) Histogram between bonding strength with the compatibility of thermal expansion of the joint partners.

The relationship of bonding strength with atomic number were much more complicated and needs more research to understand it. Fig.(3.6) represent this relation. Ignoring the effect of oxidation on the joint strength for Al and Cu metals, bonding strength seems to be reduce with atomic number, which means the difficulty of solid state diffusion process with higher atomic numbers.

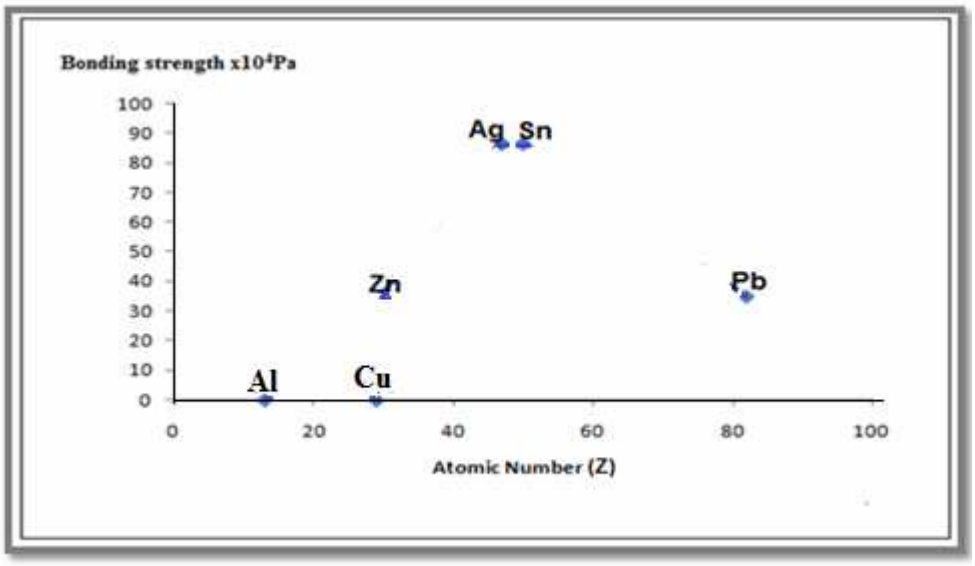


Fig.(3.6) Relationships of bonding strength with atomic number

Fig.(3.7) represent the relationships between bonding strength and mass number. Ignoring the effect of oxidation on the joint strength for Al and Cu metals, bonding strength seems to be reduce with mass number also, which means the difficulty to realize the solid state diffusion process with higher mass numbers.

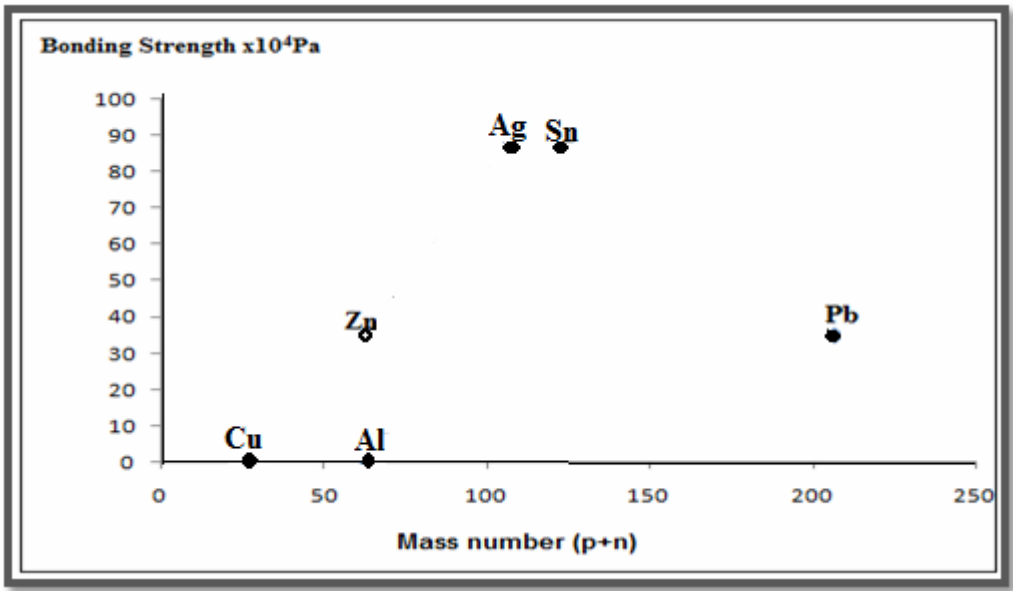
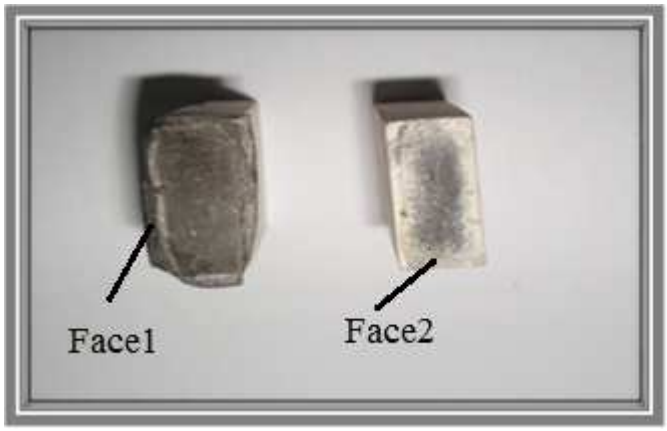


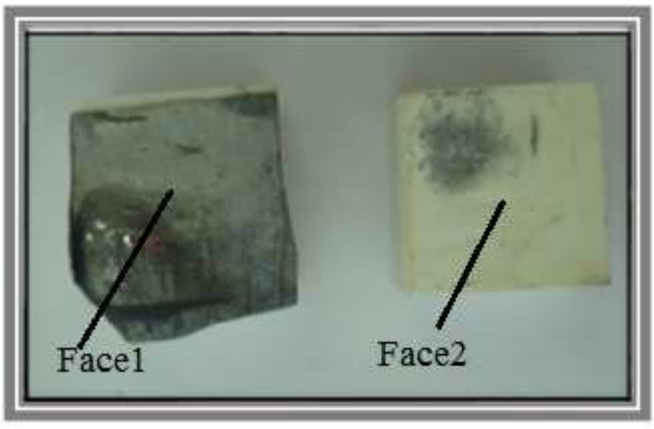
Fig.(3.7) Relationships of bonding strength with mass number

When the joint samples were tested by tensile machine the result show three types of fracture.

1.Fracture in the interface , this type is clear in the Zn , Pb samples. Fig.3.8(a, b) illustrate this type of fracture.



(a) Pb Sample



(b) Zn Sample

Fig. (3.8) Fracture in the joint interface for Pb and Zn samples

2. The fracture in the alumina part , this type of fracture is clear in Ag- Al_2O_3 joint as shown in figure (3.9).

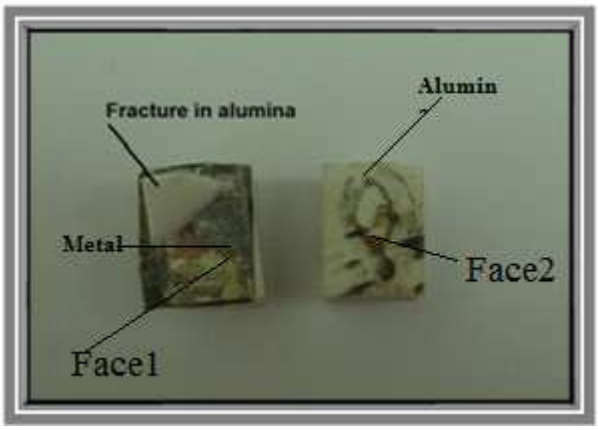


Fig. (3.9) The fracture in alumina part for Ag sample

3.The fracture in metal part , this type is clear in Cu sample when all atoms of copper are oxidized and became brittle and some of them diffused in alumina parts and covered two alumina surfaces, as shown in the previous figure (fig. 3.4).

3.3 Elemental Analysis

X-ray fluorescence technique was used for testing each face of the samples, which will prove a meaningful tool to study the diffusivity between the joint surfaces of dissimilar materials in solid state. The elemental analysis of the weak joined faces for each sample as it is illustrated in fig.(3.10), were conducted and revealed in fig's (3.11) to (3.16).

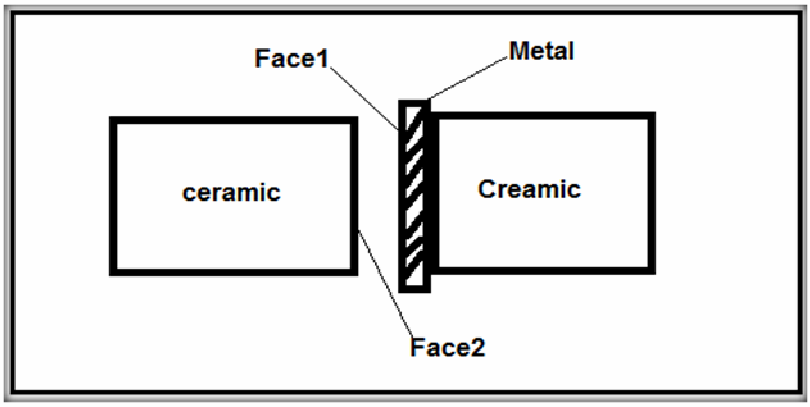


Fig. (3.10) Fracture sample faces

The X-Ray fluorescence technique was used (for each sample) to achieve the three type of fracture categories. The X-ray fluorescence for face1 for all samples are shown in figs.(3.11-3.16).

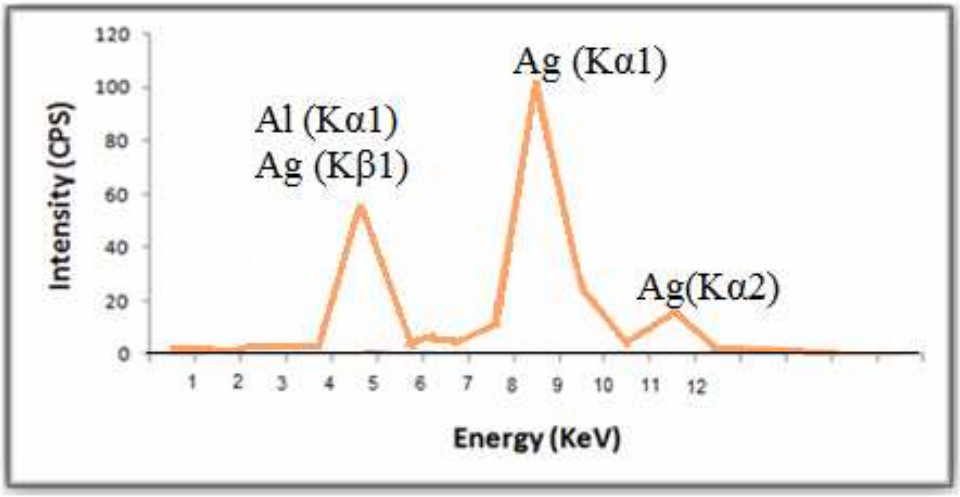


Fig. (3.11) X-ray fluorescence of Silver- Alumina sample (face-1)

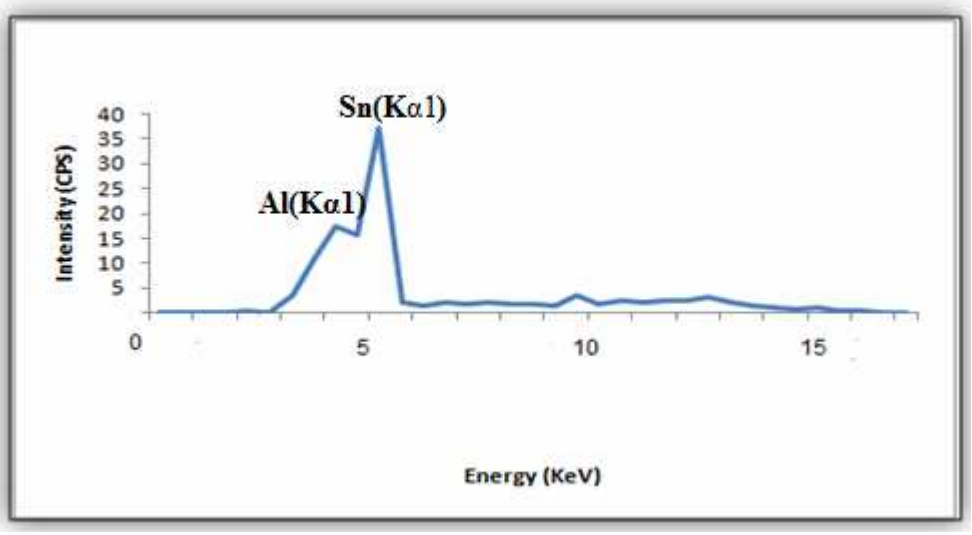


Fig. (3.12) X-ray fluorescence of Tin- Alumina sample (face-1)

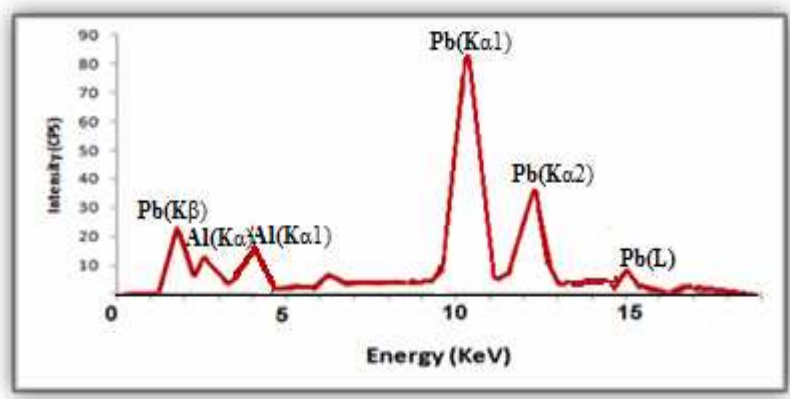


Fig. (3.13) X-ray fluorescence of Lead- Alumina sample (face-1)

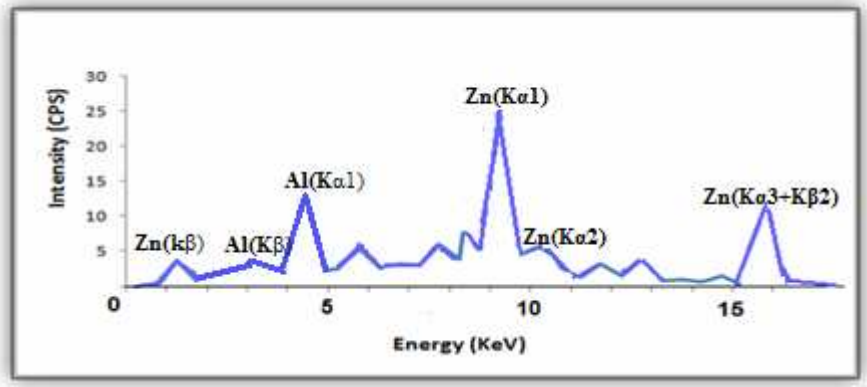


Fig. (3.14) X-ray fluorescence of Zinc- Alumina sample (face-1)

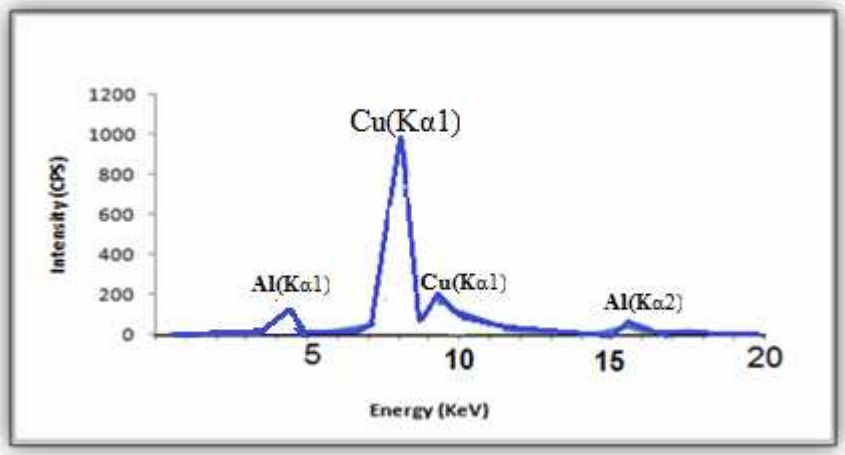


Fig. (3.15)X-ray fluorescence of Copper-Alumina sample (face-1)

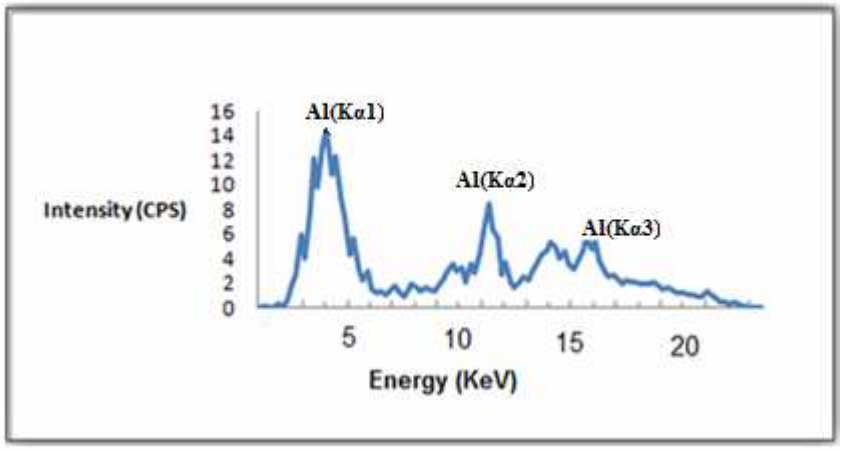


Fig. (3.16) X-ray fluorescence of Aluminum- Alumina sample (face-1)

The X-ray fluorescence for all samples of face2 as shown in figs(3.17-3.22)

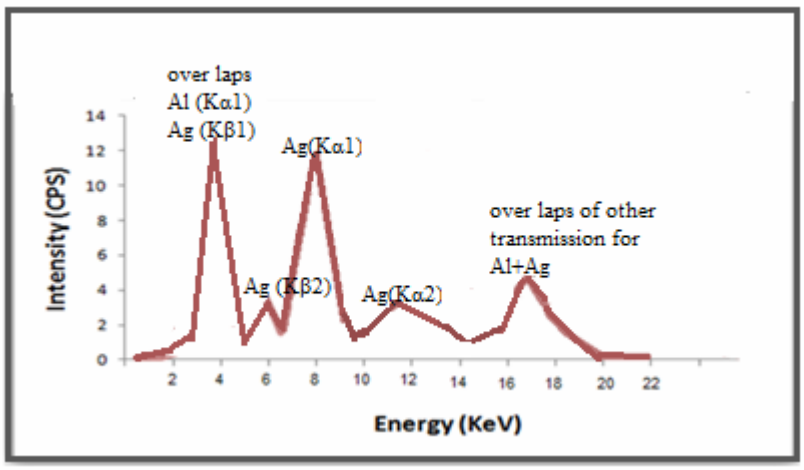


Fig. (3.17) X-ray fluorescence of Silver-Alumina sample (face-2)

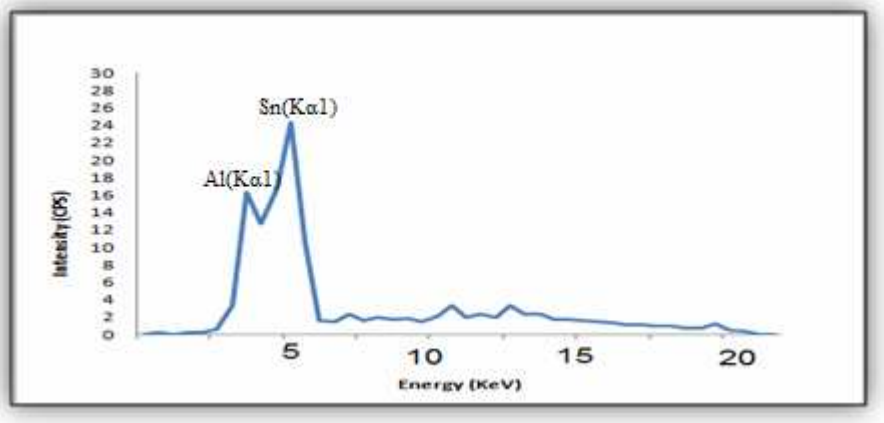


Fig. (3.18) X-ray fluorescence of Tin-Alumina sample (face-2)

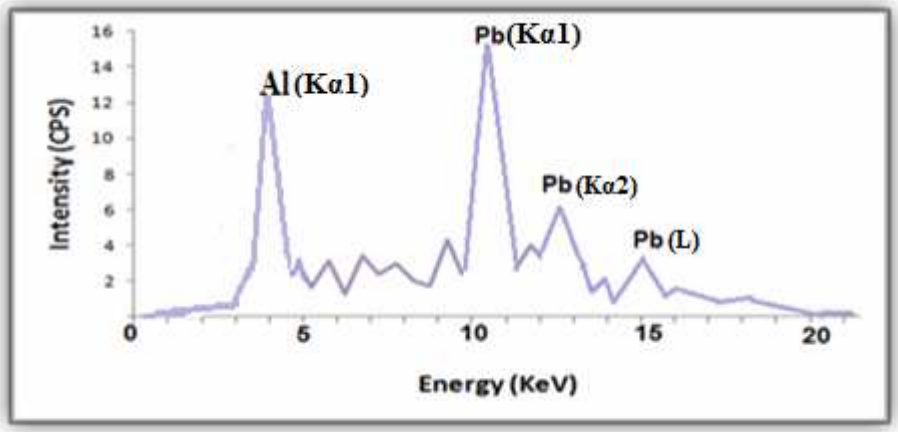


Fig. (3.19) X-ray fluorescence of Lead-alumina sample (face-2)

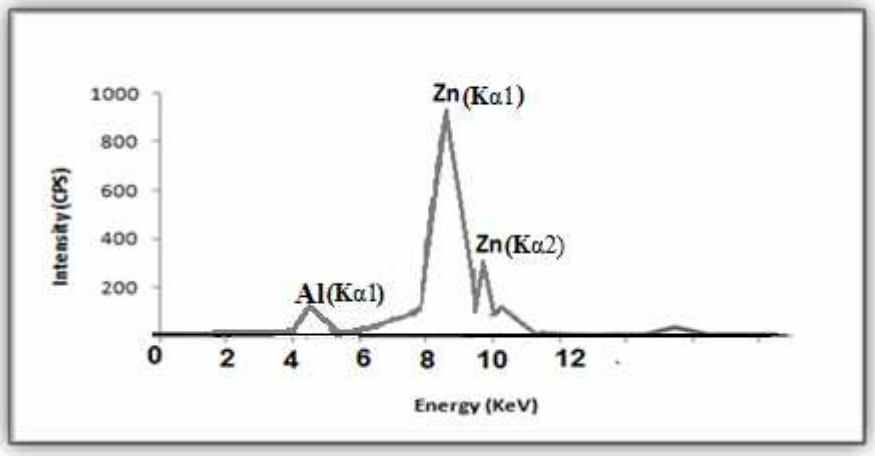


Fig. (3.20) X-ray fluorescence of Zinc-Alumina sample (face-2)

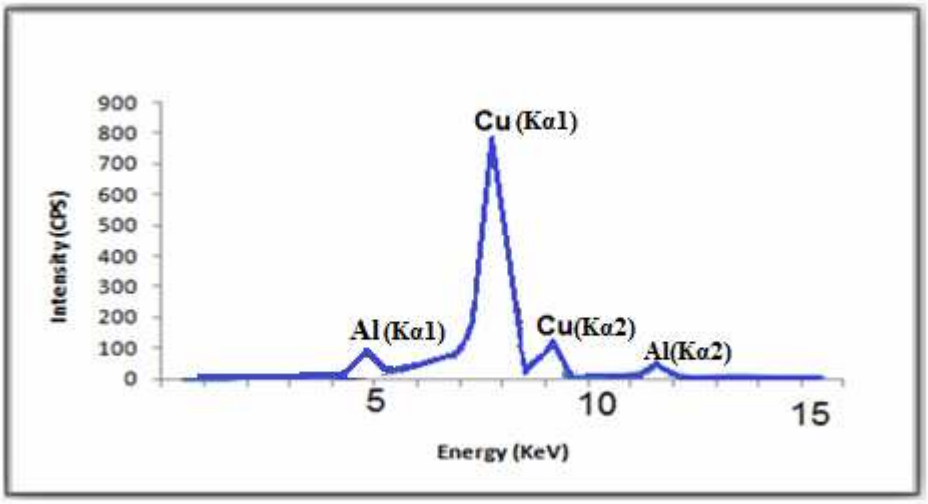


Fig. (3.21) X-ray fluorescence of Copper-Alumina sample (face2)

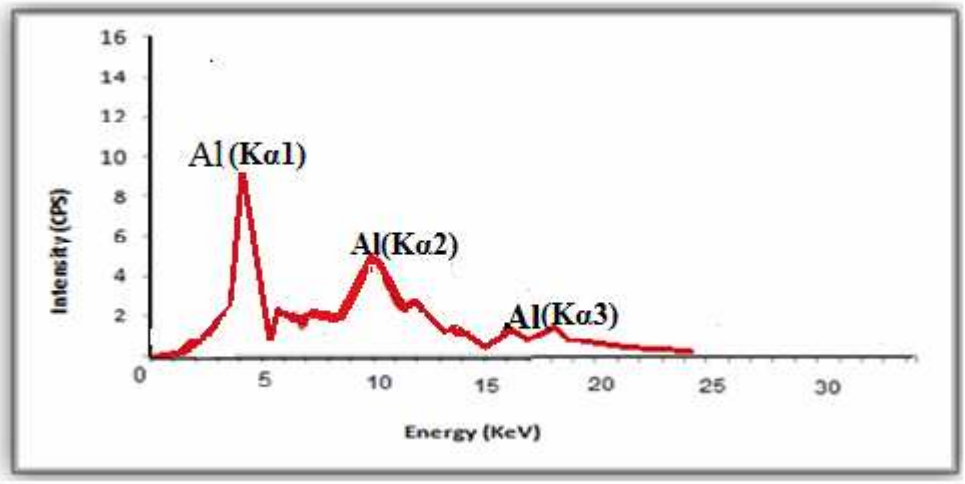


Fig. (3.22) X-ray fluorescence of Aluminum-Alumina sample (face-2)

3.4 Optical Microscopic

The identification of two fracture surfaces were carried out by optical microscope. Optical microscope gives clear evident pictures for all samples and express the causes of the weakness of the bonding, such as the effect of oxidation. The characterization of the two fracture surfaces (face 1and face2) were illustrated in figs.(3.22) up to (3.34).

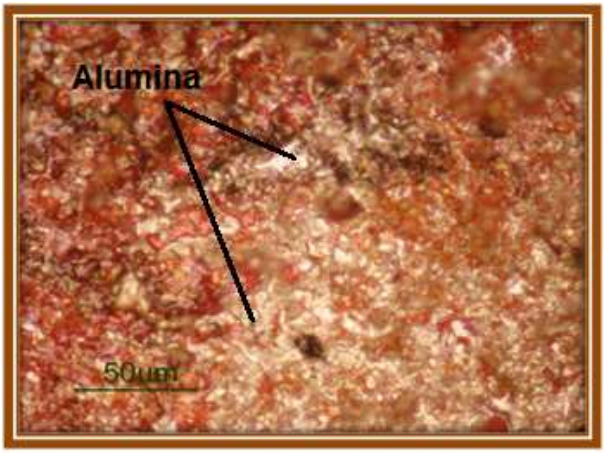


Fig.(3.23) Optical Microscope image for face1 for silver sample.

(Magnification 500x)

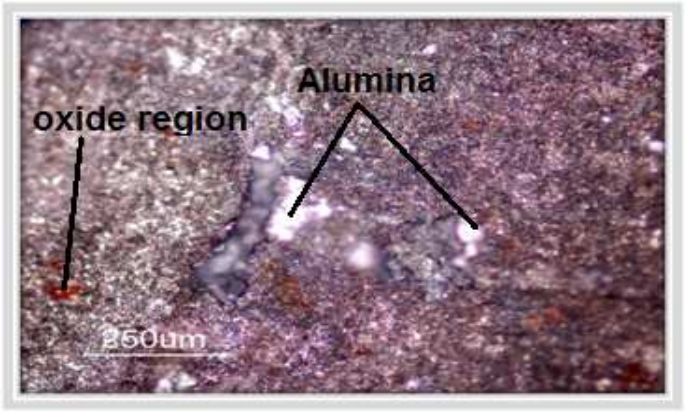


Fig.(3.24) optical microscope image for face2 for lead sample.

(Magnification100x)

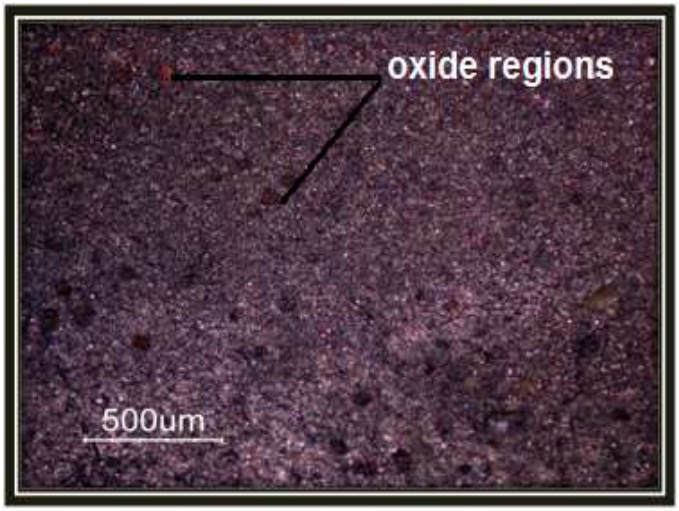


Fig.(3.25) Optical Microscope image for face2 for copper sample.

(Magnification 50x)

Figure(3.25) shows the red point regions , this regions mean the copper metal is oxide, also clear in fig.(3.24) of lead sample. The diffusivity was successes in all samples and this evident in figs.(3.20-25).

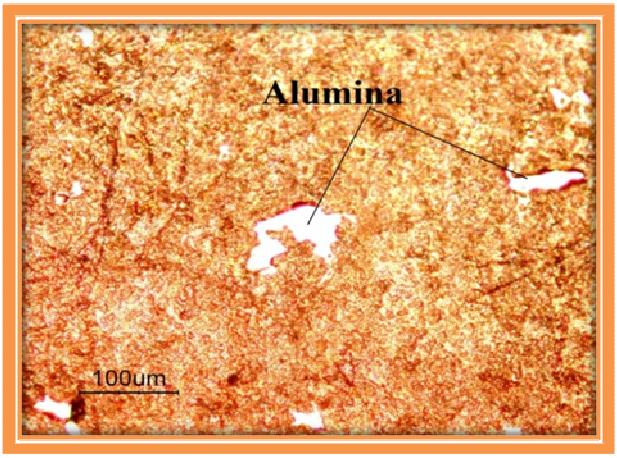


Fig.(3.26) optical microscope image for face1 for Aluminum.

(Magnification 200x).

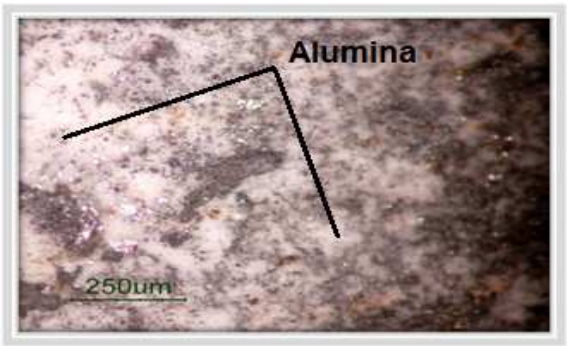


Fig.(3.27) optical microscope image for face1 for lead.

(Magnification100x).

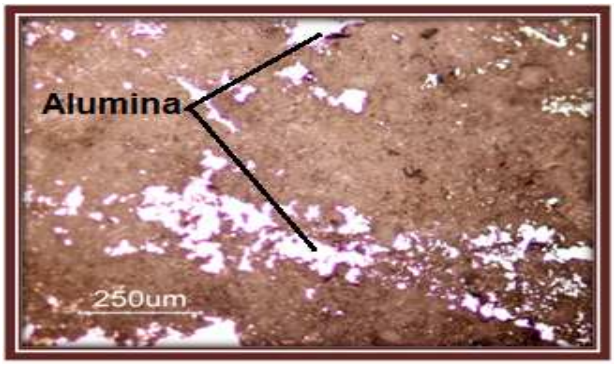


Fig.(3.28) optical Microscope image for face1 for Zinc.

(Magnification 100x)

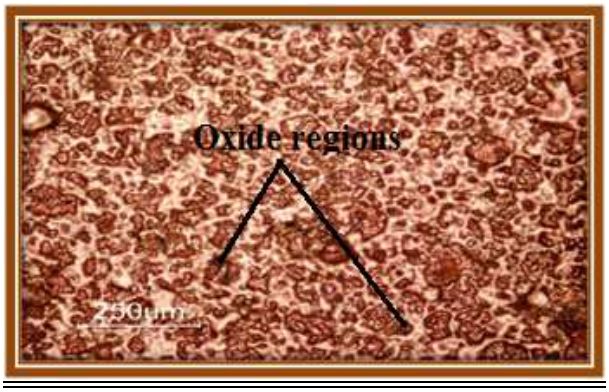


Fig.(3.29) optical Microscope image for face1 for Copper.

(Magnification 100x)

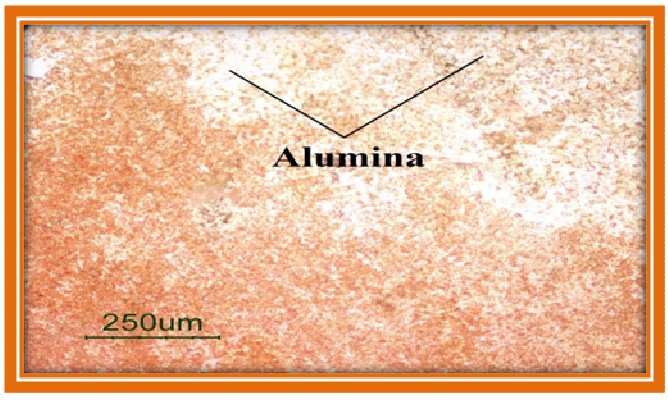


Fig.(3.30) optical Microscope image for face1 for Silver.

(Magnification 100x)

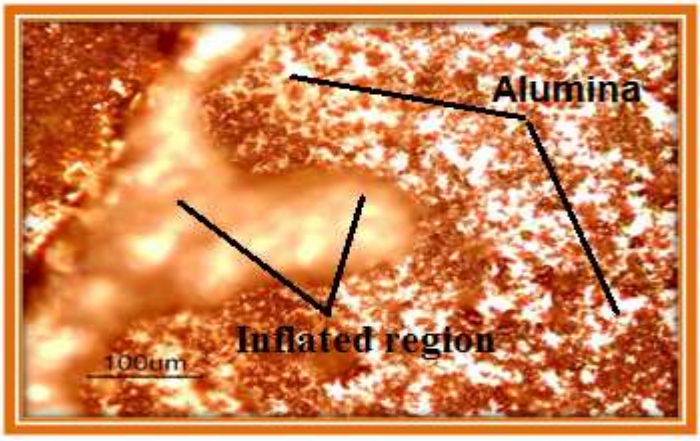


Fig.(3.31) optical Microscope image for face1 for Tin.

(Magnification 200x).

Fig.(3.32) shows the diffusivity of alumina in copper in tangential section.

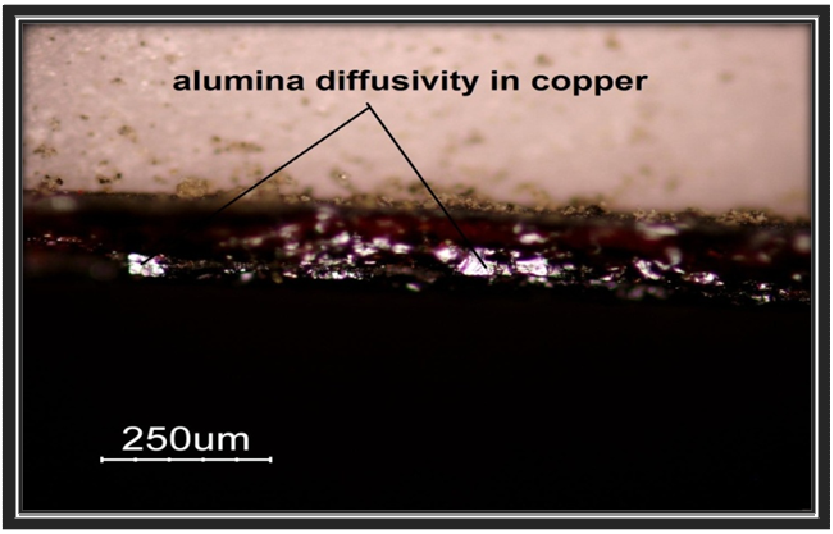


Fig.(3.32) optical microscope image for Copper-alumina in tangential section.(Magnification 100x)

Fig.(3.33) shows the inflated region in tin -alumina sample, so some region is higher than others.

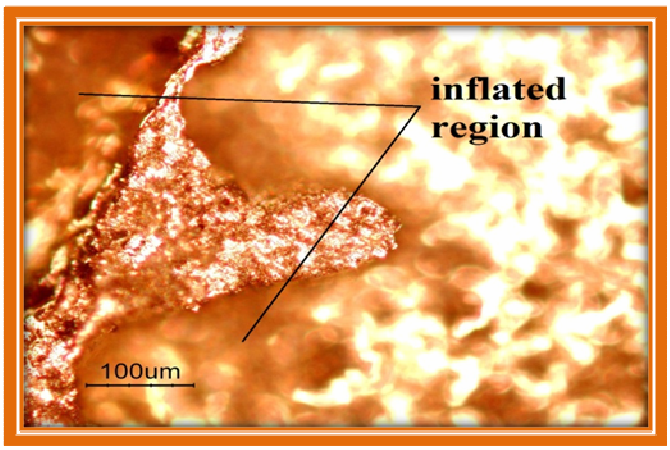


Fig.(3.33) optical microscope image for Inflated region for Tin.

(Magnification 200x)

Fig.(3.34)and (3.35) showed the crack is clear in alumina part of zinc-alumina sample.

Fig.(3.34) optical Microscope image for face2 for Zinc.

(Magnification 100x).

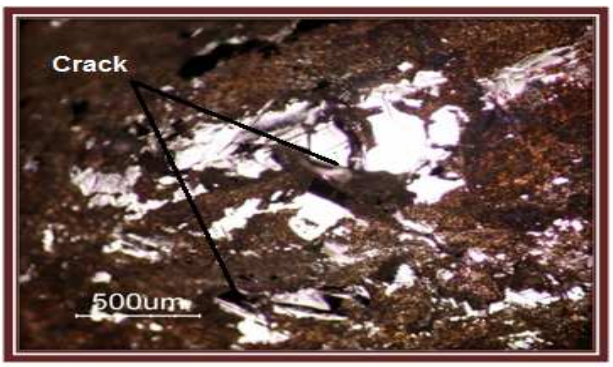
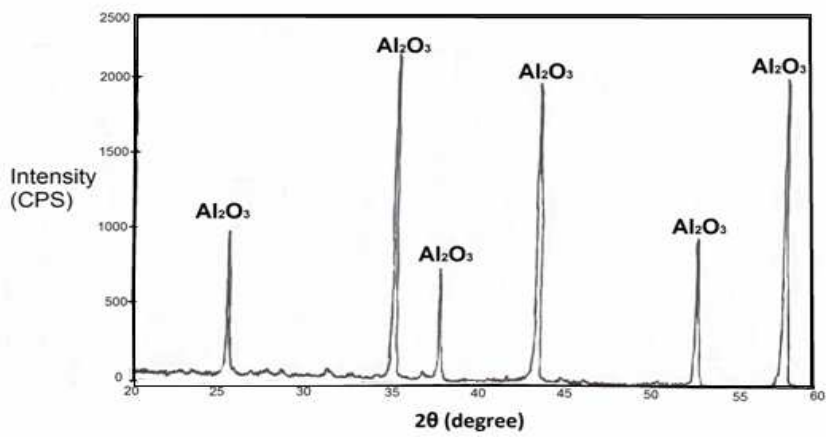


Fig.(3.35) optical Microscope image for face2 for Zinc.

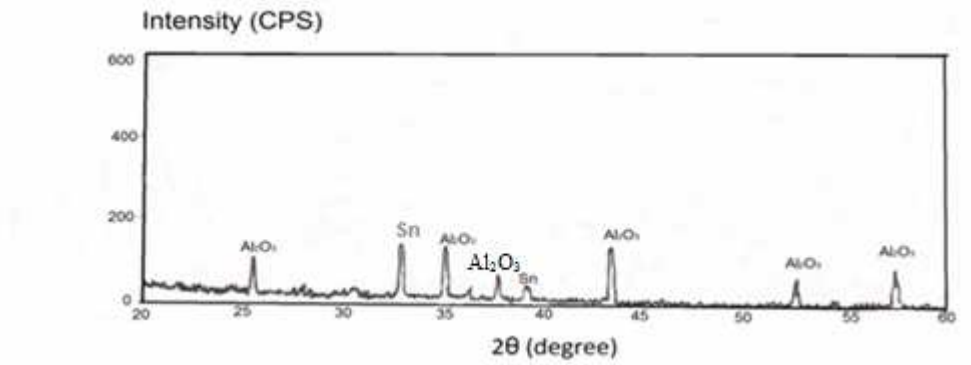
(Magnification 50x)

3.5 Phase Identification

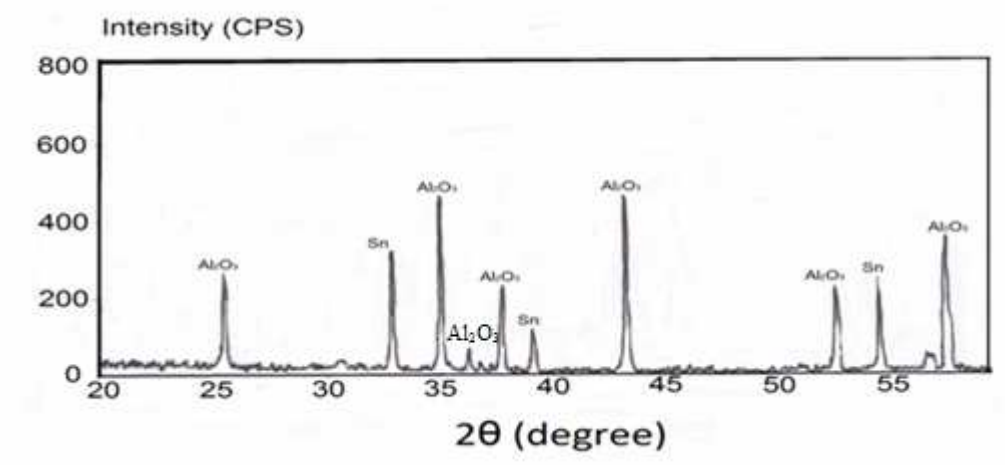
In order to detect the chemical nature of interlocking , an XRD pattern recorded for alumina which had been previously bonded with Sn for two faces (face1, face2) and alumina pure as shown in fig.(3.36)[48,49]. These patterns shows that no chemical reaction had occurred. The nature of the bonds between the metals and alumina ceramic is not yet clear whether it is of chemical or physical nature. The basic principle underlying the mechanism of bond formation between an ionic and metallic system is not yet fully understood[38].



(a)



(b)



(c)

Fig.(3.36) X-ray diffraction. (a) X-ray diffraction of Al_2O_3 pure sample. (b) X-ray diffraction of Sn sample face1. (c) X-ray diffraction of Sn sample face2.

The Conclusions and Future Work

4.1 The Conclusions

Conclusion of the present work can be summarized by the following remarks:

1. Selected metals would diffuse to alumina ceramic if the components are maintained in close contact under predetermined pressure and heated to about 90% of their melting points. The pressure that necessitate good bonding was 0.3MPa (N/m^2) for all metals.

2.The bond strength in silver and tin metals is greater than zinc and lead metals. However, no bond were detected in copper and aluminum because of oxidation effect for long bonding time(2h).

3.The X-Ray diffraction of all samples were showed no chemical reaction between ceramic and the metals.

4.2 Future Work

1.Other parameters affected on diffusivity, such as pressure and different atmospheres should be investigated.

2.Other metals foils with different thicknesses can be joined with ceramic to attempt the bond strength.

3. Fractured surfaces can be characterized by using advanced techniques such as electron spectroscopy for chemical analysis (ESCA) to obtain more surface information(elements, phases, depth of diffusion).

4.Interfacial reactions and rate of diffusivity should be studied theoretically, for different joining atmospheres.

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الخلاصة

يهدف البحث إلى دراسة متانة الربط الانتشاري بين السيراميك والمعادن في الحالة الصلبة حيث تم استخدام المعادن عالية النقاوة (الفضة والنحاس و الخارصين والألمنيوم والرصاص و القصدير) وتم تقطيعها وتهيئتها على شكل رقائق بسمك ١ملم ولمساحة تقريبة1x1سم^{2} لغرض ربطها بسيراميك الألومينا.

تم تحضير مجسمات الالومينا عالية النقاوة وبأبعاد 0.5x1x1 سم^{3} واجريت عليها عمليات صقل وتنظيف وتهيئتها للربط بالمعادن المختلفة.

تم تنظيف النماذج بطرق تنظيف معروفة وفق مواصفات عالمية معتمدة ومن ثم وضع شريحة المعدن بين قطعتي الالومينا وضغطها بقوة 0.3 ميكا باسكال تمهيدا لوضعها داخل الفرن المعد لهذا الغرض ليتم تسخين المجموعة بدرجة حرارة ٩٠% من درجة انصهار المعدن ولمدة ساعتين.

بعد إجراء عملية كسر المفصل أللحامي تبين إن متانة الربط الانتشاري للفضة والقصدير مع الالومينا عالي نسبيا 86,5 x10^{4} باسكال على التوالي، ومن جهة أخرى تبين إن متانة الربط لكل من النحاس والألمنيوم مع الالومينا كانت ضعيفة جداً.

درست نتائج الكسر مجهريا باستخدام المجهر الضوئي (OM) وكذلك باستخدام تقنية الأشعة السينية بنوعيها(فلورة الأشعة السينية (XRF) وحيود الأشعة السينية(XRD)) ومن خلال هذه الدراسة وجد إن احدى الاسباب الرئيسية المؤثر على الانتشارية وقوة الربط هو عملية التأكسد التي تحدث للمعدن إثناء اجراء عمليات الربط.

فحوص المجهر(OM) والأشعة السينية (XRD وXRF)اثبتت وجود الانتشارية بين المعدن والسيراميك وان فحص حيود الاشعة السينية (XRD)اثبت عدم وجود تفاعل كيميائي بين مواد الربط المستخدمة في هذا البحث.

النتائج العملية لطبيعة الكسر تبين وجود ثلاث هيئات للكسر وهي:

١.الكسر في السيراميك

٢.الكسر في منطقة الفاصلة بين المعدن والسيراميك

٣.الكسر في المعدن

جمهورية العراق
وزارة التعليم العالي والبحث العلمي
جامعة النهرين/ كلية العلوم
قسم الفيزياء

دراسة متانة الربط الانتشاري بين السيراميك والمعادن في الحالة الصلبة

رسالة مقدمة ألى
قسم الفيزياء/كلية العلوم/جامعة النهرين
كجزء من متطلبات نيل درجة ماجستير في الفيزياء

من قبل
أروى غازي ناجي الطائي
بكالوريوس علوم فيزياء

بأشراف

د. زاريه ازات سركيس
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رمضان -١٤٣١