

**Republic of Iraq**  
**Ministry of Higher Education**  
**and Scientific Research**  
**Al-Nahrain University**  
**College of Science**  
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# **Evaluation of the Intermediate Layer of Graphite Bonded Metal**

**A Thesis**

Submitted to the College of Science / Al-Nahrain University in a Partial  
Fulfillment of the Requirement for the Degree of Master of Science in Physics

**By**

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April  
2016

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بِسْمِ اللَّهِ الرَّحْمَنِ الرَّحِيمِ

قَالُوا سُبْحَانَكَ لَا عِلْمَ لَنَا إِلَّا مَا عَلَّمْتَنَا إِنَّكَ أَنْتَ الْعَلِيمُ  
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**Dedication**

**I dedicate this effort to**

**My mother**

**My husband**

**And to everyone who supported me**

## Acknowledgement

Above all, I thank **God** and praise him, that god has made me what I hope to complete the master's degree in physics.

I extend with great thanks and appreciation to my supervisor Assistant Professor **Dr. Thimir Abdul-Jabbar** for suggestion this project and follow up since its first steps and gave me advice and guidance help me to output this work in this way, ask God grant him the best reward.

I extend my thanks to College of Sciences departments, dean, professors and teachers and to all those who helped me in the education of undergraduate and master's stages.

I want to thank Assistant professor **Mrs. wasan** and **Mrs. Evon** for help me in testing samples.

I thank **Abu Adnan** in the glass workshop to help me in providing the quartz glass.

I want to thank **Haider (Abu Abdallah)**, a master's student in chemical engineering / Al- Nahrain University to help me in the metal cutting.

Also I would like to give my thank to **Nather Basher** PhD student in Physics Science/Al-Nahrain University to help me in general information.

I would like to express my gratitude to my friends and my colleagues who have supported me during my studies at department of physics, College of sciences / Al-Nahrain University.

Finally , I would like to give my deepest thanks to my husband and my family for their love and support all the time.

## Abstract

This work investigates the joining between graphite and stainless steel grade (410), by using active filler metal paste. Brazing technique was chosen because it is efficient, versatile, fast and economical.

High purity of metal powder of silver, copper and titanium in different concentrations were mixed carefully to prepare a filler paste. Stainless steel and graphite as a cubic shape were prepared to study both wetting and bonding behaviour as a function of essentially of furnace temperature. Decontamination processes were achieved on the all specimens under study according to the standard methods. The wettability was firstly studied, the paste drop of such filler ratio was lied on the both graphite and stainless steel surfaces. Good wettability was obtained when the filler paste consist of pure titanium at the furnace temperature of 950 °C and furnace vacuum pressure  $10^{-2}$  torr. Two Types of assembly designs were selected known butt and interlocking according to the experiments variables. For studying the bonding behaviour between the pieces to be joined, the filler paste was lied in between stainless steel and graphite as a sandwich form. Bonding processes were performed at different furnace temperatures as follows (850, 900, 950, 1000) °C with soaking time 15 min. Maximum tensile strength 15 Mpa was obtained when the filler paste consist of (75Wt% Ti + 25Wt% Ag) by using interlocking design. The fractured surfaces were examined by Non-destructive testing involved optical microscopy, Scanning electron microscopy and Energy dispersive x-ray. X-ray diffraction used for studying the intermediate structure. New structure phases were established at the intermediate layer involved (( $O_{20}P_4Ti_5$ ,  $O_5STi$  and  $CrO_9P_3$ ), ( $TiO_2$  and  $Ag_3O_4$ )).

These new phases considered enhancement the bonding strength of the assembly.



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## List of Abbreviations and Symbols

Cu	Copper
CTE	Coefficient of Thermal Expansion
$\alpha_v$	Coefficient of volume Thermal Expansion
$\alpha$	Coefficient of Linear Thermal Expansion
DSC	Differential Scanning Calorimetry
EDX	Energy Dispersive X-ray
F	Filler
$T_f$	Final Temperature
$L_F$	final length
Gr	Graphite
$L_o$	Original Length
$T_o$	Original Temperature
$V_o$	Original Volume
Ag	Silver
$C_p$	Specific heat capacity at a constant pressure
SEM	Scanning Electron Microscopy
St.St	Stainless Steel
Ti	Titanium
TG	Thermogravimetry
$\Delta V$	Volume change
XRD	X-ray Diffraction

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# **CHAPTER ONE**

**INTRODUCTION AND LITERATURES SERVAY**

## Chapter One

### Introduction and Literatures Servay

#### 1.1 Introduction

A large number of competing joining processes are in common use. It can be divided into two broad groups; cold processes and hot processes. It is this fundamental difference that forms the basis of the chart shown in figure 1.1. From this chart, only mechanical joints can be taken easily, the other joining processes leads to produce permanently joints. In many engineering application the fact that permanent joints can be produced is often the most important consideration [1,2].

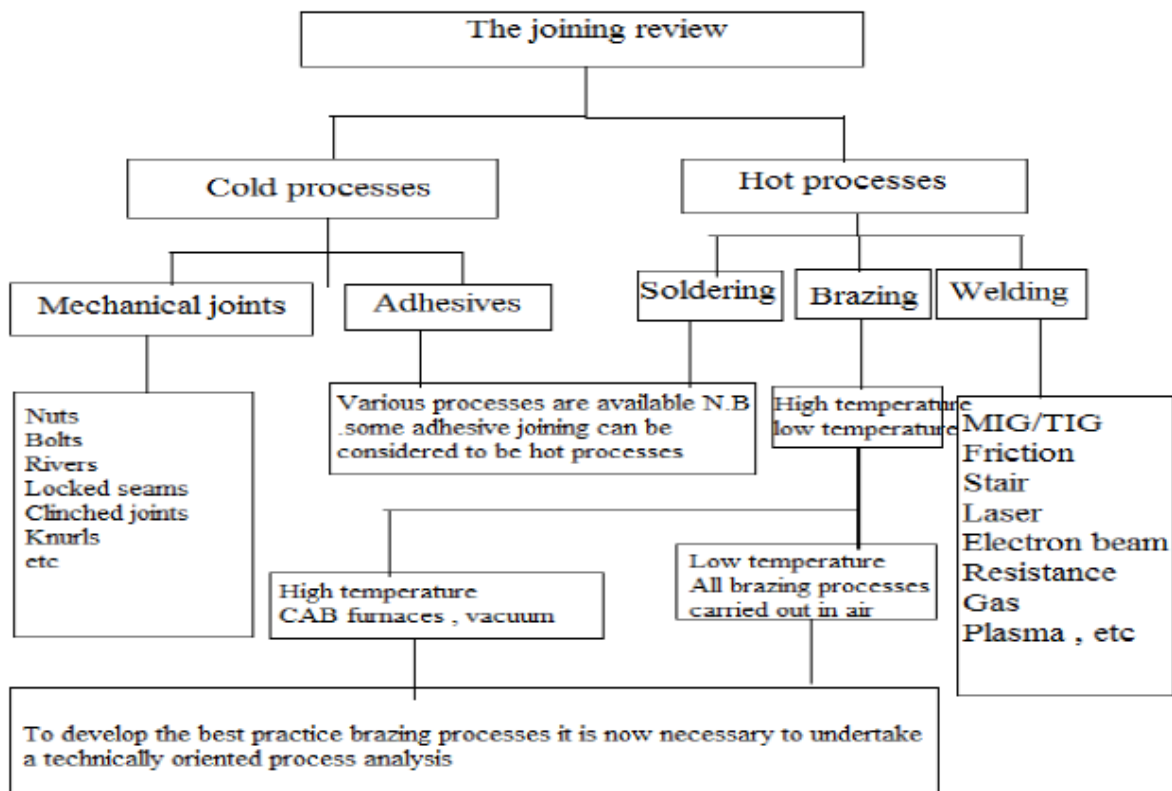


Figure1.1: classification of joining processes[1].

## 1.2 Reviewing the Brazing Process

Brazing is considered one of the best joining processes. It is necessary to undertake a process audit to determine the precise parameters that will have to be satisfied to ensure that only the best practice brazing process is employed. This is achieved by making a detailed assessment of the eight fundamental characteristics that have a direct impact on the success or failure of a specific brazing procedure. The eight generic factors that must be considered are [1,3]:

1. The parent materials that are to be joined.
2. The joint design that is to be employed.
3. The dimensions of the joint gap.
4. The choice of filler material.
5. The means of removing of oxide films from the surfaces to be joined.
6. The heating method that will be used to bring the joint area to brazing temperature.
7. The design of the fixture that will be used to support the components during the brazing operation.
8. The level of complexity needed in the production equipment to satisfy the production requirement.

The format of the audit procedure is shown in figure 1.2. All that has to be done is to follow the route from stage 1 to stage 9 in the Figure, taking account of the conditions that are known to be present in the brazing process being reviewed. Remember, since each stage is intimately linked to all the others, it is reasonable to expect that if a change has to be made in one stage it will almost certainly generate the need for changes to be made in one or more of the other stages.

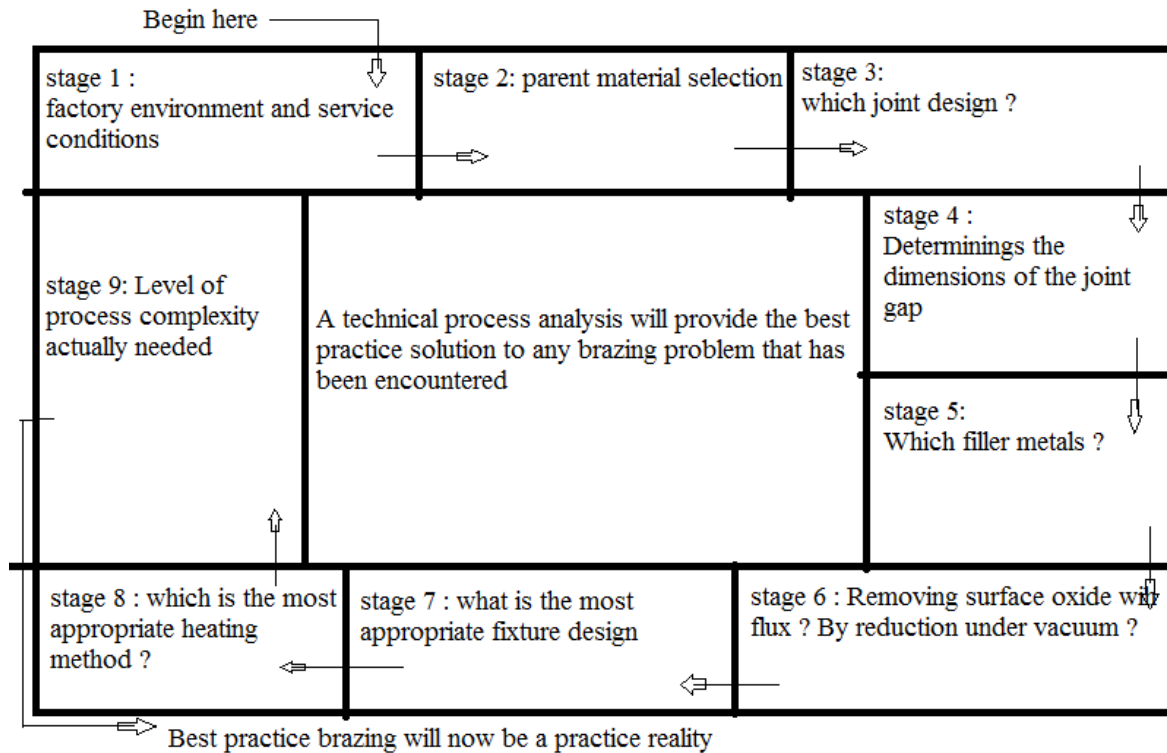


Figure1.2: Layout of analysis procedure. [1]

### 1.3 Bonding Between Dissimilar Materials

Brazing is a technique of joining two similar or dissimilar metal pieces together by heating the surface and by using a non-ferrous filler metal having its melting point above  $427^{\circ}\text{C}$  but below the melting points of metals to be brazed. The molten filler metal is distributed between the joint surfaces by capillary action. Which on cooling results in a sound joint. The main advantage of brazing process is the joining of dissimilar metals and thin sections. The process is mostly used for joining pipes and other fittings, carbide tipping on tool shanks, electrical parts, radiator, repair of cast iron parts and heat exchangers [2,3]. In brazing, bond is produced by the formation of either solid state solution (diffusion bonding ) or intermetallic compounds of the parent metal (job) and one of the metals in the filler material (brazing alloy). The strength of the brazed joint is provided by metallic bonding [2,3].

## 1.4 Soldering

The basic distinction between brazing and soldering is that brazing conducted at higher temperature (soldering process use filler metals having a liquidus not exceeding 450°C or 840°F). The historical distinction between the processes has its origin in the earliest solders which were based on tin. While brazes were based on copper-zinc alloys. Indeed, the word braze is a derivation of the old english braes, meaning to cover with brass. On the other hand, the term solder is an adaptation of the old French soudure, meaning to make solid [3,4].

Brazing and soldering require the application of a number of scientific and engineering skills to produce joints of satisfactory quality and reliability . Brazing employs higher temperatures than soldering, but the fundamental concepts are similar, particularly with respect to metallurgy and surface chemistry (see table 1.1). However, joint design, materials to be joined, filler metal and flux selection, heating methods and joint preparation can vary widely between the two processes. Economic consideration involving filler metal and process technology are also varied, particularly in relation to automated techniques, inspection and testing.

Brazing and soldering are performed in many industries, from exotic application in the electronics and aerospace field to everyday plumbing applications [3,4]. The type of metallurgical reaction between a filler and parent metal is sometimes used to differentiate soldering from brazing. Solders usually react to form intermetallic phases that is compounds of the constituent elements that have different atomic arrangement than the elements in solid form. By contrast, most brazes form solid solutions, which are mixtures of the constituents on an atomic scale. However, this distinction does not have universal validity. For example, silver-copper-phosphorus brazes react with steels to form the interfacial phase of  $Fe_3P$  in a similar manner to the reaction of tin-base solders with iron or steels to form  $FeSn_2$  other hand solid solution form between silver-lead solders and copper just as they do between the common silver- base braze and copper [3,4]. Soldering and brazing involve the same bonding mechanism, that is reaction with

the parent material, usually alloying, so as to form metallic bonds at the interface. In both situations, good wetting promotes the formation of fillers that serve to enhance the strength of the joints. Similar processing conditions are required, and the physical properties are comparable, provided that the same homologous temperature is used for the comparison [3,4].

### **1.4.1 Welding**

Welding is defined as a joining process that produces coalescence of materials by heating them to the welding temperature, with or without the application of pressure or by the application of pressure alone, and with or without the use of filler metal. In less technical language, a weld is made when separate pieces of material to be joined combine and form one piece when heated to a temperature high enough to cause softening or melting and flow together. Pressure may or may not be used to force the pieces together. In some instances, pressure alone may be sufficient to force the separate pieces of material to combine and form one piece. Filler material is added when needed to form a completed weld in the joint [3,4].

Table 1.1: Comparison of soldering , brazing , and welding

Parameter	Soldering	Brazing	Welding
Joint formed	Mechanical	Metallurgical	Metallurgical
Filler metal melting temperature °C(F)	450< (<842)	450> (>842)	450 >(>840)
Base metal	Does not melt	Does not melt	.....
Flux used to prevent oxidation of the base and filler metal and to assist in wetting of base metal surfaces	Required	Optional	Optional
Typical heat sources	Soldering iron; ultrasonics; resistance; oven	Furnace ; chemical reaction ; Induction; torch ; infrared	plasma ; electron beam ; tungsten and submerged ; resistance ; laser
Tendency to warp of burn	Atypical	Atypical	potential distortion and warpage of base metal likely
Residual stresses	.....	.....	Likely around weld area

## 1.5 Key Parameter of Brazing

### 1.5.1 Braze Joint Design

From a mechanical standpoint, the design of a brazement is not different than the design of any other part. Static loading, dynamic loading, stress concentration, and environmental factors must all be considered. In addition, some important factors specific to brazements are [4].

1. The composition of the base materials and the filler.
2. The type and design of the joint .
3. The service requirements, including mechanical performance, electrical or thermal conductivity, pressure tightness, corrosion resistance and service temperature.

In general, the strength of the filler metal in a brazed joint is lower than the strength of the base materials. the specific joint strength will vary according to



joint clearance, degree of filler – substrate interaction and presence of defect in the joint.

There are four types of joints used in brazing design, these are lap joint, butt joint, butt - lap joint and scarf joint. A variant of the butt joint, which forces loading more toward shear is called the scarf joint. These joint types are shown in figure 1.3 [5,6]. In lap joint, overlap of at least three times the thickness of the thinner member will usually yield the maximum joint efficiency. Scarf joints are attractive because they increase the joint area without increasing the thickness of the joint [4].

Joint clearance or gap, is a key parameter in the design and production of brazed joint for several reasons. First, the clearance in a joint is important purely from the standpoint of the effect of mechanical restraint. The plastic flow of braze filler is restrained by the higher strength base material only if the filler does not get too thick.

On the other hand, for purpose of strain accommodation, the braze filler layer cannot be too thin either. Second, joint clearance must be sufficient to prevent the entrapment [4,6].

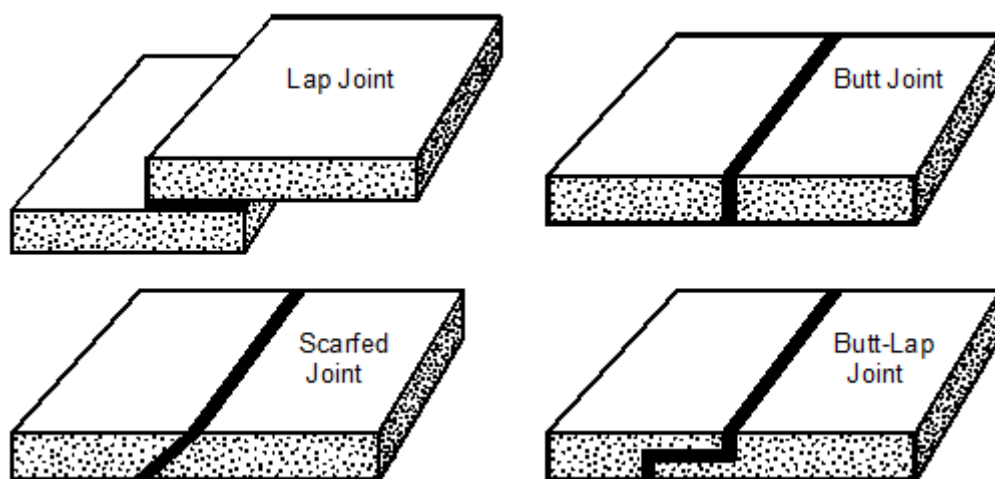


Figure 1.3: Various geometries used in brazed joints. [5,6]

Third, the relationship between joint clearance and capillary force controls proper flow and distribution of the filler. For diffusion brazing, clearance controls the amount of filler that must be diffused away. It is vitally important that the braze filler completely fills the joint and that defects are avoided during brazing. Defects to be avoided included voids (i.e. entrapped gas pockets), unbrazed areas, pores or porosity (i.e. from evolved or absorbed gases), flux inclusions, cracks, and brittle (often, intermetallic ) compounds. One of the most effective way of preventing defects of these types is to properly place filler [4,5]. Figure 1.4 shows how filler can be preplaced for maximum effectiveness, using various physical forms. Braze filler whether applied during the brazing or preplaced and whether the process is performed manually or automatically should be applied to the joint in such a way that when it flows through the joint properly, filler will appear at some point remote from the point of application .

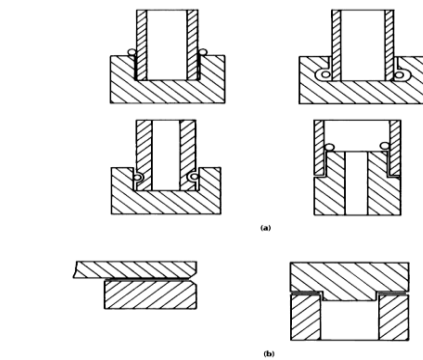


Figure1.4: Suggested methods for preplacing brazing fillers for maximum effectiveness . (a) preplaced filler metal wires ; (b) preplaced filler- metal shims. <sup>[4]</sup>

These techniques rely on the sweeping action of the filler under the influence of the capillary forces is a reasonable sign that proper flow and fill occurred [4,6].

### 1.5.2 Temperature and Time

The temperature of the brazing filler metal naturally has an important effect on the wetting action. The selection of an optimum braze temperature requires an understanding of influence of temperature on both the wetting and flow of the filler metal, because the wetting and alloying action improves as the temperature increases. Of course, the temperature must be above of the brazing filler metal and below the melting point of the parent metal. Within this range the temperature generally selected that will give the best satisfaction from overall standpoint [7].

As the temperature of the braze alloy is increased, the alloying and wetting action of the filler metal increases. In general, the brazing temperature selected must be above the melting point of the filler metal [8].

Usually the lowest brazing temperature are preferred to :

- a. Economize on heat energy required.
- b. Minimize heat effect on the base metal (annealing , grain growth, or warpage, for example).
- c. Minimize base metal /filler metal interactions.
- d. Increase the life of fixtures, jigs , or other tools.

Higher brazing temperature may be desirable so as to:

- a. Use a higher – melting but more economical brazing filler metal.
- b. Combine annealing, stress relief, or heat treatment of the base metal with brazing.
- c. Permit subsequent processing at elevated temperature .
- d.Promote base metal interactions in usually used to increase the re-melt temperature of the joint.

- e. Effectively remove surface contaminants and oxides with vacuum brazing.
- f . Avoid stress cracking [7,8].

Braze filler metal can be fairly complex alloys and their melting can take place over a range of temperatures. Melting which begins at the solidus temperature of 780°C is not complete until a temperature in excess of 850°C is reached. Within that melting range there is a 'mushy' mixture of the liquid and solid which has wetting and flow properties distinct from those of the filler metal above the liquidus temperature. Flow in the partially melted filler is much reduced and the wetting and spreading tendency of the liquid phase in the mixture leads to the phenomenon of liquidation or the tendency of the lower melting constituents of a filler metal to separate from the higher melting constituents. Liquidation results in melting point shifts and flow problems with the remaining higher melting constituents. The rate of heating through the melting temperature range and the brazing time are important parameters to avoid these type of problems[7,8]. Alloying can occur between the liquid filler metal and the base materials during the braze process. This type of interaction occurs to some extent in all braze processing, and must be controlled in some cases where the consequences degrade the process and the joint properties . In fact a number of metallurgical processes occur during the braze process that affect the solidification characteristics, grain size, and the physical properties of the brazement. Both the filler metal and the base material compositions can be significantly altered depending upon the extent of liquid and solid solubility and the chemical affinity of the elements present for each other [7,8].

Braze filler metal flow and wetting can be influenced depending on the extent to which the melting points of the filler metal and base material are affected, and new phases are formed. Significant increases and decreases in the volume of the filler, filler de-wetting and joint embrittlement can occur as a result of this alloying process. Under certain circumstances, these alloying effects are not harmful and

can be used to improve the properties of the brazement by increasing the solidus temperature and/ or toughness of the filler metal [9].

The time at the brazing temperature also affects the wetting action, particularly with respect to the distance of creep of the brazing filler metal. If the filler metal has a tendency to creep, The distance generally increases with time. The alloying action between filler metal and parent metal is, of course, a function of both temperature and time. In general, for production work, both temperature and time are kept at a minimum consistent with good quality [9].

### **1.5.3 Surface Preparation**

Oxide layers and surface contaminants can impede the wetting and flow of liquid braze filler metals and must be removed for successful brazing. When faying surfaces of parts to be brazed are prepared by blasting techniques, several factors should be understood and considered. The purpose of blasting parts to be brazed is to remove any oxide film and to roughen the mating surfaces so that capillary attraction of the brazing filler metal is increased. The blasting material must be clean and must be of a type that does not leave on the surfaces to be joined any deposit that restricts filler metal flow or impairs brazing. The particles of the blasting material should be fragmented rather than spherical so that the blasted parts are lightly roughened rather than peened. The operation should be done so that delicate parts are not distorted or otherwise harmed. Vapor blasting and similar wet blasting methods require care because of possible surface contamination. Subsequent cleaning, fluxing and the various base metals deal with the removal of unwanted surface layers. For successful processing, it is important to understand the chemical nature of both the layer being removed and the surface of the base material that will come into contact with the filler metal during brazing. Base material surface finish can be influenced in pre-braze processing and should be considered when designing the brazement and selecting the braze process [7-9].

A clean, oxide- free surface is imperative to ensure uniform quality and sound brazed joints. A sound joint may be obtained more readily if all grease, oil, dirt and oxides have been carefully removed from the base and filler metals before brazing because only then can uniform capillary attraction be obtained [8].

Chemical cleaning is the most effective means of removing all traces of oil or grease. Trichloroethylene trisodium phosphate have been the usual cleaning agents employed. Various types of oxide and scale that cannot be eliminated by these cleaners are removed by other chemical means. Objectionable surface conditions may be removed by mechanical means such as grinding, filing, wire brushing , or any form of machining, provided that joint clearances are not disturbed. In grinding of the surfaces of parts to be brazed, care also should be taken to ensure that the coolant is clean and free from impurities so that the finished surfaces do not have these impurities ground into them [9] .

Mechanical cleaning may be adequate , in which case the design must permit this during manufacture. In some cases where chemical cleaning is required, it may be followed by protective electroplating , necessitating access to the faying surface by the liquids involved [10].

#### **1.5.4 Thermal Expansion**

The coefficient of linear thermal expansion (CTE,  $\alpha$ , or  $\alpha_1$ ) is a material property that is indicative of the extent to which a material expands upon heating. Different substances expand by different amounts. Over small temperature ranges, the thermal expansion of uniform linear objects is proportional to temperature change. Thermal expansion finds useful application in bimetallic strips for the construction of thermometers but can generate detrimental internal stress when a structural part is heated and kept at constant length [11] .

Most solid materials expand upon heating and contract when cooled. The change in length with temperature for a solid material can be ex-pressed as:

$$\left(\frac{L_f - L_0}{L_0}\right) = \alpha_1 (T_f - T_0) \frac{\Delta L}{L_0} = \alpha_1 \Delta T \dots\dots\dots(1.1)$$

$$\frac{\Delta L}{L_0} = \alpha_1 (T_f - T_0) \frac{\Delta L}{L_0} = \alpha_1 \Delta T \dots\dots\dots(1.1)$$

$$\alpha_1 = \frac{1}{L_0} \left(\frac{dL}{dT}\right) \dots\dots\dots(1.2)$$

where  $L_0$  and  $L_f$  represent, respectively, the original and final lengths with the temperature change from  $T_0$  to  $T_f$ . The parameter  $\alpha_1$  is Coefficient of thermal expansion and measured in "per degree Celsius" or "per Kelvin", denoted by  $^{\circ}\text{C}^{-1}$  or  $\text{K}^{-1}$ , respectively. [11,12]

Heating or cooling affects all the dimensions of a body of material, with a resultant change in volume. Volume changes may be determined from:

$$\Delta \frac{V}{V_0} = \alpha_V \Delta T \dots\dots\dots (1.3)$$

where  $\Delta V$  and  $V_0$  are the volume change and original volume, respectively, and  $\alpha_V$  represents the volume coefficient of thermal expansion. In many materials, the value of  $\alpha_V$  is anisotropic; that is, it depends on the crystallographic direction along which it is measured. For materials in which the thermal expansion is isotropic,  $\alpha_V$  is approximately  $3\alpha_1$  [12,13].

### 1.5.5 Capillary Attraction

Capillary attraction makes leak-tight joints a simple proposition for brazing. In a properly designed joint, the molten filler metal is normally drawn completely through the joint areas without any voids or gaps, and brazed joints remain liquid - and gas-tight under heavy pressures, even when the joint is subjected to shock or vibration types of loading. Capillary action results in the phenomenon where surface tension causes molten braze filler metal to be drawn into the area that covers the parallel surfaces that are to be brazed [3].

In actual practice, filler metal flow characteristics are also influenced by dynamic considerations involving viscosity, vapor pressure, gravity and metallurgical reactions between filler metal and base metal [3].

The factors controlling the rate at which wetting , spreading, the capillary flow occur are of great practical and theoretical interest. Studies have indicated profound influences of various kinds of surface activation that cannot be explained in terms of surface energies or alterations in equilibrium contact angle [14,15]. Some of the most important of these effect have been observed in systems in which a finite contact angle is thermodynamically unstable, because the solid vapor surface energy exceeds the sum of the liquid-solid surface energies that is, a system in which thermodynamics would predict complete spreading. Spreading may or may not occur in this type of system, and the rate of spreading can be markedly dependent on surface chemistry, although the fundamental mechanisms of this dependence are not all clear [3,16].

### **1.5.6 Wetting**

If a solid is immersed in a liquid bath and wetting occurs, a thin, continuous layer of liquid adheres to the solid when it is removed from the liquid. Technically speaking, in the wetting process, the force of adhesion between the solid and the liquid is greater than the cohesive force of the liquid. In practical terms, with respect to brazing, wetting implies that the liquid filler metal spreads on the solid base metal instead of balling up on its surface figure 1.5. It has demonstrated that wetting depends on a slight surface alloying of the base metal with the filler metal [17,18].



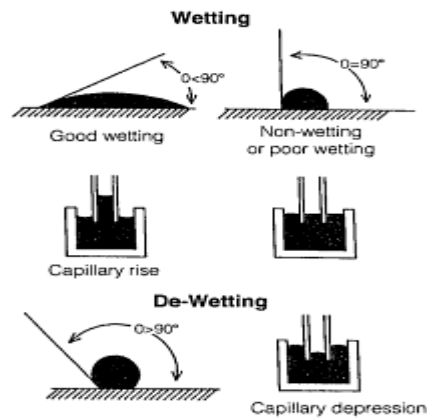


Figure 1.5: Wetting and De-wetting. <sup>[3]</sup>

Wetting is a function not only of the filler metal but also of the nature of the metal or metals to be joined. There is an evidence that in order to we well, a molten metal must be capable of dissolving, or alloying with some of the metal on which it flows.

Wetting is only one important facet of the brazing process. A very important factor effecting wetting is the cleanliness of the surfaces to be wetted. Oxide layers inhibit wetting and spreading as do grease, dirt, oil and other contaminants that prevent good contact between the filler metal and the base metal. One of the functions of the flux is to remove the oxide layer on the joint area and to expose clean base metal [17].

Good wetting and spreading of the liquid filler metal on the base metal are necessary in brazing , because the mechanics of the process demand that the filler metal be brought smoothly, rapidly, and continuously to the joint opening. If the condition within the capillary space of the joint do not promote good wetting, the filler metal is not drawn into the space by capillary attraction. It all boils down to the fact that, for successful joining of components by brazing, the filler metal selected must have a melting point above  $450^\circ\text{C}$  ( $840^\circ\text{F}$ ) and must also wet the base metal without melting it. Then the joint must be designed so that the mating surfaces of the components are parallel and close enough together to cause capillary attraction [18] .

### 1.5.7 Heat Source and Heating Rate

The heating methods available often place constraint on the designer and engineer in selecting the best type in capillary action. In principle there are many methods of heating available for brazing (see table 1.2). Effective capillary joining requires efficient transfer of heat from the heat source into the joint. The heat capillary and thermal conductivity of the assembly must be considered. The size and value of individual assemblies, the numbers required, and the rate of production necessary will influence selection of heating method. many other factors must be considered before the choice is made [3,18]. The rate of heating, differential thermal gradients, and the cooling rates, both external and internal, will vary tremendously with different methods of heating and the effects of these on dimensional stability, distortion and metallurgical structure must be considered. Getting heat to the joint can be accomplished by many ways and is best categorized by the actual method of heating [3] .

Table 1.2 Characteristic of various heating methods.

Method	Characteristics (a)					
	Capital cost	Running cost	basic output	flux requirid	versatility	operating skin required
Torch ( flame)	L/M	M/H	L	H	H	Yes
Electrical resistance	M	M	M/H	L	L	No
Induction	M/H	M	M/H	M	M	No
Furnace (atomsphere)	M/h	M/H	H	M	M	No
Furnace (vacuum)	H	L	H	M	M	No
Dip (flux bath)	L/M	M/H	L/M	L	L	Yes
Infrared	M	L	M	L	L	No
(a) H : high , M : medium L : low						

## 1.6 Advantages, Limitation and applications of Brazing

### 1.6.1 Advantages of Brazing

1. Cast and wrought metals can be joined.
2. Metallurgical properties of the base materials are not seriously disturbed.
3. Assemblies can be brazed in a stress condition.

4. Dissimilar metals can be joined.
5. Non-metals can be joined to metals, when the nonmetals is coated.
6. Materials of different thickness can be joined easily.
7. Complex assemblies can be brazed in several steps by using filler metals with progressively lower melting temperature.
8. Little or no finishing is required by the braze joint.
9. Several operation can be mechanized.
10. Almost all the common engineering materials may be satisfactorily brazed or braze welded [18,19].

### **1.6.2 Limitations of Brazing**

1. High degree of skill required.
2. Limited size of parts.
3. Machining of the joint edges forgetting the desired fit is costly.
4. Joint design is somewhat limited if strength is a factor [18].

### **1.6.3 Applications of Brazing**

The applications of brazing include the following:

1. Parts of bicycle such as frames and rims.
2. Pipe joints subjected to vibrations.
3. Band saws
4. Tipped tools.
5. Nipples and unions to M.S and copper tubing [19].

## **1.7 Filler Metal Forms**

Brazing filler metals are available as rods, ribbons, powders, pastes, creams, wires, sheets and performs (stamped shapes, washers, rings or shaped wires) shaped to fit a particular part. Depending on the joint design, heating method , and level of automation, the filler metal can be preplaced before the heating cycle starts

or face-fed after the work is heated .high production brazing . such as furnace or induction brazing, which typically involves a high level of automotive,usually required preplacement of the filler metal [20].

Rod and wire forms are usually used for manual face-feeding. Many special shapes and forms designed for specific application are utilized as preplaced or preformed filler metals. Such pre-placing of the filler metal ensures that there is a uniform amount of filler metal in the correct position on each assembly. Preplaced filler metal may be useful with manual brazing but is usually a fundamental part of any mechanized brazing procedure. Where joint areas are large, filler metal may be located between the lying surfaces. Brazing rings are sometimes inserted into grooves machined into work. Totally enclosed rings in such grooves may be necessary for long sleeve joint or for salt bath dip brazing , where it is desirable to avoid melting of the ring before the work is heated to the brazing temperature [20]. Competitive forces have pressured metal working manufactures to reevaluate their manufacturing processes. As a result, demands on brazing operations have increased to improve efficiency, produce higher quality products, and reduce costs. There are a number of factors that can affect the performance of the brazing operations. Of particular importance is the proper selection of the correct form of filler metal. There are three general categories of filler metals in production applications, they are [19,20]:

1. Non-fabricated wire and strip. Available on spools that contain a specified amount of filler metal at standard size.
2. Fabricated wire and strip forms. commonly known as performs, this filler metal can be fabricated as rings, washers, disc, shims or other engineered shapes.
3. Paste filler metal. A combination of atomized filler metal powders.

## 1.8 Properties of Materials

Some physical properties in which have important parameters affecting on the reliable joint between any dissimilar pieces are listed in table 1.3 The matching between the thermal expansion coefficient of pieces (stainless steel and graphite) are clearly noticed and its within 10% differences.

Table 1.3: The properties of materials

<b>Materials</b>	<b>Thermal expansion (<math>10^{-6}</math> m/mk)</b>	<b>Melting point °C</b>	<b>Density g/cm<sup>3</sup></b>
Stainless steel 410 [21,22]	9.9	1480 – 1530 °C	7.74
Graphite pure (carbon) [21,22]	2-6	3730 °C	2.09–2.23
Silver [25]	19.5	961.78 °C	10.49
Titanium [24]	8.6	1668 °C	4.506
Copper [25]	16.6	1084.62 °C	8.96

## 1.9 Types of Bonding

In the solid state, atoms are closely surrounded by neighboring atoms. The forces that bind atoms together are electrical in nature. Their type and intensity are largely responsible for the physical and chemical properties of crystals. These electrical forces between atoms are referred to as bonds. Depending on the electronic structure of an atom and its nearest neighbor, the types of bonding forces can vary. In general, in the solid state at low temperature, Where thermal vibration is only moderate, four types of bonding are distinguished. In metallic bonding some outer electrons have been removed from the atoms and move freely within the structure [26]. The attractive force between the positively charged atoms and

the negatively charged electrons cloud holds such structures together. The van der waals bond is a weak overall attraction between neutral atoms; it arises because the electro distribution in atoms is not uniform. ionic bonding relies on electrostatic attraction between atoms of different charge , where electrons have been removed (positive charge) or added (negative charge). In covalent bonding, single electrons are shared between two atoms in a common orbital. In reality all bonding forces are active in a crystal, but in different minerals some forces can dominate. For example, in halite (NaCl) bonding is largely ionic, whereas in diamond (C) it is covalent. Bonding can be mixed, and different types of bonding may exist between different atoms in a minerals structure. For example, the important Si-O bond involves covalent and ionic forces. In case of sulfur, some S-S bonds are largely covalent, whereas others are of the van der waals type. A very schematic triangular representation with examples of mineral species for different types of bonding (excluding van der waals bonding ) is shown in figure 1.6 [25,26].

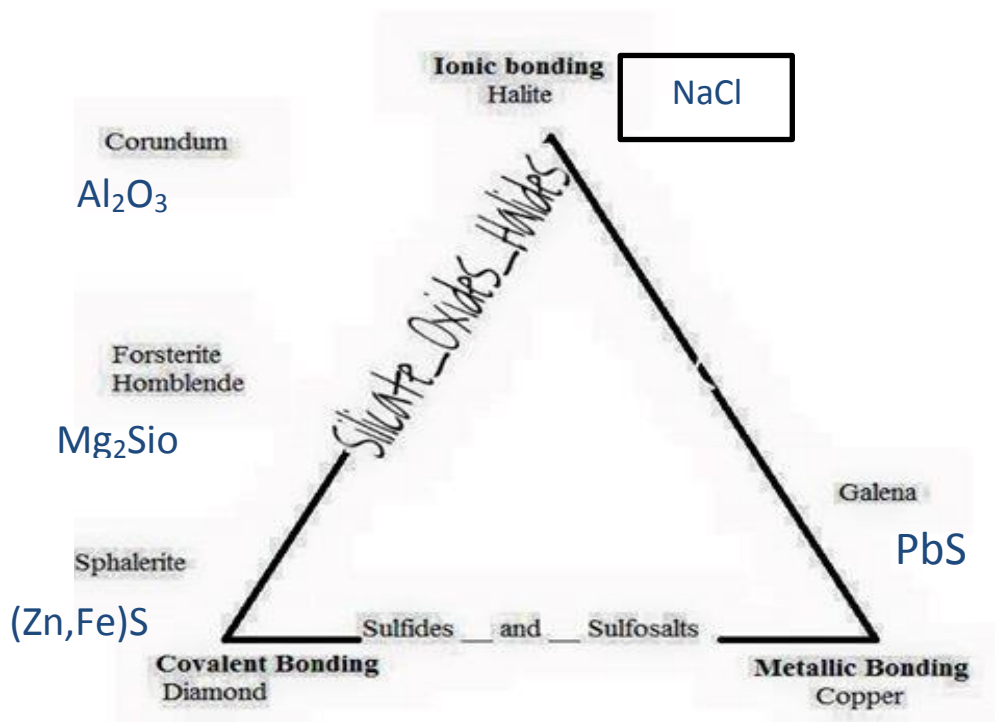


Figure 1.6 Triangular representation of ionic, covalent, and metallic bonding with some mineral representative [26].

### 1.9.1 Van der Waals Bond

The simplest examples of van der Waals bonding are the inert gas solids. The spherically symmetric filled – shell electronic configurations of isolated inert gas atoms are very stable and little affected when the atoms come together to form the solid. The interaction energy of two inert gas atoms depends only on distance between them and is represented by the well known interatomic potential curve of figure 1.7 the force between the atom is the negative of the slope of this curve. The attractive force at large separation arises because even a spherically symmetric atom has a fluctuating electric dipole moment in the other atom and the two dipoles than attract each other. The attractive force is known as the van der waals or London force. The repulsive force at small separations is associated with the overlap of the outer electron shells on the two atoms. An important contribution to the repulsion arises because of the Pauli principle, which prevents two electrons from occupying the same quantum state; overlap of the filler shells means that electrons must be promoted to higher atomic energy levels in order to comply with this requirement. The repulsive force increases very rapidly with increasing overlap, and this explains why inert gas atoms behave like attracting hard spheres and form close structures [27] .

$$\sqrt{r} = 4 \varepsilon \left[ \left(\frac{\sigma}{r}\right)^{12} - \left(\frac{\sigma}{r}\right)^6 \right] \dots\dots\dots(1.4)$$

Where  $\varepsilon$  Depth of potential well

$\sigma$  Distance at which the inert- particle potential reaches zero.

$\left(\frac{\sigma}{r}\right)^{12}$  Repulsive term of short range forces.

$\left(\frac{\sigma}{r}\right)^6$  attractive term of long range forces

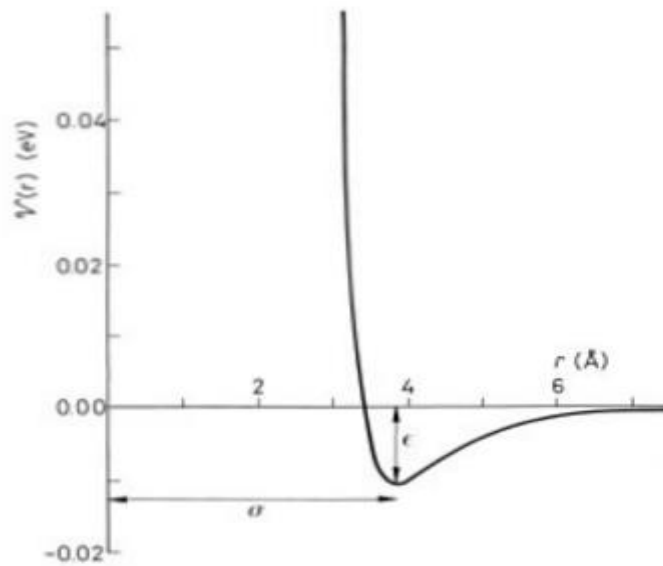


Figure 1.7 Lennard-jones potential interaction of two argon atoms. The parameters  $\epsilon$  and  $\sigma$  which specify the potential were obtained from measurements on gaseous argon [27].

The binding energies, interatomic forces and related properties of the inert-gas crystals can be calculated with reasonable accuracy by assuming that the interacting of any two atoms within the crystal is given by an interatomic potential of the form of fig 1.7 [26,27].

### 1.9.2 Ionic Bond

A similar situation to that in the inert gas solids arises in ionic solids such as sodium chloride . Transfer of an electron from a sodium atom to a chlorine atom produces  $\text{Na}^+$  and  $\text{Cl}^-$  ions with the stable electronic configurations of the inert gases neon and argon respectively. The electron states are little affected by the coming together of the ions to form the solid and the interaction of any two ions within the solid can be represented by the interionic potential curve of two isolated ions [27] .

Properties of ionic solids:

1. Ionic crystals are rigid
2. Ionic crystals have high melting and boiling points
3. Ionic crystals are insulators because of non availability of free electrons.



4. Ionic solids are easily soluble in polar solvent like water
5. Ionic compounds are soluble in non-polar solvent like benzene.

### 1.9.3 Covalent Bond

The covalent bond is usually formed from two electrons one from each atom participating in the bond. The electrons forming the bond tend to be partly localized in the region between the two atoms joined by the bond. The spins of the two electrons in the bond are antiparallel as Pauli principle. The covalent bond has strong directional properties. Covalent bonds result when atoms share electrons.

Properties of covalent solids:

1. Covalent compounds are mostly gases and liquids
2. Covalent crystals tend to be hard and brittle
3. The melting point and boiling point are usually low as compared to those of ionic crystals.
4. Most of covalent substances do not conduct electricity because of non-availability of free electrons or charged ions to carry the current
5. Covalent substances are insoluble in polar solvent like water.

### 1.9.4 Metallic Bond

Metal atoms have fewer than the four valence electrons required to form a three-dimensional covalently bonded structure. One way of looking at metallic bonding is as a type of covalent bonding in which some of the bonds are missing [27]. The valence electrons, which hold the atoms together, are not bound to individual atoms or pairs of atoms but move freely throughout the whole metal. Atoms of their same elements or different elements give their valence electrons to form an electron cloud or say electron gas throughout the space occupied by the atoms. Thus atoms in metallic crystal work as ions in space.

Properties of metallic solids:

1. Bonding energies and melting point temperature for some metals have a wide range.

2. Bonding may be weaker or stronger
3. Due to the symmetrical arrangement of the positive ions in a space lattice, metals are crystalline.
4. Metallic bond being weak, metals have a melting points moderate to high
5. Metallic crystals have high electrical conductivity.

### 1.10 Literature Servay

1. Crispin et al. (1976). investigated the effect of concentration on the wetting and bonding behavior between nickel alloys and AL23 alumina or sapphire substrates. The addition of 1 wt% chromium or 0.1 wt% yttrium produces a decrease in contact angles at 1570° C but not sufficient to cause wetting. Additions of up to 1 or 2 wt% chromium or 0.02 or 0.08 wt% yttrium improves the bonding between nickel and AL23 or sapphire respectively [29].

2. Mizuhara et al. (1985). conducted the brazing test by using an alumina substrate. A brazing filler metal perform was placed on top of the alumina, and 3 strips of Kovar were placed on top. Additional tests were carried out with silicon nitride brazed to copper [30].

3. Naka et al. (1987). used the sessile drop technique to evaluate the equilibrium contact angle and work of adhesion of molten Al-Cu alloy on Si<sub>3</sub>N<sub>4</sub> [31].

4. Hammond et al. (1988). demonstrated titanium vapor deposite on the ceramic surface and using Ag-Cu-N filler metal to produce ceramic-metal brazement at low temperature. The composition and structure of the ceramic-metal bond zone in the zirconia- to-nodular iron joints were analyzed by electron probe microanalysis and x-ray diffraction [32].

5. Mcdonald et al. (1989). have conducte the wettability tests for nineteen brazing filler metal on molybdenum and thirty two brazing filler metal on the alloy 0.5 Ti-0.08Zr-Mo(TZM) over a wide range of temperatures. By using wettability

coupons, 3.17 X 50.8 X50.8mm specimen were fabricated from commercially pure molybdenum and TZM plate stock. A 1.58-mm diameter by 12.2-mm deep thermocouple hole was drilled at the edge of each specimen. coupons were prepared with filler metals and placed in a standard, cold wall vacuum furnace. The furnace was evacuated to  $5 * 10^{-5}$  torr [33].

6. Moret et al. (1993). used direct brazing to join the ceramic to metal. The production of important components like first walls of thermonuclear power reactors advanced automotive or aeronautic turbines and heat exchangers depends on the control of ceramic to metal joining techniques especially direct brazing [34].

7. Ohmura et al. (1994). used a new brazing technique for joining graphite to itself or to metals such as W, Mo, Cu with conventional brazing filler metals has been developed. Essentially, it is impossible to braze graphite with Cu filler metal because no wetting occurs . The shear strength of a graphite/ graphite joint with a 0.12-mm (0.0047- in.) thick Fe foil at room temperature was about 32 MPa (4.6 ksi). Further, the bending strength of the graphite/graphite and graphite /Cu joints at 873 K (1112°F), as measured using the four-point bending test, was 35 and 11 MPa (5.1 and 1.6 ksi), respectively [35].

8. Lee et al. (1996). studied the effects of various active elements such as ( Titanium , vanadium , niobium and chromium on the wetting, microstructure and bond strength. In wetting, the brazed joints of Cu-2 at % Nb alloys show a high bend strength of 154 Mpa, although the wetting angle is a little higher, at about 60° [36].

9. Pintschovius et al. 1999. plates of cemented carbide were joined to steel by brazing. The structure was designed in limitation of a rock drill. The residual stresses resulting from the different thermal shrinkage and elastic–plastic behavior of the materials as well as from the different phase transformation behavior of the steels during cooling of the samples from the brazing temperature (1100°C) to room temperature were investigated by neutron diffraction with special attention to

the regions near the interface between steel and cemented carbide. Additional measurements were performed by X-ray diffraction on selected surface areas [37].

10. Jarvis et al. (2002) demonstrated markedly enhanced adhesion between oxide ceramics and conducting metals via promoting open-shell electronic structure at the interface, by using a more covalent oxide or doping with early transition metals [38].

11. Augusto et al. (2003). studied the electrochemical degradation of the interface formed between commercially pure Ti and Al<sub>2</sub>O<sub>3</sub> produced by diffusion bonding, in contact with a physiological solution. Chemical composition and morphology of samples and corrosion products were evaluated by SEM and EDS analysis [39].

12. Ricardo et al. (2005). prepared thirty cylinder specimens were cast from alloys of nickel-chromium bonded with three system of ceramics to evaluate the shear bond of ceramic to metal porcelain build up and shear strength testing were accomplished using a stainless steel mold. The Noritake ceramics system used together with Viron alloys presented the highest strength to shear forces, while Duce ram bonded to Vera bond presented the lowest bond strength [40].

13. Morscher et al. (2006). Joining titanium (Ti) and carbon- carbon (C/C) composites can be quite challenging due to thermo-elastic mismatch. Two types of joined structures were analyzed; Ti tubes joined to flat pieces of C/C and flat pieces of Ti joined to flat pieces of C/C. The former was used to determine the tensile load carrying ability of the joints for a curved surface. [41]

14. Fadhel A. Hashim et al. (2007). used four types filler metal alloys which these are (CuZnNi),(CuZnSn),(CuZnAgCd ) and pure copper for brazing plates of stainless steel L304 (thickness:10mm, 1mm) and plates of pure titanium(99.9%)(thickness 0.15mm). All these joints showed good joining results with Cu-base filler metals, but (CuZnNi) gave high joint because that filler create

an intermetallic compounds between Ta and the filler at the joint interface(NiTa) [42].

15. Salazar et al. (2007). "evaluated the shear bond strength of Co - Cr and Ni - Cr metal alloys and specific ceramic", submitted to different thermocycling immersion times. Sixty metal-ceramic samples were confectioned and standardized in cylindrical format. three thermocycling conditions were evaluated: without thermocycling, 3,000 cycles (5 degrees C/55 degrees C+/-1) with 30s of immersion time and 3,000 cycles (5 degrees C/55 degrees C+/-1) with 60s. The shear bond strength was preformed by using a special device to concentrate the tension at the metal/ceramic interface during the test. It was concluded that the thermocycling with of one minute immersion time influenced shear bond strength values for the Ni-Cr/porcelain and Cr-Co/porcelain systems [43].

16. Muna K . Abbas and Hassien M . Habeb. (2008). Studied the factors effecting the brazing of stainless steel (AISI 316L) with carbon low steel (CK15)by using three type of copper base filler metal: (DINL-Ag20Cd),(AWS RB Cu Zn-C)and (AWS RB Cu Zn-D)alloys in furnaces with inert gas atmosphere. The samples were inserted in special container called (Retort) filled by inert gas(argon) during brazing process with flow rate (1-2 litter / min). brazed joints by filler metal alloy (RBCu Zn-D) showed the best shear strength for stainless steel (316L)and low carbon steel at 5,10 min [44].

17. Bahaa S.Mahdi. (2008). investigate the brazing by using electrical arc as a heating source to join stainless steel type( 304L) to itself. The brazing process was accomplished by using filler metal composition alloy (Ag-Cu-Zn-Cd). After finishing brazing there were three destructive testings (tensile, micro-hardness and micro examination) to find the mechanical and phase transformation which happened during operation. The diffusion between the base metal and filler metal very little compare with brazing by using furnaces the main reason for this was time factor [45].

18. Fadhel A .Hashim et al. (2008). conducted more than 15 brazed joint are tested for graphite butt joint by using active filler metal alloy, which contained copper and titanium as active elements. The filler metal was used as a paste with percentage of titanium ranging from 2%wt to 50%wt. Bonding is observed by using optical microscopic for bonding region and by seeking for bonding phases at the metal/ filler interface by using XRD technique. The joints which brazed with titanium content less than 20Wt% are not strong , while that brazed by filler metal alloy of 40% content of titanium give shear stress of 14Mpa [46].

19. Ozcan M et al. (2009). estimated the effect of thermal and mechanical cycling on the shear bond strength of three fusing glassy matrix dental ceramics to commercial titanium (cpTi) when compared to conventional feldspathic ceramic fused to gold alloy. After thermal-cycling for 6000 times and mechanical-cycling for 20,000 times, Triceram-cpTi combination presented the least decrease among other ceramic-alloy combinations when compared to the mean bond strength results with Au-Pd alloy-Vita Omega 900 combination [47].

20. Adnan S. Jabur. (2009) prepared lamellar filler of (copper-low carbon steel-copper)foils as new technique for brazing of graphite with its self or with metal to overcome the poor wettability of graphite. Material used in this investigation were graphite as a substrate ,copper foil(50 $\mu$ m)and carbon low steel(200 $\mu$ m) as filler metal. after preparation of joint materials and insert in retort, it was sealed and the argon was pumped at (330 $^{\circ}$ C) until reaching the temperature(1125 $^{\circ}$ C).The best brazing time was 15min at 1125 $^{\circ}$ C brazing temperature, which gave a maximum shear strength [48].

21. Ahmed O. Jasim et al. (2009). studied the effect of overlap joint and joint thickness on shear strength of epoxy adhesive and torch brazing for joining gray cast iron to carbon steel. the brazing filler metal was L-Ag55Sn as per Din 8513, AWS/ASME SFA-5.8 B-Ag7. The liquids and solidus temperature of the BAg7 filler metal 652 $^{\circ}$ C and 618 $^{\circ}$ C respectively. brazing with ductile filler metal (Ag-base

alloy) and epoxy adhesive are more reliability to join carbon steel / gray cast iron with high shear strength and more uniform stresses with less defects [49].

22. Fadhel A. Hashim. (2009). discussed the possibility for analysis of bonding phase of joins in brazing by using X-ray diffraction testing for several planes in the bonding region .graphite / graphite active brazing process by using active filler metal (copper base alloy) containing 25w%of titanium in copper base. By comparing between three XRD chart, he conclude bonding phase which is titanium carbide (TiC) [50].

23. N.V. Moutis et al. (2009). A graphite sheet has been successfully brazed to a Nimonic 105 superalloy using a commercial TiCuSil paste. A chromium layer was deposited on the graphite surface by sputtering and controlled heat treatments were employed in order to develop a suitable microstructure. Scanning electron microscopy measurements showed rough, crack-free interfaces between the filler metal and both the graphite and nimonic parts. From metallographic examination a well defined layered structure of the metallic elements close to the filler/graphite interface has been found. The metallic elements transport from the interface to the carbon bulk where they fill all the graphite pores up to a depth up of 50  $\mu\text{m}$  and form a layered structure within the pores [51].

24. Stella Crosara et al. (2009). evaluated the metal-ceramic bond strength (MCBS) of 6 metal-ceramic pairs (2 Ni-Cr alloys and 1 Pd-Ag alloy with 2 dental ceramics) and correlate the MCBS values with the differences between the coefficients of linear thermal expansion (CTEs) of the metals and ceramics [52].

25. Moutis et al. (2010) produced c/c ceramic composites have been brazed to nimonic alloy using a TiCuSil filler metal. The C/C surface was metalized through the deposition of a chromium layer [53].

26. Fadhel A. Hashim and Q.J Al-Hussniyoy. (2011). studied the possibility of joining copper(ASTM, C1020100) to low carbon steel (ASTM A36) using the



shielded metal arc welding (SMAW) and the gas tungsten arc welding process by eight type of electrodes; ERNiCrFe-5, ERNiCu-7, ECu, ECuSn-A, E309L, E 312, E7018, and ENi-1. Three types of solidification were recognized; the first is epitaxial from the copper side, the second is nonepitaxial from the steel side, and the third type is competitive which shown in the two sides. Solidification cracks on the steel side are shown, and these cracks are filled by brazing process. All tested specimens in tension and bending show the copper properties [54].

27. Thamir Abdul- jabber. ( 2012). studied effect of furnace vacuum pressure on the joining strength of alumina bonded kovar by prepared the active filler alloy consist of purity powdered metal of silver, copper and additives of titanium .Active filler alloy was in between kovar strip and alumina as a sandwich form. A fixture was used to hold the specimen, vacuum furnace operated under two conditions  $2 * 10^{-4}$  torr and  $2 * 10^{-6}$  torr respectively. A strong joint between kovar alloy and alumina was detected by using shear test technique. A maximum value of joint strength between alumina and kovar was obtained (130Mpa)under condition of  $2 * 10^{-6}$ torr of vacuum furnace [55].

28. Mohd Afendi et al. (2012). studied the effect of bond thickness upon the shear strength of epoxy adhesively bonded joint with dissimilar adherends is addressed. the bond thickness is controlled to be ranged. As a result, shear strength of adhesive joint reduces with increasing bond thickness. The strength of shear adhesive joint is also dependent on [56].

29. Abdulla A. Mushari et al. (2012). studied the microstructure of the bonding zone between the low carbon steel (ASTM A36) and the pure copper (OFHC) by two welding process, brazing and soldering. the microscopic examinations showed that there is a clear bonding phase and interface in brazed joints, while welding by soldering have no such features[57].



30. Ray et al. (2013). attempted to improve the mechanical strength of graphite to stainless steel brazed joint. Active filler alloy Ag-Cu-Ti was used to brazed the substrates. shear strength of approximately 35 Mpa for graphite-304SS brazed joint produced at 900 °C was demonstrated [58].

31. Omer Saad Salih. (2014). used furnace brazing to produce joints between two different material, oxygen free high conductivity copper OFHC type ISO C10300 and silicon steel type ASTM 36F145 by using two group of filler metals: Silver group type DIN L-AG20Cd and copper group type AWS BCuP-1 with flux type AWS FB3-A to remove oxide and protect the joint. The brazing temperature for both filler was 790°C and 940°C respectively then soaking at different time for each filler metal. The maximum double shear strength of dissimilar brazed joints was 47MPa by using filler metal AWS BCuP-1 at 30min. soaking time, while using filler metal DIN L-Ag20Cd give rise in double shear strength to 98MPa at 30min [59].

32. Zhang et al. (2014). investigated the effect of brazing temperature on microstructure and mechanical properties of graphite / copper joints by using Ni-Cr- P- Cu filler alloy at 1173\_ 1253 K for 10 min. The maximum shear strength of the joint could reach 60 Mpa when the brazing temperature was 1223 K [60].

He Wang et al. (2015). used Ag as a beneficial alloy element in no matter solders or brazing filler metals. Obviously, the addition of Ag has a positive function on melting temperature , Wettability, mechanical property and conductivity of filler metals [61]

33. Chen et al. (2015) studied the microstructure of the AlN/AlN joints. An Au - Pd Co - Ni -V brazing alloy was designed for AlN ceramic joining. Its wettability on AlN was studied with the sessile drop method. The results showed that the contact angle was decreased gradually with increasing temperature and the prolonging of holding time. Sound AlN/AlN joints were achieved with the brazing alloy at 1170° C for 10 min. The AlN/AlN joints brazed with the AuPdCoNiV brazing alloy

exhibited three-point bend strength of 162.7 MPa at room temperature, and under the bend test the fracture of the joint occurred at the AlN ceramic substrate [62].

34. Wei-yuan Yu et al. (2015). The microstructures and formation mechanism of the interface between pure aluminum and graphite were studied when performed the active brazing in atmospheric conditions using assistive ultrasonic vibration. The application of ultrasonic vibration can remove the surface oxidation films of the base metals and molten solders; hence, ultrasonic vibration can enhance the wettability of liquid solder on the surface of the base metals and accelerate the element diffusion rates of interfacial reactions [63].

35. Tian et al. (2015). Ag–Cu–Ti/Cu mesh/Ag–Cu–Ti composite foils were designed to braze ZrB<sub>2</sub>–SiC–C (ZSC) ceramic and GH99 super alloy, and sound brazed joints were obtained. The interfacial microstructure and mechanical properties of the brazed joints were investigated. By comparing joints brazed with Ag–Cu–Ti filler alloy and Ag–Cu–Ti/Cu foil/Ag–Cu–Ti composite foils, the mechanism by which Ag–Cu–Ti/Cu mesh/Ag–Cu–Ti composite foils improves the shear strength of the joint was assessed. Using Ag–Cu–Ti/Cu mesh/Ag–Cu–Ti composite foils, the maximum shear strength of the brazed joints increased to 121 MPa, which is 560% of the joints brazed with Ag–Cu–Ti filler alloy (21 MPa) [64].

36. Wang and Song-bai Xue. (2016). studied the effect of Ag on the properties of solders and brazing filler metals by using three kinds of typical solders (Sn–Ag–Cu, Sn–Zn and Sn–Bi) and brazing filler metals (Ag–Cu–Zn, Cu–P, and Zn–Al) have been chosen for illustration. This article summarizes research status on the studying of Ag-contained solders and brazing filler metals, also analyses influence rules of Ag addition on the change of filler metals' physical property, microstructure as well as mechanical property [65].

37. Yangwu Mao et al. (2016). Brazing of graphite to Cu has been successfully realized with Cu50TiH2 (Containing 50Wt% Cu) based fillers including Cu50TiH2 filler and Cu50TiH2 + C composite filler in vacuum at 1223 K . The average strength is 10.8 Mpa for the joints brazed with Cu50TiH2 filler [66].

### **1.11 Aim of the Work**

The objective of the work described in this research is to extend the range of available information about the ability for joining dissimilar materials by using active filler paste. The purpose can be achieved by the followed steps:

- 1- Examining the wetting behavior of different active filler paste on both stainless steel and graphite.
- 2- Determining the better condition for obtaining a reliable bonding strength.
- 3- Studying the fractured surfaces for determine the factors affecting on the obtaining reliable joint .
- 4- Examining the filler paste efficient comparing with other active filler.

# CHAPTER TWO

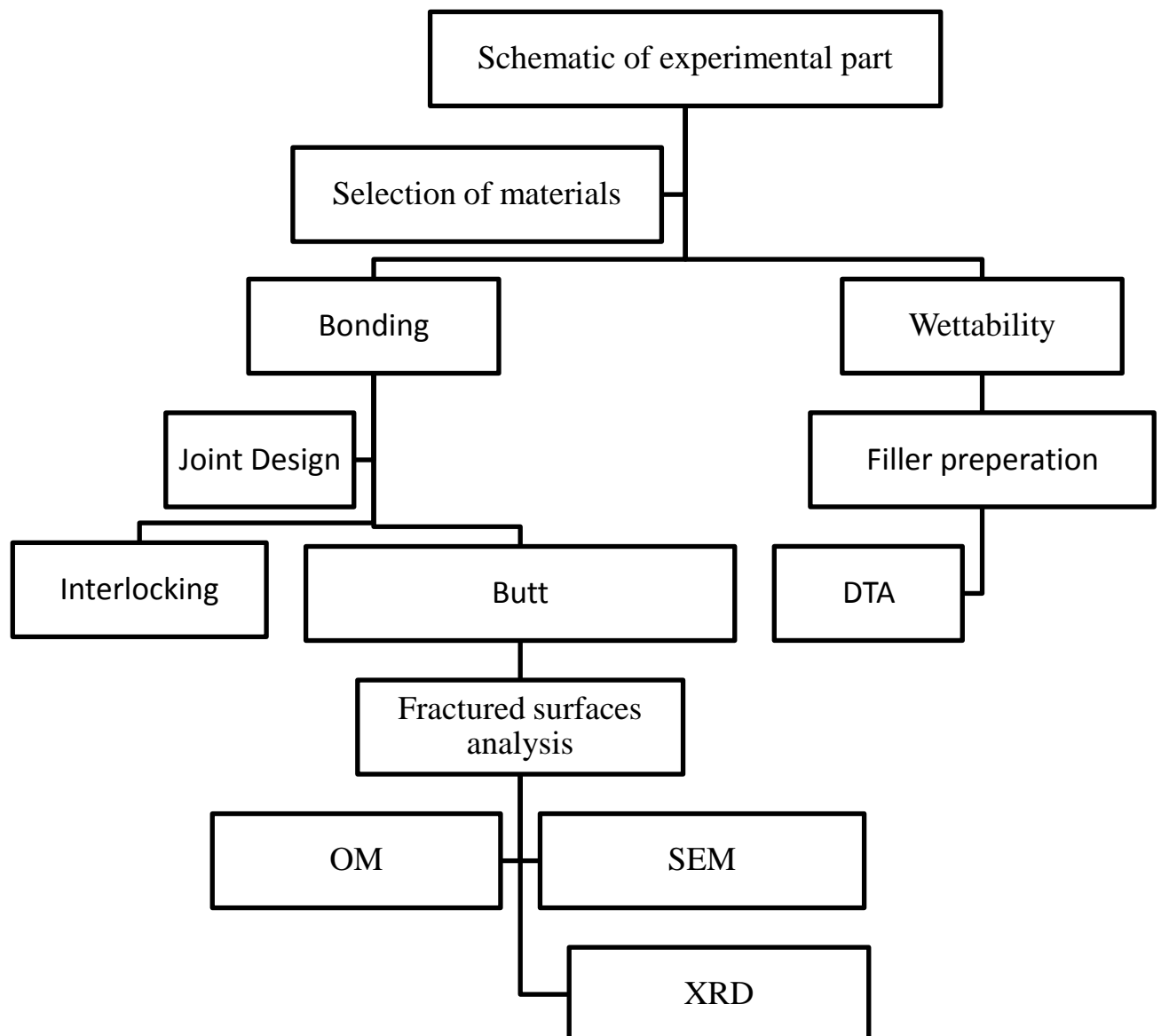
EXPERIMENTAL PART

## Chapter Two

### Experimental Part

#### 2.1 Introduction

This part of project includes the materials , sample preparation, decontamination, tests apparatus and test techniques in which were used along achievement of this project. System setting and operation also were mentioned in this chapter. The experimental program of the current study is sketched in the flow chart as below :



## 2.2 Selection of Materials

### 2.2.1 Stainless Steel

The stainless steel rod of grade 410 disk of 25 mm diameter and 5mm thickness were cut from a rod of the same material as shown in figure 2.1. The chemical composition of Stainless steel is shown in table 2.1



Figure2.1: Stainless steel 410 specimen

Table 2.1: The chemical composition of the stainless steel 410.

<b>Elemental composition</b>	<b>Weight percentage (%) .</b>
C	$\leq 0.15 \%$
Cr	11.5 – 13.5
Mn	$\leq 1 \%$
Ni	$\leq 0.5 \%$
P	$\leq 0.04 \%$
S	$\leq 0.03 \%$
Si	$\leq 1 \%$

### 2.2.2 Graphite

Graphite was cutting into cubic form with dimensions of  $(10 \times 10 \times 10)\text{mm}^3$  as shown in figure 2.2. Hard shaft of graphite china made, was prepared to the suitable form for studying wetting and bonding.

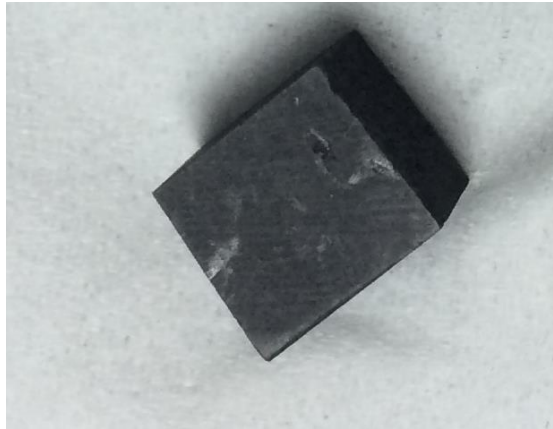


Figure 2.2: The cubic graphite sample.

### 2.2.3 Active Filler Metal

High Purity powdered metals of copper (Cu), titanium (Ti) and silver (Ag) (Fluka company with 99.98% purity) were used to prepared the mixture active filler of paste with certain percentage of powdered are set out below in table 2.2 and table 2.3. All sets of mixture were for examined both wetting and bonding

Table 2.2: The active filler paste with different concentrations(Ti-Ag).

<b>Titanium(Ti) of wt.%</b>	<b>Weight (gm)</b>	<b>Silver (Ag) of wt.%</b>	<b>Weight (gm)</b>	<b>Filler code</b>	<b>Total weight (gm)</b>
100	0.25	/	0	F1	0.25
75	0.18	25	0.07	F2	0.25
60	0.15	40	0.1	F3	0.25

Table 2.3 The active filler paste with different concentrations(Cu-Ag)

<b>Copper (Cu) wt.%</b>	<b>Weight (gm)</b>	<b>Silver (Ag) wt.%</b>	<b>Weight (gm)</b>	<b>Filler code</b>	<b>Total weight (gm)</b>
60	0.15	40	0.1	F4	0.25
50	0.125	50	0.125	F5	0.25
65	0.162	35	0.087	F6	0.25
55	0.137	45	0.113	F7	0.25

Experimental procedure for preparation the filler paste regarding the following steps

- 1- weighted each element by four digital balance (Stanton 462AL made in UK.
- 2- mixed the metal powders into mold by hand for half hour then added some glycerin to make the mixture as a paste form.

Table 2.4 (a and b) the number and dimensions of specimens of stainless steel and graphite for wetting and bonding.

a. Purpose of wetting

<b>Set No.</b>	<b>Sample code</b>	<b>Set No.</b>	<b>Sample Code</b>
2	St.St / F1 at 1200° C	2	of Gr /F4 at 850° C
2	Gr / F1 at 1200° C	2	of St.St/F6 at 850° C
2	St.St / F2 at 920° C	2	of Gr /F6 at 850° C
2	Gr / F2 at 920° C	2	of St.St/F7 at 850° C
2	St.St/F4 at 850° C	2	of Gr /F7 at 850° C



## b. Purpose of bonding

Set no.	Sample code	Set No.	Sample code
2	St.St/F1/Gr at 1000° C	2	St.St/F2/Gr at 900° C
2	S.S/F1/Gr at 950° C	2	St.St/F2/Gr at 850° C
2	St.St/F1/Gr at 900° C	2	St.St/F3/Gr at 950° C
2	St.St/F1/Gr at 850° C	2	G/F5/Gr at 920° C
2	St.St/F2/Gr at 950° C		

- The dimensions of samples designed for interlocking shape as following :  
Graphite cubic was tapered with diameter 5mm and Stainless steel cubic was drilled with diameter 6mm

### 2.3 Preparation of Samples

Stainless steel samples were cutting into shape of dimension  $(10 \times 10 \times 4)$  mm<sup>3</sup> by used suitable cutter machine. The grinding process was performed by used metallographic paper of grade (800,1000,1200) and then polishing proces (by used wet cloth) processes were achieved on a stainless steel specimens to obtain a proper surfaces for wetting and bondin processes. Graphite samples were cutting into cubic form of dimension  $(10 \times 10 \times 10)$  mm<sup>3</sup>. Graphite surfaces were conducted by blasting using sand stone of Sic for enhance the interlocking bonds.

### 2.4 Decontamination

Decontamination was carried out by immersing the St.St in thinner for 15 minutes and followed immersing in acetone for 10 minutes to remove any hydrocarbon material such as grease, oil ... etc, then washing in distilled water and finally kept the stainless steel samples in the desiccators until the time will use [66].

To remove any contaminants materials from the surface of graphite, an atmospheric furnace setting on the temperature of 450°C was performed, then the adherent contaminants pieces were burned and fly out.

## 2.5 Vacuum furnace Set Up

The Vacuum furnace of pressure  $10^{-2}$  torr was used to investigate this work. Vacuum furnace type GSL-1700X-S-UL-60 is TUV standard compact tube furnace, made in Germany, the current location of this furnace

Al-Nahrain university, collage of sciences.

The programming of this furnace is listed below:

1. press and hold button ▲ for 3 second, the word **stop** will appear , and then the program will end
2. For a new program press the button ◀, the word **CO1** will appear, it represent the initial ( primary ) temperature ( $0^{\circ}\text{C}$ )
3. press the button ← once, the word **TO1** will appear and it represent the time required to reach the first step.
4. press the button ← once again, the word **CO2** will appear, it represent the temperature ( $^{\circ}\text{C}$ ), that you wish to reach in the first step **TO1**.
5. press the button ← , the word **TO2** will appear and it represent the time required to reach the second step.
6. press the button ← , the word **CO3** , it represent the temperature ( $^{\circ}\text{C}$ ) that you wish to reach in the first step **TO2**.
7. press the button ← , the word **TO3** will appear and it represent the time required to reach the second step.
8. press the button ← , the word **CO4**, it represent the temperature ( $^{\circ}\text{C}$ ) that you wish to reach in the third step **TO**.
9. Repeating the same process sequent
10. press and hold the button ▼ the word **RUN** will appear in the screen.



Figure 2.3: Vacuum tube furnace

### 2.5.1 Operation Mechanism

For examination the wetting and brazing behavior, vacuum tube furnace of vacuum pressure  $10^{-2}$  torr was used as shown in figure 2.3. Many steps should be done before starting and operation as following :

The first step was putting the specimen under study into tube furnace. The second step was pumping the nitrogen gas of purity ( 99%) into a tube furnace through a regular gauge to determine the flow rate of nitrogen at 0.5 L/ min for two minutes. The purpose this step is to release the remaining oxygen from the tube furnace. The third step was starting heating according to the convenient temperature – time schedule shown in figure 2.4. The third step involved vacuum operation until reach up to  $10^{-2}$  torr. This operation sequences was done along each experiments of wetting and brazing.

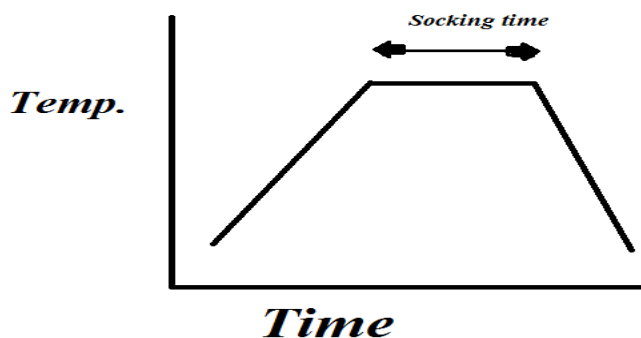


Figure 2.4: heat – time schedule .

## 2.6 The Wettability Tests

Wetting experiments was carried out by placing a small amount of each ratio of filler paste (0.25gm ) on the both graphite and stainless steel surfaces, and heating it at different temperature (700, 800, 850,900 and 950) °C in the tube furnace of vacuum pressure  $10^{-2}$  torr. After the previous step, the sample was took out until the furnace reaches to the room temperature. It is necessary known that the melting point of each element reacts is up of the experiment temperature. The contact angle (young angle), which is the angle the tangent makes with the solid surfaces, was measured from the profile of the filler drop shape. The contact angle was measured by new technique, where by using a computer program to transfer two dimension image to 3D. Figures 2.5 a,b and c demonstrate the different shapes of active filler on the fraphite surfaces.

These ratios of pure powders were used into the form of paste mixed with sticky substance (glycerin) to configure active filler alloy. The weight of this paste almost 0.25 g. Formed a paste of pure copper powder mixed with sticky substance (glycerin ), placed a drop on both graphite and stainless steel to test the wettability of the filler metal on the base metal. The sample was placed inside the furnace at different temperature (700, 800, 850 and 920), below melting point of filler metal. When the temperature of furnace reached to room temperature, the sample was took out. This process was repeated with other filler mentioned in table 2.3.

The process of wettability attempt of filler metal on the base metal (St.St, G ) was repeated where the filler paste of copper and silver with different concentration were mixed with glycerin and put a drop on both G and St.St as shown in figure 2.5 then placed inside the furnace at 920° C for 15 min.



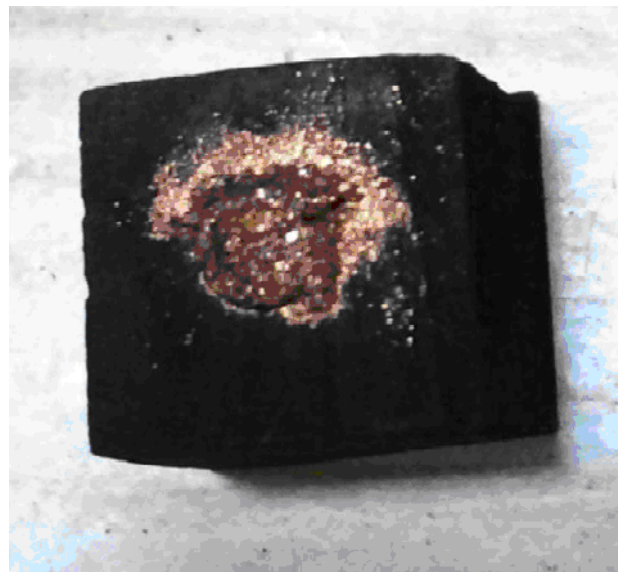
65% Cu  
35% Ag

(a)



60% Ag  
40% Cu

(b)



*50 % copper - 50 % silver*

(c)

Figure 2.5: different concentration of filler paste on the graphite surface.

Titanium powders in different concentration was used as filler paste instead of copper paste for enhanced wettability and bonding strength.

Formed a paste of filler metal consisted of pure powdered of titanium (F1) and placed on both samples of graphite and stainless steel to attempt the wettability of titanium on the base metals( St.St , G ) and placed it into furnace at a temperature 1200 °C less than melting point of titanium which equal to 1600 °C. the assembly of F1 and St.St was examined by X-ray diffraction.

## 2.7 Bond Strength Tests

After attempting the degree of wetting and spreading on the pieces surface, which considered one of the fundamentals of brazing technique. Brazing of graphite to the stainless steel was carried out for 18 sets differs in concentration ratios as mentioned in table 2.4. In order to obtain the optimum brazing conditions, many experiments involved temperature – time relationship were carried out. A stainless steel fixture was used to held the assembly before lying in the furnace as shown in figure 2.6.

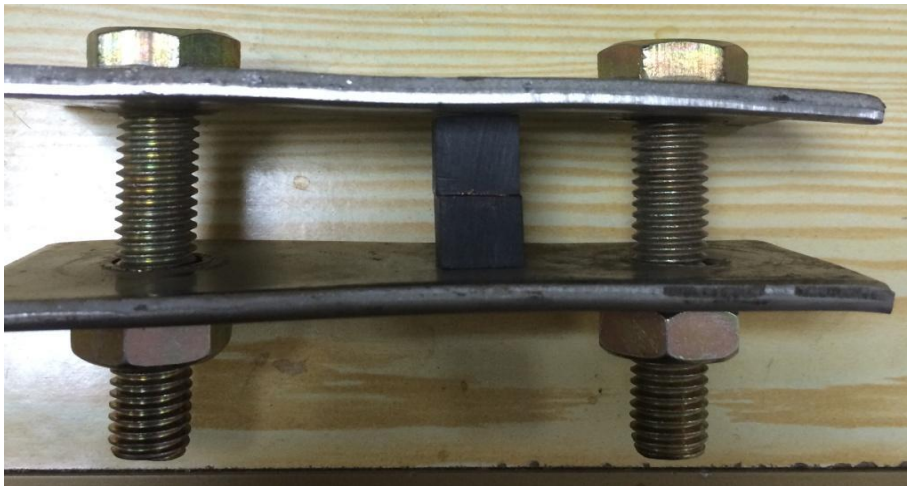


Figure 2.6: Stainless fixture hold the assembly (G/F5/G)

The vacuum furnace of pressure  $10^{-2}$  torr was used instead of atmospheric furnace to avoid the oxidation. The correlation between furnace temperature – time was clearly noticed on figure 2.7.

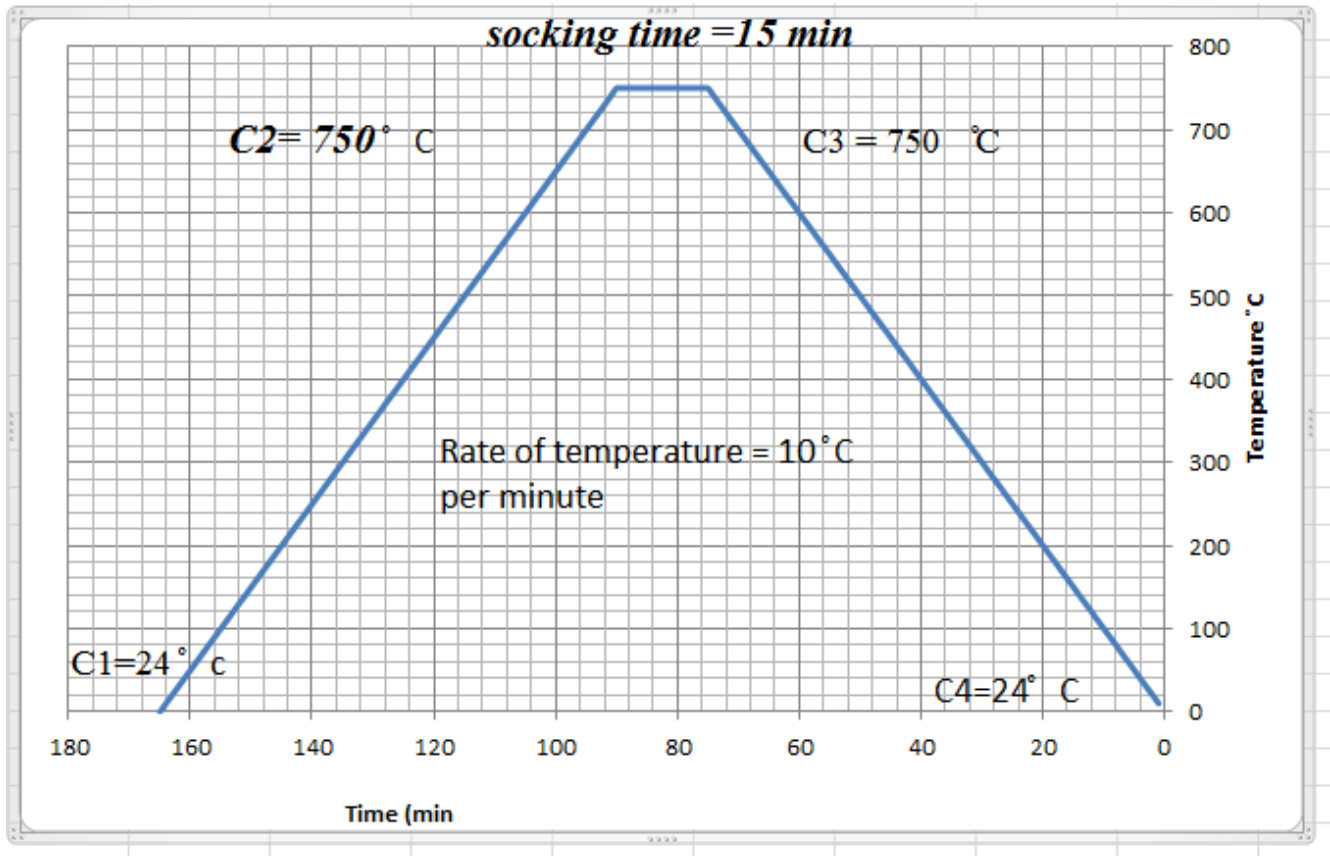


Figure 2.7: The schedule of Temperature – time of tube furnace.

Starting room temperature up to brazing temperature  $750^{\circ}C$  and holding the temperature for 15 min as a soaking time. The raising temperature was  $10^{\circ}C / \text{min}$  is the same reducing temperature until arrival to the room temperature. Repeating the process of brazing with another filler paste ratios for examining the favorable ratio to be used in order to obtain the optimum brazing strength. Two braze designs were used, butt and interlocking for examining the favourable design in which considered the best one gives a reliable joint. Because the butt joint design was failed corresponding with some ratios of active filler may be either due to unwettable surfaces or mismatch of thermal expansion coefficients between the pieces to be joined. For this reason interlocking design was substituted instead of butt in order to obtain a reliable joint. For interlocking design requirement, the graphite cube was turned to half with diameter 5mm, while the stainless steel was drilled a hole of diameter 6mm to insert the graphite pin into stainless steel hole as shown in figure 2.8.



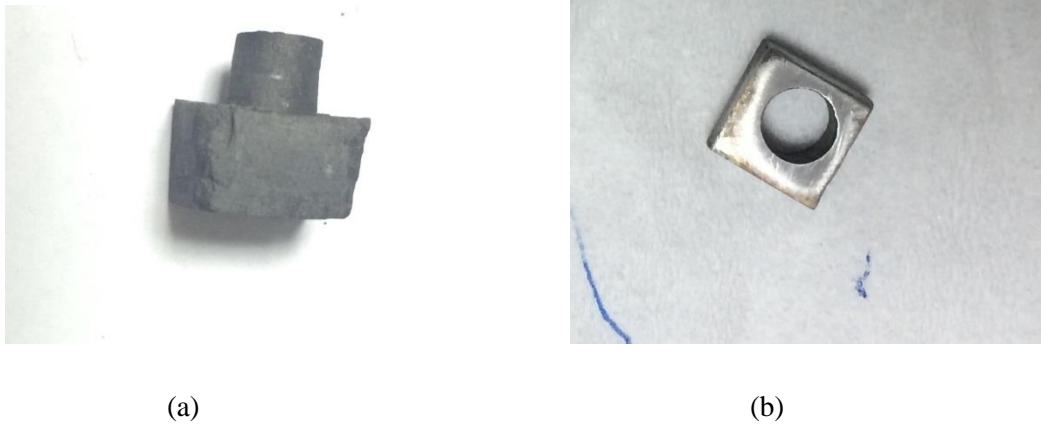


Figure 2.8: The interlocking design of samples graphite and stainless steel.

## 2.8 Fracture Tests

The graphite – stainless steel joint deal with in this work require the application of tensile stress vertically to the interface to test the extent of its strength. After verification of strength joint, the epoxy was used for adhesive screw at the tensile test as shown in figure 2.9 tensile test was carried out according to the ASTM F19 – 61 CLM. For this test, mechanical machine (Bongshin), model Dsck, was used in this investigation. The tensile force was applied at the joint with cross head speed of 3mm/ min. The fracture stress measured in ( $\text{N/m}^2 = \text{Pa}$ ) was calculated from the force applied vertically on the joint divided by the fractured real area.

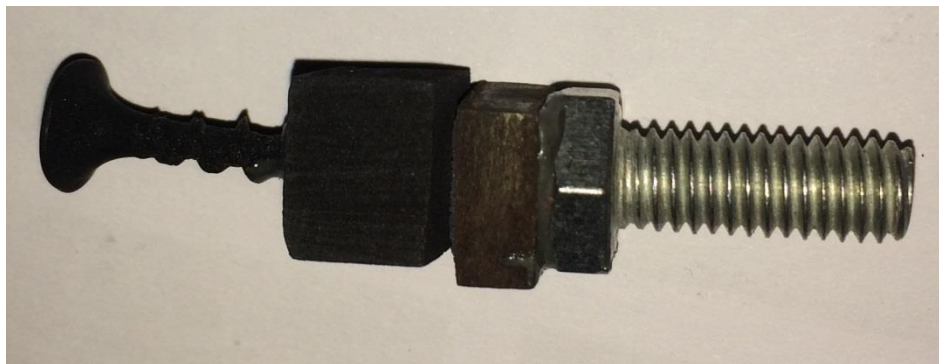


Figure 2. 9: the bonding of screw to the samples on both face.



## 2.9 Optical Microscopy

The optical microscope, often referred to as the "light microscope", is a type of microscope which uses visible light and a system of lenses to magnify images of small samples. Optical microscopes are the oldest and simplest of the microscopes. Digital microscopes are now available which use a CCD camera to examine a sample, and the image is shown directly on a computer screen without the need for optics such as eye-pieces. Other microscopic methods which do not use visible light include scanning electron microscopy and transmission electron microscopy[68,69]. The model of the microscope that to be used is mm0087000m-6307, Winona Ave, St. Louis, MO 63109-2124 U.S.A located at ALNahrain University-College of Science, with magnification 100x, 125x, 400x, DIN Objective 40x.

## 2.10 X-ray Diffraction

X-ray powder diffraction (XRD) is a rapid analytical technique primarily used for phase identification of a crystalline material and can provide information on unit cell dimensions. The analyzed material is finely ground, homogenized, and average bulk composition is determined [70,71,72]. Phase identification of the active filler and fractured surfaces had been achieved by using ( match program ). The type of the XRD system is PW3040/60-PANalytical B.V., Lelyweg 1, 7602 EA ALMELO, The Netherlands, system ID 202747 with the following conditions:

The x-ray tube target is Cu with monochromatic for  $K_{\alpha 1}$  radiation operated at 45kV and 30 mA. The  $2\theta$  range was taken from  $20^\circ$  \_\_\_\_\_  $80^\circ$ . The current location of this apparatus Al-nahrain university, collage of sciences.



Figure2.10: X-ray apparatus

## 2.11 Scanning Electron microscopic (SEM)

A scanning electron microscope (SEM) is a type of electron microscope that produces images of a sample by scanning it with a focused beam of electrons. The electrons interact with atoms in the sample, producing various signals that can be detected and that contain information about the sample's surface topography and composition. The electron beam is generally scanned in a raster scan pattern, and the beam's position is combined with the detected signal to produce an image. SEM can achieve resolution better than 1 nanometer. Specimens can be observed in high vacuum, in low vacuum, in wet conditions (in environmental SEM), and at a wide range of cryogenic or elevated temperatures. The most common SEM mode is detection of secondary electrons emitted by atoms excited by the electron beam [73,74]. The model of the system to be used is FEI inspect S50, low-vacuum scanning electron microscope (SEM) platform with high-resolution capabilities using thermal emission electron optics. While the low-vacuum settings are particularly useful for inspection and characterization of non-conductive and heavily contaminated materials, the system also minimizes sample preparation. The current location of this apparatus Al-nahrain university, collage of sciences.

## 2.12 The Differential Scanning Calorimetry (DSC)

The Differential Scanning Calorimetry is the most popular measurement technique to detect endothermic and exothermic transitions like the determination of transformation temperatures and enthalpy of solids and liquids as a function of temperature. Therefore the sample and reference are maintained at nearly the same temperature throughout the experiment and the heat flux will be measured [75].

## 2.13 Thermogravimetry (TG)

Thermogravimetry is a technique in which the mass of the sample is monitored (balance) against time or temperature (thermal) while the temperature of the sample, in a specified atmosphere, is programmed. This technique serves the determination of material compositions. It is a common analysis method in the chemical and pharmaceutical industry. Thermogravimetric analysis (TGA) is performed on polymers, food, pharmaceuticals as well as many other materials [76]. The current location of this apparatus Al-nahrain university, collage of sciences.



Figure 2.11: The Differential Scanning Calorimetry - Thermogravimetry (TG) apparatus



# **CHAPTER THREE**

## **RESULTS AND DISCUSSION**

## Chapter Three

### Results and Discussion

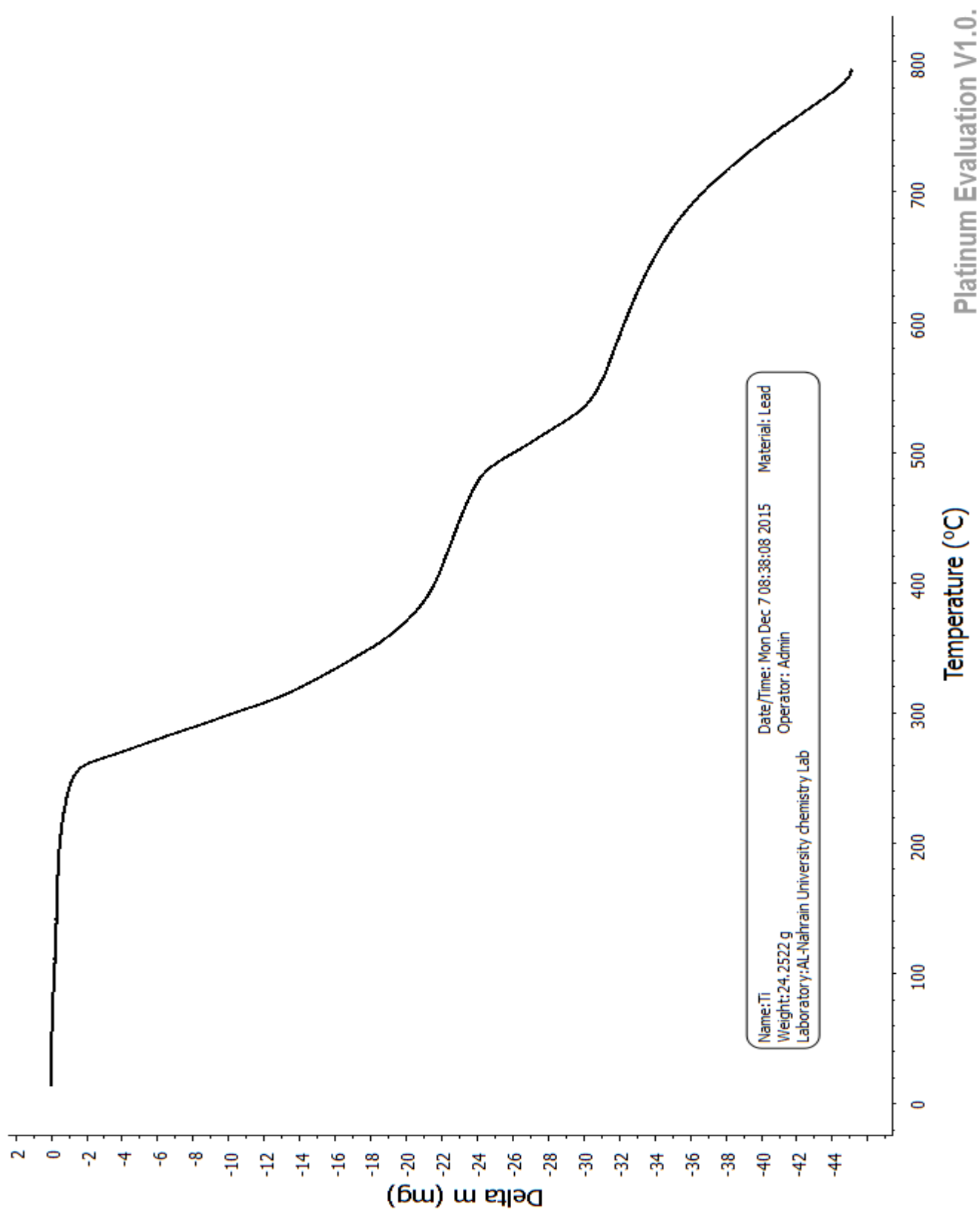
#### 3.1 Introduction

This chapter includes the attempts of such wettability of filler paste on both graphite and stainless steel. As well as the physical properties of each type of filler which were used on this investigation. Bonding between graphite and stainless steel were also attempted in different design.

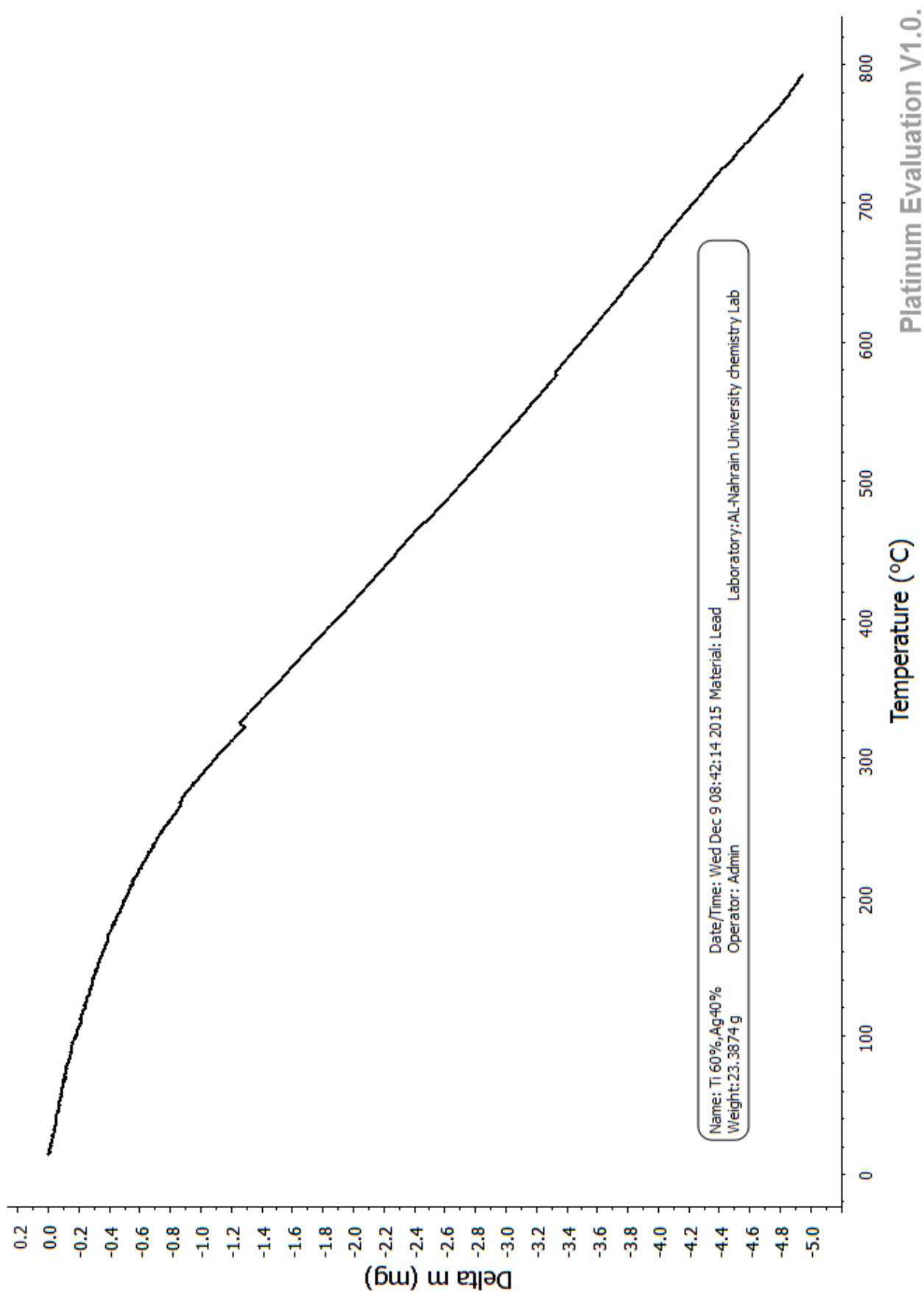
#### 3.2 Active Filler Tests

Three categories of active filler were subjected to the thermal analysis. /the constituents of active filler were mainly Cu and Ag in different ratios. The main advantage of active thermal analysis are to determine melting point of each filler and endothermic – exothermic states. Thermal analysis showed the change in a property of a sample, which is related to an imposed change as a function of temperature. The active fillers were separated from the base material and then subjected from the base material and then subjected to the test. Dehydration, decomposition, desorption and oxidation process are associated with Tg analysis. The area under the endothermic or exothermic is related to the enthalpy of the thermal reaction  $\Delta H$ . Figures 3.1 a,b,c illustrate the heat behaviour of the filler under test. This figures reveals the correlation of mass variation as a function of temperature. The reduction of mass was associated starting with increasing temperature along experiment time providing heat. The figure 3.1 c, the sample mass was increased at the temperature about 730 °C, this increases may be due to the oxidation. The  $TiO_x$  phases were confirmed by x-ray diffraction and may form as a result of an exothermic reaction of the titanium consisting with oxygen fed by diffusion from the metal and graphite substrates. The recording of the change in the sample heat with respect to the reference as a function of temperature was plotted in figures 3.2 a, b, c. This figures illustrate the measured of heat content

(enthalpy). In addition, the down peaks indicates to exothermic state while the peaks indicate to endothermic state.

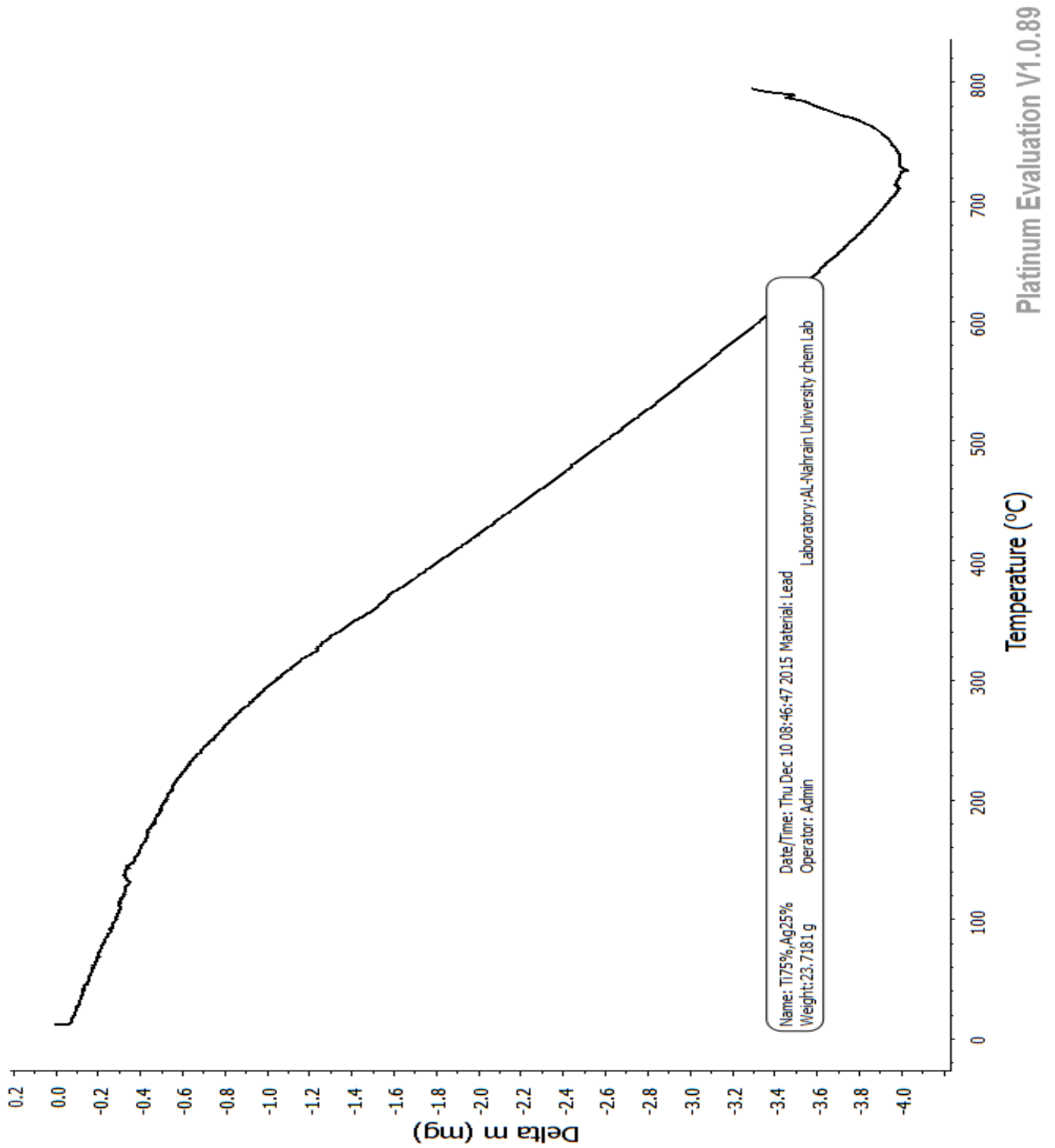


(a)



(b)  
51





(c)

Figure 3.1: the Thermogravimetric Analysis (TG) for Filler Paste

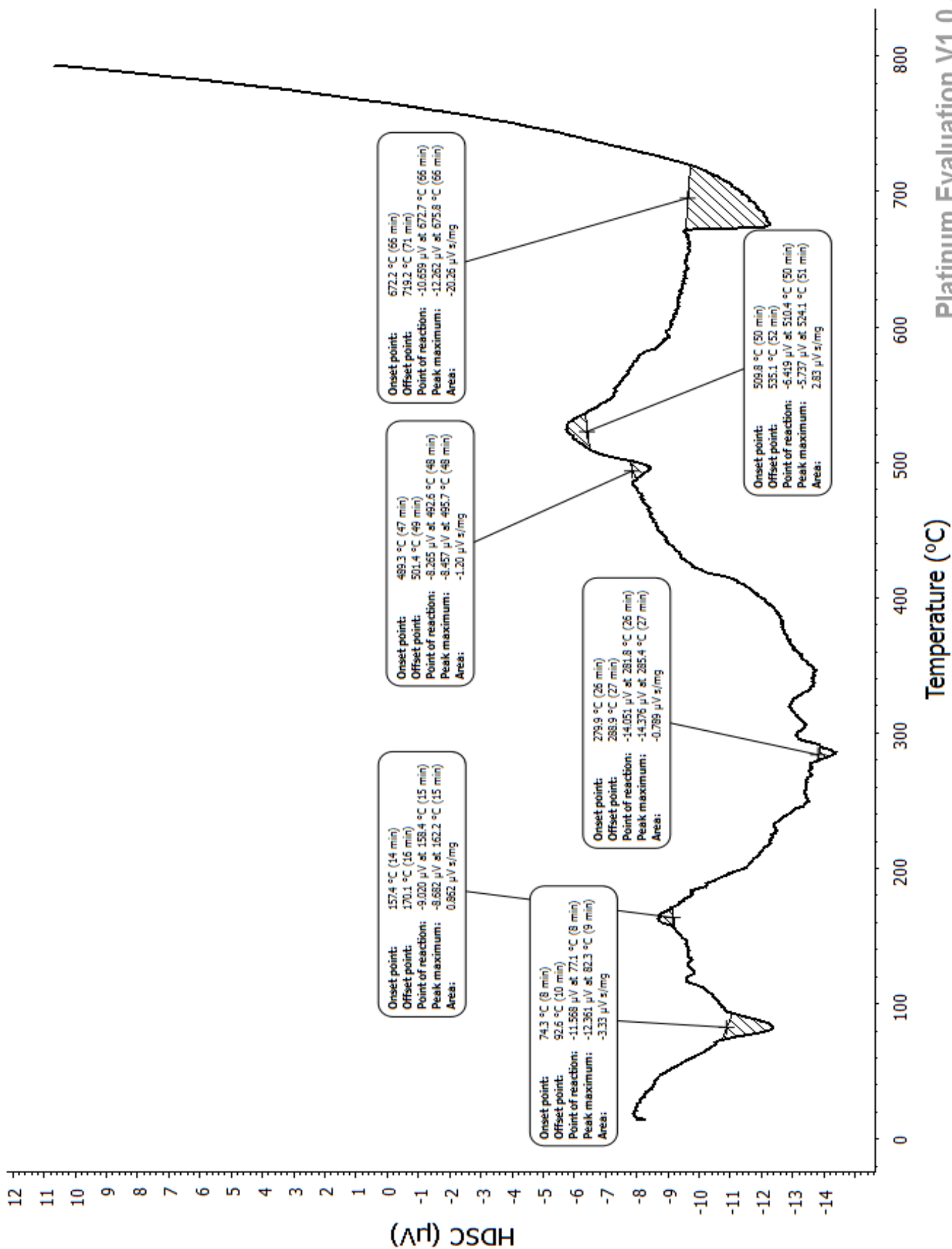
- (a) Titanium Filler paste.
- (b) 60% Titanium , 40% silver filler paste.
- (c) 75% Titanium ,25% silver filler paste

### 3.2.1 Thermal Analysis of Active Filler Alloy

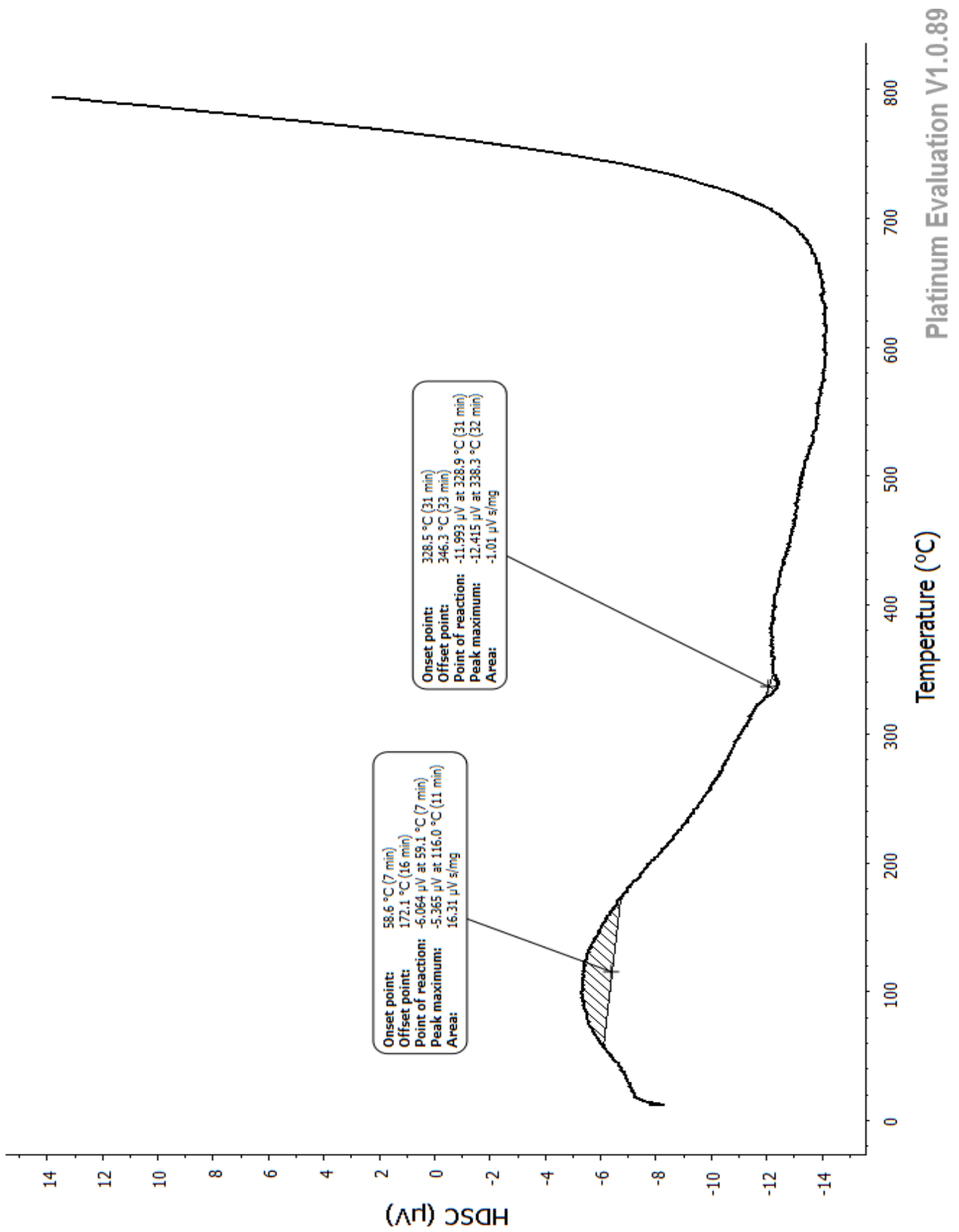
Thermal analysis showed the change in a property of a sample, which is related to an imposed change in the temperature of the sample. Figures 3.1a, b and c represent the thermogravimetric analysis (TG) test for three types of active fillers (pure Ti), (60%Ti +40%Ag) and (75%Ti +25% Ag) respectively. TG measures mass change in a material as a function of temperature under a controlled atmosphere. Dehydration, decomposition, desorption and oxidation processes are associated with TG analysis. Both endothermic and exothermic reactions take place within the sample. The area under the endotherm or exotherm is related to the enthalpy of the thermal reaction  $\Delta H$ . As can be noticed from the figures that all reactions are associated with loss in mass of the samples. The main advantage of thermal analysis is to determine the melting point of each active filler, and therefore to determine furnace temperature concerning bonding between the partners to be joined.

### 3.2.2 Differential Scanning Calorimetry

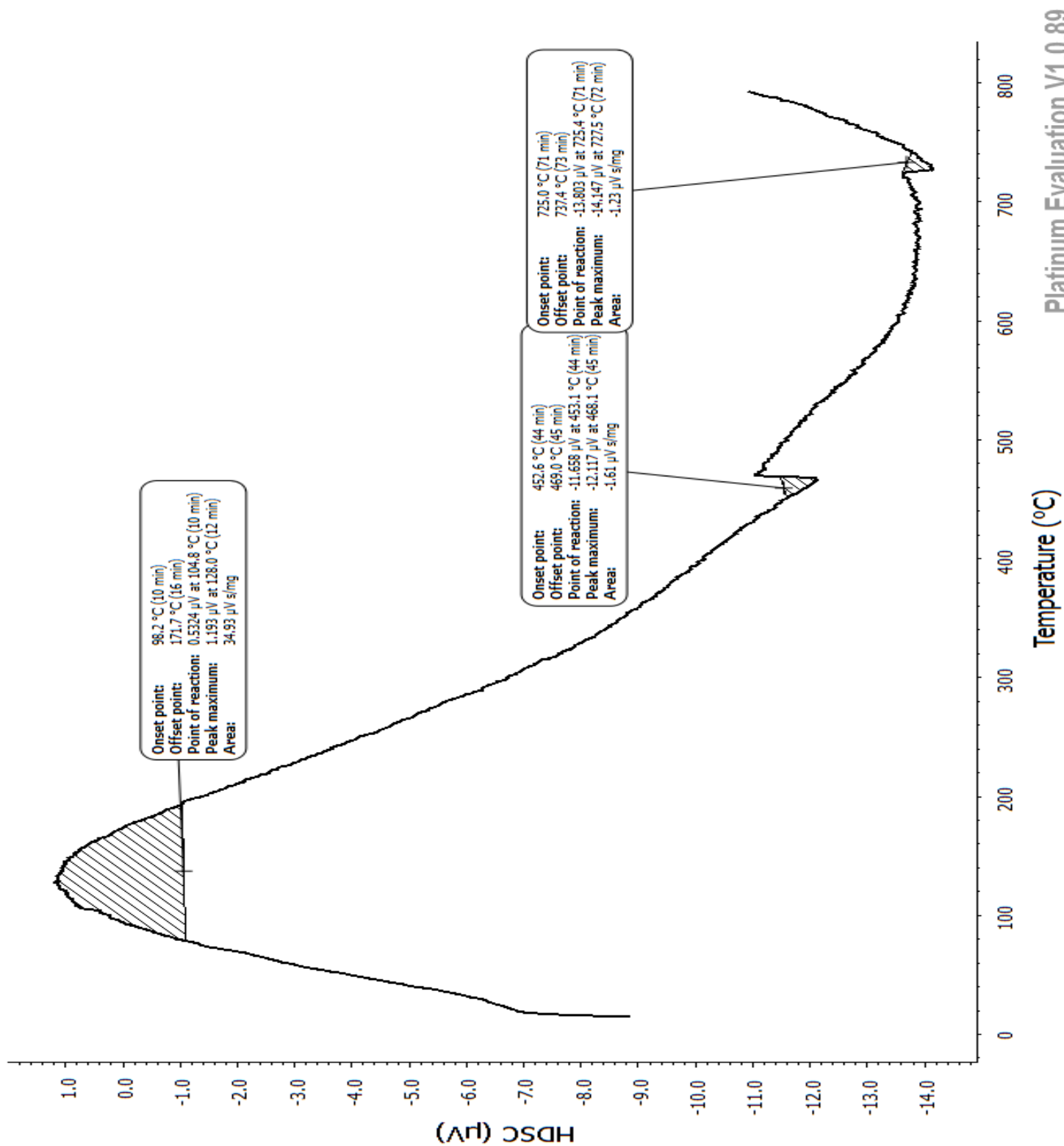
The objective of this technique is to record the change in sample heat with respect to reference as a function of temperature. This measures the heat capacity and specific heat capacity at a constant pressure  $C_p$ . As shown in figure 3.2.



(a)



(b)



(c)

Figure3.2; the Differential Scanning Calorimetry for Filler Paste

- (a) Titanium Filler paste.
- (b) 60% Titanium , 40% silver filler paste.
- (c) 75% Titanium ,25% silver filler paste.

### 3.3 Wetting and Spreading Tests:

#### 3.3.1 Wetting of Pure Copper Filler Paste.

Several experiments of both wetting and spreading were carried out on the surface of graphite and stainless steel in vacuum furnaces under pressure of  $10^{-2}$  torr at different temperatures. Accordingly, pure copper filler when became solid state seem incapable to wet graphite surface as noticed in figure 3.3 a, b. This result is in agreement with other researcher [46]. In the same time the active copper filler reveal the ability wet the stainless steel surface as shown in figure 3.3 a, b. Due to the plastic form of copper filler, there is lower spreading of the filler on the substrate (stainless steel). Because of the mismatch of thermal expansion coefficient, the copper filler was separated partially from the stainless steel surface when the furnaces temperature reduced to the laboratory. Another variable is the surface finish of the stainless steel . Considered a reason of filler separated. These previously effects considered sufficiently reasons that make copper filler could not verification a permanent joint with stainless steel.



(a)

(b)

Figure 3.3: the wettability of filler metal of copper on the graphite and stainless steel.

### 3.3.2 The Wetting of Silver Filler

Other experiments of wetting and spreading were carried out using silver filler paste. The experiments improved the ability of silver to wet both graphite and stainless steel surfaces as shown in figure 3.4. Same problem of separated filler from the base pieces was found soon they lefted the furnace. The reason of separation may be due to the mismatch in coefficient of thermal expansion between the pieces to be joined. The second reason may be due to oxides growth on the joint surface, where in pure reactive metal oxide systems, the oxide – reduction reaction is generally used to discuss weak reactivity [34].

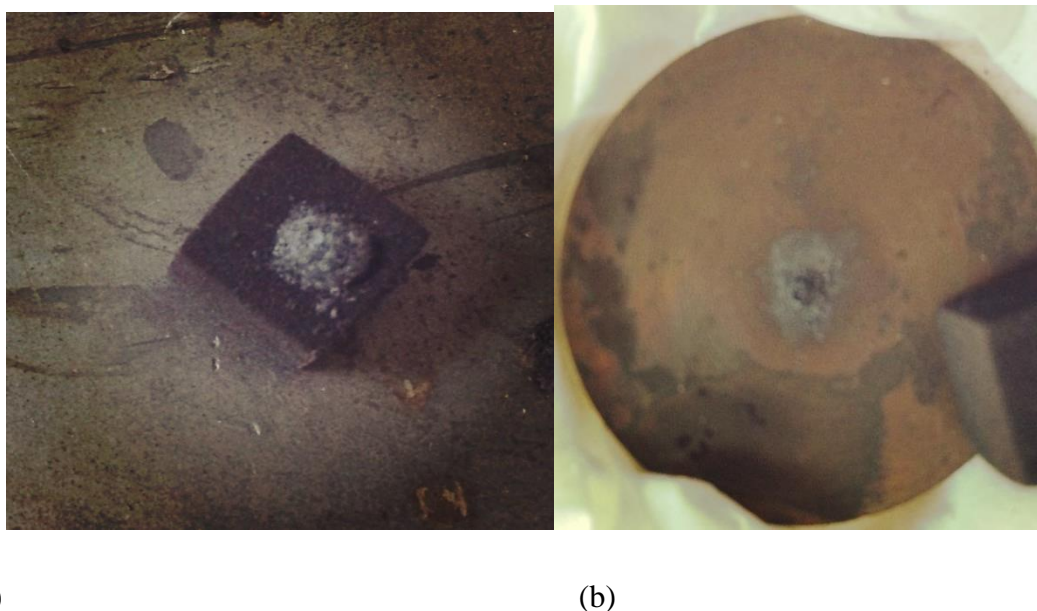


Figure 3.4: the wettability of filler metal of silver on the graphite and stainless steel

### 3.3.3 The Wettability of Copper – Silver Filler Test

Three ratios of active filler paste in different concentration of copper – silver were prepared and examined using vacuum tube furnace of pressure  $10^{-2}$  torr at different temperature (800, 850, 900 and 950) $^{\circ}$ C. Figure 3.5, illustrate the behaviour of filler wettability, so one can deduce that increasing ratio of silver in the mixed paste will be enhanced the wettability on both surfaces of stainless steel and graphite. The young angle on both graphite and stainless steel surfaces was found equal  $80^{\circ}$  obtained where the filler composed of (60 wt % Ag + 40 wt %



Cu). The young angle was measured by using a suitable computer program by transfer the two dimension image into three dimension image, then the contact angle was measured by protractor level between the drop tangential and the x-axis of the substrate as shown in figure 3.7. These results led to the conclusion that the wettability of ( Ag+ Cu) filler is increased as increases of silver ratio in the mixture. Accordingly, the spreading of the active filler composed of (60% Ag + 40% Cu) when becomes alloy, has a favorable state from other ratios of Ag and Cu under the same operating condition. This result is in agreement with other researcher [46].

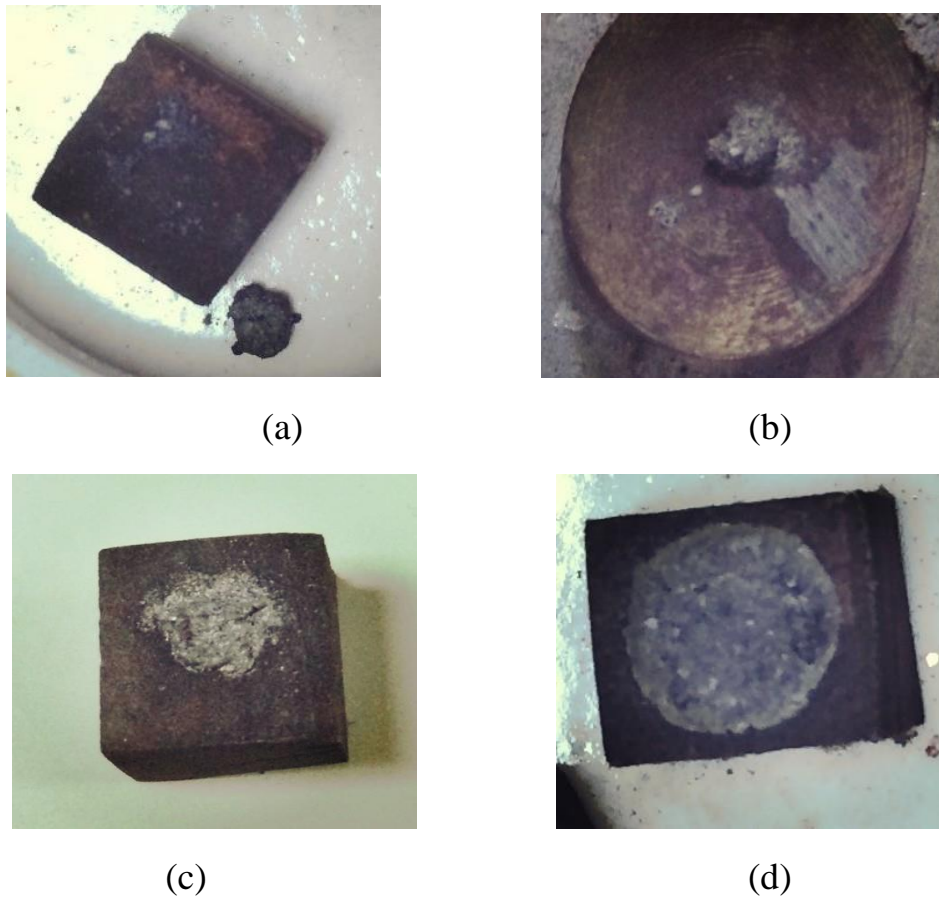


Figure 3.5: the wettability attempt on both graphite and stainless steel in different concentrations of filler paste.

(a) Graphite specimen with drop of filler paste (35% Ag , 65 % Cu ).

(b) Stainless steel specimen with drop of filler paste (35% Ag ,65 % Cu ).

(c) Graphite specimen with drop of filler metal (consist of 50%copper and 50% silver) on the graphite.



(d) graphite specimen with drop of filler paste ( 60 % Ag , 40 % Cu ).

### 3.3.4 Wettability of Titanium

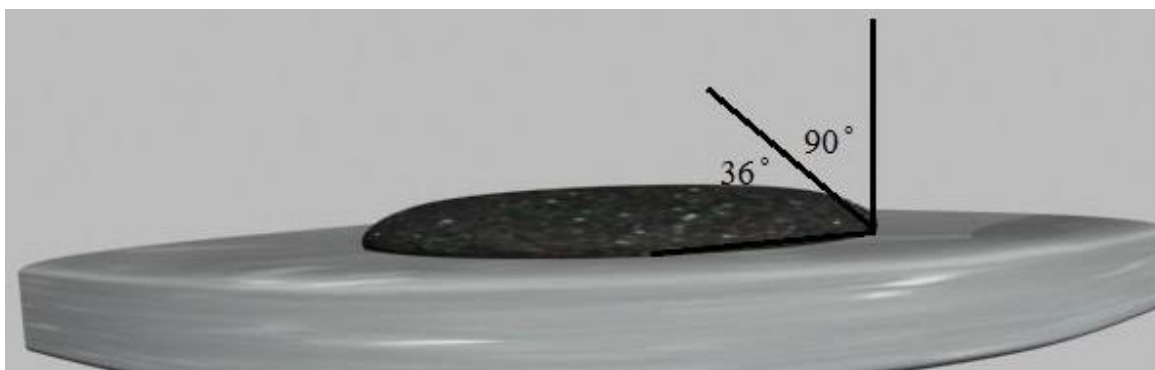
The ability of active filler contains titanium to wet graphite and stainless steel surfaces can be notice on the figure 3.6. As the Ti increases in the filler, the wettability is increases too, because the titanium is often chosen as solute element in filler metals to improve the wetting on pieces surface. The contact (Young) angle of the filler drop tangential with x- axis of substrate are found equal  $67^\circ$  and  $36^\circ$  as shown in figure 3.7 on graphite and stainless steel respectively. According to this experimental real, a strong improvment of wetting of a non- reactive metal can be obtained adding active element like titanium, that leads to achieve good bonding between the pieces to be joined, On the other hand, one can notice the titanium considered reduce element for the spreading state. This experimental fact is in agreement with other researcher and theoritical part mentioned in chapter one. The correlation of titanium concetration with the contact angle for graphite and stainless steel are illustrated in figures 3.8 , 3.9. Both figures demontrated the effects of titanium concentration with respect contact angle. This experimental attempt was enhanced by the figures 3.6.



Figure 3.6: The wettability of titanium on both graphite and stainless steel.

From the above Figures one can noticed that the wettability of Ti better than that of any filler paste which were used in this investigation. The good wettability

give us more hope to obtain good bonding between graphite and stainless steel . The contact angle of Ti on graphite is about ( $67^\circ$ ) while ( $36^\circ$ ) on the stainless steel AS shown in figure 3.7.



(a)



(b)

Figure 3.7: the contact angle of wettability of filler metal on base metal by using software program to convert image from 2D to 3D.

- (a) . The contact angle of wettability of filler metal (titanium) on stainless steel.
- (b) . The contact angle of wettability of filler metal (titanium) on graphite.

The figures 3.8, 3.9 shows the schematics of the relation between the concentrations of titanium against the contact angle for graphite and stainless steel respectively.

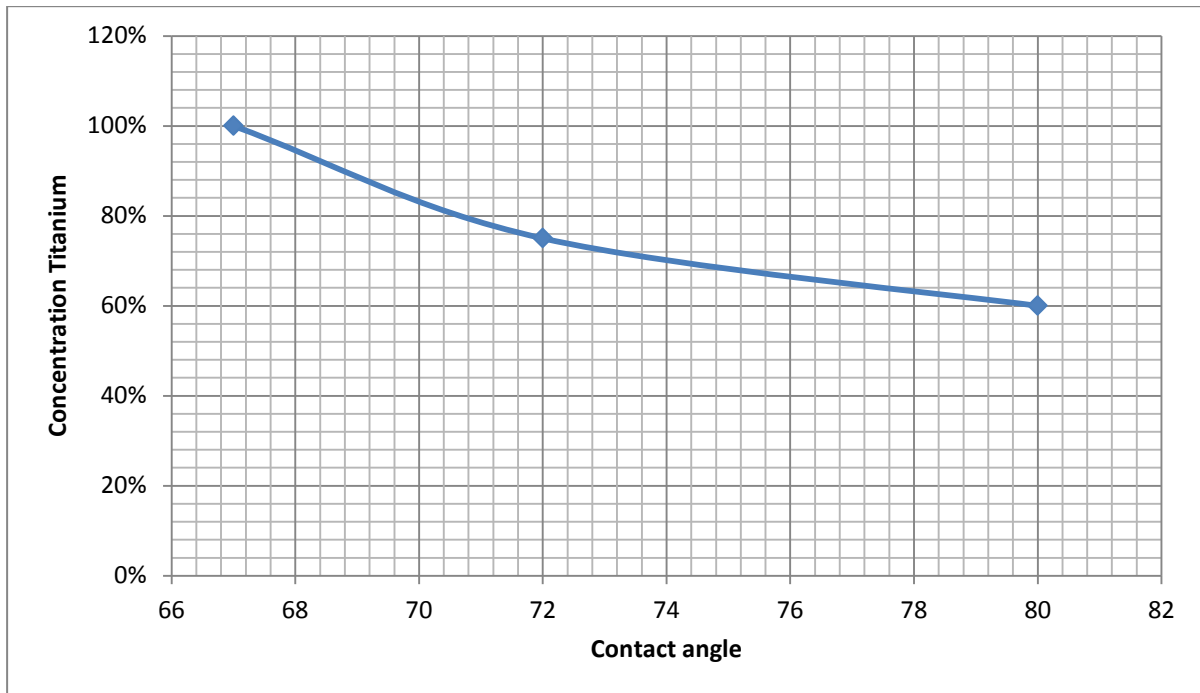


Figure 3.8: relation between the concentration of titanium against contact angle for graphite.

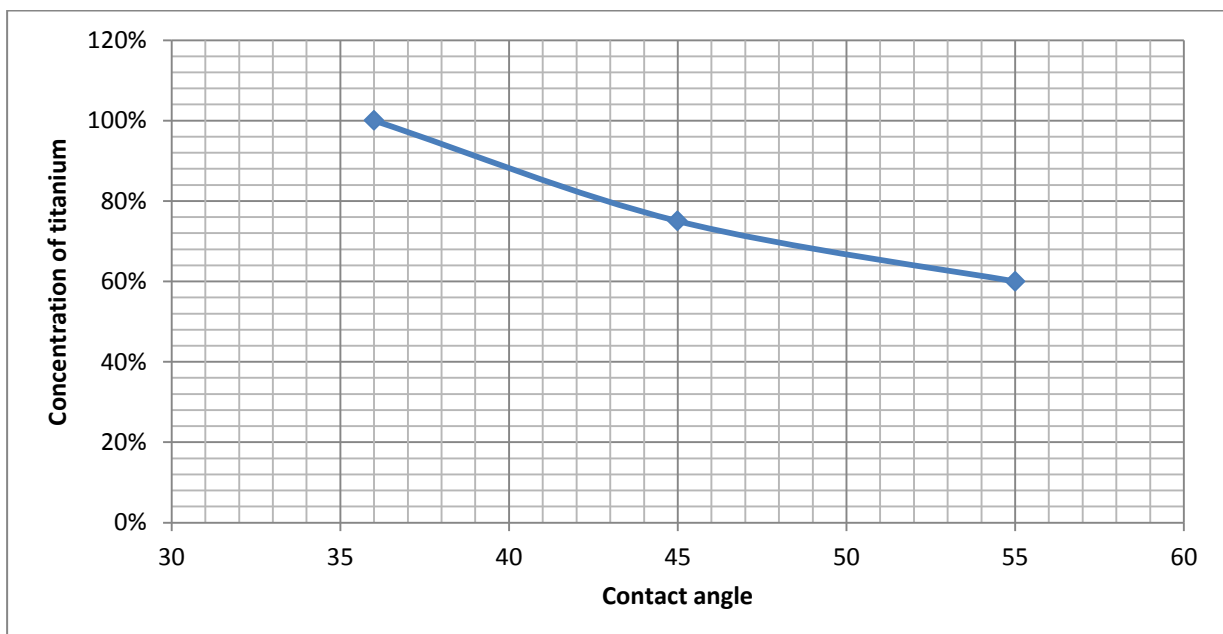


Figure 3.9: relation between the concentration of titanium against contact angle for stainless steel.

### 3.5 Joint Strength

Several experiments of bonding were performed to investigate the joint reliability of the graphite bonded stainless steel by brazing mechanism. The first attempt was carried out using an active filler composed of (50wt% Cu + 50 wt % Ag). The filler paste lied in between graphite – stainless steel. Under the operation condition of furnace vacuum  $10^{-2}$  torr and furnace temperature  $850^{\circ}\text{C}$ . The butt design was used for joining the pieces. It was clear that could not obtained a reliable joint as shown in figure 3.10 where the graphite pieces were separated soon the assembly left from the furnace. It was notice some of the adherent filler on the graphite surfaces. This encourage us to repeat more experiments through using another bonding design. Another way was take place by instead the smooth graphite surface by roughed surface using a compressed air for applied pressure on the Sic stone, where the compressed Sic was striking graphite surface for achieved roughning process. The target of this procedure is for increasing the bonding surface area and then increasing the interlocking bonding and hence obtained a reliable joint.



Figure 3.10: Thin layer of filler metal on the two samples of graphite.

### 3.5.1 The Bonding by Pure Titanium

New active filler paste of pure titanium was used as an active brazing filler for verification bonding graphite – stainless steel. The brazing was failed because of low furnace temperature (1050 °C compared with the melting point of titanium  $\approx$  1668 °C and may be the butt brazing design was used as shown in figure 3.11. It was noticed some of titanium filler was adhere strongly with stainless steel surface as shown in figure 3.11. This phenomena was happened due to the chemical reaction between stainless steel – titanium filler was achieved under the experimental condition.



Figure.3.11: Fracture surfaces of graphite – stainless steel

### 3.5.2 Bonding by Ag – Ti Filler Paste

The brazing of graphite – stainless steel was carried out using active filler paste composed of (75 wt% Ti + 25 wt% Ag) and butt – lab design was used for this investigation as shown in figure 3.12. The bond strength of joint obtained was 15 Mpa. The bonding results listed in table 3.7 involved different ratios of Ti, Ag elements which composed active filler and different furnace temperatures used for the goal receiving the optimum condition of a reliable brazing joint. One can notice the more fractured mode happened from the filler layer. The fracture stress applied on the brazing joints considered involved a consequence of the integrity in the joining conditions include brazing temperature, furnace vacuum, soaking time

and type of active filler. These are presents the answer of question arises of what status exsited to bring fracture stress to its maximum value. Accordingly titanium element was used in the mixture of active filler for enhancing bonding strength. The titanium element was segregate to the brazing elements and then consists new compound such  $TiO_x$  or  $TiC$  at intermediate layer. The correlation between the fracture tensile stress and the ratios of bath Ti and Ag in active filler were plotted in figures 3.13 and 3.14 respectivley. The use of silver element in brazing active filler is justified by the special balance of properties that its inclusion provides, such properties are the ability to the wet the graphite and stainless steel low melting temperature compared with other elements. These properties was found enhanced the wettability and hence bonding strength of graphite to be bonded stainless steel. On other hand, Titanium is chosen as solute element in filler alloy to enhance and improve the bonding strength.

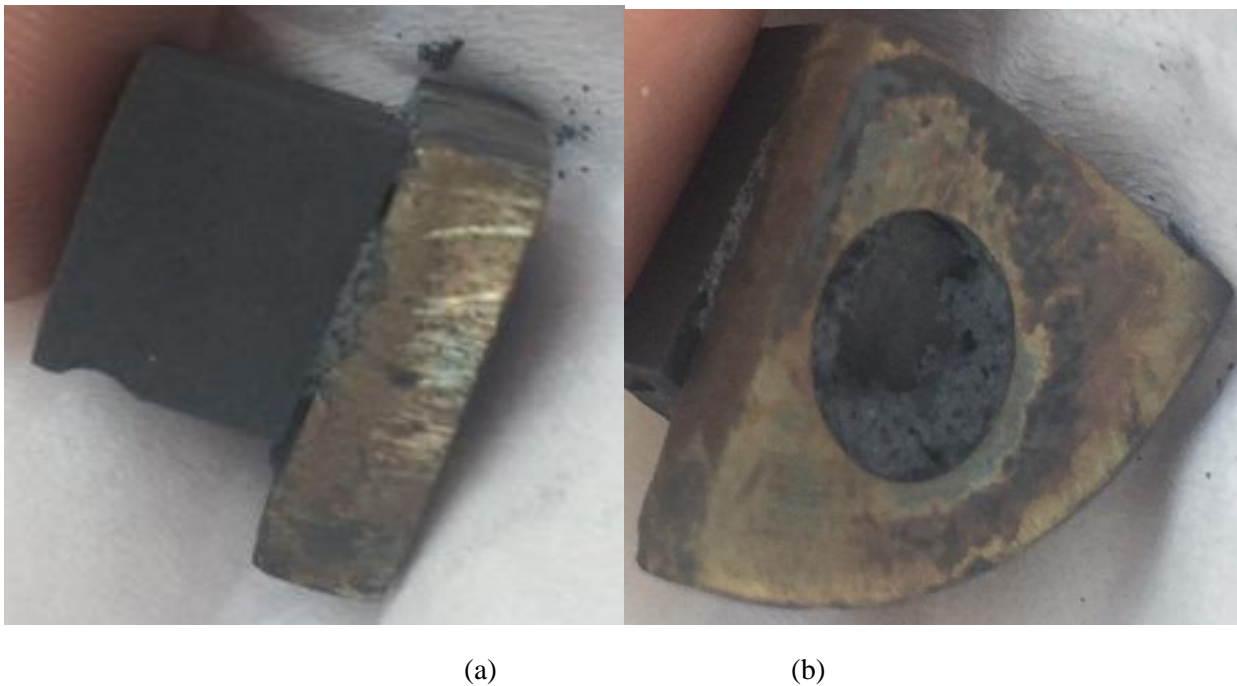


Figure 3.12: Butt - lab design of graphite – stainless steel.

Table 3.1: Represent the bonding strength relative to type of active filler

Set no.	Filler type	Fracture mode	Fracture stress (Mpa)	Temperature ° C
1	75wt% Ti + 25wt% Ag	Filler	15	900
2	75wt% Ti + 25wt% Ag	Filler	0.75	950
3	100wt% Ti	Filler and part of graphite	3.5	900
4	100wt% Ti	Filler	3	950
5	100wt% Ti	Filler	Null	1000
6	60wt% Ti + 40wt% Ag	Filler	2	950
7	100wt% Ti	Filler and part of graphite	0.5	950
8	60wt% Ti + 40wt% Ag	Filler	Null	950
9	75wt% Ti + 25wt% Ag	Filler	1	850

The figures 3.13, 3.14 shows the schematics of the relation between the concentrations of titanium against the fracture stress and the concentrations of silver against the fracture stress in :



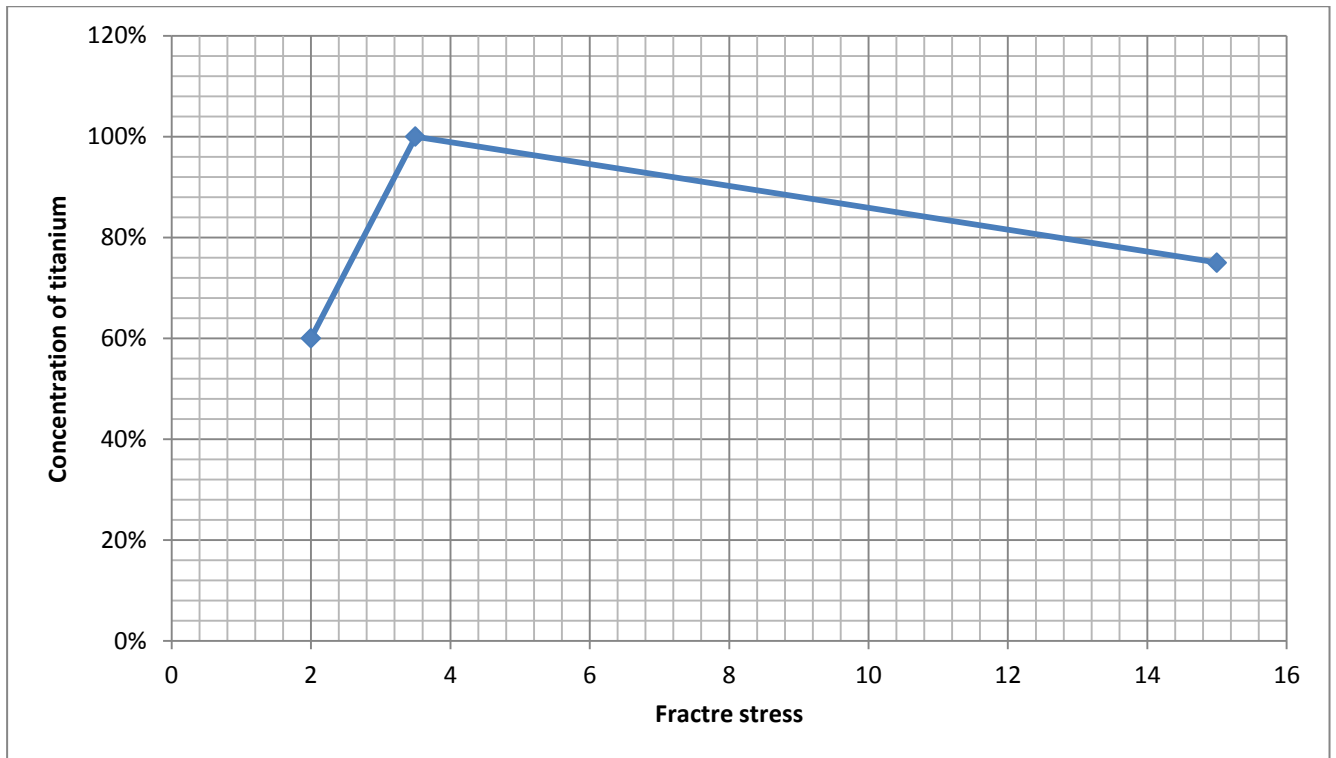


Figure.3.13: relation between the concentration of titanium against fracture stress.

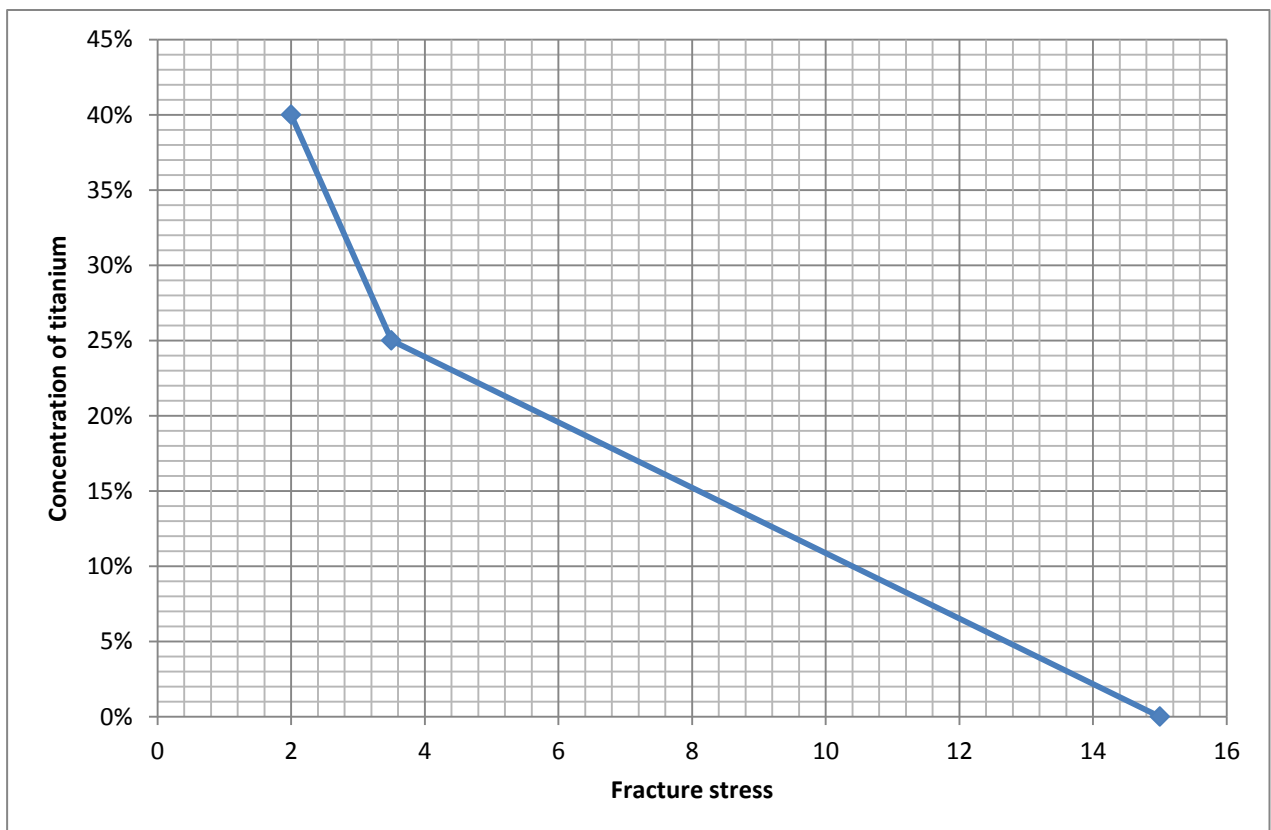


Figure.3.14: relation between the concentration of Silver against fracture stress.



### 3.6 The Factors affecting of Bonding Strength Between Graphite \_ Stainless Steel

- 1- The good wettability of Titanium on both graphite and stainless steel give good bonding.
- 2- Nitrogen gas was pumped into furnace before applied pressure for releasing oxygen.
- 3- The temperature of furnace should be 70 % of melting point for filler metals (plastic form).
- 4 –Interlocking design were used instead of butt design.
- 5- bonding surface area has proportional to the bonding strength.

### 3.7 Microscopic Analysis

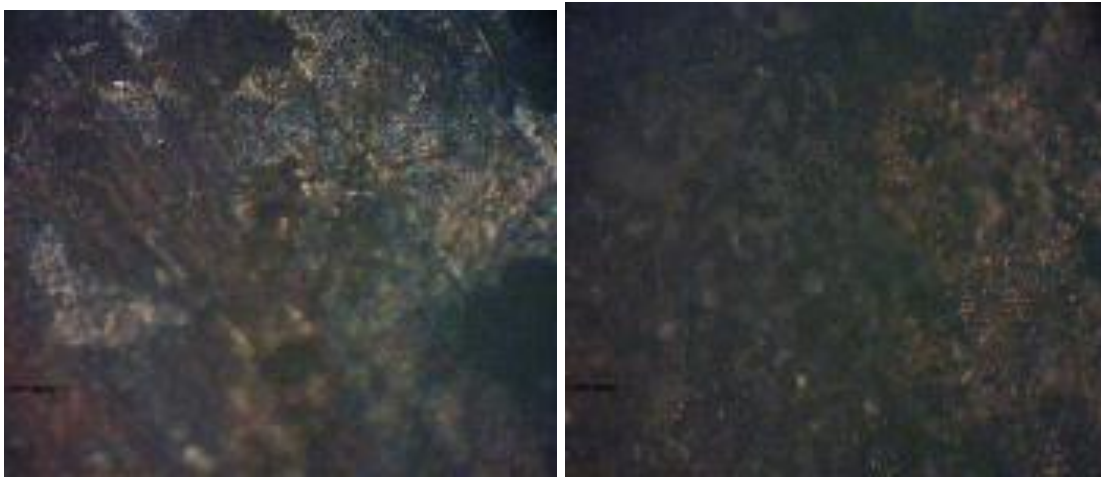
Three main parameters affecting the analysis of surface topography. These parameters are suitable illumination, adequate magnification, and contrast test area figure 3.15 reveal the ability of pure copper wet and adhere with stainless steel surfaces as a bright colour area shown on the right side of the image. The dark area was found free of active filler, this is due to unhomogenous distribution of filler to cover all the brazed area. Figures 3.16 and 3.17 showed the ability of pure titanium to wet graphite and stainless steel surfaces. It is clear small regions of stainless steel surfaces was covered by Ti filler, this is may be due to insufficient furnace temperature ( $\approx 1000$  °C) Compared with the melting point of Ti. As can noticed spots area of graphite surface were covered by Ti filler. This is because the weak ability of Ti to wet graphite surface as known from the theoretical part and other researchers experiments [46]. More detail about wetting and bonding behaviour were obtained when using SEM technique. Figure 3.18 b .



(a)

(b)

Figure 3.15: Optical image of pure (copper) on stainless steel (magnification x 40).



(a)

(b)

Figure 3.16: Optical image of pure (titanium) on stainless steel (magnification x 10).

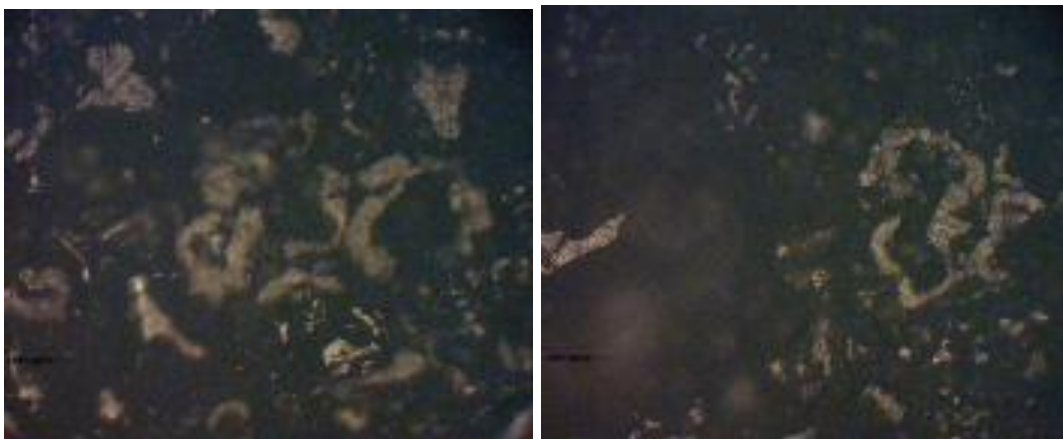
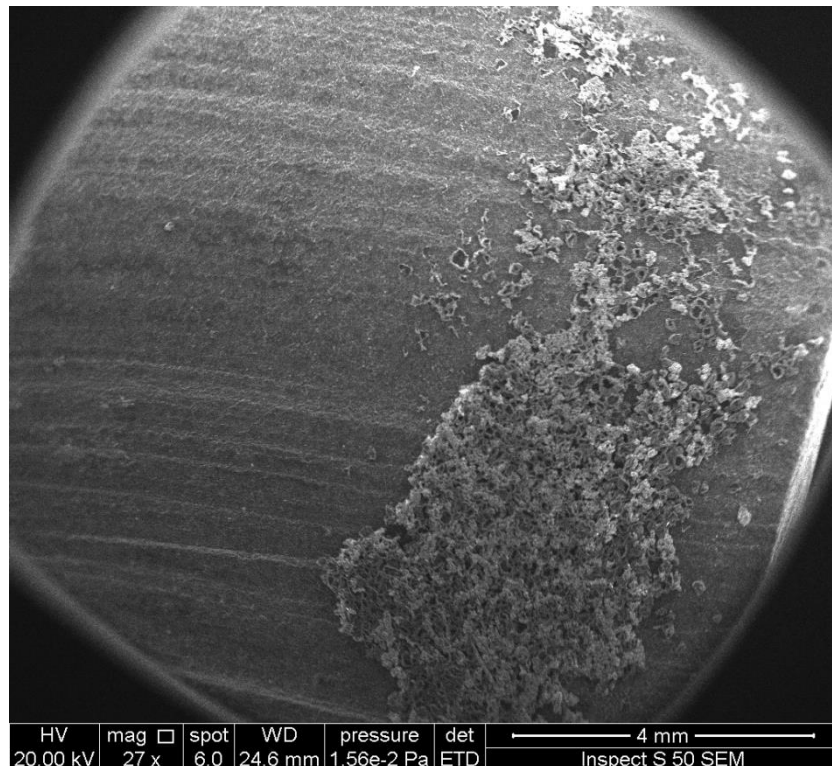


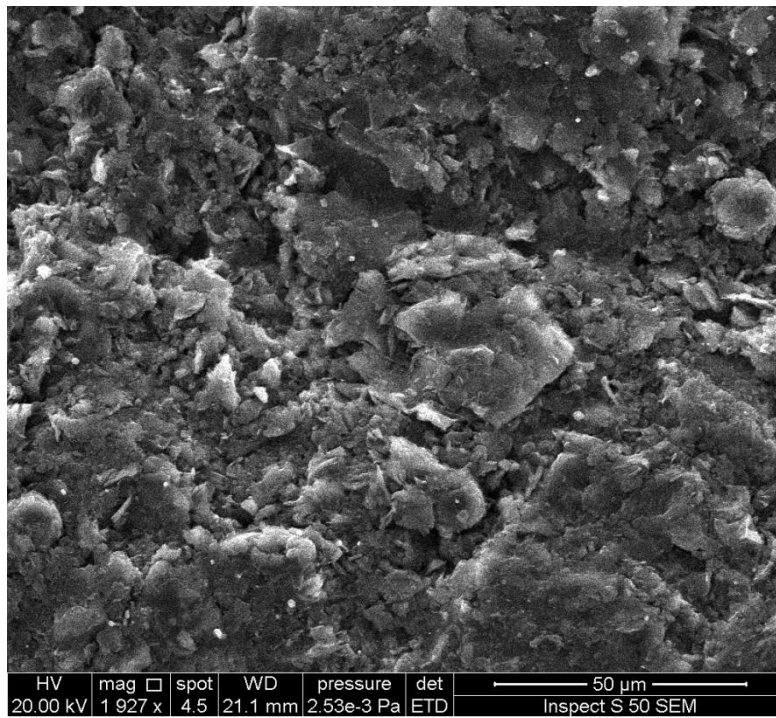
Figure 3.17: Optical image of pure (titanium) on graphite( magnification x 10) .

Figure 3.18-b reveal of the lamella of adherent filler on the graphite surface, due to this adherent filler is contributed in bonding mechanism where the electrons occupies the valance bond in the metal filler and stainless steel will be joint together in order to compose orbital (may be ionic or covalent bonds ) figure 3.16-crepresents the bright spots consist of titanium filler on the graphite substrate while more texture of titanium filler is clearly noticed as figure 3.19-a. The regular distribution of active filler on the graphite surface will be gives an indicate about the good bonding between the partners. The micrograph illustrate in figure 3.19–b represent the fracture surfaces; one can noticed there are many dark regions were found built on the active filler, this regions due to graphite fracture mode and this region pointed to the good bonding between titanium (filler) and graphite surface.

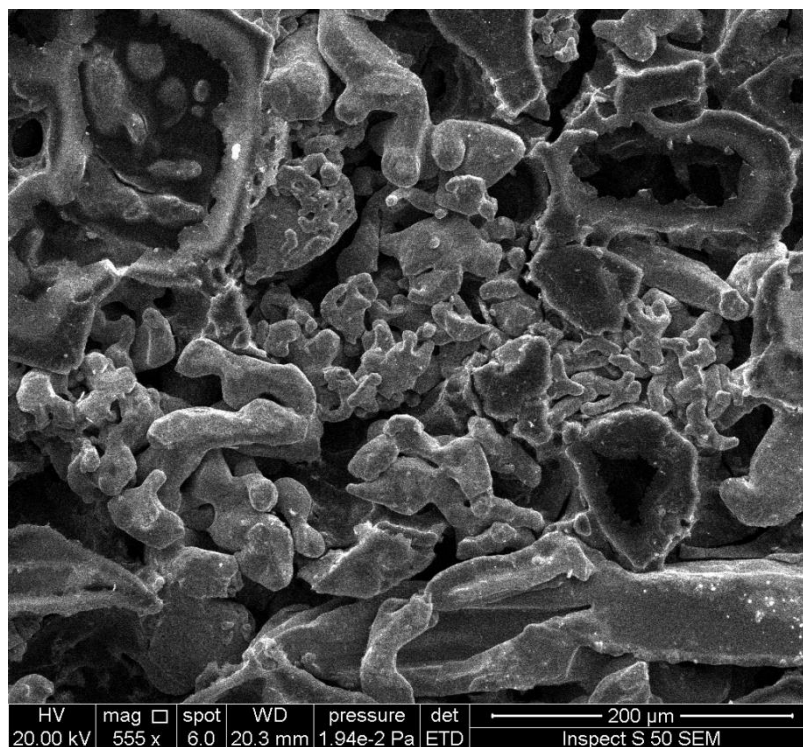


(a)



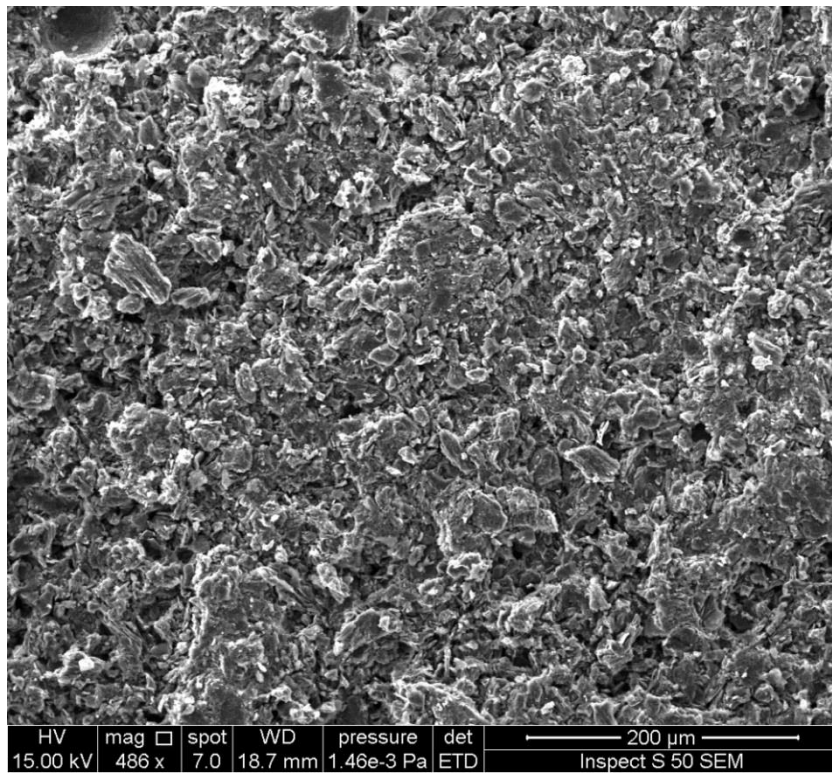


(b)

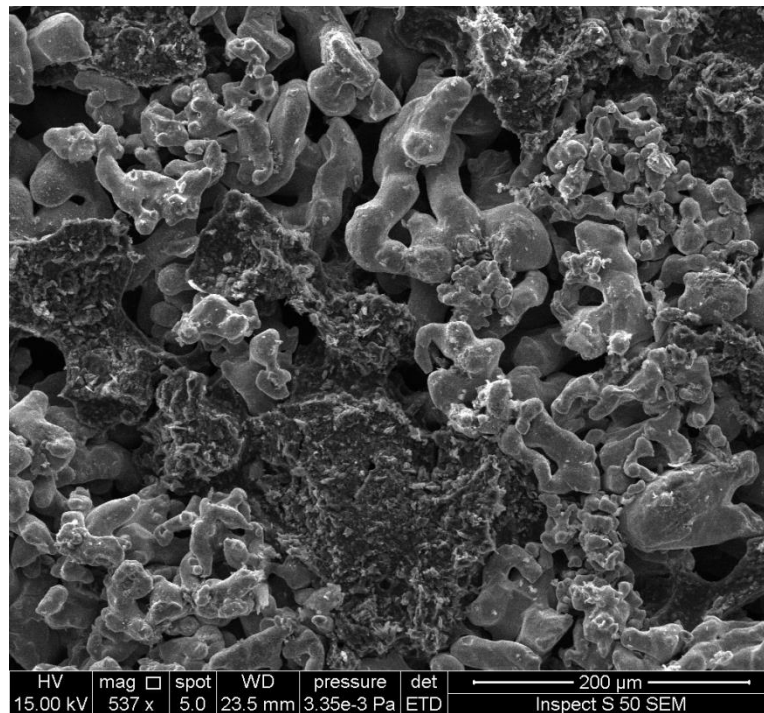


(c)

Figure 3.18: The scanning electron microscopy for surface of graphite after bonding with titanium filler paste.



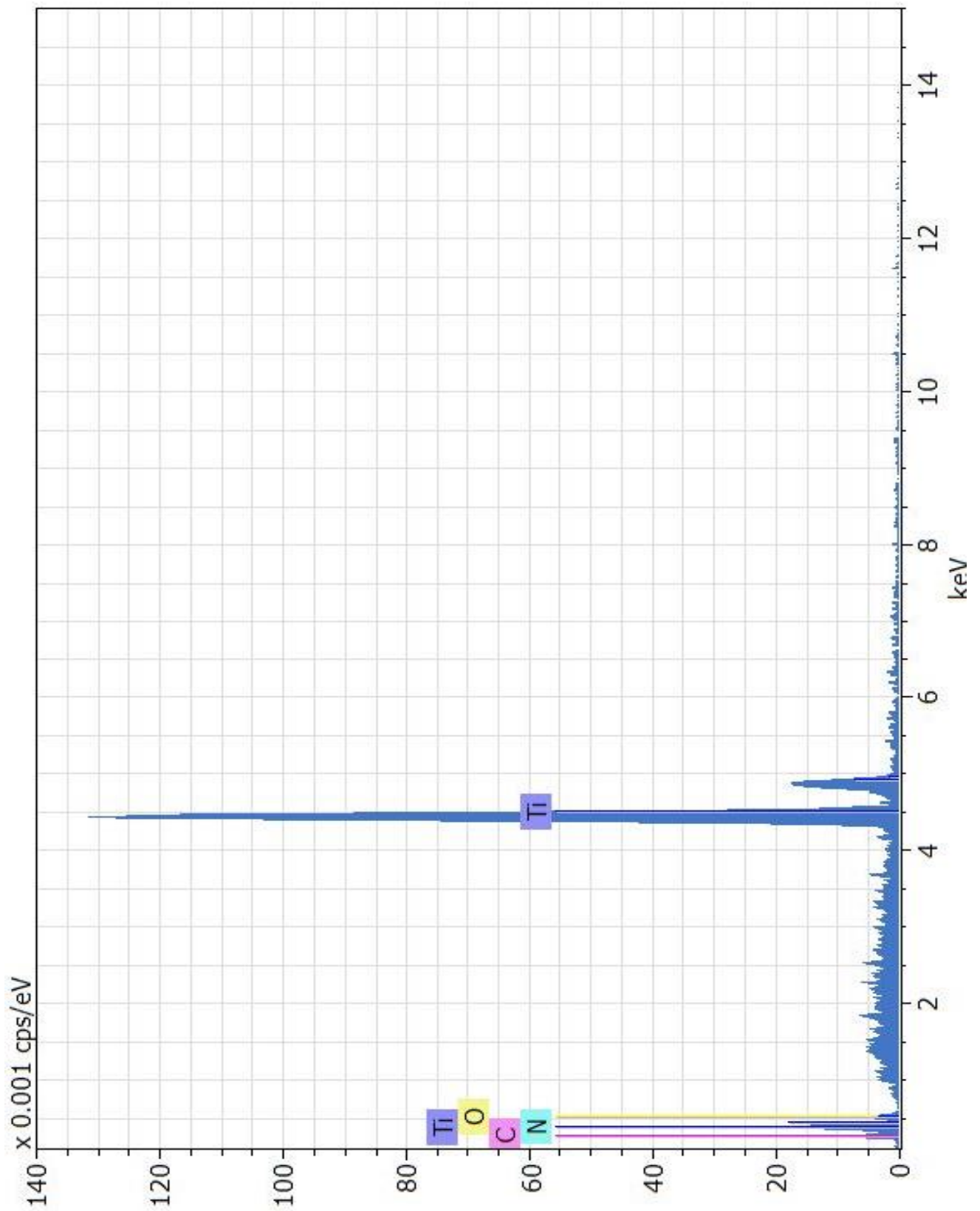
(a)



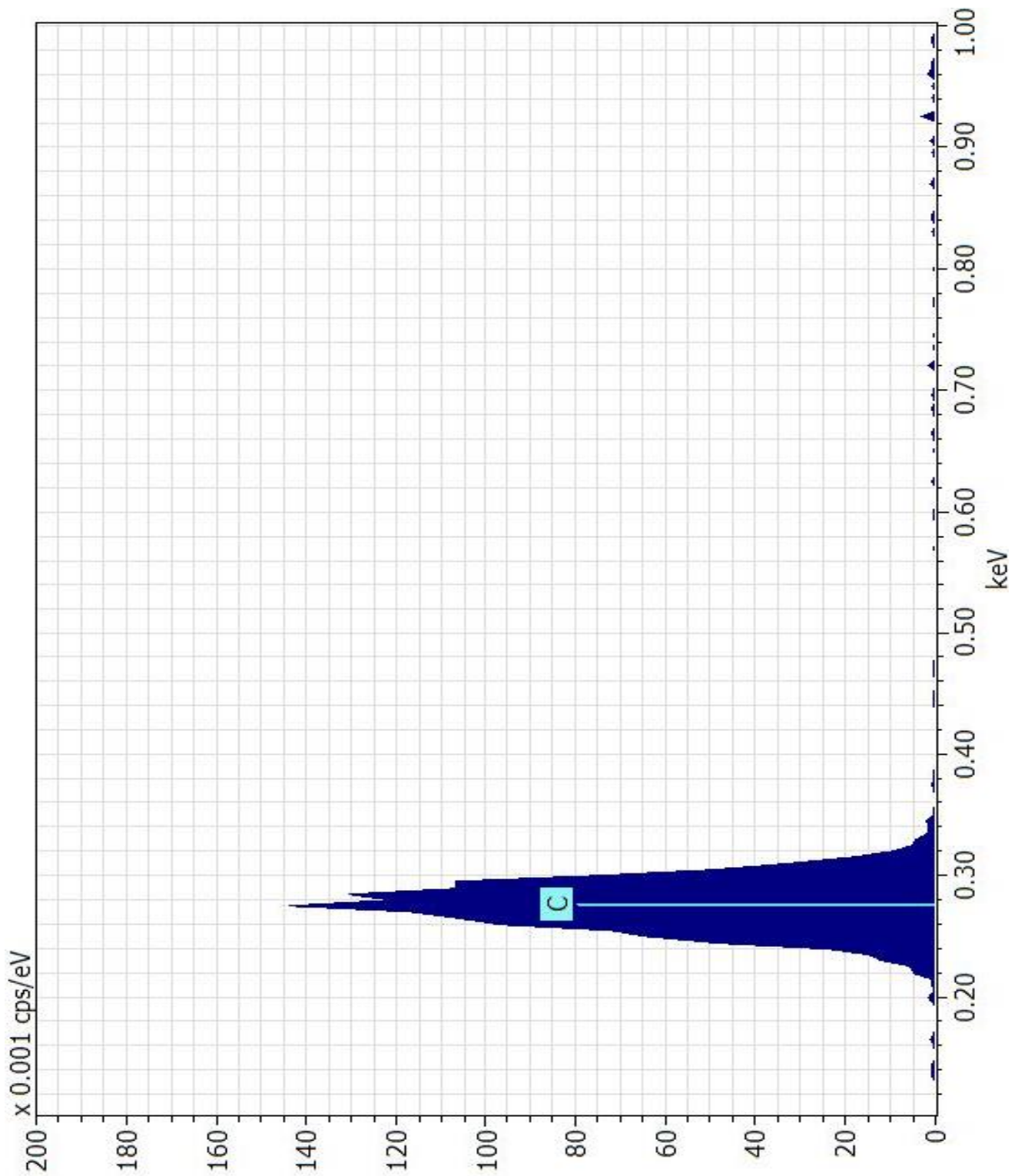
(b)

Figure 3.19: The scanning electron microscopy for surface of stainless steel after bonding with titanium filler paste.





(a)



(b)

Figure 3.20: The energy dispersive x-ray for the fractured surface.

The EDX technique was used to investigate the fracture surface component. Figure 3.20 presents the Edx chart, which indicates the existance of pieces and filler elements and their concentration.

### 3.8 Phase Identification

The x-ray diffraction patterns of fractured surfaces were revealed in figure 3.21 and figure 3.22. New structural phases were found at the intermediate layer graphite – stainless steel. These new phases were listed in table 3.2 The formation of new structural phases gives an indicates for the exsistance a chemical bonding between the braze pieces elements espically when the Ti elements was bonded together with brazing elements to gets the new phases listed in table 3.2. The formation of new components phases was found practically enhancement and improvment of the bonding strength

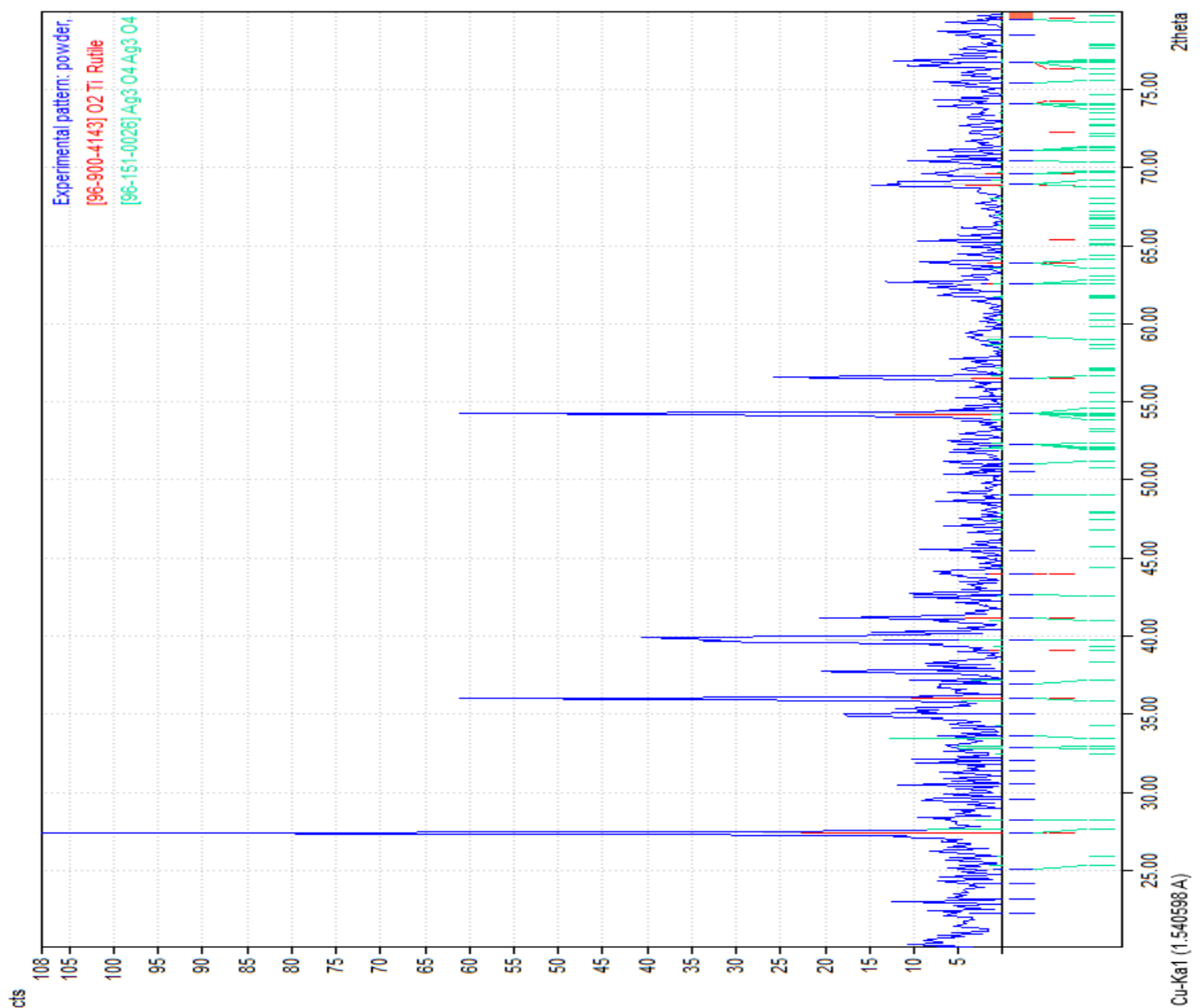


Figure 3.21: X-ray diffraction pattern of the fractured surface for the titanium – silver filler metal after bonding with stainless steel.



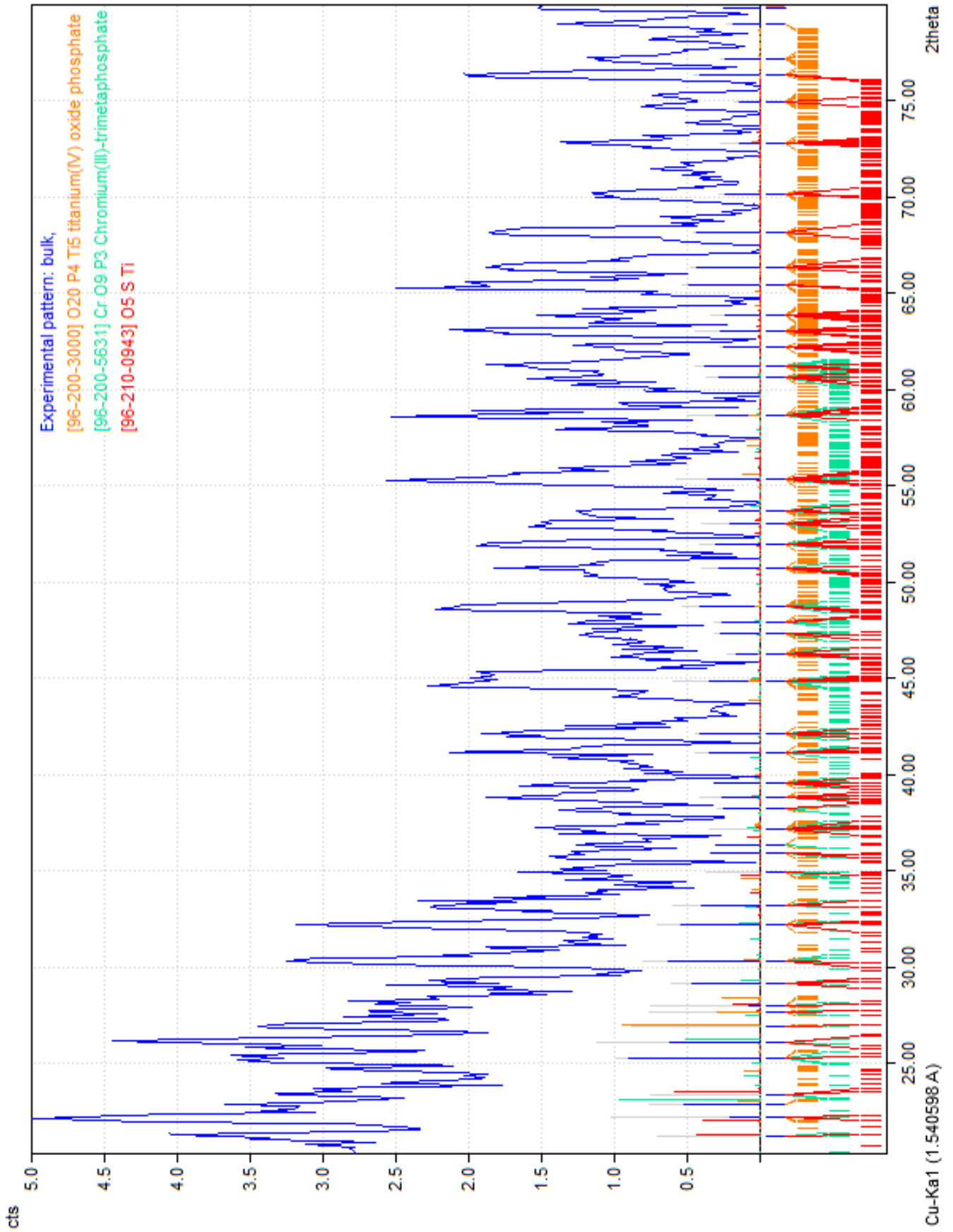


Figure 3.22: X-ray diffraction for stainless steel by using Ti-Ag filler metal.

Table 3.2 Illustrate the miller indices and 2 theta of phases.

Phases	Miller indices	2theta
$O_{20}P_4Ti_5$	412	37.1
$O_5STi$	218	36.8
$CrO_9P_3$	202	37.4
$TiO_2$	101	36
$Ag_3O_4$	012	34

### 3.9 Conclusions

The main conclusions can be summarized as follows:

- 1- The surface preparation of base materials is very important to obtain a reliable joint.
- 2- The wetting is one important factor in bonding process , the copper paste filler incapable to wet graphite surface so did not get bonding with copper filler paste.
- 3- The matching of thermal expansion of materials are necessary to avoid the separation of filler paste from base materials.
- 4- The small value of contact angle of titanium filler metal indicated to good wettability on the surface of stainless steel and graphite.
- 5- The joint bonding strength increases as concentration of titanium increases in filler paste.
- 6- When pure titanium in filler paste was used , the temperature of furnace should be 70% of melting point of titanium for enhanced wetting bonding between partners.
- 7- Maximum tensile strength was obtained with filler metal ( 75%Ti +25%Ag) 15Mpa at condition of furnace of high vacuum pressure should be selected, at temperature 900 and soaking time 15 min.

### 3.10 Suggestions for Future Work

- 1- For obtaining an efficient wetting & hence bonding between metal – graphite one can suggest using a furnace of high vacuum up to  $10^{-2}$  torr.
- 2- Several metals and alloys can be suggested for achieving bonding with graphite at a limited conditions.
- 3- Different filler constituents and form can be used to verify a reliable joint.
- 4- Roughness of graphite surfaces can be conducted to obtain multi shape behavior for increasing contact area.
- 5- Studying the fractured surfaces by using TEM technique for revealing the intermediate surface more obviously.

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## الخلاصة

يبين هذا العمل امكانية الربط بين معدن الكرافيت وسبيكة الفولاذ المقاوم للصدأ صنف 410 بأستعمال معجون حشوة معدنية مألثة. تم اختيار تقنية اللحام بالمونة لعدة اسباب منها, كفاءة وبارعة وسريعة واقتصادية. تم خلط مساحيق عالية النقاوة من الفضة والنحاس والتيتانيوم بنسب مختلفة لتحضير معجون الحشوة المألثة. تهيئة مكعبات من المواد المزعم ربطها وهي الكرافيت والفولاذ المقاوم للصدأ صنف 410 لدراسة كلا من التبليل والربط كدالة لدرجة حرارة الفرن. تم ازالة المواد الملوثة العالقة على سطوح المواد المزعم ربطها وطبقا لطرق قياسية. تم دراسة قابلية التبليل اولا وذلك من خلال وضع قطرة من معجون الحشوة بنسب مختلفة على سطوح كل من الكرافيت والفولاذ. حصلنا على افضل قابلية تبليل باستخدام حشوة مألثة من التيتانيوم عند درجة حرارة 950 م° وضغط فرن فراغي 10<sup>-2</sup> تور. تم اختبار نوعين من تصاميم الربط وهما النوع الطرفي والنوع التشابكي واعتمادا على متغيرات التجربة. تم وضع معجون الحشوة بين سطحي الكرافيت والفولاذ لغرض دراسة متانة الربط بينهما. انجزت عمليات الربط تحت ظروف تشغيلية مختلفة من درجة حرارة الفرن ( 850 , 900 , 950 , 1000 ) م° مع زمن اسقاء مقداره 15 دقيقة. تم الحصول على اعظم متانة ربط 15 ميكاباسكال عند استخدام حشوة مألثة (75Wt% Ti + 25Wt% Ag) بأستخدام التصميم التشابكي ثم فحص سطوح التكسر بتقنيات غير اتلافية تتضمن المجهر الضوئي والمجهر الالكتروني الماسح ومنظومة حيود الاشعة السينية ومنظومة تشتت الاشعة السينية. استخدمت تقنيات حيود الاشعة السينية لدراسة المنطقة البينية وقد اظهرت تكون اطوار تركيبية جديدة وهي

(TiO<sub>2</sub> and Ag<sub>3</sub>O<sub>4</sub>), (O<sub>20</sub>P<sub>4</sub>Ti<sub>5</sub>, O<sub>5</sub>STi and CrO<sub>9</sub>P<sub>3</sub>) وهذه الاطوار الجديدة قد عززت من

متانة ربط الكرافيت بالفولاذ المقاوم للصدأ.



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## تقييم السطح البيني لمنطقة الربط الكرافيت بالمعدن

رسالة

مقدمة الى كلية العلوم /جامعة النهرين  
وهي جزء من متطلبات نيل درجة الماجستير في الفيزياء

من قبل

مريم بديع عبود

بكالوريوس 2013

اشراف

أ.م.د. ثامر عبد الجبار جمعة

نيسان

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