

BRAZING OF GRAPHITE/GRAPHITE USING A NEW FILLER TECHNIQUE

Dr. ADNAN S. JABUR
Engineering College - University of Basrah

ABSTRACT

Brazing is one of the best methods of graphite to graphite or to metals joining. But the major problem associating the graphite brazing is the poor wetting by the conventional molten fillers. For this reason, scientists have produced a special filler metal based on active elements which interacts with graphite to form carbides. Also, recently another technique to overcome the wetting problem was introduced by H. Ohmura and T. Yoshida . It included inserting an intermediate layer of pure iron foil inside the copper filler. In the present work, another filler combination of Cu/ steel/Cu foils is proposed as new filler technique for graphite brazing. It was found that, it produced a succeeded joint with a good properties consisted of a columnar phase which resulted from the partially dissolution of iron in molten copper. Additionally, the increasing of brazing time caused reducing the thickness of the steel central layer and increasing the thickness of the columnar phase layers. The x-ray diffraction test developed that, the joints contained two carbide types, iron and copper free elements.

لحام المونة للكرافيت بالكرافيت باستخدام تقنية جديدة للتحمية

د. عدنان شمشي جبر
كلية الهندسة - جامعة البصرة

الخلاصة

لحام المونة احد افضل الطرق في وصل الكرافيت مع الكرافيت او مع المعادن. لكن المشكلة الرئيسية التي ترافقه هي قلة تبلل الكرافيت بالمحسوات المعدنية السائلة التقليدية. لذلك اتبع العلماء حشوة معدنية خاصة تتضمن عناصر ثقالة تتحد مع الكرافيت وتكون كريدات. ومؤخرا اقترح اومورا و يوشيدا تقنية اخرى لحاوم مشكلة التبلل. وتتضمن حشوة طبقة وسطى من الحديد النقي بشكل رقاقة داخل حشوة النحاس. تم في هذا البحث تجربة التركيب المكونة من طبقات (نحاس/فولاذ/نحاس) كحشوة مقترحة جديدة للحام الكرافيت. ولقد وجد بانها اتحت وصلة ناجحة ذات خواص جيدة تتكون من طور عمودي ناتج من التوبان الجزئي للحديد في النحاس السائل. اضافة لذلك فان زيادة زمن اللحام سببت نقصان سمك طبقة الفولاذ المركزية وزيادة سمك طبقات الطور العمودي. فحوصات جرد الانسجة السنية الطورت ان الوصلة تألف من نوعين من الكريدات وعنصري الحديد والنحاس بصورة حرة.

KEYWORDS: brazing, graphite, ceramic joining, wetting problem, columnar phase

INTRODUCTION

A prime difficulties associating the brazing of graphite to graphite and to common structural metals is that the poor wettability of graphite by conventional molten fillers such as the copper base type. In addition to the general requirements of good brazing alloys such as the few differences in coefficient of expansion between the graphite, the brazing alloys and the structural metals^[1-3].

R.G. Donly and G. M. Slaughter [4] developed filler alloys to obtain a good joint with acceptable properties. These alloys which contain strong carbide forming elements such as (Ti and Zr) were found to readily wet and flow on graphite. They appear to be useful in fabricating graphite assemblies for a wide variety of elevated

temperature applications, aerospace and nuclear reactors.

Another brazing technique for joining graphite to itself or to metals with conventional filler metals has been developed by H. Ohmura et al [5,6]. This technique is performed by placing pure iron foil inside the copper filler. During brazing, the columnar phase is formed at both graphite faces and grows towards the iron foil. As a result, both graphite base materials are united by the columnar phase through the iron foil.

This work aims to evaluate a lamellar filler of (copper-low carbon steel-copper) foils as a new technique in the efforts series to overcome the wetting problem accompanied with graphite brazing.

EXPERIMENTAL WORK

Materials used in this investigation were graphite as a substrate material, copper foil (50 μm thickness), and low carbon steel foil (200 μm thickness) as filler metals. Table (1) shows the chemical analysis of copper and steel foils.

Three joint types were employed to evaluate the present filler technique. They were: single butt joint (figure 1a), fillet butt joint (figure 2a), and double butt joint (figure 3a). The first type was utilized to study the microstructure, microhardness and x-ray diffraction test, while the other two types were used for shear testing.

The graphite was cut to the desired dimensions according to the joint design, then the two joint side surfaces were ground with (180, 500, 800, 1000, 1200) emery papers.

All joints were fixed by a simple fixture of two sheets of graphite to hold the joint pieces in between and tilted with four stainless steel bolts (figures 1b, 2b, and 3b).

To keep the brazing processes carrying out under inert atmosphere a stainless steel retort (figure 4) was employed to hold the brazing samples and fixtures.

After preparation of joints materials, they were assembled in the proper fixtures and inserted in the retort. Then it was sealed and the argon was pumped at 350°C until reaching the temperature of (1125°C), so the joints were held for different times (2.5, 5, 10, 15, 20, 30 min.), then the assemblies were left to cool in the furnace, and the argon closed at 400°C. Figure(5) illustrates the thermal cycles of these brazing processes.

Figure (6) shows the brazing joint samples of fillet and double butt types.

For evaluation the present filler technique, several inspections were performed in this work. They include a microstructural study with an optical microscope, x-ray diffraction, and shear tests which are divided into the following:

I) Fillet Butt Joints Shear Testing

They were inspected by single shear testing using the die shown in the figure (7). The assembly of joints with die was held down in the INSTRON compression machine to obtain load-displacement output charts with crosshead speed of 0.5 mm/min.

II) Double - Butt Joints Shear Testing

The double brazed shear tests were carried out using the die shown in figure (8). The assembly of the graphite joints with die was held down in the INSTRON compression machine at the same previous crosshead speed.

RESULTS AND DISCUSSION

I) Microstructural Evaluation

Figure (9) contains a micrograph of graphite/graphite joint brazed at 1125°C and holding time of 2.5 min. It shows an ideal joint consisting of three zones; graphite substrate, columnar phase, and steel center. In figure (10) other observations from the same joint conditions. They develop uneven thicknesses of columnar phase and steel layers in the same micrograph and in different micrographs. This inhomogeneity in layer thicknesses may be attributed to many factors such as; unequal pressure applied by four bolts, the change of pressure at elevated temperatures due to the loss occurring by expansion, and inhomogeneous distribution of temperature in the retort.

Figure(11) contains a combination micrographs of before brazing assembly (Graphite-Cu-steel-Cu-Graphite), and after brazing the (same joint). It shows a high reduction in the steel layer thickness and growth of columnar phase through all of the copper, graphite and part of steel layers.

The effect of brazing time on the phase change and interface movement of both filler and base materials is shown in figure (12). The steel layer reduced in thickness and the columnar phase initiated and expanded through the copper layers, graphite substrates and central steel layer with increasing time. So at 2.5 min. the large thickness of steel layer is clear, but at 30 min. of brazing time it has almost reduced. A very important observation is the aggression of the graphite which, is transferred into the columnar phase towards the iron. This aggression increased clearly with increasing time.

II) X-Ray Diffraction

X-ray diffraction tests were carried out for five specimens in different brazing times (2.5, 5, 10, 15, 30 min.) as shown in figure (13). The main deduction; is that, the predominant existence of two iron carbide phases (Fe_3C , FeC), which indicates a clear diffusion and interaction between the graphite and the iron atoms across the two liquid copper layers. Also, the charts assessed the presence of both free copper and iron in the joints at all brazing times except at 30 min., so they disappeared completely. The poor sensitivity of the instrument reduced an opportunity of identification of a columnar phase and intermetallic compounds peaks in the charts.

III) Shear Test Results:

a- Single Butt Fillet Shear Testing:

The values of single butt fillet shear strengths are plotted versus the brazing time in figure(14). It shows a fluctuation in this relation, that cannot develop a clear role of brazing time in maximum shear strength. Figure (15) includes photographs of the fractured (after testing) samples. It shows that all six samples were fractured through the graphite substrate which means some rotation effect resulted from the moment which initiated due to unbalanced sample design. Many attempts were done to avoid this problem such as cutting the upper edge and make the distance between the upper edge and the joint region only (3 mm) and (2 mm) but the same results were obtained. For this reason the usage of double brazed shear tests to have reliable tests and results became required.

b- Double Butt Shear Testing

Figure(16) develops good reliability and the effect of time was obvious. The joint shear strength increased with increasing the brazing time from 2.5min. until 15min. This can be due to the increasing of diffusion and interaction processes with time advance. The fracture behavior supports this explanation since it occurred totally through the joint as shown in figure (17). After 15 min., the shear strength began to reduce gradually until 30 min., so this descent was attributed to the growth of brittle carbide phases and disappearing of the tough copper and iron concurrently. This explanation is supported by the transition of the fracture style more towards the graphite substrate.

IV) Microhardness test

The results of microhardness tests are shown in figure (18). Generally there are no big difference between the curves of different brazing times. The maximum hardness was obtained at the center of the joints and it decreased gradually through the columnar phase until the graphite substrates which have the minimum values.

CONCLUSIONS

- 1-The present filler technique succeeded in producing an excellent joint.
- 2-The liquid copper played as a path to the carbon atoms to move towards the steel center and formation of iron carbides.
- 3-The best brazing time was (15min.) at 1125°C o brazing temperature, which gave a maximum shear strength.
- 4-The maximum hardness was at the center of the joints.

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Table (1) The chemical composition of copper and steel foils

	Al	As	Bi	Ni	Pb	Zn
Copper Foil	0.034	0.005	0.006	0.023	0.073	0.045
	C	Si	S	P	Ni	Cu
Lead Steel	0.09	0.0049	0.0163	0.0173	0.0086	0.0021

*Balance are Cu and Fe

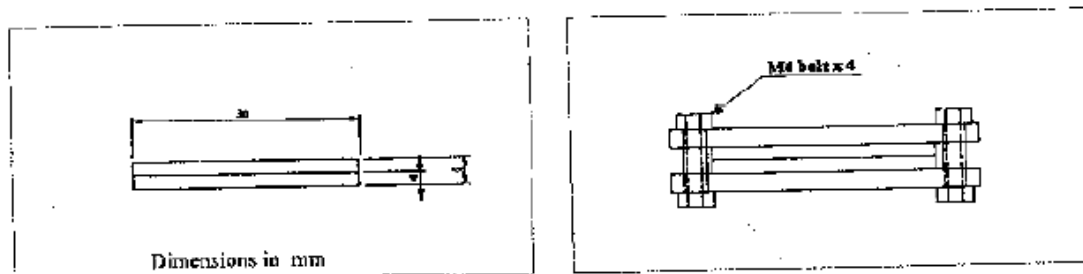


Figure (1) a- The joint design and dimensions of single butt joint b- The fixture of the single butt joint

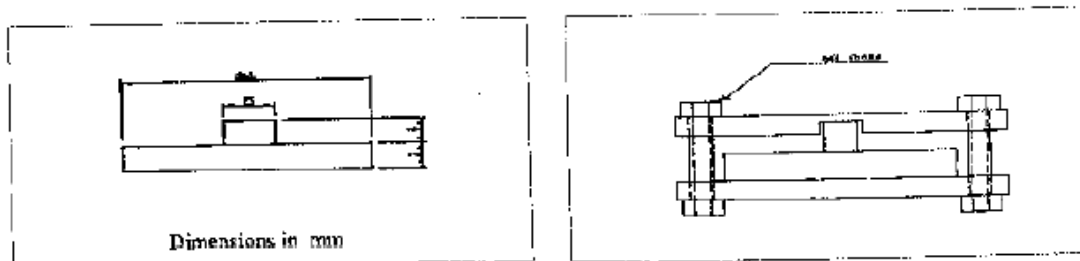


Figure (2) a - The joint design and dimensions of fillet butt joint b- the fixture of the fillet butt joint

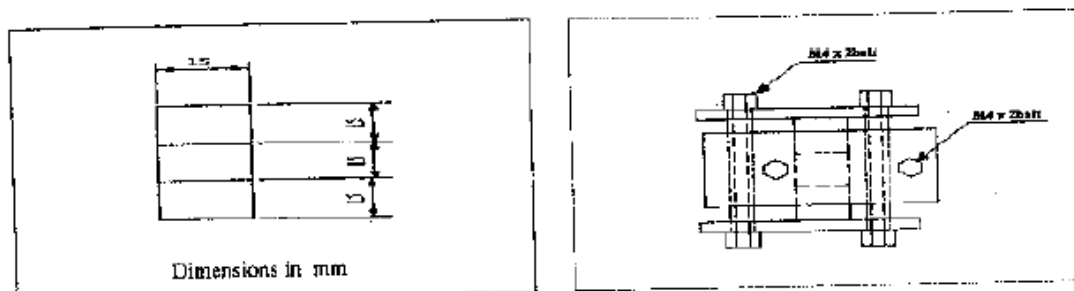


Figure (3) a- The joint design and the dimensions of double butt joint b- The fixture of the double butt joint

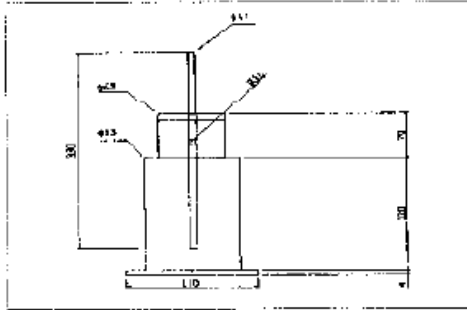


Figure (4) the retort which used in brazing processes

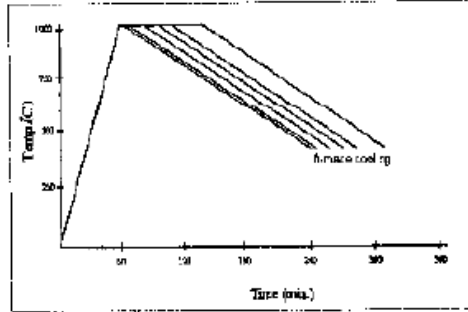


Figure (5) Thermal cycles of brazing processes

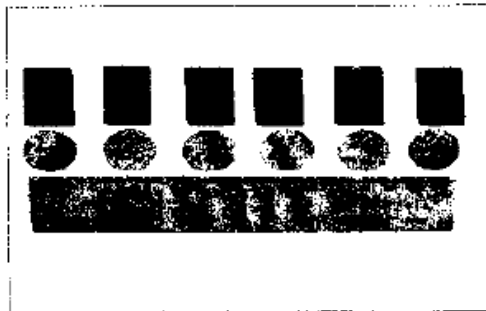
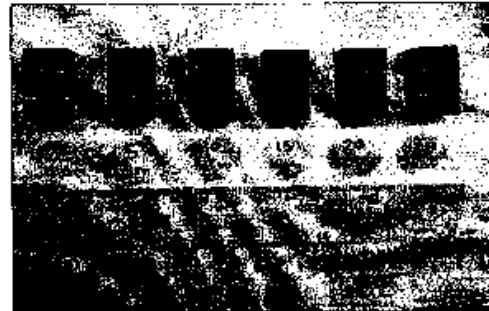


Figure (6) a-A photograph of brazed fillet butt joint samples



b-A photograph of brazed double butt joint samples



Figure (7) A die of fillet butt joint shear testing

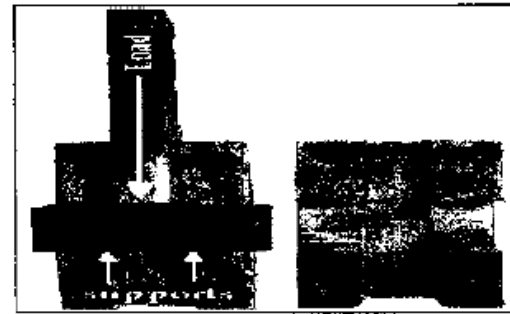


Figure (8) A die of double butt joint shear testing

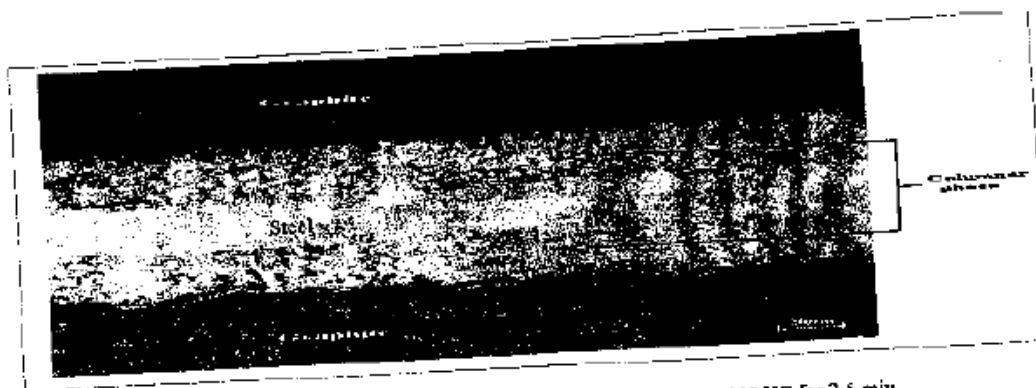


Figure (9) A microstructure of the joint at brazing conditions ; 1125°C for 2.5 min.

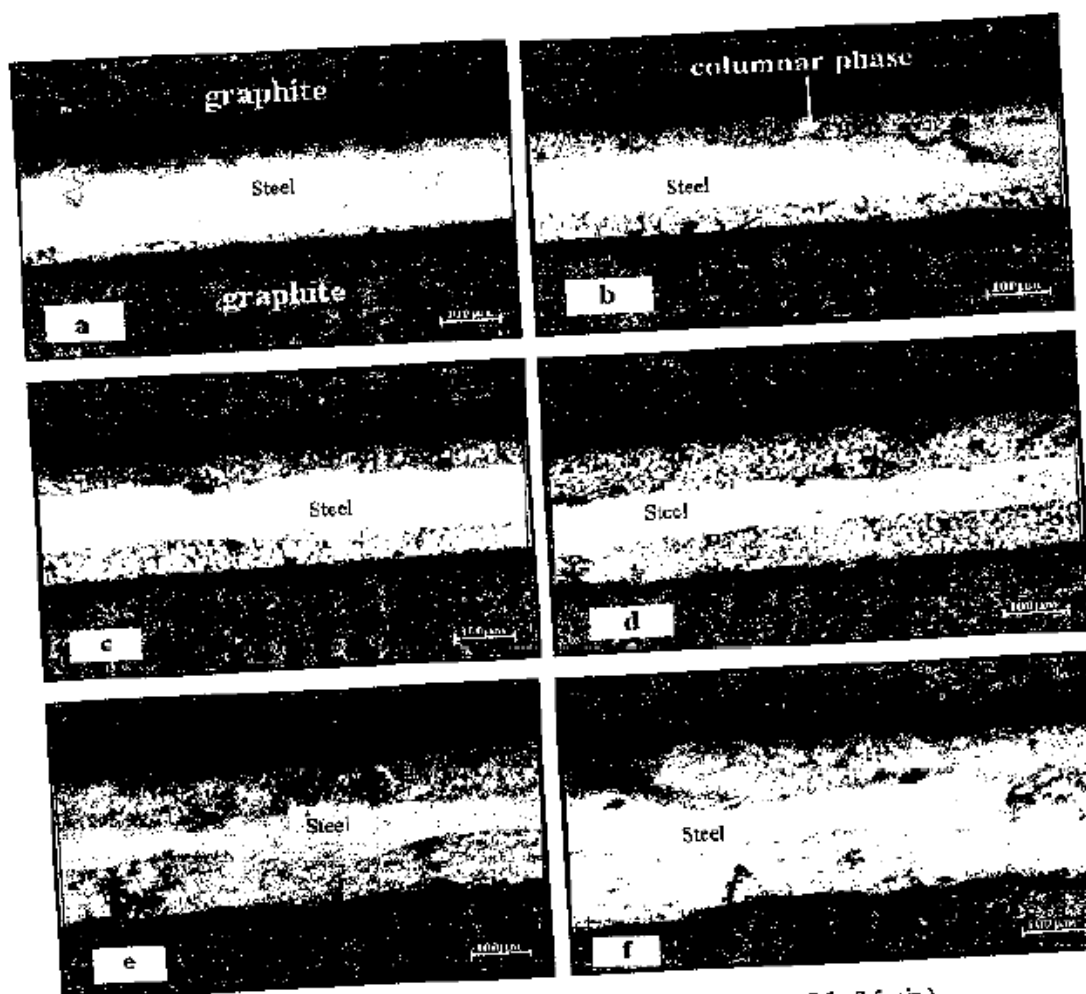


Figure (10) Microstructure of joints of same brazing conditions; (1125°C for 2.5 min.)



Figure (11) The variation in microstructure before and after brazing

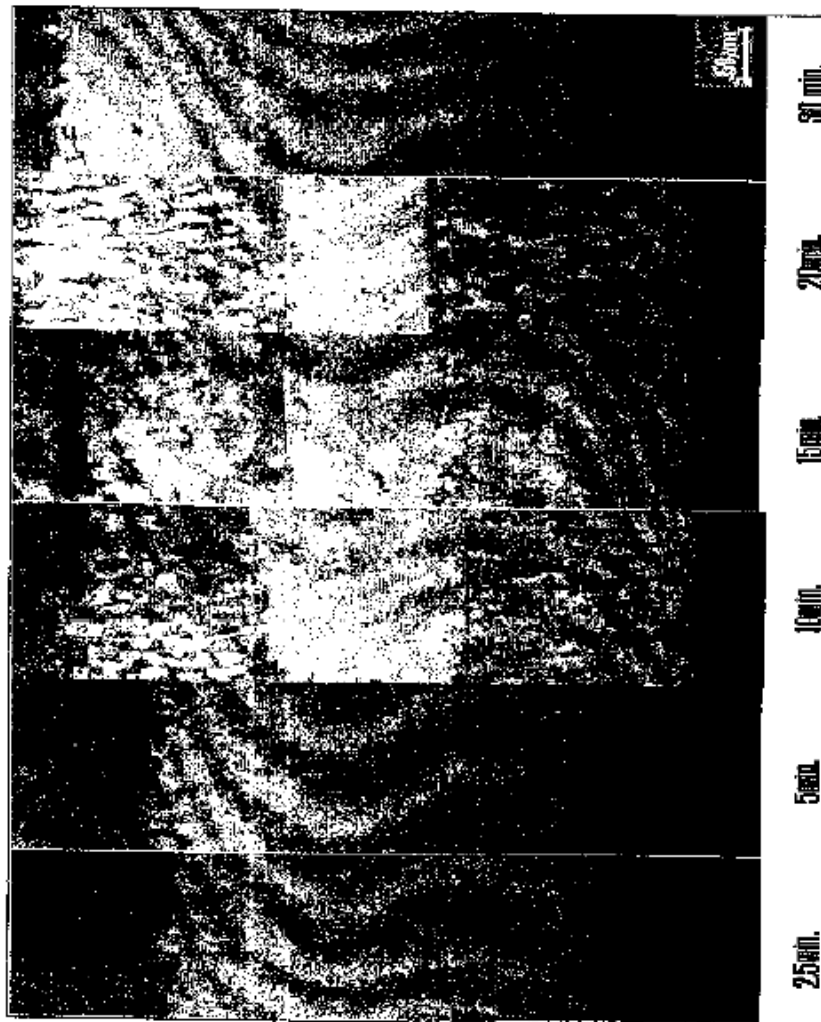
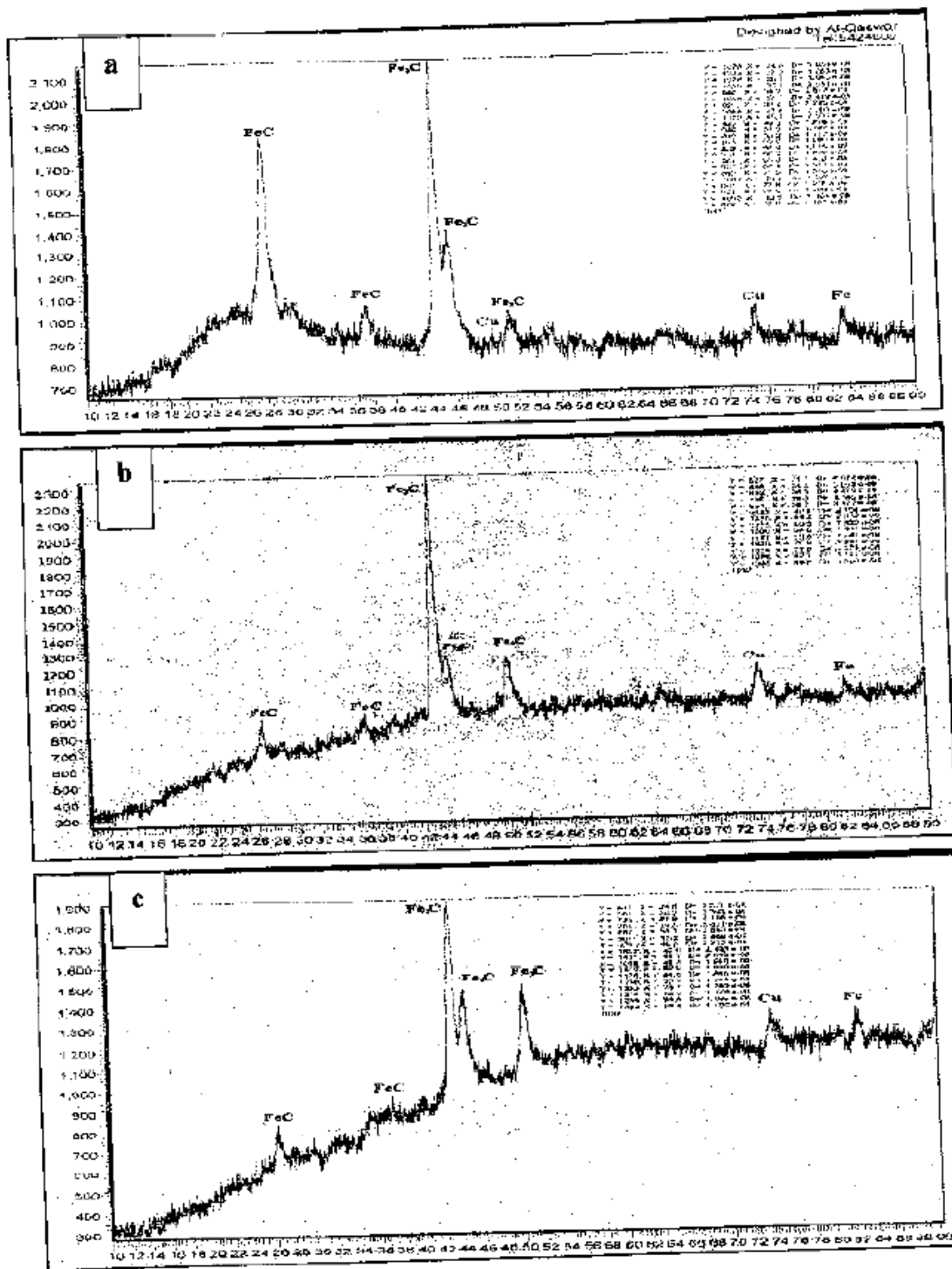
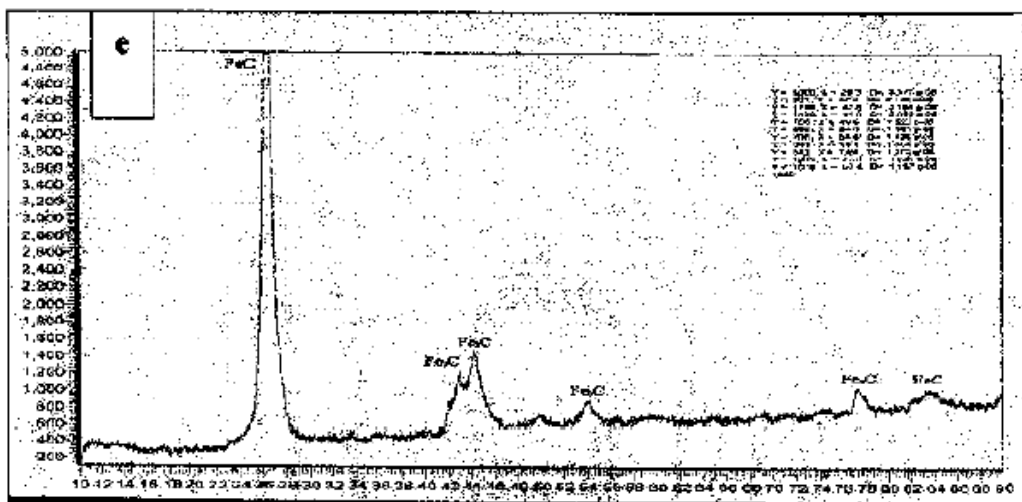
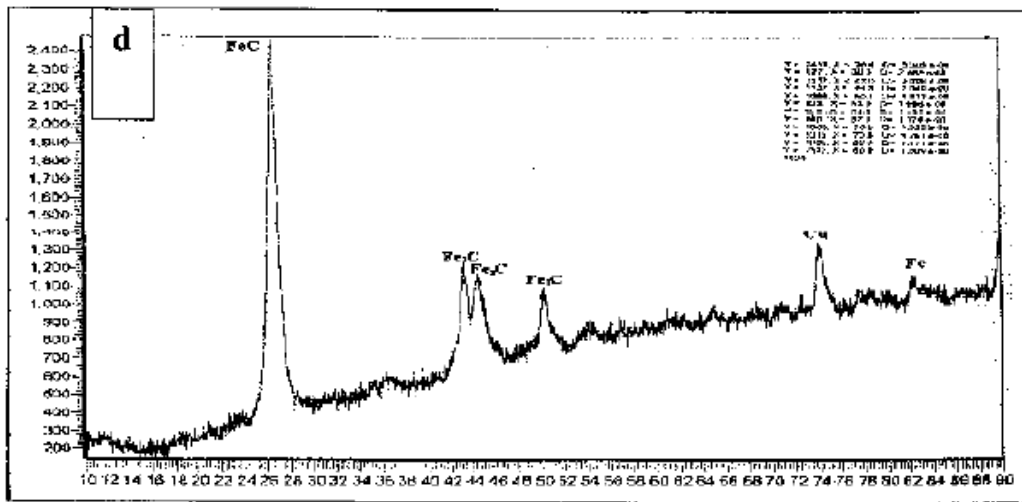


Figure (12) The effect of brazing time on the joint microstructure



Figure(13) The charts of x-ray diffraction charts of brazing samples at different brazing times; a) 2.5 min. b) 5 min. c) 10min. d) 15min. e) 30min.



Continuous of previous figure (13)

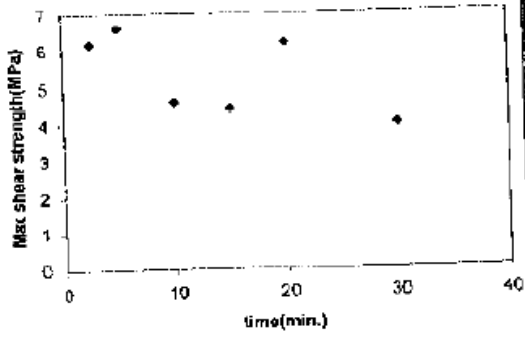


Figure (14) The effect of brazing time on max. shear strength of single butt fillet joints



Figure (15) A photograph of fractured single butt fillet joints

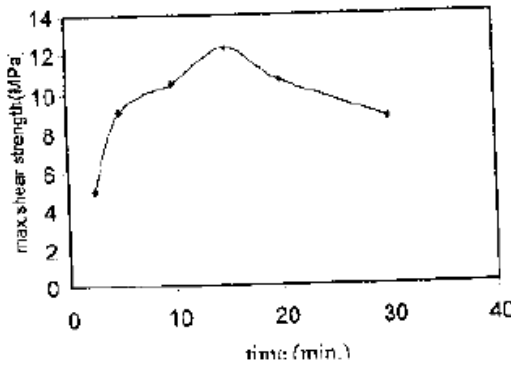


Figure (16) The effect of brazing time on max. shear strength of double butt joints

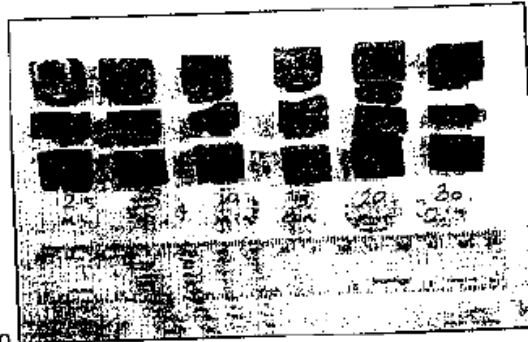


Figure (17) A photograph of double butt joints

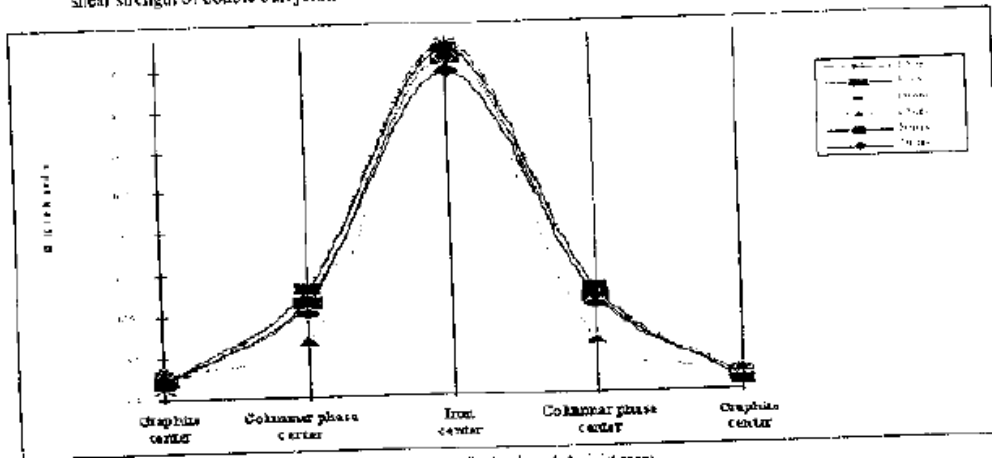


Figure (18) Hardness distribution through the joint zones