Alumina-to-Alumina Actively Brazed Using Cu-Ti, Cu-Zr, and Eutectic Ag-Cu-Ti Filler-Metal Alloys

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Abstract

Al_2O_3-Al_2O_3 brazing systems were produced using a one stage actively brazed technique based on Cu-Ti, Cu-Zr, and Ag-Cu-Ti alloys. Single and double butt joints were used for micro-structural and mechanical properties studies respectively. The joints that brazed by using Cu-Zr filler-metal alloys (2% , 4% , 6% and 8% Zr weight percent) have been showed low shear strengths at the ZrO_2 interface. Higher shear strength were obtained by using Cu-Ti filler-metal alloys (2% , 4% , 6% and 8% Ti weight percent), and eutectic (Ag-26%Cu-4% Ti). As judged by the phases formed at the interface, $Cu_2(AlTi)_4O$ is more effective to wet and both alumina to alumina.

لحام المونة الفعالة للالومينا/الالومينا باستخدام حشوات المونة Cu-Ti ، Cu-Zr وسبيكة اليوتكتيك Ag-Cu-Ti .

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الخلاصة

وصلات لحام من الالومينا/الومينا تم الحصول عليها بواسطة استخدام طريقة اللحام من نوع المونة الفعالة بخطوة واحدة دون الحاجة الى طرق اضافية والتي تعتمد على استخدام حشوات المونة Cu-Zr، Cu-Ti، Ag-Cu-Ti. تم لحام وصلات تناكبية احادية وثنائية لدراسة التراكيب المجهرية والخواص الميكانيكية على التوالي. اظهرت الوصلات الملحومة باستخدام نظام ال Cu-Zr (2%،4%،6%،8% نسب وزنية من الزركونيوم) متانة قص واطئة عند الحد الفاصل الحاوي على ZrO_2 ، بينما

اظهر نظام من نوع Cu-Ti (%2،%4،%6،%8 نسب وزنية من التيتانيوم) متانة عالية يعول عليها في الربط، وكذلك اظهر ذلك النظام الايوتكتك Ag-Cu-Ti حيث تم اعتماد الاطوار عند الحد الفاصل بين الالومينا ومنطقة الربط والطور السائد هو $Cu_2(AlTi)_4O$.

Introduction

The development of a new techniques to joint the structural ceramics have the potential of structure significantly importing the advanced ceramic industry. The most commonly used method is the moly-manganese process[1-3]. This process is well-established and produced highly reliable joints between ceramics and alloys. However, the moly-manganese process which requires two processing steps of metalization the ceramic and brazing is a time consuming. Furthermore, the metalization process is conducted at very high temperature $(\sim 1500^\circ C)$. In addition, joint properties are sensitive to process variables and hence, precise process control is required to obtain the reliable joints, as a result, this process is very expensive.

High shear strength direct brazing between ceramic and ceramic or metal is the focus of studies in an effort to select the successful active filler-metals[4-10]. Because this is a one-step process, it is simple and economical. Although active metal brazing has been investigated since 1940, it has not been widely accepted because of inconsistent joint properties[11].

There are several methods of the active metal brazing one method involves a sheet of titanium (the most commonly used active element) that can be cladded by two sheets of the conventional brazing metal[6]. Another technique uses titanium hydride powder mixed with powders of conventional brazing metals[5]. The most economical method utilizes a filler-metal where an active element forms true alloy with the base filler-metal[11].

This work concentrates on the microstructural and phases formed through the interface between the alumina and filler metal alloy. The research has also focused on the selection and design the brazing cycles and shear test.

Experimental Procedures

Alumina substrates of cubic shape (14 x 14 x 10 mm^3) were used for brazing. The contact surfaces were polished with a 1200 silicon carbide paper then cleaned ultrasonically in acetone. After cleaning, 0.15g of the brazing paste was painted onto the surface to a thickness of ≈0.1 mm. The alumina joints were assembled as shown in Fig. 1, and placed in the vacuum furnace. The brazing temperatures were 1100°C and 980°C for Cu-Ti, Cu-Zr, and the eutectic filler-metal alloy respectively. The samples were held at the brazing temperature for 5, 10, 15, 30, 60 and 90 minuts. The furance was evacuated to the pressure of $5x10^{-5}$ torr before heating up to the brazing temperature.

The transverse sections of brazed samples were examined by optical and scanning electron microscopies, with and without prior grinding and polishing, Fig. 2 shows a typical transverse section of brazed samples. There will show the general features of bonding. X-ray diffraction analysis were performed to identify the reaction product the ceramic/filler interface by the Philips PW 1050 diffractometer from (Fig. 3). The shear test provides important information on the mechanical properties of the joint, namely shear strength. In this investigation the double brazed shear test DBS[11] is a new type of the shear test. The basic principle of the DBS test is illustrated schematically in Fig. 4, using the test apparatus shown in Fig. 5.

Experimental Results and Discussion

Heating Procedures

The main objective of actively brazed ceramics is to produce a joint, essentially wetted by the filler-metal alloys by careful selection of heating cycle to produce the suitable interfacial reactions. It was observed that the heating procedure is an important parameter under vacuum atmosphere as shown in Figs. 6 and 7. By using the common procedure under the argon atmosphere, the heating cycle was about 10°C/min and the holding time at the brazing temperature rate was about 90 min[11]. This means long time for volatilisation of brazing element such as copper or silver will take place by using vacuum atmosphere. The effective procedures for brazing are consistent with fast heating rate of about 40°C/min or even higher, at a shortest souking time of about 10 min. The main goal of these procedures is to maintain the brazing element in vacuum atmosphere to enhance the integral bond between the alumina and the active brazing alloy (ABA).

Bonding Mechanism

It was observed from the transverse sections that extensive reaction between ceramic and the filler metal alloys which is characterize areas by of light and dark regions as shown in Fig. 8 for Cu-Ti filler-metal alloy and Fig. 9 for eutectic filler-metal alloy. They are clearly observed that these joints contain two phases (the region between the two alumina pieces). The first phase is the interface and the second phase which is at the middle region in between the two interfaces. The second phase may call as lace-work phase which means that this phase will supply the first phase by the active bonding element (Fig. 10). Figures 11 and 12 compose the x-ray diffraction for the actively brazed Cu-Zr and Cu-Ti. The main phase in both layers is the phase I corresponding to ZrO_2 for Cu-Zr brazed and $Cu_2(AlTi)_4O$ for Cu-Ti. It was found that the low shear strength is due to the formation of ZrO_2 phase in Cu-Zr system. On the other hand, the formation of $Cu_2(AlTi)_4O$ resulted high shear strength. The present results obtained from phases analysis can be used effectively to proposed the bonding mechanism for actively brazed ceramic-to-ceramic as shown schematically in Fig. 13.

Shear Strength

The shear strength of the effective brazed ceramics as evaluated from the double butt joints for different types of filler-metals (Fig. 14) are in the range of 15 to 42 MPa (Table 1) for Cu-Ti and eutectic Ag-Cu-Ti filler-metal alloys, while it is less than 2.5 Mpa for Cu-Zr filler-metal alloys. The load-displacement for the fractured specimens (Fig. 15) is shown in Fig. 16. From these results it can say that the CuZr filler-metal alloys was formed a bad bonding mechanism because of the interface reaction created zirconia. It was not bonded with the matrix due to residual stresses formed from high to low transformation (t to m). Therefore, brittle weak bond resulted from Cu-Zr filler-metal system when joining the ceramics. While Cu-Ti and Ag-Cu-Ti filler-metal alloys showed good shear strength due to interface structure of $Cu_2(AlTi)_4O$ bonded with the matrix.

Conclusions

1- The heating cycle of joining alumina-to-alumina by brazing process is an important point to be selected and controlled to obtain high performance joining strength.

2- The actively brazed alumina-to-alumina using Cu-Zr paste filler-metal alloys consisted primarily of m-ZrO_2 phase which was formed via a diffusion process with low shear strength.

3- The formation of $Cu_2(AlTi)_4O$ phase developed during actively brazed using both Cu-Ti and Ag-Cu-Ti filler-metal alloys were resulted in improved of shear strength of the alumina-to-alumina joints.

4- The phases formed at the interfaces of ceramics are due to dissolve of the filler elements to react with the ceramic elements to form a new phases.

References

1- H. Pulfrich, "Ceramic to Metal Seals", U.S. Pat. Nos. 2163407 and 2163410, June, 1939.

2- H.J. Nolte and R.F. Spurch, "Metallizing and Ceramic Sealing with Manganese", U.S. Pat. Nos. 2667432 and 2667427, Jan., 1954.

3- A.J. Chick and L.J. Speck, "Fabrication of Metal-to-Ceramic Seals", U.S. Pat. No. 2708787, May, 1955.

4- C.S. Pearsall, "New Brazing Methods for Joining Non-Metallic Material to Metalls", Mater. Methods, 30, 61-62 (1942).

5- F.C. Kelly, "Metallizing and Bonding Non-Metallic Bodies", U.S. Pat. No. 2570248, Oct. 9, 1955.

6- H. Mizuhara, "Process of Making a Composite Brazing Alloy of Titanium, Copper and Nickel", U.S. Pat. No. 3561099, Feb. 9, 1971.

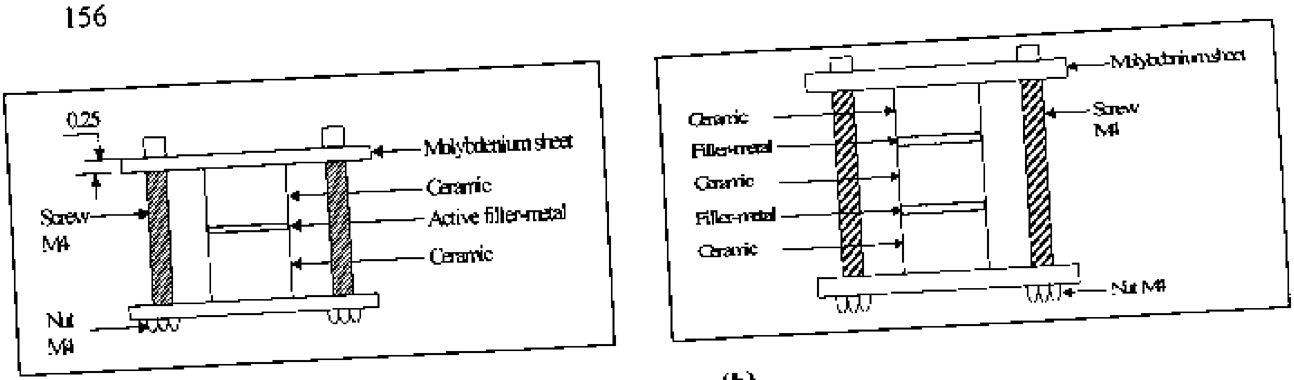
7- H. Mizuhara and K. Mally, "Ceramic-to-Metal Joining with Active Brazing Metal", Weld. J., 64 (10) 27-32 (1985).

8- E. Lugscheider, H. Krappits, and H. Mizuhara, "Joining of Non-Metallized Ceramic with Metals by Insert of Ductile Active Brazing", presented at the Second International Colloquium, Joining of Ceramic, Glass, and Metals. Bad Nauheim, FRG, 1985.

9- A.J. Moorhead and H. Keating, "Direct Brazing of Ceramics for Advanced Heavy-Duty Diesels", Weld. J., 65 (10) 17-31 (1986).

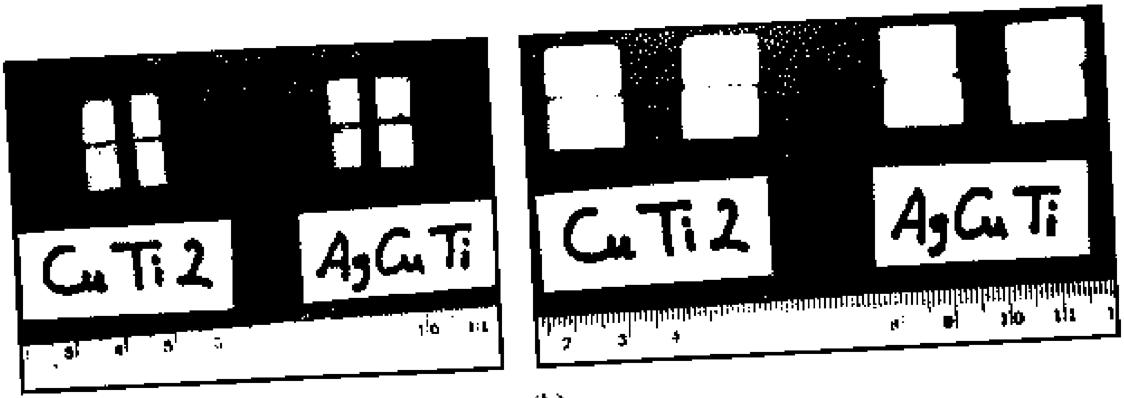
10- H. Mizuhara and E. Huebel. "Joining Ceramic to Metal with Ductile Filler Metal", Weld. J., 65 (10) 43-51 (1986).

11- H. Mizuhara, E. Huebel, and T. Oyama, "High-Reliability Joining of Ceramic to Metal", Ceramic Bulletin, 68 (9) 1591-1599 (1989).



(a) (b)

Fig. 1 Fixtures for (a) single butt joint and (b) double butt joint .



(a) (b)

Fig. 2 Two samples of ceramic–to–ceramic single-butt joint actively brazed using CuTi2 and eutectic AgCuTi as a paste filler-metal, (a) the two samples were cut by diamond wheel perpendicular to the filler-metal cross-section and (b) showing the interface line.

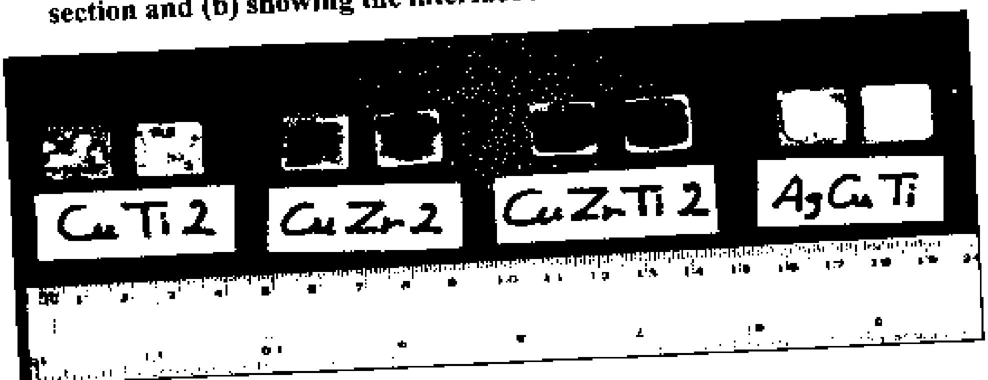


Fig. 3 Photograph shows different types of single-butt joints, using different active filler-metal alloys. CuTi2 and CuZr2 brazed using true filler-metal, while CuZrTi2 and Ag-Cu-Ti were brazed using paste filler-metals.

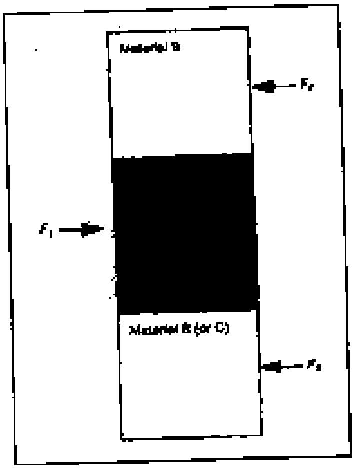
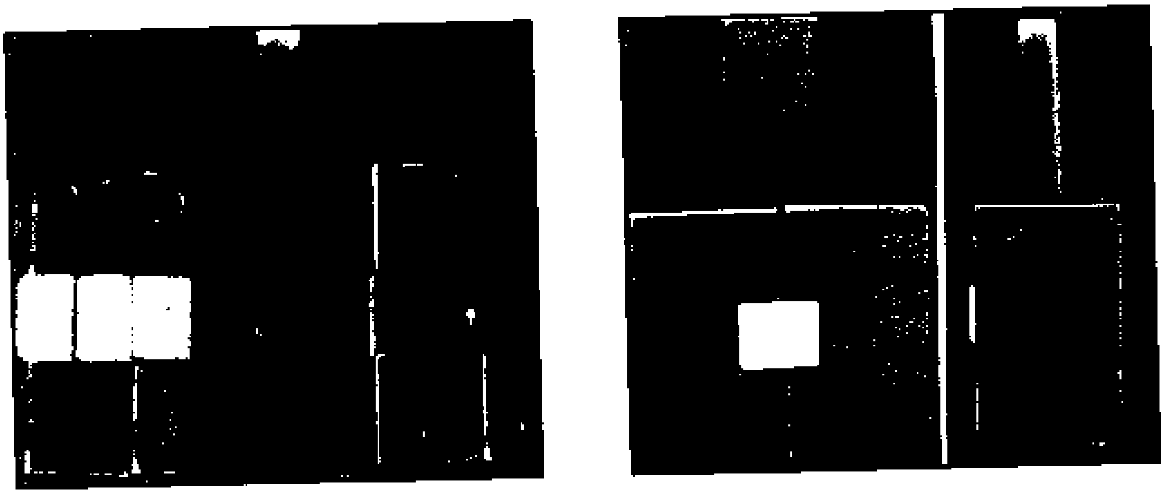


Fig.4 Schematic illustration of the double brazed shear test.[11]



(a) (b) (c)

Fig. 5 Fixture used for double brazed shear (DBS) test.
(a) Fixture opening, (b) Fixture assembly front view, and
(c) Fixture assembly side view.

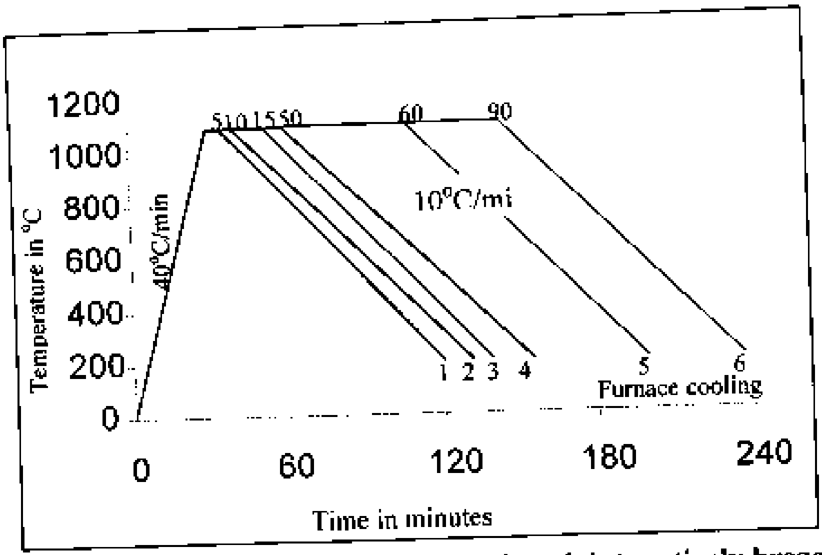


Fig. 6 Heating procedure for double butt joints actively brazed by using paste filler metal type CuTi2.

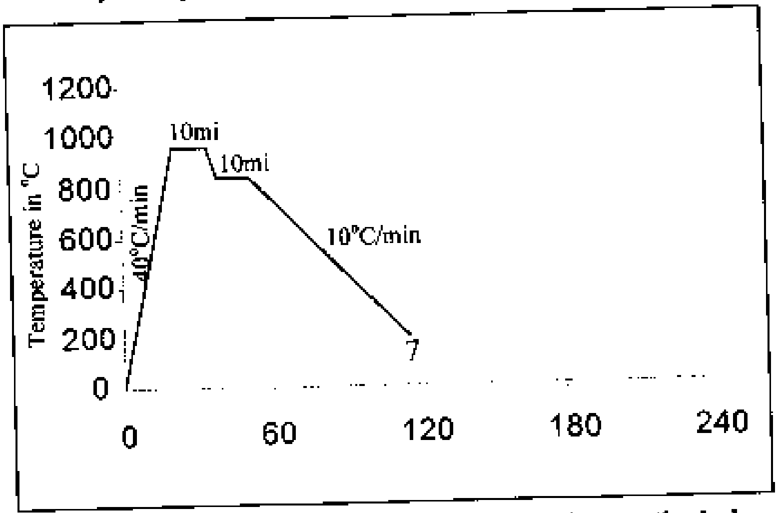


Fig. 7 Heating procedure for double butt joints actively brazed by using paste filler metal type AgCuTi.

Fig. 8 Bonding line showing ceramic-to-ceramic bonded by active brazing filler-metal alloy type CuTi2, showing two phases as remarkable. Phase I represents the interfering and phase II represents the matrix phase, X280.

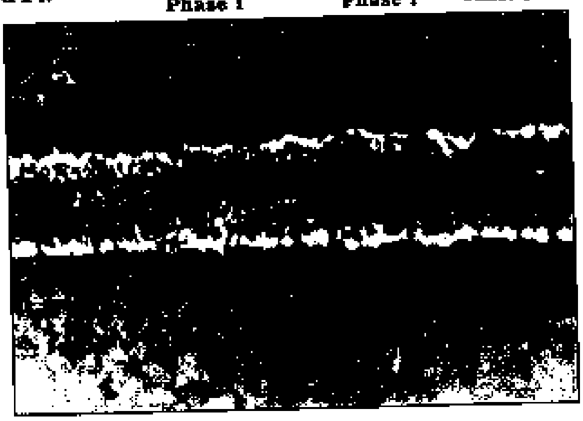


Fig. 9 Bonding line showing ceramic-to-ceramic bonded by active brazing filler-metal alloy type AgCuTi. It shows also two phases, Phase I represents the interfering phase and Phase II represents the matrix phase, X280.

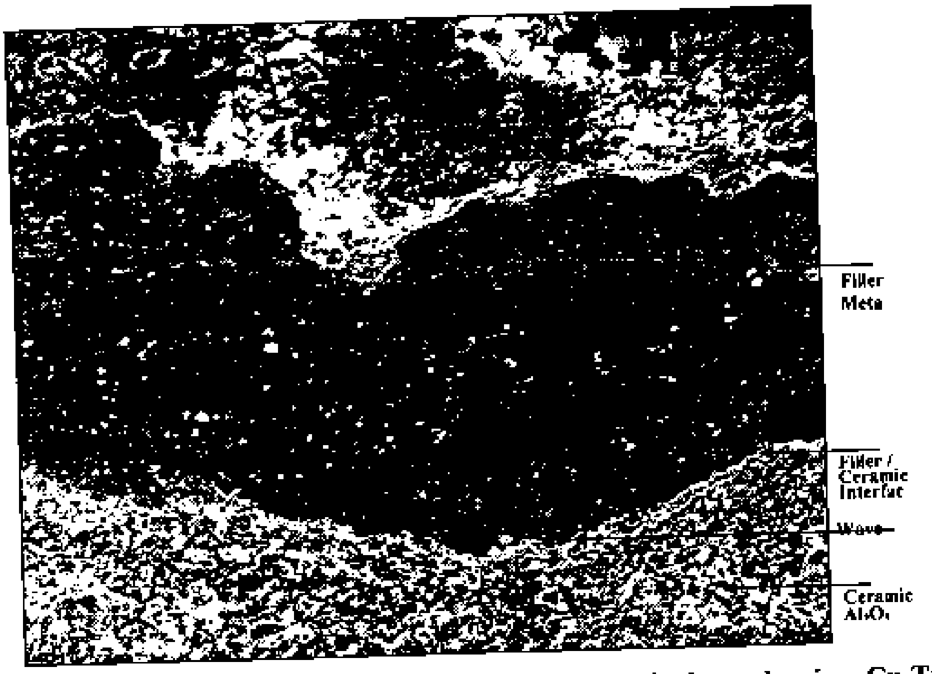


Fig. 10 SEM photograph for ceramic-to-ceramic actively brazed using Cu-Ti paste filler metal alloy showing interfering and waving between filler and ceramic. X300

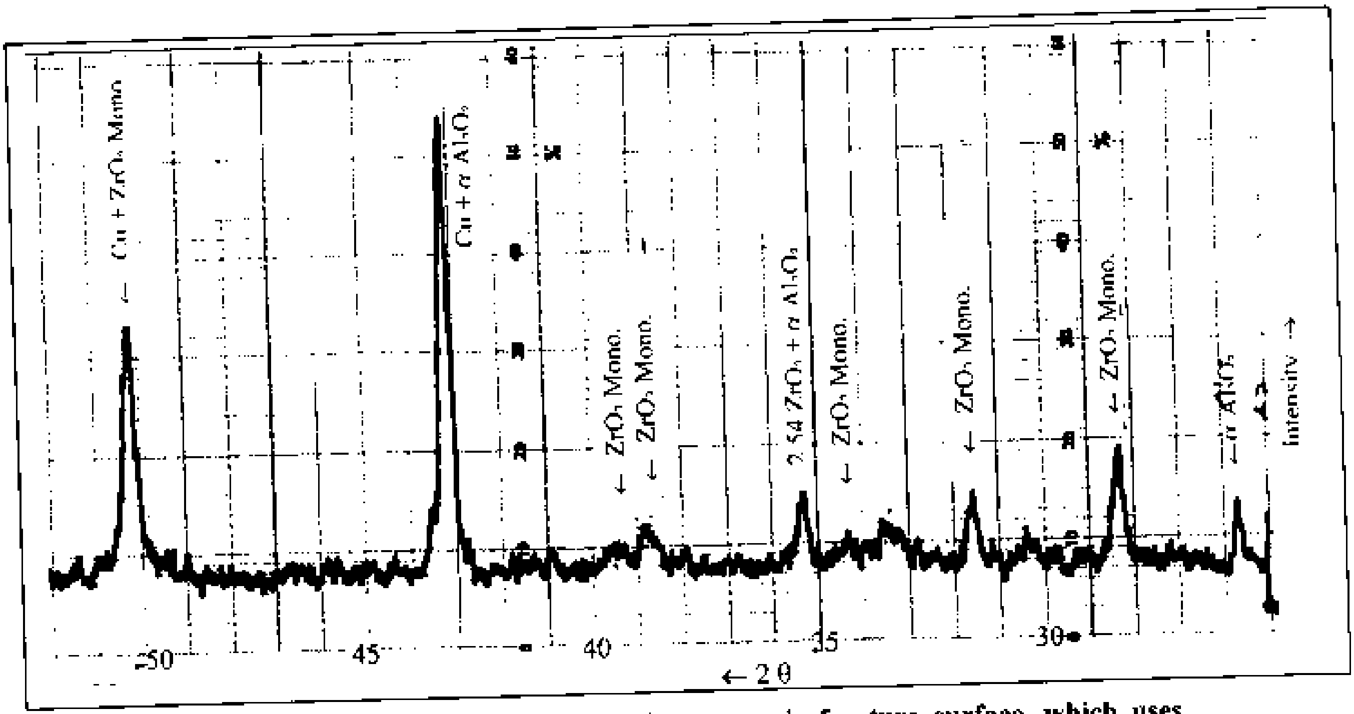


Fig. 11 The XRD pattern of ceramic-to-ceramic fracture surface, which uses CuZr2 as active paste filler-metal, using heating procedure regime No.2

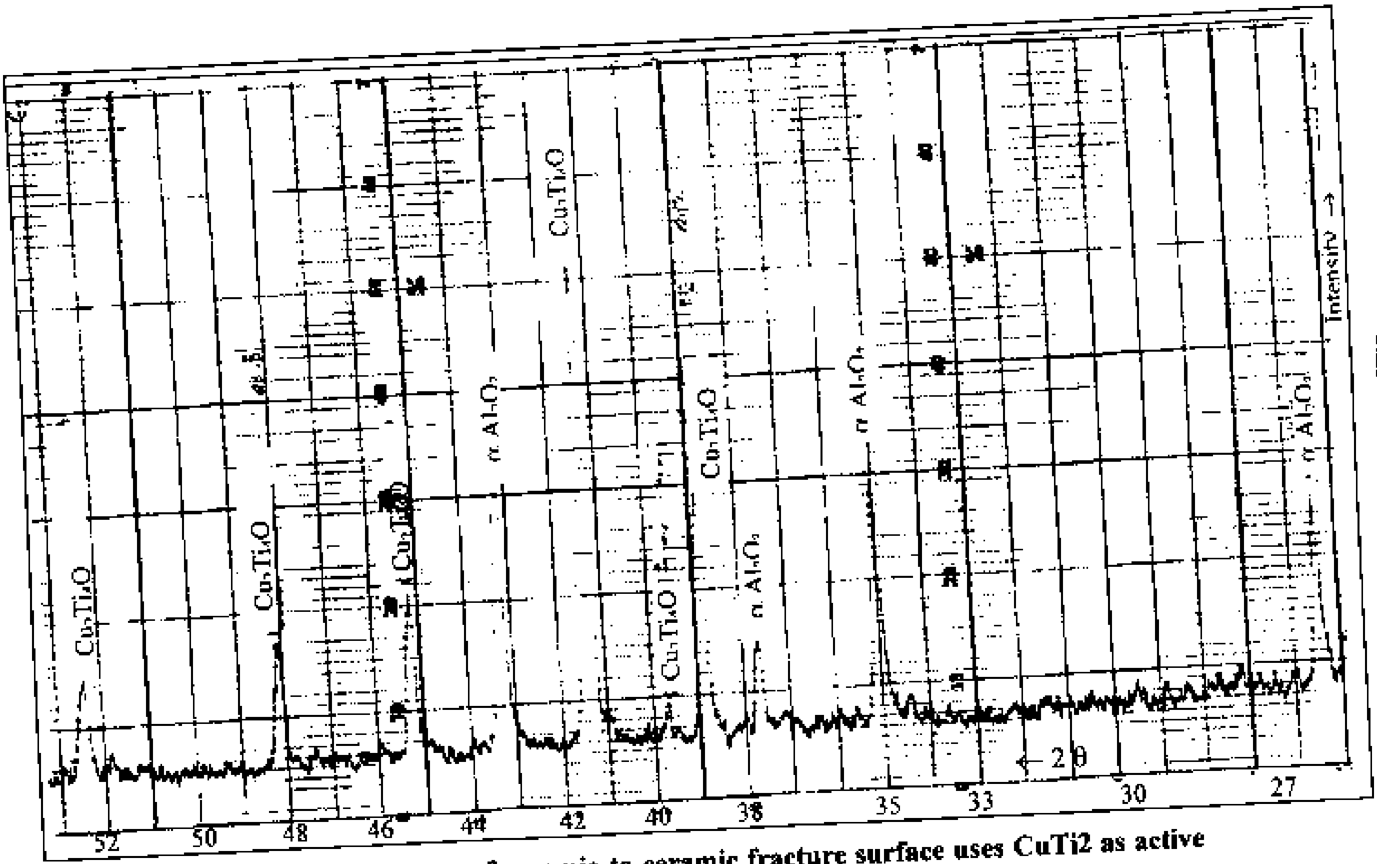


Fig. 12 XRD pattern of ceramic-to-ceramic fracture surface uses CuTi2 as active paste filler- metal, using heating procedure regime No.2

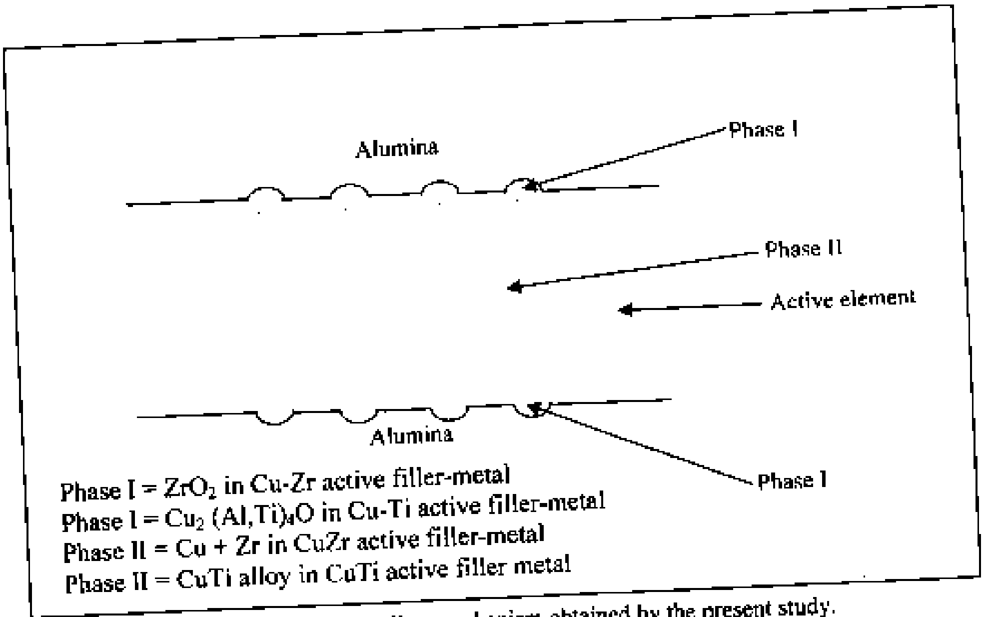


Fig. 13 Suggested bonding mechanism obtained by the present study.

Table 1 Shear strength obtained by using DBS testing for different paste filler-metal of Cu-Ti system and eutectic.

Paste filler-metal types	Shear strength, MPa	Standard deviation MPa
CuTi1	15.0	2.5
CuTi2	10.7	7.0
CuTi3	19.2	2.77
CuTi4	16.1	3.88
CuTi5	21.0	7.32
AgCuTi	42.25	3.35

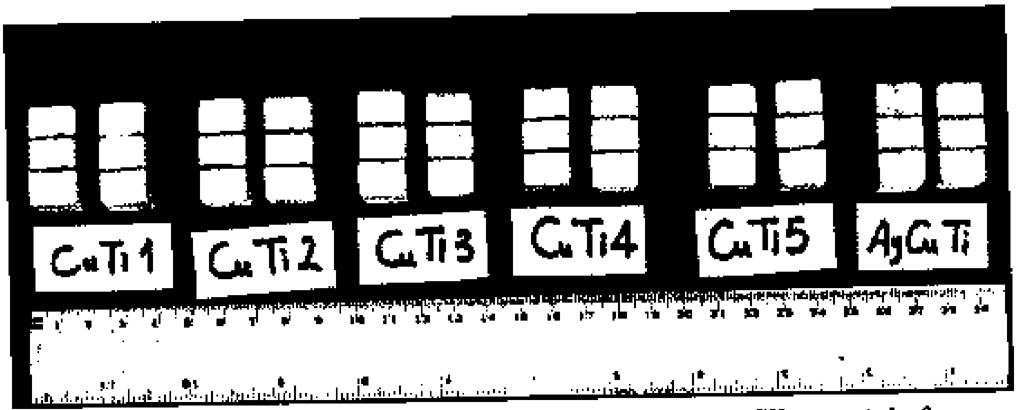


Fig. 14 Double butt-joints actively brazed using paste filler-metal of Cu-Ti alloys and eutectic Ag-Cu-Ti alloy.

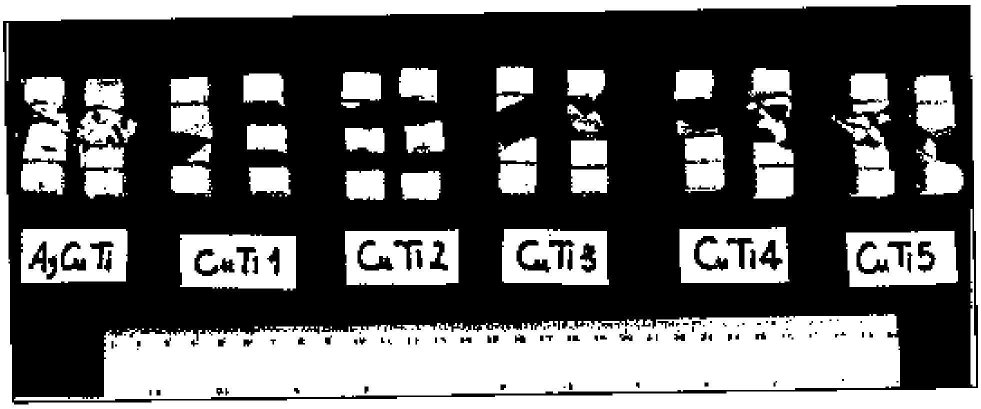


Fig. 15 The DBS fracture pictures for the filler-metal alloys types Cu-Ti and Ag-Cu-Ti systems.

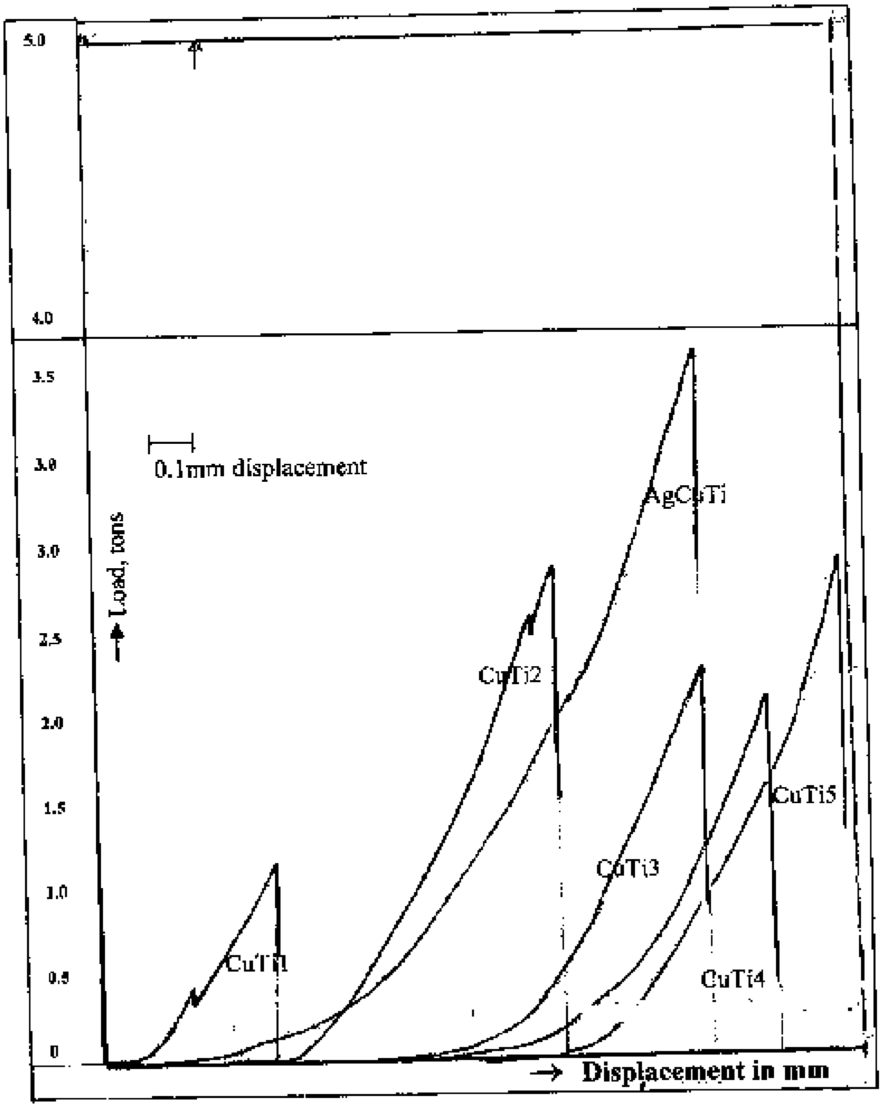


Fig. 16 Load-displacement curves for five DBS tests.