Bonding Behavior of Brazing Al_2O_3 / ZrO_2 Composite to Kovar Alloy

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Abstract:

The production of a reliable joint between alumina composite and kovar alloy by brazing technique using an active filler foil consist of (Ag 70%wt., Cu 28%wt., and Ti 2%wt.). Decontamination processes were achieved over all research specimens. Wetting and bonding behavior were attempted at different condition. Active filler is well brazed with the kovar alloy where the Young angle is (33), while poor wettable achievement appear on the ceramics surfaces where the values of Young angle are between (70°) to (80°). Good bonding obtained with the assembly of (K / 2a / AZ_{15}) where the optimum bonding strength obtained is (90 Mpa), and the fracture occurred from the ceramic and ceramic – filler interface. New structural phases are established at the interlayer of bonded area when subjected to the XRD technique. The new phases enhancement the bonding strength of the kovar bonded alumina composite. The fractured area was subjected to the test of SEM technique then notice the active filler was diffused into alumina composite. This phenomenon gives indicate that the reliable joint was due to the diffusivity of active filler into alumina composite and then enhanced the joint strength.

Keywords: Brazing, dissimilar material, Alumina toughing, Bonding strength, Wettability.

Introduction

Joining ceramic to metal using an active brazing filler alloy is simple as well as it is one step joining process and required a minimum of resources. Brazing of ceramic – metal involves three major partners; alumina composite, kovar alloy strip, and active filler foil. The properties of each joint partner influence the resultant joint properties. Effects of joining variables, including joining material, joint design, surfaces condition of both ceramic and metal, decontamination procedures, furnace type, filler behavior, and brazing process are regarding and evaluated to obtain reliable joints between the partners to be joined [1]. Thermal compatibility of the metal- ceramic pair could play a significant role in metal–ceramic bonding strength (MCBS) because it constituents the main physical requirement to avoid stress at the interface [2]. The coefficient of linear thermal expansion (CTE) of the metal and ceramics must be similar in order to avoid thermal stress. Two situations can be found when comparing the CTE differences between metal and ceramic, when there is a positive difference of CTE, the CTE of alloy is higher than the CTE of ceramic, where ceramic is compressed and metal is under tension when the difference is negative, the CTE of ceramic is higher the CTE of alloy, where ceramic is under tension and metal is compressed[3], usually, a variation ranging from 0.5 to 1.0* 10^{-6} ℃$^{-1}$ between the CTE of alloy and ceramic is considered adequate when the metal coefficient is higher than that of the ceramic. It keeps the ceramic compressed, increasing the life time of restoration [3,4]. The contaminated materials such as carbon promotes a formation of carbon monoxide during the ceramic baking, creating bubbles and porosities, which could be partly responsible for undesirable outcomes, such as cracks or fractures and leads to reduce the bonding strength as a results of these procedures[1,4]. The toughening of alumina by dispersing zirconia particles was first encouraged by the development of the partially stabilized zirconia. The presence of zirconia grains in the alumina matrix as a discrete second phase enables the former to behave in an intrinsic manner [5-7]. The objective of this article is to extend the range of available information about the effects toughening alumina on the joint strength of alumina composite – kovar alloy. As well as the ability of the active filler to wet surfaces of different types of alumina is attempted and hence, for evaluate the joint strength of the assembly.

Experimental Part

High purity of alumina was mixed with partially stabilized zirconia with different concentration (5%wt., 10%wt., and 15%wt.). The physical properties of each type of ceramic stated in Table (1). The PVA binder was added to ceramic matrix to be molded in a steel die of dimension $(40*20*5)mm^3$, in form of a rectangular bar shaft of green composite. The green bar shaft was compacted by pressing machine under (5 tons) for a necessary time within (3 min.). The green samples have been fired and sintered in an atmospheric furnace at 1650℃[5-6]. The standard active filler alloy which consist of Ag(70%wt.), Cu(28%wt.), and Ti(2%wt.),was prepared and conducted by rolled into a suitable thickness of (0.5mm) as a flat form. More information about the physical properties of the standard active filler is listed in the reference [1]. The second partner will be joined to the alumina composite is kovar alloy. A standard strip of kovar alloy was prepared and modified into thickness of 0.8mm. Shape and thickness of kovar strip should have matched to the dimension of alumina composite specimen. The partners will be bonded together should have free of any contaminated materials, and those surfaces should be flat according to overlap design. For this reasons, cleaning, polishing and decontamination processes should be done by using a standard methods [8,9]. Active filler foil was lied in between ceramic – kovar assembly. The assembly constituents alumina composite and kovar strip were held into stainless steel fixture for applying a proper pressure. Due to the matching of the thermal expansion coefficients, lap design was chooses for the purpose of testing the assembly. Vacuum furnace under pressure of $2*10^{-6}$ torr was setting under operation condition of 850℃ at a socking time 20 min. Shear test technique was selected for examined the joint bond strength by using tension machine with crosshead speed of 3mm/min., as illustrated in Fig.(1). The fractured surfaces were evaluated microscopically using an optical microscope, scanning electron microscope and x-ray diffraction techniques.

Table (1)
Presents the physical properties of the ceramic materials.

Formula	Thermal expansion K^{-1}	Molecular weight	Melting point	Density(g/cm^3)
Al_2O_3	$8*10^{-6}$	101.92	2015 °C	3.97
ZrO_2	$5.5*10^{-6}$	123.22	2677 °C	5.56

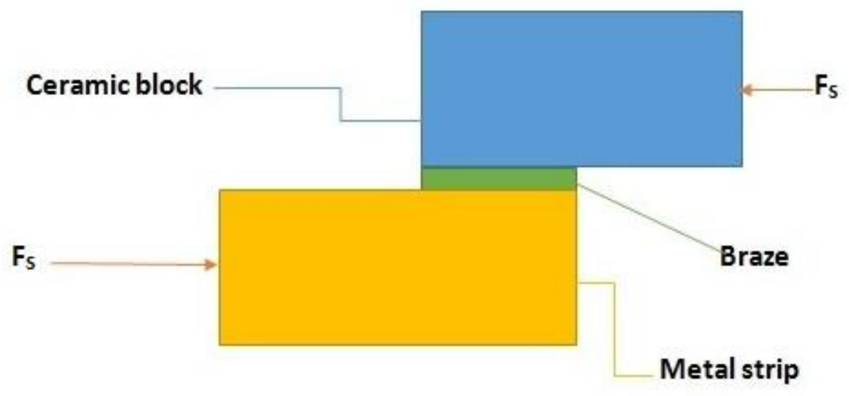


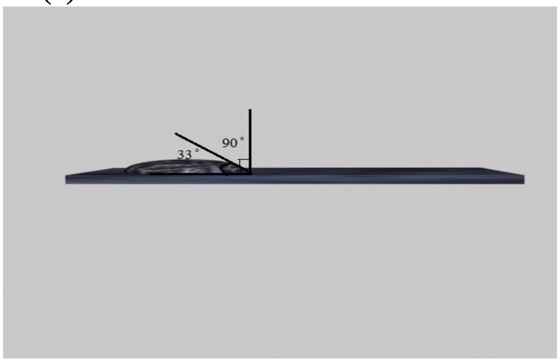
Fig.(1): Schematic illustration of shear test on lap brazed joint.

Results and Discussion

Although increasing ratio of the active element (Ti) in active filler mixture will be promotes the wettability for both surfaces of alumina composite and kovar alloy. In the same time, increasing of titanium concentration up to 2%wt. considered incompatible for wetting behavior. In this research, the existence of the active element titanium enhanced wetting and hence bonding strength. Fig.(2) revealed the satisfied evidence for the ability of filler to wet both alumina composite and kovar alloy. The contact (Young) angle of active filler to the kovar surface is nearly (33°) while the contact angle of such filler to the alumina composite is in between (70°) to (80°). The contact angle was measured by transfer the assembly image of two dimension into three dimension image by using a suitable program to achieve this issue, then the contact angle was measured by protractor bevel between the drop tangential and the x-axix as shown in Fig.(3). These results are agreements with some other researchers [2,6,10]. In general, alumina toughening by zirconia will be enhanced the density and compatibility [7,11]. Accordingly, a thin layer of filler metal after melts at the joining temperature and diffused into both ceramic and kovar alloy. The reaction between the active filler especially the element titanium with the alumina composite will be reacts with ceramic oxides and then the resultant is composed a new structural phases such as TiO_X, gets a reliable joint bond strength [2,10-12]. It can be noticed the best average joint strength is (90 Mpa) when the joint composed of (K/2a/ Al_{15}), i.e. when the alumina toughed by 15% zirconia. This means the increases of a limited ratio of zirconia will be increases the joint strength [11]. The bond information and fractured modes are listed in Table (2) and illustrated in Fig.(3). The failure at the ceramic- filler interface is more happened than other fracture modes. This is because the principle obstacle to be overcome in brazing ceramics is the fact that most metals or alloys cannot easily wet the surface of most ceramics especially which has smooth and pores free surface, although the active filler incorporate an active element titanium in the filler alloys [12,13]. During the brazing process the titanium segregates to the ceramic surface and reacts with ceramic constituent oxides to form a wettable surface and hence, to obtain a reliable joint. On the other hand, the active filler was brazed carefully with kovar alloy surface and composed an efficient reliable joint as illustrated in Fig.(4). The mechanism of adhesion between a metal and ceramic contains oxides is assured by the electron transfer from the metal surface into oxides of alumina composite valence band which is not completely filled with electrons at brazing temperature, and is enhanced and promote bonding when these electrons transfer at the metal / oxide interface is increased [2,13]. The fractured joint was subjected to the XRD technique to investigate the structural intermediate layer due to previously brazed. Figs.(5-7) presents the XRD patterns of scanning the fractured area. It can be seen the several peaks pointed to the original material such α- alumina, copper and silver, while the other peaks which are pointed to the new phases composed by reaction between the constituents of the assembly be joined. Some of these new phases such as TiO_X, CuO, Cu_3Ti, and Cu_4Ti are enhanced the bond strength and gives a reliability of the joint. The existence of Ti element and it's compound at the intermediate layer are promotes the bonding strength between the kovar to respect the alumina composite. As the intensity of the new phase increased will be followed the bonding strength was increased too. This result was agreements with other researchers [1,7,11-13].

(a) Active filler on to kovar surface.

(b) Active filler on to ceramic surface.



(c) Contact angel of Filler on the Kovar

(d) Contact angel of Filler on the ceramic

Fig.(2): Wetting behavior on the both Kovar and ceramic.

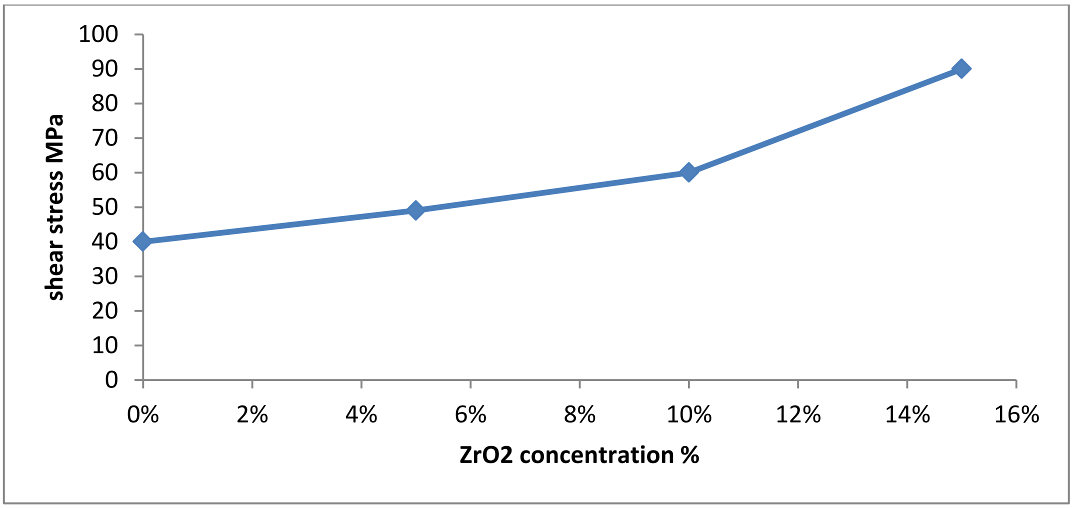


Fig.(3): Represents the Correlation between Shear Stress and Zirconia Concentration.

Table (2)

Presents the fracture modes and bonding strength of the joint.

Joint Type	Fracture mode	Fracture strength(Mpa)
K / 2a / PA	Ceramic - filler mode Ceramic fracture mode Ceramic - filler mode	37 41 42
K / 2a / AZ_5	Ceramic fracture mode Ceramic – filler mode Ceramic – filler mode	45 52 47
K / 2a / AZ_{10}	Ceramic – filler mode Ceramic - filler mode Ceramic – filler mode	90 92 88
K / 2a / AZ_{15}	Ceramic mode Ceramic mode Ceramic mode	60 64 67

Where K= kovar, 2a = 2%Ti in filler alloy, PA = pure alumina, AZ_5 = Alumina with 5% zirconia, AZ_{10} = Alumina with 10% zirconia, and AZ_{15} = alumina with 15% zirconia.

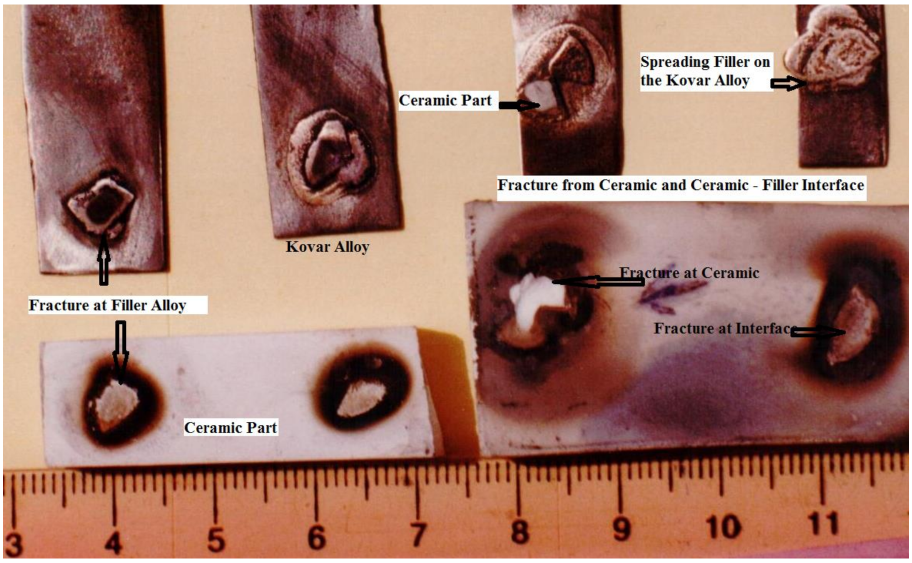


Fig.(4): Represents the Fracture Modes of Different Joints Figure.

Table (3)
Presents the XRD test information of the active filler.

2θ	d (interlayer spacing)	Phase name
38.4	2.34	Ag
41.6	2.16	Cu_3Ti
43.2	2.09	Cu
44.5	2.03	Ag + Cu_3Ti
45.5	1.99	Cu_4Ti
50.5	1.80	Cu

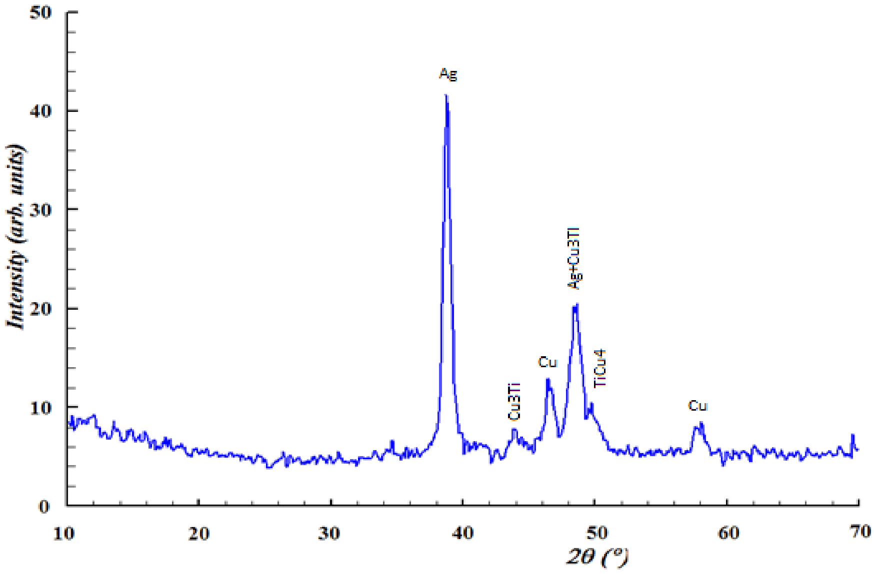


Fig.(5): Represent the XRD Pattern of the Filler Alloy.

Table (4)
Presents the XRD test information of the fractured surfaces.

2θ	d (interlayer spacing)	Phase name
25.2	3.53	TiO_2
25.4	2.34	A- alumina
35.0	2.36	A- alumina
35.5	2.52	CuO
36.8	2.44	Zirconia
37.7	2.38	A- alumina
38.0	2.35	Ag
38.7	2.32	CuO
48.7	1.87	CuO

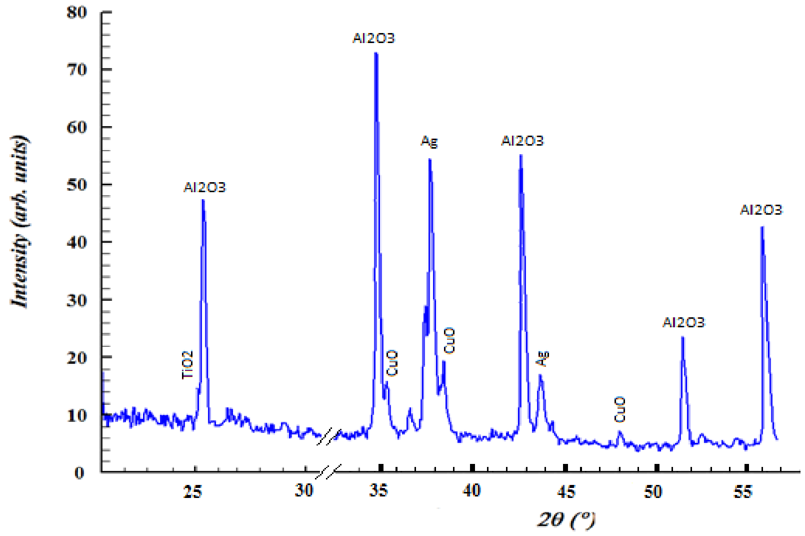


Fig.(6): Represent the XRD Pattern for the Fractured Area.

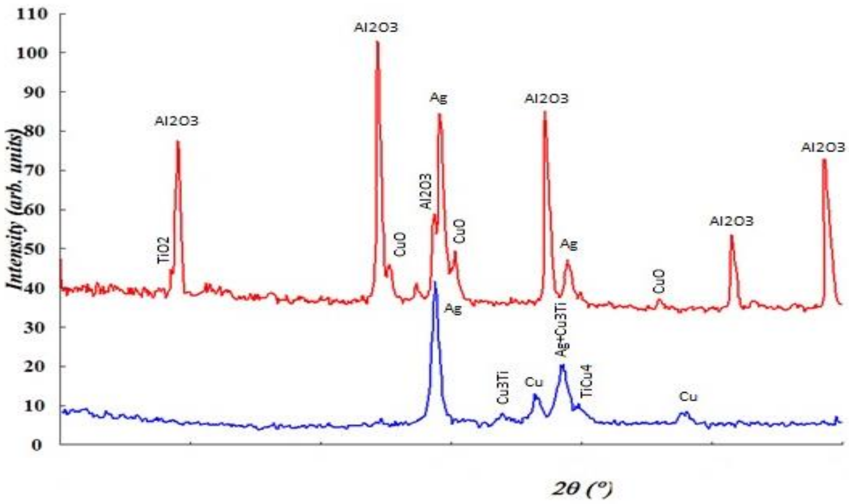


Fig.(7): Represents the XRD Pattern for both Active Filler and Fracture Area.

Microscopic examination: More information about the joint reliability can obtained from the micrographs of SEM technique. It can be seen the diffusivity of the active filler into ceramic part due to some porosities existence on the alumina composite surfaces. As well as the diffusivity of active filler into alumina composite by the mechanism of capillary action also contributed in the joint bond strength [14,15]. Also one can notice from the same Fig.(8), the components of filler are brazed carefully with each other and with the kovar alloy. Fig.(8) shows cross-sectional views of the resultant joint. Three regions are obviously noticed, ceramic, kovar, and the interface region between them. The active filler diffused in the ceramic part and reacts with ceramic oxides to get an aggregate spots in the ceramic part. This aggregate spots may be enhanced the bond strength between the assemblies. Also one can conclude the distribution of the active filler on whole the contact area in which had cracks free. These reasons contributed to good bond strength of the assemblies are joined. Some other researchers concluded are agreements with the our noticed and results [10-16].

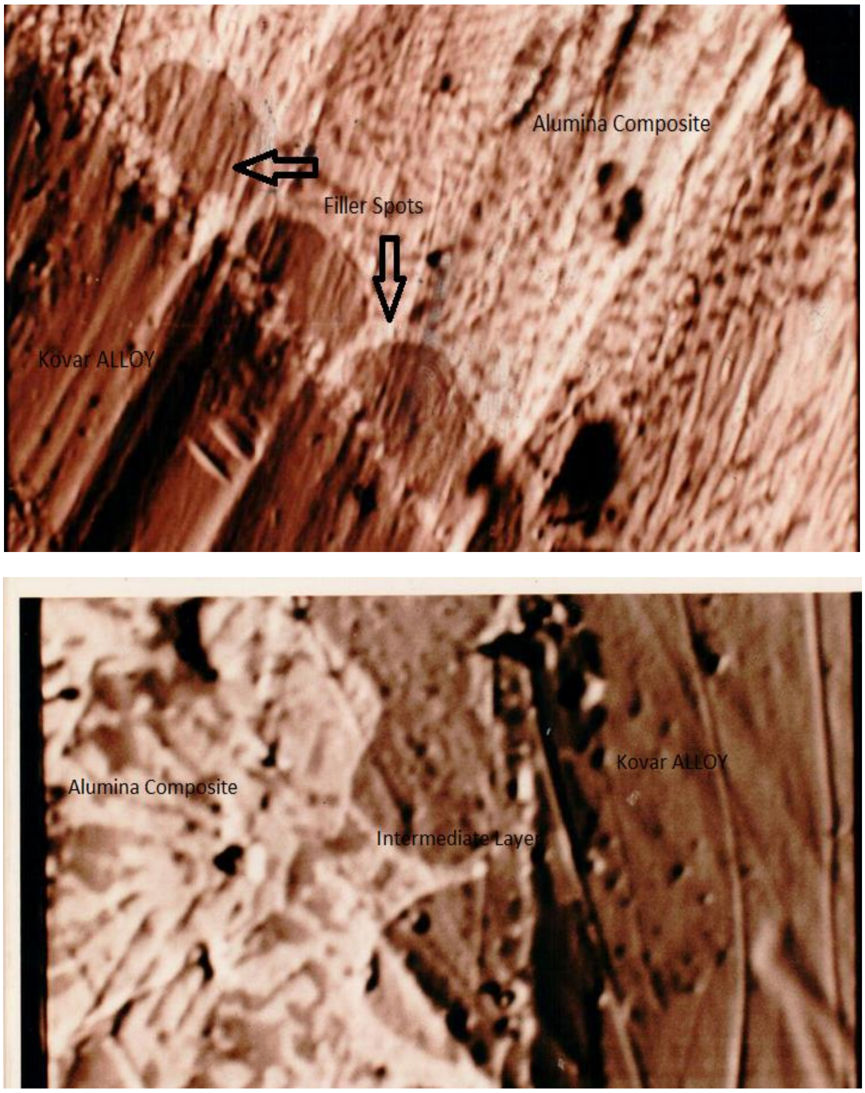


Fig.(8): Represents the SEM Micrographs of Intermediate Layer of the Joint (x= 2000 , and bar scale = 10 µm, 15kV).

Conclusion

1- Best wettability of the filler alloy to respect alumina composite and kovar alloy is achieved when alumina toughed by 15% zirconia.
2- High reliability joining of alumina composite to kovar alloy requires determined carefully a variable process condition such brazing temperature and furnace vacuum.
3- The new phases which are composed at the intermediate layer will be gives enhancing bonding strength of the assemblies are joined.
4- Decontamination processes should have achieved on the specimens surfaces in order to gives a reliable ceramic – kovar joint.

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الخلاصة

يتضمن مشروع البحث تصنيع مفصل لحام متين بين متراكب الالومينا وسبيكة الكوفار باستخدام تقنية لحام المونة المكونة من 70% فضة و 28% نحاس و 2% تيتانيوم. اجريت عمليات ازالة الملوثات عن سطوح المواد المزمع لحامها وتهيئتها لعملية الربط. تم اختبار عملية التبليل والربط بين المواد باستخدام ظروف عمل مختلفة؟. تبين ان الحشوة الفعالة لها قابلية تبليل جيدة لسبيكة الكوفار حيث حصلنا على زاوية يونك مقدارها 33 درجة بينما اظهرت الحشوة المالئة قابلة تبليل واطئة نوعا ما لكافة سطوح السراميك وكانت قيمة زاوية يونك تتراوح بين 70 درجة و80 درجة. ان افضل مقاومة ربط حصلنا عليها (90 ميكا باسكال) عند استخدام المجمع اللحامي المتكون من سبيكة الكوفار وحشوة مالئة ومتراكب الالومينا الذي يحتوي على 15% زركونيا. لوحظ كذلك ان المفصل اللحامي قد كسر اثناء فحصه من منطقة السيراميك وايضا من المنطقة البينية بين السيراميك والحشوة الفعالة. عند فحص منطقة اللحام بوساطة منظومة حيود الاشعة السينية تبين وجود اطوار جديدة ضمن البنية التركيبية للمفصل اللحامي وهو ما يعزز متانة الربط التي حصلت بين متراكب الالومينا وسبيكة الكوفار. اجريت فحوصات بوساطة المجهر الالكتروني فتبين وجود انتشار لمكونات الحشوة الفعالة داخل المنطقة السحية لمتراكب الالومينا وهذا بدوره قد عزز الربط المحكم بين المواد المترابطة.