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# **Review Paper on Joining of Alumina to Metal through Brazing**

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#### ABSTRACT

Joining of ceramics is a viable alternative manufacturing route to these of processing and shaping of large ceramic components with often complex geometries. Further, joining can offer cost and reliability advantages. Similarly ceramic-to-metal joining is usually essential for the fabrication of components that have ceramic inserts in otherwise metallic structures. Alumina ceramic was brazed to different metals such as Ti, Ni, Nb, Fe Different reaction products were formed at the alumina- metal interfaces, including solid solutions and intermetallic compounds. The alumina-Nb joint exhibited the highest bond strength value among the different couples, because Nb produces the lowest residual stress among the alumina-metal joints. In this Article different articles Review to join ceramic materials (Alumina) with metals.

Key word: Brazing, Ceramics, Interfacial oxidation, Ultrasonic-assisted brazing, Laser brazing

## Introduction

The development of new techniques to join structural ceramics has the potential of significantly broadening the applications for advanced ceramics. A commonly used method to join structural ceramics to metals is known as the moly manganese process [1]. However, the moly-manganese process requires two processing steps, namely, metallization of the ceramic, which is conducted at a high temperature (1500 8C), and subsequent brazing, and is therefore time consuming. Furthermore, joint properties are sensitive to process variables and hence precise process control at high temperatures is required to obtain reliable joints, consequently this process is expensive [2]. There are several methods of active metal brazing. One method involves a sheet of titanium (the most commonly used active element) clad between two sheets of a conventional brazing metal [3]. Another technique uses titanium hydride powder mixed with powders of conventional brazing metals such as Cu and Ni [4]. However, the most economical brazing methods utilize an alloy of a base filler metal and an active element [5]. With such an alloy the brazing becomes a simple and economical one-step process.

Alumina ceramic offers high strength, good thermal conductivity, excellent electrical properties, a low cost and high density interconnect: as a result, it is the most popular ceramic material for multilayer packages [6]. In many structural applications, ceramics are selected for critical components forming parts of a total system that is largely metallic, and therefore depend for their success on the fabricator being able to provide good quality ceramic–metal joints. Several techniques have been developed to join ceramics to metals, including diffusion bonding, soldering, and brazing with or without ultrasonic waves [7], although active metal brazing has been investigated since 1940, it has not yet been widely accepted for joining structural ceramics because of inconsistent joint properties [8]. However, research on joining of metals to ceramics using active metal brazing has produced some encouraging results [9]. It has been demonstrated that active brazing joints require the careful selection of the brazing alloy, brazing temperature, heating rate, cooling rate, soaking time and brazing atmosphere [10] and of these the most important factor is the brazing alloy.

Ceramics have a wide range of applications in electric vehicles, aerospace industries, nuclear and chemical-power plant applications. Irrespective of their potential in various applications, utilizing ceramics in required complex shapes imposes serious manufacturing and economic limitations in industrial sectors [11]. The idea of ceramic to metal joining was considered as a promising approach that resolves the existing difficulties in the manufacturing process. Joining metals to ceramics provides complementary properties that cannot be achieved with either ceramics or metal alone; however, achieving a good joint interface between ceramic and metal has a lot of hurdles to overcome [12]. Moreover, the overall properties rely on the joint interface strength. Ceramics and metals have distinct differences in their bonding nature and chemical and physical properties. Metals have a metallic bond with free electrons; in contrast, ceramics have a covalent or ionic bond with extremely stable outer-shell electrons. Hence, molten metal does not wet the ceramic surface, thus limiting the prerequisite condition for joining, i.e., establishing intimate contact between ceramic and metal surface [13].

Engineering ceramics are covering a growing application field due to their specific properties such as high temperature stability, corrosion and wear resistance. In order to construct ceramic parts and components these materials have to be machined which can be a difficult and time consuming task with respect to their high hardness and brittleness. For the integration of ceramic components into complex shapes it can be, therefore, advantageous to use joining techniques such as brazing [14]. Different routes or procedures have been studied to join ceramic to ceramic and metal to ceramic. Conventionally, the brazing process is conducted at high temperature within a furnace chamber in vacuum or inert gas atmosphere [15]. As an alternative method to the uniform heating of the component within a furnace laser beam brazing [16] has been studied which utilizes a localized heat input onto the joining zone.

There are two key problems associated with the brazing process of metals and ceramics. These are the poor wetting behavior of liquid metals on a ceramic surface and the commonly large mismatch in the thermal expansion coefficient and the elastic modulus. Both result in metal–ceramic joints with low mechanical strength. The first problem can be overcome by applying active filler braze materials which utilize elements like Ti or Zr for surface activation in order to improve the wetting behavior. The second problem can be solved by using intermediate metal layers between the metal and ceramic [17]. Within this paper we review different articles on joining of alumina to metal through brazing, as well as the results of brazing alumina ceramic to different metals such as Ti, Ni, Nb, Fe, and AISI 304 stainless steel will be presented. The interface structure and joining mechanism will be discussed.

#### Literature Review

Different Authors worked on joining of Alumina and different material in different way, everyone has its own prose and coins following we review his achievement one by one to find out his gap and define future work for further researchers.

A systematic microstructural characterization of alumina joined to Hastelloy C22<sup>®</sup> by means of a commercial active TiZrCuNi alloy, named BTi-5, as a filler metal is reviewed and discussed. The contact angles of the liquid BTi-5 alloy measured at 900°C for the two materials to be joined are 12° and 47° for alumina and Hastelloy C22<sup>®</sup> after 5 min, respectively, thus demonstrating good wetting and adhesion at 900 °C with very little interfacial reactivity or inter-diffusion. The thermo-mechanical stresses caused by the difference in the coefficient of thermal expansion (CTE) between the Hastelloy C22<sup>®</sup> super alloy (~15.3 × 10<sup>-6</sup> K<sup>-1</sup>) and its alumina counterpart (~8 × 10<sup>-6</sup> K<sup>-1</sup>) were the key issues that had to be resolved to avoid failure in this joint. In this work, a circular configuration of the Hastelloy C22<sup>®</sup>/alumina joint was specifically designed to produce a feed through for sodium-based liquid metal batteries operating at high temperatures (up to 600 °C). In this configuration, adhesion between the metal and ceramic components was enhanced after cooling by compressive forces created on the joined area due to the difference in CTE between the two materials [18].

Sri Harini Rajendran and Seung Jun Hwang [19], investigate the synergic effect of Ti and Sn in the active metal brazing of Al2O3 ceramic to copper brazed, using the multicomponent Ag-Cu-Zr filler alloy. Numerous fine and hexagonal-shaped rod-like ternary intermetallic (Zr, Ti) 5Sn3 phase (L/D = $5.1 \pm 0.8$ , measured in microns) were found dispersed in the Ag-Cu matrix of Ag-18Cu-6Sn-3Zr-1Ti alloy, along with the ternary CuZrSn intermetallic phases. An approximate 15° reduction in contact angle and 3.1 °C reduction in melting point are observed upon the incorporation of Ti and Sn in Ag-18Cu-3Zr filler. Interestingly, the interface microstructure of Al2O3/Cu joints brazed by using Ag-18Cu-6Sn-3Zr-1Ti filler shows a double reaction layer: a discontinuous Ti-rich layer consisting of (Cu, Al)3 (Ti, Zr)3O, TiO, and in-situ Cu-(Ti, Zr) precipitates on the Al2O3 side and continuous Zrrich layer consisting of ZrO2 on the filler side. The shear strength achieved in Al2O3/Cu joints brazed with Ag-18Cu-6Sn-3Zr-1Ti filler is 31% higher, compared to the joints brazed with Ag-18Cu-6Sn-3Zr filler. Failure analysis reveals a composite fracture mode indicating a strong interface bonding in Al2O3/Ag-18Cu-6Sn-3Zr-1Ti filler/Cu joints.

M. Rohde, I. Su<sup>\*</sup>dmeyer, A. Urbanek, and M. Torge [20], studied a laser supported method to join ceramic materials with metals Using a CO2-laser and an active braze filler material, Al2O3- ceramics have been brazed to steel. The microstructure of the interface has been examined and also the mechanical strength of the brazed joint using bending tests. Typical processing times are of the order of several minutes, which is faster than furnace brazing.

Al2O3 to Al2O3 joints were produced using a one stage active brazing technique based on CuTi, CuZr and AgCuTi active brazing alloys. Single and double-butt joints were used for microstructural (light and scanning electron microscopy and X-ray diffraction) and mechanical property (double shear test) studies, respectively. The joints produced with CuZr filler alloys (containing 2%, 4%, 6% and 8% Zr, wt %) showed low shear strengths (0.2–0.4 MPa) due to the presence of ZrO2 at the braze–substrate interface and failed in mode I crack opening. Higher shear strengths of 15–24 and 42 MPa were obtained by using CuTi filler alloys (containing 2%, 4%, 6%, and 10% Ti, wt %) (Mode II/mode III dependent on the Ti content) and the eutectic (Ag–27 wt% Cu–5 wt% Ti) alloy, respectively. The high shear strengths were attributed to the small amount of Cu2 (AlTi) 4O at the braze–substrate interface and led, in the case of the ternary alloy, to the failure of the substrate rather than braze (mode III axial splitting [21].

K. M. Hafez, M. H. El-Sayed and M. Naka [22], worked on joining of the Alumina ceramic to metals Alumina ceramic was brazed to different metals such as Ti, Ni, Nb, Fe, and AISI 304 stainless steel using the Zn–5 mass-%Al brazing filler alloy. Different reaction products were formed at the alumina– metal interfaces, including solid solutions and intermetallic compounds. The alumina–Nb joint exhibited the highest bond strength value among the different couples, because Nb produces the lowest residual stress among the alumina–metal joints. The high mismatch in thermal expansion coefficient between the brazing filler and the AISI 304 stainless steel yielded the lowest bond strength among the joints.

Kirsten Bobzin, and Thomas Schläfer [23], investigated the thermochemistry of reactive air brazing as a joining technology for SOFCs and membranes. X1CrTiLa22, X15CrNiSi25-20, 3YSZ, BSCF and Al2O3 were tested as base materials, Ag8Cu, Ag8Cu0.5Ti and Ag4Cu4Ni as braze alloys. Both DSC

tests and microstructural analysis showed strong reactions between the braze alloys and BSCF as well as the braze alloys and both steels. Further tests such as X-ray diffractometry will help to analyses the reaction products, e. g. mixed oxides, and to understand the reaction mechanisms in detail. Good correlation between both test procedures was achieved. These reactions were analyzed by differential scanning calorimetry tests to get a better understanding. Therefore, three braze alloys (Ag8Cu, Ag8Cu0.5Ti and Ag4Cu4Ni) and five base materials (alumina, 3YSZ partially stabilized zirconia, BSCF perovskite ceramic, X1CrTiLa22 and X15CrNiSi25-20) were investigated.

Linlin Yuan, and Xiaomeng Huang studied to characterize the AgCuInTi filler paste, and to study the direct brazing of Al2O3 ceramics and copper. T microstructure and melting properties of AgCuInTi powder was analyzed using scanning electron microscopy (SEM) and differential scanning calorimeter (DSC). The brazing process was carried out on vacuum brazing furnace at 700°C for 10 min. The joint microstructures were studied by means of SEM and EDS [24].

Wei Cui, and Shuqi Li, show a novel ultrasonic-assisted brazing method for making sapphire (mono-crystalline  $\alpha$ -alumina) joints using a high strength aluminum alloy of Al–4.5Cu–1.5Mg as a filler metal. The bonding between the sapphire alloy and the Al–4.5Cu–1.5Mg was realized through an interfacial oxidation of metallic elements, resulting in formation of MgAl2O4 at the interface. Ultrasonic wave was used to promote the interfacial oxidation during brazing and an annealing heat treatment was used during cooling. Nominal shear strength of the joints made with Al–4.5Cu–1.5Mg was 125 MPa, 2.7 times higher than that made with pure Al. Finite element method (FEM) was used to study the difference on fracture behaviors of the joints made with the Al–4.5Cu–1.5Mg alloy and pure aluminum. The actual bonding shear strength at the sapphire/Al–4.5Cu–1.5Mg interface was estimated as high as 230 MPa [25].

Besides the well-known application as circuit boards and housings, multilayer low-temperature co-fired ceramics (LTCC) offer a flexible and temperature-stable platform for the development of complex sensor elements. Commercial LTCC qualities are usually available with a matching set of metallization pastes which allow the integration of various electrical functions. However, for the integration of ceramic sensor elements based on LTCC into standardized steel housings it is necessary to compensate the mismatching thermal expansion behavior. Therefore balancing elements made of Kovar® (Fe–29 wt % Ni–17 wt % Co) and alumina ceramic (Al2O3) can be used. These components have to be joined hermetically to each other and to the LTCC sensors. In this study, brazing experiments were performed for combinations of Kovar–Al2O3 and Kovar–LTCC with Ag–Cu–Ti- and Ag–Cu–In–Ti-based commercial braze filler metals, Cusil-ABA® and Incusil®-ABA, respectively. For both active braze filler metals, optimized processing parameters were investigated to realize hermetic Kovar–Al2O3 and Kovar–LTCC joints [26].

S U Thielk, R Agustsson, evaluates a novel low-loss alumina AO479U, provided by Kyocera, and a reactive sputtering process suitable to deposit a 10-20 nm thick TiN coating on a large diameter window. Conventional RF vacuum windows are made of metalized ceramics, hermetically brazed to a pillbox cavity. High-power windows, operating in ultra-high frequency (UHF) band, require the fabrication of ceramic disks with diameters on the order of 9". Furthermore, a Titanium Nitride (TiN) multipactor suppression coating must be applied to the ceramic surfaces. The large size and complex internal geometry of these windows create challenges in validating the coating in the fully fabricated assembly [27].

## **Result and Discussion**

Andrea Baggio, Fabiana D Isanto find out that The liquid BTi-5 filler metal from the TiZrCuNi alloy family was demonstrated to have excellent chemical compatibility with both alumina and Ni-superalloy Hastelloy  $C22^{\oplus}$ , which exhibited contact angles of 12° and 47°, respectively. The interdiffusion of some elements between the substrates and the brazing alloy, even though limited, promoted excellent adhesion in the Al<sub>2</sub>O<sub>3</sub>/Al<sub>2</sub>O<sub>3</sub> and H-C22/H-C22 joints and showed homogeneous and crack-free joining areas after a thermal treatment conducted at 900 °C as the maximum temperature. The significant difference in CTE between the metal and ceramic substrates made it difficult to achieve a reliable joint between the planar alumina and the H-C22 substrates. However, in this study, it was possible to overcome this issue, and a sound joint was obtained between such dissimilar materials in a circular configuration, ideal for feed through to be used in the harsh environment found in sodium-based liquid metal batteries (high-temperature and corrosive environment). It was also demonstrated that the compressive forces applied by the outer metal ring onto the joining area favor adhesion, thus making it possible to achieve good joints between ceramic and metal substrate with different CTEs up to  $7 \times 10^{-6} \text{ K}^{-1}$ .

Sri Harini Rajendran and Seung Jun Hwang study that Al2O3/Cu joint was successfully brazed at 840 °C, for 15 min, using multicomponent Ag-Cu-Sn-Zr and Ag-Cu-Sn-Zr-Ti fillers. The results can enhance the understanding of elements interaction in the multi-component fillers, as well as their interaction in ceramic/metal interface. Conclusions are drawn as follows:

The microstructure of Ag-Cu-Sn-Zr filler consists of CuSnZr ternary phase and blocky Cu4AgZr IMC in the Ag-Cu matrix, whereas the microstructure of Ag-Cu-Sn-Zr-Ti filler consists of CuSnZr ternary phase and micron-sized ternary (Zr, Ti) 5Sn3 IMC dispersed in the Ag-Cu matrix.

The peak melting temperature of Ag-18Cu-6Sn-3Zr is 782.9 °C, which is 3.1 °C higher than Ag-18Cu-6Sn-3Zr-1Ti filler. The contact angle of Ag-18Cu-6Sn-3Zr-1Ti filler is 15° lower compared to Ag-18Cu-6Sn-3Zr filler. The presence of Ti in Ag-18Cu-6Sn-3Zr filler proved to enhance the spreading.

Al2O3/Cu joint brazed by using Ag-18Cu-6Sn-3Zr-1Ti filler at 840 °C for 15 min produced a Ti-rich region on the Al2O3 side, followed by m-ZrO2 reaction layer on the filler side (Al2O3/Ti-rich region consisting of Cu3Ti3O, TiO and Cu-(Zr, Ti) /mZrO2/filler/Cu), whereas the Ag-18Cu-6Sn-3Zr filler resulted in a typical m-ZrO2 reaction layer.

Al2O3/Cu joints brazed by using Ag-18Cu-6Sn-3Zr-1Ti filler exhibits a higher shear strength and stronger bonding interface, as compared to Al2O3/Cu joints brazed by using Ag-18Cu-6Sn-3Zr filler. The presence of the Ti-rich region reduces the residual stress build-up on Al2O3 substrate and presence of consisting of Cu3Ti3O, in situ Cu-(Ti, Zr) IMC dispersed in reaction layer enhances the fracture toughness of m-ZrO2 reaction layer.

M. Rohde, I. Su<sup>-</sup>dmeyer, A. Urbanek, and M. Torge, studied and they find out that alumina can be brazed to steel using a laser beam. Typical processing times are of the order a few minutes compared to hours in conventional furnace braze. Furthermore, since a closed process chamber is not needed for the laser process, the joining procedure is highly flexible and can be easily adapted to complex component geometries. The results of the mechanical tests show that the failure of the brazed metal–ceramic joint occurs within the ceramic close to the interface between the braze filler metal and the ceramic part. Thermally induced stresses may lead to cracks within the ceramic, which initiates the failure under mechanical loading. The typical bending strength varies between 40 and 80 MPa with a Weibull modulus ranging from 4.3 to 6.1. However, one important result of this study is that the average joint strength and the Weibull modulus are lower than that of the original ceramic. This behavior can be attributed to the thermally induced stresses, which occur during the laser process and upon cooling down to room temperature. Therefore, the laser process has to be optimized with the focus on a reduction of stresses since this appear to be the main reason for the failure of the joint.

K. Mohammed Jasim, F.A. Hashim, and R.H. Yousif studied that Selection of the correct heating cycle for joining alumina to alumina by a brazing process is critical and a short holding time of 10 min at the brazing temperature was found to be appropriate for the alloys studied in this investigation. The interface in actively brazed alumina to alumina joints using CuZr paste filler alloys consisted primarily of monoclinic ZrO2, which resulted in failure at the interface and low shear strength (0.2–0.4 MPa). The formation of Cu2 (AlTi) 4O at the interface of joints developed during actively brazing using both CuTi and AgCuTi filler alloys was beneficial for mechanical performance. The intermediate shear strength (15–24 MPa) exhibited by CuTi brazes led to the crack deviating into the substrate in the later stages of the test. A consequence of the higher shear strength of at least 42 MPa of the eutectic AgCuTi joint was that failure was largely confined to the central alumina substrate. The binary and ternary Ti-containing systems show promise for producing alumina to alumina joints by active brazing.

K. M. Hafez, M. H. El-Sayed perform that Alumina ceramic was brazed to different metals such as Ti, Ni, Nb, Fe, and AISI 304 stainless steel using the Zn–5 mass-%Al brazing filler alloy. Different reaction products were formed at the alumina– metal interfaces, including solid solutions and intermetallic compounds. The total effect of ultrasonic waves can be seen from Fig. 13a and b, which shows the difference between the metallising of alumina with and without ultrasonic waves; it can be seen that ultrasonic waves enhance the mechanical interaction between the filler alloys and alumina, and therefore accelerate the wetting by the filler alloys at the alumina surface. Note that the ultrasonic waves accelerate the interaction between filler alloy and metal without any additional flux. As the thermal expansion coefficient of the metal increases, the joint bond strength decreases. And the alumina–Nb joints were the strongest among those investigated, whereas the alumina–AISI 304 stainless steel joints were the weakest. This is largely due to the effect of the thermal stresses at the joints.

Kirsten Bobzin, and Thomas Schläfer investigated the thermochemistry of reactive air brazing as a joining technology for SOFCs and membranes. X1CrTiLa22, X15CrNiSi25-20, 3YSZ, BSCF and Al2O3 were tested as base materials, Ag8Cu, Ag8Cu0.5Ti and Ag4Cu4Ni as braze alloys. Both DSC tests and microstructural analysis showed strong reactions between the braze alloys and BSCF as well as the braze alloys and both steels. Further tests such as X-ray diffractometry will help to analyse the reaction products, e. g. mixed oxides, and to understand the reaction mechanisms in detail. Good correlation between both test procedures was achieved. The results give information on how the reactions can be minimized between the braze alloys and the steels, which are supposed to cause inadequate mechanical properties of the brazed joints, especially by using the braze alloy Ag4Cu4Ni. When brazing X1CrTiLa22 with Ag4CuNi4, the DSC results imply that a reduction in the brazing temperature will also reduce the reactions between Ni and the steel seems to prevent the later strong exothermic reactions. Consequently, these results will contribute to improving the RAB process for joining SOFCs and membranes. The results help to explain the reaction mechanisms and allow optimized selection of filler metals and brazing temperature.

S U Thielk, R Agustsson, P Carrier, studied the impact of vacuum brazing on a TiN multipactor suppression coating for use on RF windows was evaluated. The coating was deposited on Kyocera AO479U alumina and sapphire coupons and analysed with profilometry, optical microscopy, SEM, and RBS before and after exposure to a mock braze cycle. Coating thickness uniformity and repeatability was demonstrated for windows up to 225 mm (9") in diameter. Despite a change in visual appearance, no significant changes in coating thickness, composition or morphology was noted. How-ever, the brazing process introduced various microscopic FOD that presents contamination concerns.

J. Schilm, and A. Goldberg, his work focused on the joining process of Kovar with alumina and LTCC as part of on approach to integrate LTCC-based sensors into steel connects. While using commercially available active braze filler metals (Cusil ABA, Incusil-ABA, Incusil-25-ABA) under certain conditions, both ceramic types were hermetically sealed to Kovar. Hermetic joining of Al2O3 to Kovar was possible with Incusil-ABA and with Cusil-ABA for all investigated temperatures. Additionally, brazing of LTCC to Kovar was possible and shown for the first time. At 755 °C with Incusil-ABA hermetic LTCC–Kovar joints were realized. The higher indium content of Incusil-25-ABA would enable lower brazing temperatures, but the strong interaction with the Kovar metal and the porous brazing seams result in unreliable joints. With Cusil-ABA, joints were hermetically sealed at brazing temperatures > 810 °C. In all cases microstructural analysis revealed the development of intermetallic compounds that might be brittle, but their influence on the joint strength is unclear and will be investigated in the future. The combination of this metal-to-ceramic brazing step with additional joining processes allows the hermetic integration of a ceramic LTCC pressure sensor into steel housing with an adapted standardized thread.

Wei Cui, Shuqi Li, and Jiuchun studied that the high strength aluminum alloy of Al-4.5Cu-1.5Mg was used as a filler metal for successfully joining alumina. The interfacial oxidation occurred and MgAl2O4 crystal grains formed at the metal/sapphire interface during the ultrasonic-assisted dipping.

As the sonication time was shorter, the MgAl2O4 grains were scattered on the sapphire substrate. As the sonication time was long enough, the MgAl2O4 grains covered the overall surface of sapphire, forming a dense transient layer between the metal/sapphire. The lattice orientation of the grains was the 111 ð Þ plane, indicating a quasi-epitaxial growth on the sapphire substrate. A robust bonding was performed between the sapphire and the Al–4.5Cu–1.5Mg alloy. The shear strength of the joints made with the Al–4.5Cu–1.5Mg alloy was 125 MPa. The fracture occurred in the sapphire blocks, rather than inside the Al–4.5Cu–1.5Mg alloy or at the interface. The actual shear strength at the Al–4.5Cu–1.5Mg/sapphire interface was estimated as high as 230 MPa by FEM analysis, showing a very strong bonding nature.

#### Conclusion

The present work focused to study and review different Researcher and his work on the joining of alumina through brazing and different techniques with different condition. Everyone has its own prose and coins. Some authors join the brazing ceramics and metal through air some used filler metal for joining alumina. Some others used Ultrasonic waves for joining of alumina and metal, and someone used to brazed alumina to steel using laser beam. Every researcher his its own target.

In the future scope we will suggest that these findings will be helpful towards the development of high entropy brazing fillers in the future. The results help to explain the Different reaction mechanisms and allow optimized selection of filler metals and different brazing temperature

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