

## Fabrication of Reliable Joints of Alumina Ceramics by Microwave-Assisted Reactive Brazing Technique

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Microwave-assisted reactive brazing technique was utilized for joining of alumina ceramics at 950°C and 1050°C for 20 min in argon atmosphere using TICUSIL (68.8Ag-26.7Cu-4.5Ti in mass%) paste as the braze alloy. Conventional heating technique was also employed for comparison purpose only. The microwave and conventionally brazed joints were characterized by X-ray diffraction, scanning electron microscopy, energy dispersive X-ray analysis and Vickers microhardness measurement. X-ray diffraction data showed that the Ti-based compounds were formed at the substrate-filler alloy interfaces of the microwave and conventionally brazed joints. Scanning electron microscopy exhibited the formation of thicker reaction region in the case of joints microwave brazed at higher temperature. Energy dispersive X-ray analysis determined the elemental compositions across the joint cross-section. Vickers microhardness measurements indicated more reliable performance of the joints microwave brazed at lower temperature. Hermeticity of the microwave and conventionally brazed joints was evaluated by Helium leak test and found to be acceptable for actual applications. [[doi:10.2320/matertrans.M2015350](https://doi.org/10.2320/matertrans.M2015350)]

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### 1. Introduction

Joining provides a means of manufacturing complicated structures. As compared to metals the brittleness, high melting temperatures and thermal expansion mismatch properties of ceramics have created problems in finding the most suitable joining technique.<sup>1)</sup> Brazing<sup>2)</sup> is a joining technique, which does not involve any melting of the base metal. In this process coalescence of the filler alloy is produced by heating to a temperature higher than 450°C. The filler alloy flows into the gap between the mating surfaces under capillary action resulting in joint formation. Active metal brazing<sup>3)</sup> is one of the most extensively used joining techniques for joining of ceramics. Since the wettability of ceramic materials is very low bonding between the ceramics can be promoted by using an active filler alloy. The active metal reacts chemically with the ceramic material because of its affinity and forms certain compounds at the interfaces which makes sufficient contact angle<sup>4)</sup> to be able to wet the ceramic material.

Microwave heating is a novel technique for processing of materials, which is fundamentally different from the conventional processing method.<sup>5,6)</sup> During conventional processing thermal energy is delivered to the surface of the material by radiant and/or convection heating that is transferred to the bulk of the material via conduction method whereas energy is delivered directly to the material through molecular interaction with the electromagnetic field in case of the microwave processing. Microwave processed ceramic materials exhibit quality improvement and cost savings over conventionally processed ones. Since microwaves can penetrate into the material so heat can be generated throughout the volume of the material resulting in volumetric heating.<sup>7-9)</sup> Uniform heating is essential during processing of materials. Usually, non-uniform temperature distribution can lead to unanticipated results.

To solve this type of non-uniformity, microwave hybrid heating (MHH)<sup>8)</sup> has been proposed by the researchers. In this method some susceptors that are good absorbers of microwave energy at a specific temperature range, used to provide extra heat flux to the sample by conduction and radiation of heat energy. Microwaves can be transmitted, reflected and/or absorbed depending upon their interaction with the materials. Metals reflect microwaves whereas oxide materials such as Al<sub>2</sub>O<sub>3</sub> and SiO<sub>2</sub> are transparent to microwaves at room temperature. Carbon and silicon carbide are good absorbers of microwaves. The transparent materials can absorb the microwaves by the addition of conductive phases and/or by using hybrid heating.<sup>10)</sup>

Microwave heating technique has great potential for joining both oxide and non-oxide ceramics as a cost-effective method. The first report on microwave joining was published by Meek and Blake in 1986<sup>11)</sup> that was followed by significant work performed by Palaith and Silbergitt.<sup>12)</sup> Joining of oxides, carbides and nitrides was achieved in both single and multimode applicators wherein filler alloys were used that are better microwave absorbers than the substrate materials.<sup>13-16)</sup> Case and Crimp<sup>17)</sup> joined ceramic materials using spin-on interlayer technique by the application of a liquid to the surface of the material to be joined. Subsequently, the liquid is dispersed over the specimen surface by high speed spinning resulting in formation of thin interlayer needed for joining of ceramics. Aravindan and Krishnamurthy<sup>18)</sup> joined sintered alumina-30% zirconia ceramic composites by MHH with sodium silicate glass powder as an interlayer.

High-purity alumina ceramic joints have great potential for use in microwave tube applications<sup>19)</sup> and proton synchrotron application<sup>20)</sup> due to their excellent combination of properties e.g. high electrical resistance, excellent tensile strength, superior thermal resistance, high flexural strength and high radiation resistance. We have already carried out investigations on the alumina-alumina, alumina-graphite and alumina-

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monel super alloy conventionally brazed joints for electron tube applications.<sup>21,22</sup>) Joining of alumina ceramics using TICUSIL filler alloy has been widely investigated.<sup>23–25</sup>) In the present work, microwave hybrid heating (MHH) principle was utilized to produce reliable joints of alumina ceramics for suitable use in electron tube and rapid cycle proton synchrotron machine.

## 2. Experimental Procedure

Pure alumina powder (Alcoa, USA; 99.99% purity) was cold isostatically pressed (EPSI NV, SO, 10036 Belgium) to form cylindrical shaped compact by using 150 MPa pressure. The green powder compacts were dried and subsequently calcined in an electrical furnace at 800°C for 1 h (ELECTROHEAT, Model No. EN170QT, Naskar & Co., Howrah, India). The calcined powder compacts were cut and finally sintered at 1600°C for 2 h. The sintered alumina (Dia.–10 mm, thickness–2 mm) surfaces were ground in a grinding machine (BAINLINE Belt finishing machine, Chennai Metco Limited, Chennai, India) and polished in a polishing machine (SS1000, LECO Corporation, MI, USA) with 6  $\mu\text{m}$ , 3  $\mu\text{m}$  and 0.25  $\mu\text{m}$  diamond pastes (Buehler USA). Average surface roughness of polished alumina ceramic specimen was 0.05  $\mu\text{m}$ . Samples were cleaned ultrasonically with acetone for 15 min before joining operation. TICUSIL paste (68.8Ag–26.7Cu–4.5Ti in mass%, WESGO Inc., Hayward, CA 94544 USA) was used as filler metal. In order to apply a paste thickness of  $\sim 0.07$  mm TICUSIL filler paste of 0.15 g was painted onto the prepared alumina surface using an artist's brush.

Alumina samples were joined conventionally in a high vacuum furnace (Hindhivac Private Limited, Bangalore, India) at 950°C for 20 min with a vacuum of  $5 \times 10^{-6}$  mbar. According to our earlier paper<sup>22</sup>) heating and cooling rate was maintained. The total processing time for the conventional brazing operation was  $\sim 9$  h. The microwave-assisted joining was conducted in a multimode microwave furnace with a magnetron having frequency of 2.45 GHz and maximum output power in the range of 0.3 to 3 kW (Enerzi Microwave Systems Pvt. Limited, Bangalore, India). Schematic of a typical microwave furnace is shown in Fig. 1. The specimens were brazed by microwave heating at 950°C and 1050°C for 20 min with a heating and cooling rate of 25°C/min in argon atmosphere. Microwave brazing of alumina ceramics required total  $\sim 2$  h processing times. Principle of MHH was employed using SiC powder as susceptor so as to initiate coupling of the microwaves with the alumina specimen. The alumina fiberboard was used as the insulator.

Phase identification of the interfacial reaction products for all the alumina joints was conducted by X-ray diffraction (XRD, PW 1710, Philips Research Laboratory, Eindhoven, Netherlands) using Cu K $\alpha$  radiation (45 kV, 35 mA). Microstructural observations were performed by scanning electron microscopy (SEM, Phenom ProX desktop SEM, Phenomworld, Eindhoven, Netherlands) and elemental composition was determined by Energy dispersive X-ray analysis (EDX Phenom pro desktop SEM, Phenomworld, Eindhoven, Netherlands). Vickers hardness tester (ESEWAY, 410 series, Bowers group, U.K.) was utilized to measure microhardness

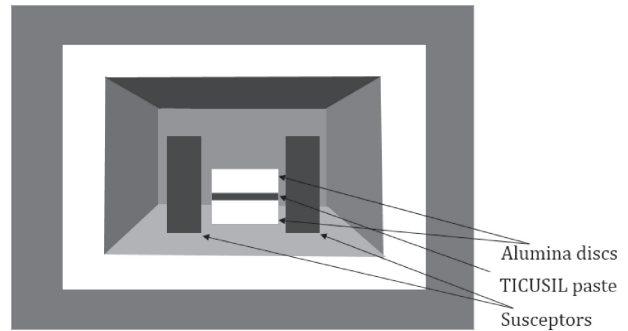


Fig. 1 Schematic diagram of the a typical microwave furnace.

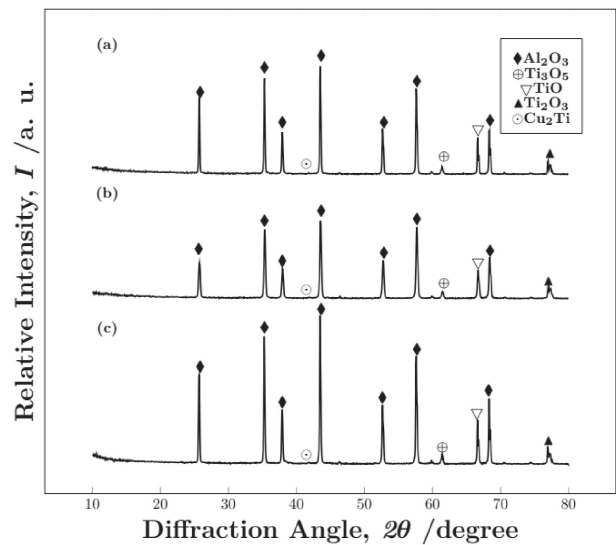


Fig. 2 XRD analysis of alumina joints (a) conventionally brazed at 950°C and microwave brazed at (b) 950°C and (c) 1050°C.

across the joint cross-section at a load of 100 g with 30 s loading/unloading times. Five specimens were examined for a particular specimen. 25 numbers of data were taken for each specimen. Large numbers of data were taken to avoid wide scattering in the data. Helium leak testing of the microwave and conventionally brazed joints was performed by using a helium leak detector (Adixen, ASM 142, France).

## 3. Results and Discussions

During microwave and conventional brazing active element of the filler alloy induced substantial interfacial reaction with the alumina substrates and formed several brittle reaction products.<sup>26</sup>) XRD analysis revealed the presence of Ti-based compounds such as TiO, Ti<sub>2</sub>O<sub>3</sub>, Ti<sub>3</sub>O<sub>5</sub> and Cu<sub>2</sub>Ti as interfacial reaction products at all the interfaces of the joints microwave and conventionally brazed at 950°C and 1050°C (Fig. 2). This was in good agreement with our earlier reported data and other published data.<sup>21,22,26</sup>)

SEM images of the conventional and microwave-assisted brazed joints of alumina ceramics (Fig. 3) showed that the interfacial regions were devoid of any crack or other defects. In case of joints conventionally brazed at 950°C for 20 min the reaction layer of  $\sim 8$   $\mu\text{m}$  thickness was generated. On the other hand, the reaction layer thickness was  $\sim 5$   $\mu\text{m}$  for the

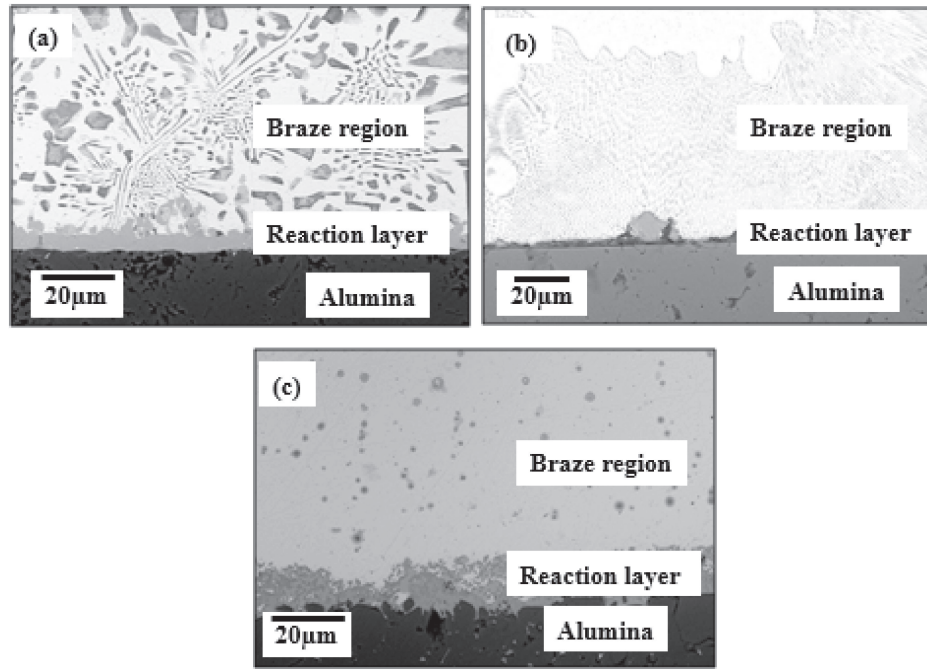


Fig. 3 SEM micrographs of alumina joints (a) conventionally brazed at 950°C and microwave brazed at (a) 950°C and (b) 1050°C.

microwave brazed joint under identical processing conditions. It was noted that the reaction layer thickness of microwave brazed joint enhanced to  $\sim 16\ \mu\text{m}$  with increasing brazing temperature from 950°C to 1050°C while soaking time was made constant. Similar observation has been already noted by other researchers also. They have observed that gradual increase in the brazing layer thickness takes place with increasing the brazing time.<sup>27)</sup>

Substantial amount of time and energy is emaciated to heat up the interfaces by conduction of heat energy through the substrates during conventional brazing of ceramics. On the other hand, brazing alloy in fine metal paste form can absorb the microwave radiation very rapidly. Therefore, during microwave brazing fine powdered metallic paste was not only heated by means of conventional heat conduction method but also by the microwaves directly resulting in hybrid heating. Compared to conventional brazing method microwave heating technique produced relatively thinner reaction layer within a microwave brazed joint under identical processing conditions. Although SiC susceptor facilitated the heating of substrate materials still faster microwave heating ( $\sim 2\ \text{h}$ ) did not allow much heat conduction to the interfaces. Therefore, enlargement of reaction layer depended considerably on the microwave interaction with the filler alloy and thereby, generated thinner reaction layer. In contrast, prolonged heating ( $\sim 9\ \text{h}$ ) during conventional brazing led to slightly wider reaction layer than that obtained for the microwave brazed joint. Increment in reaction layer thickness with the rise of brazing temperature during microwave brazing could be attributed to that fact that significant reaction occurred between the substrate materials and metallic filler alloy with the increase in brazing temperature due to enhanced interaction of the filler alloy with the microwaves leading to intense volumetric heating as well as additional conduction of heat to the interfaces through the substrates resulting in thicker reaction layer.

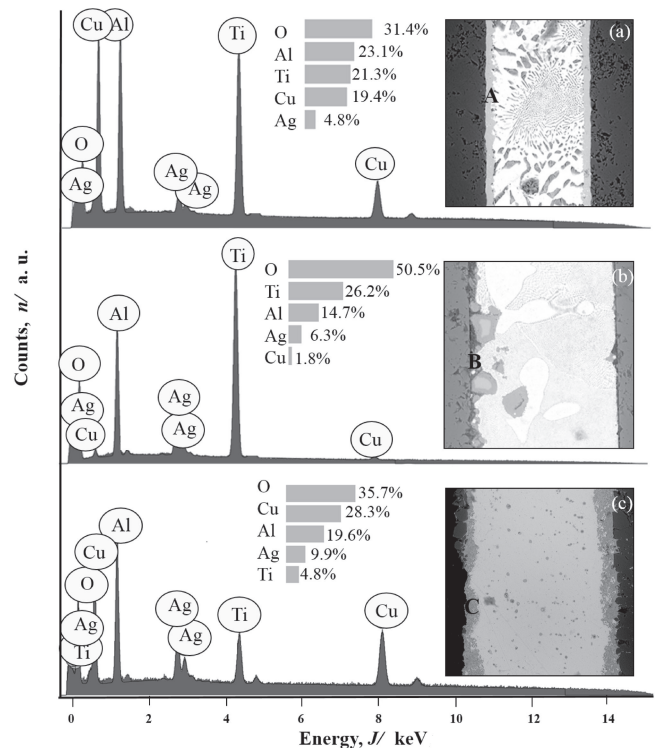


Fig. 4 EDX analysis of alumina joints (a) conventionally brazed at 950°C and microwave brazed at (a) 950°C and (b) 1050°C.

The presence of Ag, Cu, Ti, Al and O elements were detected by EDX analysis at the interfacial regions of the microwave and conventionally brazed joints (Fig. 4), which supported the XRD results. The region marked as 'A' of the conventionally brazed joint (shown in Fig. 4(a)) was consisted of high amount of Ti element (21.3 mass%). However, higher Ti concentration (26.2 mass%) was noted in the region marked as 'B' of the microwave brazed joint

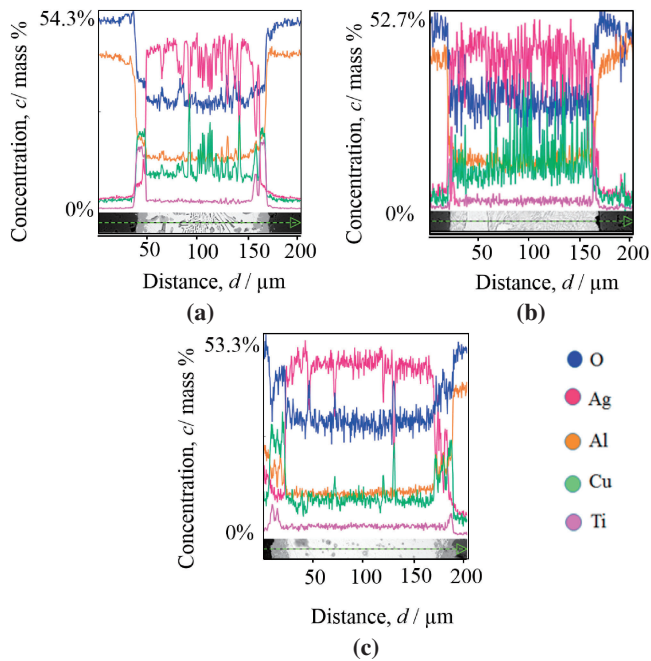


Fig. 5 Line EDX of alumina joints (a) conventionally brazed at 950°C and microwave brazed at (a) 950°C and (b) 1050°C.

(shown in Fig. 4(b)). On the contrary, the region marked as 'C' showed the presence of lower amount of Ti (4.8 mass%). This can be ascribed to the fact that thin reaction layer of joint microwave brazed at 950°C contained Ti that was concentrated in the 'B' region whereas Ti content was distributed in the thicker reaction layer of the joint microwave brazed at 1050°C leading to lower concentration of Ti at the 'C' region (Fig. 4). Conventionally brazed joint exhibited transitional Ti concentration as the reaction layer thickness was intermediary among three types of brazed joints.

Line EDX analysis along the cross-sectional regions of the microwave and conventionally brazed joints confirmed the point EDX analysis (Fig. 5). It was also noted from Fig. 5 that Ag element was homogeneously distributed in the braze region in the case of the microwave and conventionally brazed joints. However, Cu was mainly concentrated in the reaction layer of the joint microwave brazed at 1050°C whereas in the microwave and conventionally brazed (950°C, 20 min) joints Cu was distributed throughout the braze region as well as appeared at the interfacial regions. Presence of aluminium and oxygen elements in the braze regions of all the brazed joints could be attributed to the diffusion of these elements towards the joint area (Fig. 5).

It is well established that soft filler alloy accommodates the stress through plastic deformation when load is applied on the joint of alumina ceramics.<sup>28)</sup> However, deformation is restricted at the interfaces of substrate–filler and consequently, local stress is concentrated at the interfacial regions.<sup>29)</sup> Microhardness distribution across the alumina brazed joint implies the extent of microhardness differences between the filler alloy and the interfaces on each side. Whenever load is applied stress concentration is generated at or near the both interfaces due to large amount of hardness difference resulting in formation of crack. Vickers microhardness measurement across the joints (Fig. 6) revealed

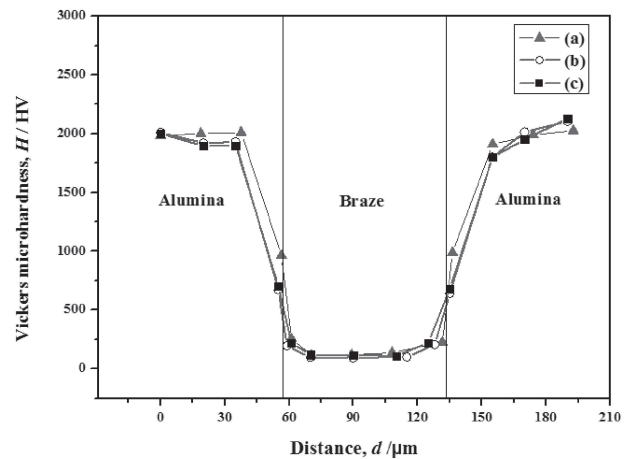


Fig. 6 Microhardness versus distance curves for alumina joints (a) conventionally brazed at 950°C and microwave brazed at (a) 950°C and (b) 1050°C.

good transition between the substrate materials of the brazed joints, indicating reliable joint performance during service. Vickers microhardness value for alumina substrate region was measured to be  $1921 \pm 175$  HV. Conventionally brazed (950°C, 20 min) joint showed Vickers microhardness value of  $976 \pm 20$  HV and  $170 \pm 85$  HV for the interfacial region and braze region, respectively. The interfacial Vickers microhardness values were  $656 \pm 15$  HV and  $692 \pm 10$  HV for the joints microwave brazed at 950°C and 1050°C for 20 min, respectively. Vickers microhardness values of the braze regions for the joints microwave brazed at 950°C and 1050°C for 20 min were  $138 \pm 68$  HV and  $158 \pm 65$  HV, respectively.

The microhardness measurement showed that the microhardness of the braze region was quite lower than those of alumina substrate and interfacial region. Therefore, stress relaxation of the microwave brazed joints occurs through plastic deformation of the soft braze region.<sup>29)</sup> The microhardness values of alumina-braze interfacial regions were intermediate with respect to those values of alumina and braze region, which might be caused on account of the formation of Ti-based compounds. However, the gradual change of microhardness across the interfaces suggested the absence of significantly hard brittle phase at the braze-substrate interfaces leading to low stiffness of the microwave and conventionally brazed alumina joints and thereby, indicating reliability of the joints during actual applications.<sup>29)</sup> The amount of brittle reaction products relatively increased at the alumina-braze interfaces with increasing the brazing temperature leading to enhancement in interfacial microhardness slightly. Lowest interfacial hardness and braze region hardness values were observed with the microwave brazed (950°C, 20 min) joint indicating most reliable performance from that joint. Helium leak test demonstrated that all the microwave and conventionally brazed joints could withstand up to  $1.3332 \times 10^{-7}$  Pa pressure without any leakage, which was indicative of excellent hermeticity of the microwave and conventionally brazed joints.

#### 4. Conclusion

In the present study, alumina ceramics were successfully

brazed by conventional and microwave-assisted reaction brazing techniques. Intensive chemical reactions occurred at the interfaces of the microwave and conventionally brazed joints. The reaction layer was found to be slightly thinner in case of the joint microwave brazed at 950°C than that of the joint conventionally brazed under identical brazing parameters. Reaction layer thickness enhanced significantly by increasing the microwave brazing temperature. Gradual transition of microhardness values was observed across the cross-section of all the joints indicating reliable performance during service. Helium leak test indicated good hermiticity for the microwave and conventionally brazed joints. However, it was observed that most reliable performance can be obtained in case of the joint microwave brazed at lower brazing temperature. The experimental results demonstrated that microwave heating is a promising cost-effective technique for joining alumina ceramics to fabricate very high power electron tube components as well as ultra-high vacuum chambers of a proton synchrotron machine.

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