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Microwave Joining of Ceramics: An Overview

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Abstract

Joining is an integral component of the construction industry. The joint is often the weakest part of any manufactured product. Thus, researchers are trying to increase the strength of the joint. Joining of ceramics is becoming an important manufacturing step slowly but steadily. The evolution of joining processes has allowed ceramics to be used in combination with metals or other ceramics in a number of hybrid devices from traditional light bulbs and seals to improved cutting tools, modern monitoring and measuring electronic devices. New joining methods have been developed for improving the reliability and making the joint interfaces capable of withstanding high temperature with minimum residual stresses. Brazing is the most widely used joining technique on account of its low-cost and possibility to join intricate geometries for large-scale production. Recently, microwave processing has emerged as a novel processing technique for the processing of various materials such as ceramics, polymers, composites and even metals. Microwave processing can afford savings in energy and provides a product with unique properties that cannot be achieved through conventional methods and therefore, it has been adopted by several industries. An overview on recent investigations in the area of microwave joining of ceramic materials is presented herein.

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Introduction

Joining is a process used to bring separate parts or components together to produce an integrated structural entity. It is an important step in the construction of engineering structures such as formation of a building or assembly of an automobile. However, joining is also a crucial process in the formation of many components that are later joined to produce these buildings and automobiles. The joining process also extends beyond mechanical joints formed by the use of fasteners e.g. nails, bolts and clamps. A significant number of products contain components that are joined by chemical reactions. Joining is the preferred method for manufacturing the complicated shaped products [1]. All types of materials such as ceramics, metals, polymers and composites have been joined to one another for a huge number of applications. The applications that involve the joining of ceramics are extensive. The joining challenges are growing faster than the knowledge of the process. The need for joining has increased with respect to time. Ceramic-ceramic joints are followed because they provide an opportunity to improve the final properties of the product. Ceramic-ceramic joints can provide an increased resistance to operating temperature as compared to the metallic joints. They are more resistant to oxidation than most of the metallic joints. The enhanced resistance to degradation, oxidation and corrosion of ceramics over metals at elevated temperatures can be used to increase the efficiency of heat exchangers and turbine engines [2, 3].

Brazing [4] is widely used as a joining technique, which does not involve any melting of the base metal. In this process, coalescence is produced by heating to a temperature higher than 450 °C by using a filler metal [5] that must have a liquidus temperature higher than 450 °C and below the solidus temperature

of the base metal. During brazing, assembled parts and filler metal are heated to a temperature high enough to melt the filler metal but not the parts [6]. The molten filler metal spreads into the joint and must wet the base-metal surfaces. After cooling the filler metal is held in the joint by capillary attraction and fastens the part together [7]. Active metal brazing is most commonly used method, which is based on the use of active metal like Ti, Zr, Nb, Cr, or Y in the filler alloy particularly for Al₂O₃. The active metal is incorporated into the braze alloy in order to aid wetting by forming an oxide and another compound (e.g., carbide, boride) through reaction of the active metal with the ceramic materials [8–13].

Until recently, in case of the joining of materials most of the works have been performed using conventional heating. The specimens were heated in either a resistance or radio frequency (RF) heated furnace for joining. Radio frequency heating is restricted to materials that are electrically conductive and capable of supporting an induced current. Some works have been done using lasers to heat the work pieces during joining. Recently, microwaves have been used to provide the energy necessary to heat the specimens to temperatures suitable for joining. Researchers have joined alumina using microwave energy and established an understanding of the joining process [3]. Microwaves are electromagnetic radiations with wavelengths ranging from about 1 mm to 1 m in free space and frequencies between 300 GHz to 300 MHz. The most common microwave frequency used for research is 2.45 GHz, which is same for the domestic microwave ovens. The other allowable frequencies are 915 MHz, 30 GHz and 83 GHz for some specific applications. Since microwaves can penetrate the material and supply energy, heat can be generated throughout the volume of the material resulting in volumetric heating. Hence, it is possible to achieve

and uniform heating of the thick materials. Therefore, the maximal gradient in the microwave-processed material is the reverse of that in the material processed by conventional heating methods. In case of microwave heating, molecular level energy transfer has some advantages. Microwaves will selectively couple with the higher loss tangent material when it is in contact with materials having different dielectric properties leading to selective heating of the materials [14, 15]. Beale et al. [16–18] developed a control system for microwave joining of a variety of ceramics. Various researchers worked on designing of microwave apparatus [19–22].

Industry is becoming aware of the potential of microwave joining in case of fabricating ceramic joints. However, microwave joining of ceramics to ceramics/metals is relatively new. Due to rapid heating by microwaves, the microwave joining method can be many times faster than the conventional joining methods. The current paper provides an overview of recent developments in the area of microwave joining of ceramics.

Joining

Much of the earlier work until the mid-1950 was focused on glass-metal joining. In the earlier years sealing technology remained as an art based on hard earned experience rather than scientific technique. Consequently, a review of earlier literature shows an apparently infinite number of processes and techniques [23]. Nascimento et al. [24] has summarized the progress of metal-ceramic brazing. Kingery et al. [25], Armstrong et al. [26–28] and Sutton [29] have examined the metal-ceramic reactions and the effects of certain elements to aid the formation of the ceramic-metal bonds. Naka et al. [30] indicated that the formation of a titanium oxide on the Al_2O_3 surface led to the strong joining between alumina and the Cu-Ti filler metal. More investigations concerning the bonding mechanisms and characteristics of Al_2O_3 -metal joints using the active Ag-Cu eutectic alloys (i.e. Cusil-ABA, Incusil-ABA, and Ticusil) have been reported [31–33].

Apart from alumina ceramics that is the most commonly used structural ceramic, zirconia ceramics is gaining importance [34]. Zirconia ceramics exhibits high strength and fracture toughness due to their transformation toughening abilities, particularly at temperatures below 300 °C and it is also a good ionic conductor at high temperatures. Zirconia has potential applications for wire drawing dies, cutting and machining tools to oxygen sensors and fuel cells. Zirconia was joined to stainless steel [35] and to itself. Rathner and Green [36] studied the brazing of yttria-tetragonal zirconia polycrystal (y-TZP) to itself with pure Al and Al-5.8 wt% Zr alloy. The joint brazed with Al-Zr alloy exhibited higher strength because of the formation of needle like Al_3Zr precipitates that helped in strengthening of the joint. Moorhead [37] developed various filler metals for brazing MgO and Y_2O_3 stabilized PSZ. Ag-30Cu-21Ti and Cu-20Au-18Ti filler alloys produced ceramic-ceramic joints with room temperature flexural strengths greater than 100 MPa.

Microwave Processing

In 1946, Percy Spencer found a candy bar was melting in his pocket during his experiments on the MW generation tube and discovered the MW heating. Since then, MW heating has been applied to various fields. Microwaves are a part of the electromagnetic spectrum like visible light, radio waves and X-rays. Microwaves frequency extends between 300 MHz and 300 GHz corresponding to wavelengths ranging from about one meter to less than one millimetre. In the past, microwaves were used extensively in the field of telecommunication such as radar, sensing and so on before and during the World War II. Recently, microwave processing has been emerging as an innovative sintering method for ceramics, ceramic composites as well as

polymer and polymer composites. [38–41]. Joining, melting, fibre drawing, development of functionally gradient materials, reaction synthesis of ceramics, synthesis of ceramic powder, phosphor materials, whiskers, microtubes and nanotubes, sintering of zinc oxide varistors, glazing of coating and coating development have been done using microwave heating. In addition, microwave energy is being utilized for the sintering of metal powders also [42].

Microwave furnace consists of the source, the transmission lines and the applicator. The source generates electromagnetic radiation and the transmission lines deliver the electromagnetic energy from the source to the applicator. The energy is either absorbed or reflected by the material in the applicator. The type of applicator used depends on the materials to be processed. The single mode applicator and the traveling wave applicator are successful in processing materials of simple geometries. However, the multimode applicator has the capability to produce large and complex components. Any electromagnetic losses in the materials used were considered detrimental. Recent introduction of microwave ovens occurred due to the realization that electromagnetic losses could be used for cooking. In 1985, Meek and Blake [43] used the electromagnetic loss induced heat in materials not containing water. They were able to heat a ceramic material for melting purpose. Clark et al. [44] wrote an excellent review based on the theory behind the interactions of microwaves with materials in which he discussed about different types of polarization.

Microwave hybrid heating incorporates a strong microwave absorbing material into the surrounding insulation. This absorbing material absorbs more microwaves than the material that is being processed. It is beneficial to use microwave hybrid heating (MHH) [45] to heat most ceramic materials in a multimode microwave oven. Hybrid or microwave-assisted heating continues to be an important processing method and its application in the commercial sector and new research areas are reported as well. Silicon carbide is the most common microwave absorbing material. It is an excellent and inexpensive microwave absorber used in air at elevated temperatures. Above 900 °C, the silicon carbide will oxidize to form a protective silica layer and the oxidative weight gain increases with increasing the temperature. The combination of the absorbing material and the insulation is usually called the microwave susceptor. The susceptor assists the heating of the material to the processing temperature. Microwave processing of low loss materials such as high-purity alumina using multimode microwave ovens has become possible by using microwave susceptors [44, 46]. Although many of the potential advantages of microwave processing of ceramics have been recognized for over 30 years, yet during the past 10-15 years potential of this processing technique have been attracting widespread attention. The increasing benefits associated with microwave processing are noted from enormous numbers of research papers presented at technical meetings and symposia.

Application of MW heating to the energy and environmental fields has also been attempted e.g. incineration of medical wastes, devulcanization of rubber tire, treatment of sewage sludges, regeneration of spent activated carbon and chemical residues of petrol industries. MW de-nitration for producing MOX (Mixed Oxide) nuclear fuels has been investigated by re-disposal of the used Plutonium [47]. Vitrification of the nuclear wastes by MW heating had been proposed by Morita et al. [48]. MW application to polymer has started earlier such as curing of thermosets [49]. It was recognized during MW drying of Al_2O_3 castable that apart from drying they can be heated as well by MW. MW heating of the ceramic powders above 1400 °C made it possible to sinter ceramics in a microwave field [50, 51]. Later, it was investigated that MW sintering enhances the diffusion rate. Bykov et al. [52]

discussed microwave processing of materials from the physical aspects. He included the fundamental concepts regarding the absorption of electromagnetic waves, heat transfer, and the electrodynamics of single and multimode microwave cavities in his paper. Some formulas, estimates and illustrations were given in the paper. Further, he mentioned the advantages of using millimeter-waves over microwaves.

Microwave Joining

In the past several years extensive use of microwave energy was noted for the processing of materials. Microwave energy provides rapid heating resulting in an overall reduction of the processing time [15, 53–55]. Microwave joining of ceramics is a relatively new field. Commercial applications for microwave brazing include machine tools, twist drills, drill bit cutters, abrasives, ceramic substrates for electronic devices, turbocharger rotors, turbine blades, automotive engine components, nuclear fuel rods, heating elements, heat exchangers, and fuel cells [56].

First of all, a glass interlayer was utilized for microwave joining purpose using a home microwave oven. Meek and Blake [43] had used a home microwave oven to join alumina to itself and to metal using a borosilicate glass interlayer. In 1988, it was first reported by two groups that closed joints fabrication was possible with no interlayer in a Materials Research Society Symposium on microwave processing of materials. One of these groups was led by Drs. R. Silbergliitt and D. Palaith of Technology Assessment and Transfer, Inc., and the other group led by Dr. H. Fukushima at Toyota Central R & D Laboratory in Japan. Published results of these groups displayed that alumina, mullite ($3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$), and silicon nitride can be joined directly by microwave heating [57]. Fukushima et al. [58] joined several different purities of alumina (92 - 99 wt. %) and silicon nitride. Joining was carried out at a temperature ranging from 1400 °C to 1850 °C and pressures up to 2.4 MPa. Microwave joining of SiC to SiC was accomplished by Silbergliitt et al. [59] with interlayer materials composed of Si, C and Ti. A new joining method for superconductors by microwave heating was developed by Cai et al. [60]. Bars of microwave-joined Bi-Pb-Sr-Ca-Cu-O ceramic superconductors showed good mechanical integrity. Di Fiore et al. [61] used a thin interlayer to join ZnS bars using microwave hybrid heating. Arunajatesan et al. [62] used electron probe microanalysis (EPMA) and transmission electron microscopy (TEM) to examine the SiC/Al interfaces in the microwave joined parts.

Cozzi et al. [63, 64] joined 99.5 wt. % pure alumina ceramics using 94 % pure alumina as the interlayer material. The interlayer material was cut from a rod into discs approximately 2 mm thick. Ahmad et al. [65–68] joined reaction bonded silicon carbide without any interlayer or applied pressure. Sato et al. [69] joined Al_2O_3 and MgO within a single mode cavity microwave furnace. MgAl_2O_4 layer was formed at the interfacial region between Al_2O_3 and MgO. The hardness of the joined region was higher than other regions. Binner et al. [70, 71] used microwave heating technique to directly join Reaction Bonded Silicon Carbide (RBSC) to itself and zirconia to zirconia with total processing times of about 30 min wherein the bonding time was 10 min or less. Su et al. [72] carried out simultaneous sintering and joining of ceramics using microwave energy. Al-Assafi et al. [73, 74] joined alumina ceramics of different purities using alumina gel as an interlayer and microwave energy as the heating source. Direct joining and joining with gel were carried out and comparative studies were made between two types of joints.

Suryanarayana et al. [75] brazed diamond and tungsten carbide using NiTi as the filler. Lee et al. [76, 77] joined silicon carbide and magnesium fluoride using conventional and microwave heating. A spin-on interfacial layer was utilized in case of both

conventional and microwave joining of the SiC and MgF_2 , which allowed joining with low external applied pressures. Bykov et al. [78] joined ZrO_2 and Al_2O_3 ceramics via nanostructured interlayer. Case and crimp [79] joined Al_2O_3 / PSZ specimen using spin-on interlayers. The specimens were joined at 1500 °C for 20 min using a silica interlayer. The spin-on interlayer technique is the application of ceramic precursor liquids by spinning on the adherent surface. This technique provided thin and strong joints between similar and dissimilar ceramic materials. Liu et al. [80] joined alumina ceramics using microwave heating. SiC was added in the interlayer as a microwave-absorbing material. Joining of alumina was conducted by raising temperature of SiC itself that aided microwave-absorption of the oxides.

Bruce et al. [81] joined silicon carbide fiber-reinforced silicon carbide matrix composites for fusion applications. The composites were joined in a microwave heating environment using preceramic polymer slurry as the filler. Kondo et al. [82] joined silicon nitride by inserting glass joint between two silicon nitride pipes and silicon carbide susceptor was placed around that joint. The susceptor was locally heated through absorption of the microwave radiation. Uniform heating is important during processing of materials. In case of ceramic sintering, non-uniform temperature distributions can lead to undesired results [83]. Two methods have been used to solve this type of non-uniformity. In one method the heated material is surrounded with an insulator while the other method involves hybrid heating. Two hybrid heating methods have been proposed. One hybrid heating method is called microwave hybrid heating (MHH) [84] where some susceptors, good absorbers of microwave energy at a specific temperature range, are used to provide extra heat flux to the heated sample by heat conduction and radiation. Alumina is a poor absorber of microwaves at room temperature. With increasing temperature, alumina can absorb microwave energy significantly. By surrounding the alumina with SiC susceptors that absorb microwave energy even at room temperature, the initial temperature increase of alumina is occurred due to heat conduction and radiation from susceptors. When the temperature of the alumina ceramics increases up to 800 °C, it absorbs the microwaves directly. Generally, the researchers use a multimode cavity and microwave hybrid heating. Silicon carbide is an excellent microwave absorber and has attracted attention for microwave joining. Reaction bonded silicon carbide and hot pressed silicon carbide ceramics have different degree of microwave absorption capability. Reaction bonded silicon carbide contains residual silicon metal throughout the microstructure. MMH method is a good idea to heat some transparent and opaque materials. Uniform temperature distribution can be generated with the inclusion of an insulator. Two problems with the MMH method must be highlighted. First, the characteristic of rapid heating is compromised due to the use of heat conduction. Secondly, microwave energy does not transmit directly into heated materials during some stages of processing [85].

Aravindan and Krishnamurthy [42] had reported successful joining of sintered alumina and 30% zirconia ceramic composite using MHH at 2.54 GHz frequency and 700 W power. Samandi and Doroudian [86] used microwave plasma to rapidly heat the ceramic to a temperatures at which it started absorbing the microwaves. The microwave radiation was directly coupled to the ceramic. The plasma was turned off since energy was no longer available to ionize the gas and sustain plasma. This hybrid approach avoids the limitations of conventional microwave heating for sintering and joining of advanced materials. Similarly, Barmatz et al. [87] conceived microwave plasma assisted method and system for heating and joining materials. The invention uses microwave induced plasma to controllably preheat poor microwave absorbing specimens. The plasma preheats the

specimens to a temperature that improves the ability of the materials to absorb microwave energy. The plasma is put out and microwave energy is able to volumetrically heat the specimen. Localized heating of good microwave absorbing materials is done by shielding certain parts of the specimen and igniting the plasma in the areas that are not shielded. Zhao et al. [88] joined alumina composites in air by microwave heating using Al-Si alloy powder as the interlayer. Microwave brazing process was patented by Budinger [89] in 2008. Sallom et al. [90] joined gamma titanium aluminide using microwave heating using Ag-Ti and Ag-Cu-Ti filler metals and reducing atmosphere of Ar-H₂. The results showed that it is possible to join this alloy to itself and to other alloys such as H13 tool steel and superalloys without formation of any crack. In addition, the diffusion layers are well identified in the brazed area and thereby, indicating different phase formation with different chemical composition. Hoyt et al. [91] melted the interface filled with tin powder in glycerol suspension for metal-ceramic soldering. Joining of alumina ceramics using microwave-assisted reactive brazing technique has been reported elsewhere [92, 93] using TICUSIL (68.8Ag-26.7Cu-4.5Ti in wt. %) paste as the braze alloy. Table 1 shows the microwave joining of ceramics using different kinds of fillers.

Table 1: Microwave joining of ceramics by using different fillers

Microwave joining of ceramics	
Filler	Reference
Alumina gel	[73], [74]
Direct joining	[57], [58], [66], [70,71], [74]
Glass	[43], [82]
alumina/zirconia	[62], [63]
Si powders	[88]
Interlayers	[61], [76,77], [78], [79], [81]
TICUSIL paste	[90], [92,93]
Polymer slurry	[80]
NiTi	[75]

Conclusions

A review of recent progress in the field of joining of ceramic materials by using microwave heating technique has been presented in the current paper. During the past twenty years, there has been considerable growth and activity in research and developmental efforts resulting in significant improvements in the processing of materials in terms of techniques, equipment, control etc. as well as applying better understanding towards the processing technique, process modelling and exploring new areas of applications. It is distinct to the researchers that there is a huge scope for innovative research works using microwave technology in future.

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