

RECENT PROGRESS IN JOINING OF CERAMIC POWDER METALLURGY PRODUCTS TO METALS

M. Mazar Atabaki^{1,2*}

¹*Institute for Materials Research, the School of Process, Environmental and Materials Engineering, Faculty of Engineering, University of Leeds, Leeds, UK*

²*Department of Mechanical Engineering, Faculty of Materials Engineering, University Technology Malaysia, 81310, Malaysia*

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Abstract

Joining technologies of ceramic powder metallurgy products to metals have been advanced during recent years, especially in powder metallurgy industries. There are a lot of difficult problems to be overcome in developing joining techniques. To control the chemistry of interfaces is, of course, one of the major key points. The active metal brazing has been established for solving this problem and gives scientists high quality of joints. To construct an appropriate interlayer for reducing thermal stress originating from thermal expansion mismatch between a ceramic and a metal is another critical problem. The soft metal interlayer and the soft metal/hard and low expansion metal laminate interlayer are proved to be two of the effective interlayers. The other bonding parameters such as surface roughness of bond face and bonding pressure are also important in order to obtain a sound as well as strong ceramic/metal joints. Most ceramic/metal bonding is conducted at high temperatures, where chemical reaction is to be expected. In this article, recent developments in joining ceramics to metals considering the interfacial behavior of their bonds are discussed. Therefore, methods of joining were compromised and the most famous insert layers were introduced. In addition, recent developments in the joining of powder metallurgy products to metals in terms of case studies have been conducted in detail.

Key words: ceramic-metal joining; powder metallurgy; structural ceramics; brazing; interlayer

* Corresponding author: M. Mazar Atabaki, m.mazaratabaki@yahoo.co.uk

Introduction

With low density, high strength and excellent high temperature resistance, ceramics are widely used in areas of aerospace and powder metallurgy. Especially in high temperature techniques, ceramics and ceramic matrix composites possess more advantages than metals. However, ceramics have low toughness and are hard to manufacture in complex components. Therefore, to meet the requirements it is reasonable to manufacture composite parts of ceramics and metals by joining [1, 2]. In use of any kinds of ceramics, it is needed to overcome two of serious defects of ceramics, i.e., hard formability and brittleness, which are fatal as structural components. We should adopt simple process for production of ceramic components. For this purpose, it should be maintained reliabilities of productions with ceramic components. Recent trends in ceramic researches such as machinable ceramics and nano-composites in power amplifier packing and ceramic biomaterials like implantable microstimulators are typical solutions for these problems. On the other hand, ceramics may always be used combining with other kinds of materials. Ceramic knives have been established in this way, i.e., zirconia blades are joined to metal substrates. Over the past years, several authors using physical tests [2] have emphasized the importance of evaluating different alloys and ceramics, which have been designed [3] to reproduce the complex set of forces that act on the metal-ceramic interface [4]. In view of this, there has been an increased interest on the assessment of metal-ceramic bond strength, seeking the test that is closest to the ideal and can simulate real conditions as reliable as possible. Several tests have been used to evaluate properties such as color stability, tensile, shear, flexural and torsional strength [3,4]. In addition, despite the obvious advantage of ceramic powder metallurgy components the joining of sintered powder materials has been associated with difficulties related to their inherent characteristics, such as porosity, contamination and inclusions, at levels which tend to influence the properties of a welded joint.

In view of the above, a review of the current state of the art of welding ceramic powder metallurgy components to metal is presented. The intention is to identify preferred joining processes and identify technology gaps in joining of ceramic powder metallurgy components to different base metal alloys.

Comparison of joining methods

A summary of recent improvements on alternative approaches to ceramic-metal joining are presented herein. The present review also focuses on recent advances towards brazing ceramic powder metallurgy products and the selection of filler alloys, since in a scenario that includes joining by laser and direct bonding with transient liquid phases, brazing continues to be by far the most widely used approach to joining as a result of its low-cost and possibility to join intricate geometries for large-scale production. Examples of joined ceramic parts are illustrated in Fig. 1 [4, 5, and 6].

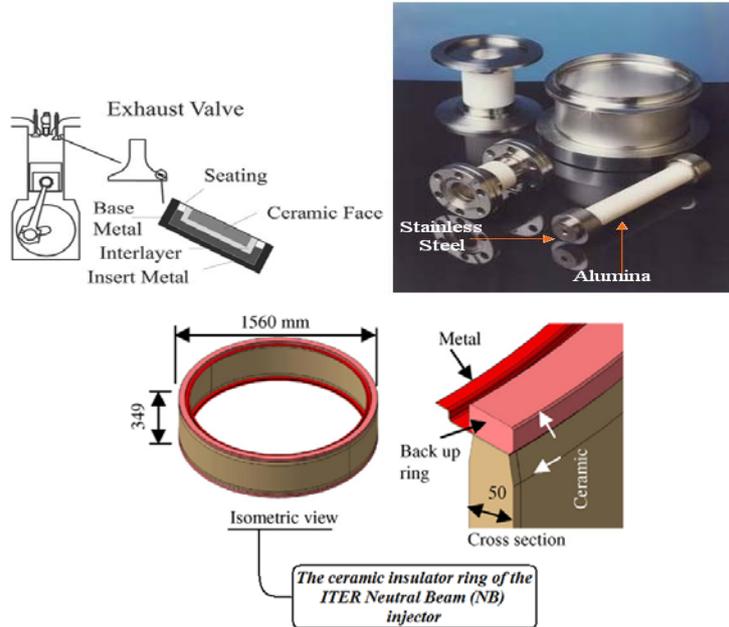


Fig.1. Ceramic-metal joint components [4, 5].

For the former approach, different routes from improving composites to modifying microstructures have been employed. The latter methodology basically involves the use of small parts made of monolithic ceramics joined together to structural metals. The development of residual stresses in joining dissimilar materials has been investigated (Fig. 2a, b). Ceramics with limited fracture toughness interface often rupture under the effect of such stresses. The key to a successful joint with dissimilar counterparts is the design of buffer interfaces capable of accommodating materials dissimilarities [3, 5].

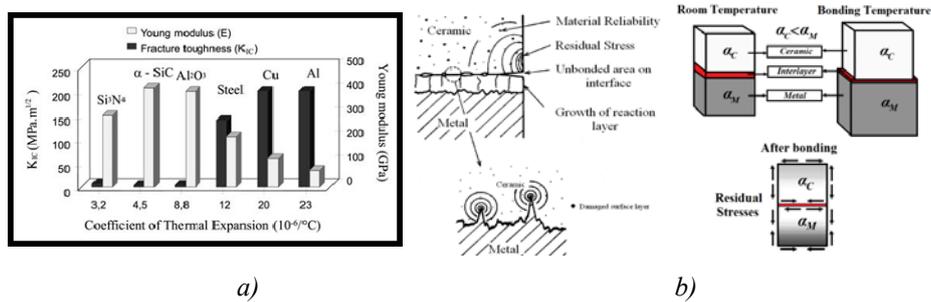


Fig.2. (a) Properties of ceramics and metals [5, 8] and (b) schematics of residual stresses developed during joining process.

Ceramics and metals can be joined together by mechanical, direct, or indirect processes [3, 7]. Table 1 presents a brief comparison of joining methods for

ceramics/metals [9]. Among the different methods of joining the ceramic powder metallurgy products, brazing and solid-state joining have excellent potentials for structural purposes because they can supply both good strength and heat resistance. Researchers, however, have been concentrated on oxide ceramics especially for application in electronic components for many years. Metallization such as so-called the Mo-Mn method, which is limited for oxide ceramics, is one of the most common processes for those applications and is widely used in the actual productions. Recent structural applications of ceramics require different type of joining techniques not only for oxide ceramics but also for non-oxide ceramics in order to produce the stronger and more reliable ceramic/metal joints, which are sometimes expected to have heat resistance superior than those obtained by the conventional metallization processes. From this requirement, a considerable number of joining techniques have been developed and reported for structural ceramics such as silicon nitride, silicon carbide, alumina and zirconia. Solid-state diffusion is a well-known example of a direct ceramic-to-metal joining process. Temperatures in excess of 1024°C are often possible for SiC-, Si₃N₄- and Al₂O₃-metal joints. Stresses are reduced by optimizing joint geometry and using a variety of interlayers such as refractory metals or functionally graded materials (FGM) [3, 8]. Friction welding is another example of solid state joining which is used extensively for this purpose [10].

Table 1. Comparison of joining methods for ceramics/metals.

Requirement	Unit	Adhesive	Cement	Mechan.	Recent Joining				
					Brazing	Eutectic	Diffusion	Fusion	Friction
Strength	MPa	20-80	<10	10-50*	100-500	200-400	100-1000	50-200	50-200
Heat Resistance		Poor	Medium	Medium good	Medium good	Good	Good	Good	Poor-Medium
Highest Temp.(°C)		177	----	427	527	927	>1027	>1027	227
Vacuum tight		Poor**	No	Good	Good	Good	Good	Good	----
Cost		Low	Low	Low-medium	Low-medium	Medium	Medium-high	Low-high	Low-medium
Problems		---	---	Stress Concentration	Oxidation resistance	---	---	Grain growth, Thermal Stress/shock	Only for Al
*This is not the interfacial strength of a butt- joint									
**e.g., due to outgassing of organic constituents.									

Application in ceramic powder metallurgy products/metal joining is still in its early stages of development. Successful ZrO₂/aluminum alloys joints have been reported [10]. Adhesive joining using organic interlayers offers suitable mechanical strength below 250°C. Glassy interlayers can also be used to improve high temperature resistance. In the latter case, ceramic powder metallurgy products are used as a protective barrier against corrosion of wear. Glassy interlayers have been long used to join Al₂O₃ to Nb in sodium vapor light bulbs [3, 8 and 11]. Brazed joints can be used in devices subjected to temperatures as high as 500°C with moderate mechanical strength (~100 MPa) [5, 8, and 11]. Structural ceramics such as Si₃N₄, SiC, Al₂O₃, AlN and ZrO₂ have been brazed to a number of structural metals and alloys [6, 12]. New and improved

filler alloys have been extensively studied aiming at lowering joining costs and producing refractory joints [12, 13]. The availability of reliable ceramic/ceramic and ceramic/metal joining processes and its effect on the expansion of the structural ceramics market to large scale use is a quite established concept by now [14,15]. Broadening the use of joining technology to new devices as well as improving joining to specific applications have always been the major goals [5, 13, and 16]. Recent improvements on ceramic rotor technology and their systematic use to reduce consumption and pollutant emission of Japanese automobiles have driven forward metal/ceramic joining and resulted in significant technological and scientific advances. Traditionally, shrink fitting and brazing have been used to join Si_3N_4 rotors to metallic shafts [12, 16]. On a more recent joint design, the ceramic rotor was shrink fitted to a metallic sleeve which was then friction welded to the shaft, resulting in improved bearing cooling and reduced residual stresses [16, 17]. Diffusion bonding with the formation of transient liquid phase and direct joining of metals to ceramics using laser are some of the most recent advances [9, 18 and 19]. Nevertheless, brazing is by far the most widely used joining process when mechanically reliable vacuum tight joints are required to operate at relatively high temperatures. Diffusion bonding has been developed specifically within the aerospace industry, and its industrial importance is such that it should be considered separately here. The process is used commercially for ceramic to metal joining, this material being one that exhibits superplastic properties at elevated temperatures within defined strain rate conditions. These conditions of temperature and pressure coincide with the conditions required for bonding, and therefore the two processes have been combined into one manufacturing operation either in sequence or together. The main advanced structural ceramics, i.e., SiC, Si_3N_4 , and Al_2O_3 have been brazed to a variety of metals and steels of engineering interest [5, 11, 16, 20, 21, and 22]. Some examples are shown in Table 3 [8, 20 and 23]. In another research, $\text{Fe}_{72}\text{Al}_{28}$ alloy was diffusion-bonded using a copper interlayer under vacuum at 1075 °C for 6 h durations at 3.2 MPa applied pressure [24] which results a sound joint because of the copper role in preventing the formation of stable aluminum oxides at the bond region. Moreover, Barrena et al. [25] successfully joined WC-Co cermet to 90MnCrV8 steel using 70Ni-30Cu with diffusion bonding in air.

Recent experimental details

Researchers such as Eager [11] have been trying to join a pair of SiC and molybdenum with vanadium foil as an insert material. The optimum fabricating conditions were a bonding temperature of 925°C, a bonding pressure of 30 MPa, and a bonding time of 10.8 ks. The joint had high shear-strength levels of 150MPa at room temperature and 52 MPa at 700°C. At the interface of SiC and vanadium, a layer of vanadium silicide, V_3Si was formed. In other research he also tried to join Si_3N_4 to nickel base super alloy (Inconel 718) with Ni interlayers attached to Ti and Cu microfoils at the side of Si_3N_4 by partial transient liquid phase bonding. It was demonstrated that this joining process can produce a more heat resistant Si_3N_4 -to-Inconel 718 joint than active brazing using Ag-Cu-Ti alloys. In addition no interfacial fracture between Si_3N_4 and filler metal occurred after cooling from bonding temperature of 900°C. It is also showed that the interfacial microstructure of the joint is unique in terms of interest combination of eutectic phases like $\text{Si}_3\text{N}_4/\text{TiN}$ and Ti_5Si_3 (~nm

thickness)/Ti₂Ni (~2μm thickness)/α-Cu (~1 μm thickness)/ CuTi₅Ni₁₄ and α-Cu and TiNi₃ (~8μm thickness)/ Ni (~498 μm thickness)/Inconel 718.

An investigation was conducted [8] to gain information about the capabilities of some filler metals for direct brazing of cubic boron steel during vacuum brazing with Ag-Cu-Ti active filler metal. Pressurization started at various temperatures (600, 700, and 800°C) and ended at room temperature during cooling. Results showed that there is an optimal starting pressurization temperature at which the maximum room temperature shear strength of joint is obtained. Scientists [26] successfully fabricated joints of Si₃N₄ and molybdenum with a vanadium interlayer using a vacuum hot pressing facility. The optimal joining conditions for producing a joint with the highest shear strength were found at 1055°C, a mechanical pressure of 20MPa and a bonding time of 5.4ks. In new approach a general method for brazing ceramics to metals using compliant metallic foam as a buffer layer has been developed [27]. Using stainless steel foams, bonds between alumina and stainless steel 316 with shear strengths up to 33MPa have been achieved. The resultant ductility enhances the resistance of the joint to thermal cycling; AlN-Inconel 600 bonds exhibited good thermal shock resistance [27]. As mentioned in this investigation, the thermal expansion of alumina and aluminum nitride is normally much smaller than those of most alloys. When forced to cool together, residual stresses resulting from the differences in thermal expansion can compromise the integrity of the joint. Thus, joints between alumina and a metal are less likely to fail due to thermal expansion mismatch than the joints between AlN and same metal. The problems can be minimized by optimizing the brazing conditions but the expansion mismatch is an intrinsic feature of the joint and cannot be avoided. As a result, buffer layers are sometimes used to make a gradual transition between the metal and ceramic. The compensation interlayer approach relies on inserting one or more interlayers with an expansion which is between that of the metal and ceramic being joined (Table 2).

Table 2. Examples of ceramic powder metallurgy product/metal joints and corresponding brazing approach.

<i>Ceramic/Metal</i>	<i>Interlayer</i>	<i>Application</i>	<i>Environment Condition</i>	<i>Strength, MPa</i>
Al ₂ O ₃ / Type 321 steel	Al	Vacuum devices, Electronic packaging	Vacuum/Argon	70
Tetragonal Zirconia Polycrystal/ Type 316 steel	Cu	Vacuum devices, automotive parts	Vacuum/Argon	52
MgO/Steel	Cu/Metal foam	Shuttle, aerospace, steel making(EAF)	Vacuum	33
Al ₂ O ₃ /Steel	BA03	Electronic packaging	Vacuum	50-150
SiC/Al	Cu-C fiber	Composite Component	770°C (Al was joined at 550°C)	---
TiN/ Mo	TiN/ Mo	CNC devices	1573,3 GPa, 30 min	80
Al ₂ O ₃ /Fe	FeO-Fe	---	1200°C, 29 MPa, 1h	---
Al ₂ O ₃ /Type 405 steel	Nb/Mo	Aerospace	1400°C, 100 MPa, 30 min	500

<i>Ceramic/Metal</i>	<i>Interlayer</i>	<i>Application</i>	<i>Environment Condition</i>	<i>Strength, MPa</i>
Si ₃ N ₄ /Steel	BA03/WC	---	610°C, 2Mpa, 30min	200
SiC/Steel	BA03/WC	---	610°C, 2Mpa, 30min	150
SiC/ Corrosion resistant iron–chromium–aluminium alloy (Fecralloy)	Thin pure Cu(12.5µm)	Spark plugs, piston heads and rocker arms in automobile engines	1100 °C, 30 min, 9.5 MPa	67.5(±5)
Sialon	BA03/WC	---	610°C, 2Mpa, 30min	300
Si ₃ N ₄ /Type 405 steel	Fe/W	---	1200°C, 10 MPa, 30 min	60
SiC/ Super alloy	Ni/Kovar/Cu	Gas and airplane Turbine	1050°C, 54 MPa, 2h	100
Si ₃ N ₄ / Super alloy	Ni/Kovar/Cu	Gas and airplane Turbine	1050°C, 54 MPa, 2h	---
SiC/ Type 316 steel	Ti/Mo	New generation of tank engine	810°C, 0 min	50
Si ₃ N ₄ /Type 405	Al/Invar(Cracking in intermetallic compound)	---	800°C, 0.15MPa, 7 min	60
ZrO ₄ /Steel	Active Metal Brazing (Ag-Cu-Ti)	Nuclear power plants	---	109-144
Al ₂ O ₃ /Type 304 stainless steel	97(Ag ₂₈ Cu) ₃ Ti: AFA	---	1000°C, 5 min	70
Alumina/Copper	Sn _{3.5} Ag ₄ Ti(Ce, Ga)	Electrical devices	>727°C	23.7
Si ₃ N ₄ /Inconel 600	Ag ₇₁ Cu ₂₇ Ti ₂	---	927-1467°C	30-100
Si ₃ N ₄ /nickel/Incoloy909, Inconel718	Au-5Pd-2Ni Ag-Cu-Ti Ni-B	Aerospace, military aims	827-1197°C	---

The use of more than one interlayer gives a smoother transition in thermal properties. A generalization of the multiple interlayer approaches involves the use of a functionally graded interlayer in which the material properties vary continuously across the thickness of the interlayer. Most of early work on metal foams was predominantly on the processing and manufacturing of aluminum foams [21, 28]. Foams of various steels and nickel-based superalloys have been also developed [29]. In this view the scientists used open-cell 316 stainless steel foam with 75% porosity as a flexible interlayer. In that study, a vacuum chamber was used and evacuated down to a pressure 10^{-4} to 10^{-5} mbar (Fig.3). Fracture surfaces of the brazed samples showed a cup and cone fracture within the ceramic after the first cycle whereas the latter failed from the

ceramic-foam interface after more than 60 cycles (see Fig.4). To summaries, mechanical and thermal tests indicated that the acceptable thickness of the foam is 0.2- 0.4 mm for the specific materials and joint configurations.

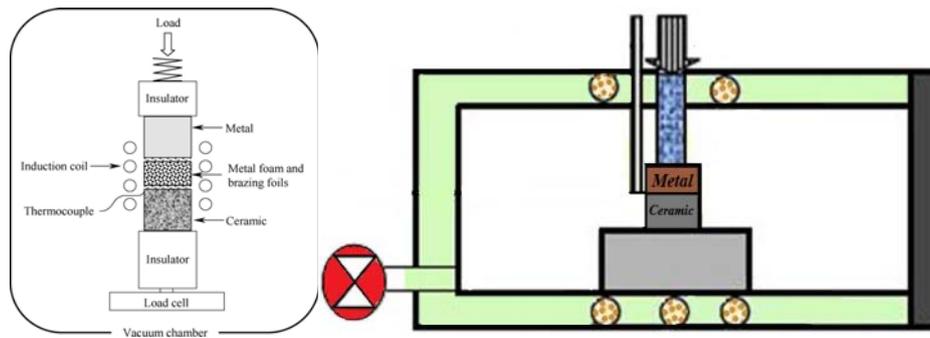


Fig. 3. Bonding set-up used for joining ceramics to metals with intervening brazing foils and foam.



Fig. 4. Fracture surfaces of joints without and with metallic foam following thermal cycling between 300 and 800°C in air [27].

It is worth noting that Ni-Ti composite was manufactured with a chemical composition of 51 % Ni – 49 % Ti (wt.%) as a mixture with an average dimension of 45µm diffusion layer, bonding was performed under argon atmosphere, with a constant load of 5 MPa, under the temperature of 850, 875, 900 and 925°C and, for 20, 40 and 60 minutes experimental time. In this case, at elevated temperatures diffusion mechanism was accelerated and diffusion period was decreased to achieve the same coalescence. In practical works diffusion bonding technology was applied to rotor core in hybrid cars, which requires both magnetic properties and mechanical properties at a time. Diffusion bonding of sintered materials and the bonding area as a factor affecting it were also analyzed. The satisfactory bond strength was accomplished by increasing the bonding area through the pressure by press fitting of the inner and outer components in addition to the difference in thermal expansion of the sintered material. On the other hand, diffusion bonding of Fe-5%Cu powder compact to wrought carbon steels was studied by Fillabi et al. [30]. The results of this study showed that a profound effect of carbon

content of the solid steel on the diffusion bonding process. It was suggested that the diffusion of carbon through the interface during sintering influences the swelling of the powder compact, thereby affecting strength of the bonding zone. A direct relationship between the depth of carbon diffusion and the maximum stress required to separate the bond interface was noticed. It was also found that there was an optimum iron particle size for maximum swelling with narrow carbon diffusion layer.

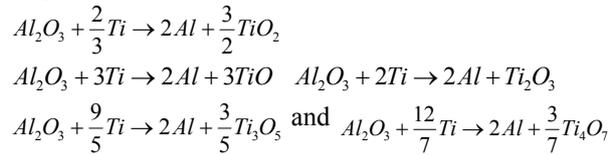
Alumina/Metal matrix composite (MMC)

Metallurgical behavior of alumina/alumina ($\text{Al}_2\text{O}_3/\text{Al}_2\text{O}_3$) joints and alumina/aluminum metal–matrix composite ($\text{Al}_2\text{O}_3/\text{Al-MMC}$) using an $\text{Al}_2\text{O}_3/\text{Al}_2\text{O}_3$ –Al composite as an interlayer was studied [20]. The $\text{Al}_2\text{O}_3/\text{Al}_2\text{O}_3$ joining procedures involved the sintering of silica or silicate glass on the surface of the Al_2O_3 , followed by the reactive penetration of liquid aluminum into the sintered glass to form an Al_2O_3 –Al composite interlayer between the two Al_2O_3 pieces. To join Al_2O_3 to Al–MMC, after the fabrication of the Al_2O_3 –Al composite layer on the surface of Al_2O_3 using the combination of sintering and reactive penetration, Al_2O_3 /composite/MMC joints were fabricated by using diffusion bonding between the Al_2O_3 –Al composite layer and the MMC. Experimental results showed that high porosity in the sintered glass layer led to the formation of a porous Al_2O_3 –Al composite layer between two Al_2O_3 pieces. The addition of cordierite ($2\text{MgO}\cdot 2\text{Al}_2\text{O}_3\cdot 5\text{SiO}_2$) to the pure silica lowered the melting point of the glass and led to the formation of a dense glass layer at the surface of the Al_2O_3 . Stronger joints were fabricated with shear strengths of the $\text{Al}_2\text{O}_3/\text{Al}_2\text{O}_3$ and Al–MMC/ Al_2O_3 joints of 105.2 and 47.6 MPa, respectively.

$\text{Al}_2\text{O}_3/\text{Cusil-ABA/Ti}$ and $\text{Al}_2\text{O}_3/\text{Cusil-ABA/Hastealloy X}$ joints

According to reference 31, the joining behavior of polycrystalline alumina sintered at different temperatures (1200–1500°C) and times (0.5–4 hr) to Ti, Hastealloy using two Ag–Cu active metal brazes (CuSil–ABA and TiCusil) containing Ti has been evaluated. Partial densification and the resulting high porosity content of Al_2O_3 substrates sintered at low temperatures led to braze infiltration of Al_2O_3 pores in 20 min contact but no penetration occurred in dense Al_2O_3 substrates sintered at high temperatures. The scanning electron microscopy (SEM) and energy dispersive spectroscopy examination of the joints showed that chemical reactions between Al_2O_3 and braze constituents formed a Ti-rich reaction layer at braze/ Al_2O_3 interface regardless of the sintering conditions and degree of densification of the Al_2O_3 [13, 31]. Interestingly, in this article authors tried to use numerical calculation for achieving good capillarity. The SEM views of the $\text{Al}_2\text{O}_3/\text{Cusil-ABA/Hastealloy}$ and $\text{Al}_2\text{O}_3/\text{Cusil-ABA/Ti}$ joints are shown in Fig. 5(a, b), respectively. Unlike the well-distinguished $\text{Al}_2\text{O}_3/\text{Cusil-ABA}$ boundary in these samples, the Cusil–ABA/Hastealloy and Cusil–ABA/Ti boundaries are rather diffused because of extensive dissolution and interdiffusion of the metal substrate in the molten braze at these boundaries. This has led to a rather gradual transition in the metallurgical structure of the joints at the Cusil–ABA/Hastealloy and Cusil–ABA/Ti interfaces. Nickel and titanium have very strong chemical affinity for each other. In addition, Ni extensively dissolves in Cu–Ag alloy melts. These factors will favor a gradual transition in the interface structure [31, 32]. The reaction layers in all the samples had particularly high concentration of Ti which

suggests the possible formation of titanium oxides at braze/alumina interface. These titanium oxides can be formed from the reduction of alumina by Ti according to the reactions given below:



Most of oxides of titanium are very stable as shown by the large negative change in Gibb's free energy, ΔG , for their formation at 850°C. In this work the values of ΔG , were calculated by the software HSC chemistry version 4.1 for the different oxides of Ti at 850°C. The relationship between infiltration depths, h of brazing infiltration time, t was written by the following formula [31]:

$$\frac{2\mu h}{\sigma} = \cos \theta \left[R_0 t - \frac{2H}{3} t^{1.5} \right]$$

where H is a parabolic reaction rate constant and R_0 is initial capillary radius at $t=0$ and μ is the viscosity, σ the surface tension and h is the penetration distance and the other term that is contact angle, θ , is well known in a number of ceramic/metal systems is decreased exponentially with time.

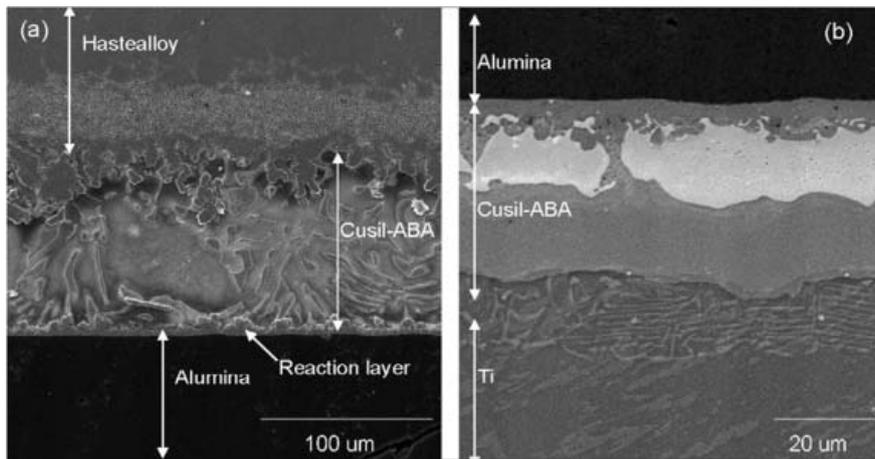


Fig. 5. (a) A Hastelloy/ Al_2O_3 joint made using Cusil-ABA and (b) a titanium/ Al_2O_3 joint made using Cusil-ABA. The Al_2O_3 substrates were sintered at 1400°C for 2 h [31].

Zirconia/Stainless Steel

In other experimental work, ZrO_2 ceramic/stainless steel joints were produced by pressureless brazing with Ag-Cu filler metals and TiH_2 powders. The results showed that there existed three zones with a distinguished difference in microstructure crossing the brazing interlayer. A double-layer structure including a reaction layer and a sublayer was formed at the ZrO_2 /filler interface, where Ti^{4+} , Ti^{2+} and Zr^{2+} were located based on the X-ray Photoelectron Spectra (XPS) [33]. It was further found that Ti originated from

TiH₂ coating diffused into the whole interlayer. The high activity of neonatal Ti caused the reactions of Ti/ZrO₂ and Ti/Cu, resulting in the interfacial phases such as Ti₃Cu₃O, CuTi₃ and Zr [34].

Al₂O₃/Ti-base filler metal/Ni system

The observation of the microstructure of Al₂O₃/Ti/filler metal/Ni system revealed that Ni-Cu solid solution has formed near the Ni/ filler metal interface. The composition of 75.72 at.% Ni, 17.91 at.% Cu, 2.92 at.% Ag and 3.3 at.% Ti with small amount of Al has been detected. This composition is feasible because Cu and Ni are soluble in each other in the solid as well as in the liquid state in the whole range of composition [35]. A number of intermetallics of the Ti-Cu system (TiCu, Ti₂Cu and Ti₂Cu₃) were observed. In most cases, the intermetallics showed some solubility for Ag, which replaces Cu in the superlattices of the Ti-Cu binary system. Two intermetallics, namely, Fe₂Ti and FeTi of the Fe-Ti binary system were also observed. Cu has been observed to replace Fe in the binary intermetallics of the Fe-Ti. Two intermetallics of the Ti-Ni system, namely, Ni₃Ti and Ti₂Ni were observed when more concentration was made on the faying surface. The formation of Ni-Cu solid solution has depleted the filler metal with Cu. A two-phase layer consisting of Ni₃Ti and (Ag) has formed just below the Ni solid solution layer. A dark layer of Ti₂Cu₃ (Ni) has been seen in this study. This layer was associated with large dispersed islands of silver below the Ti₃Sn⁺ (Ag) layer, consistent with the (Ag⁺ Ti₂Cu₃) phase field reported by Kubaschewski [36] in the Ag-Cu-Ti ternary system. The observed composition of this intermetallic is 39.23 at.% Ti, 44.86 at.% Cu, 15.7 at.% Ni and small amounts of Ag, Al and Sn were also present [18, 27]. The (Ag) solid solution has shown a composition of 86.94 at.% Ag, 10.65 at.% Cu and 1.48 at.% Ni with small amounts of Al, Sn and Ti [35, 38 and 39]. The Ti₂Cu phase shows a composition of 3.47 at.% Ag, 18.87 at.% Cu, 10.26 at.% Ni and 66.87 at.% Ti. Ni and Ag appear to substitute for Cu in the Ti₂Cu intermetallic [40]. Recently, D.Y. Chung et al. [41] used induction brazing process to join anode-supported tubular solid oxide fuel cells (SOFCs) to ferritic stainless steel using nickel based brazing alloys (BNi₂, BNi₄) modified by reactive TiH₂. The brazing alloy with TiH₂ showed good wettability with Y₂O₃ stabilized ZrO₂ (YSZ) electrolytes due to a formation of TiO_x layer. Pure alumina (99.99% grade compositions) also was brazed to 4J33-Kovar alloy using partial transient liquid phase bonding technique with nickel and titanium interlayers [42]. In the bond area, α-Ti solid solution, Ni₂Ti₄O and Ti₂Ni intermetallics were formed at two sides of the bond. The joint shear strength showed the highest value of about 65MPa.

In the end, it should be noted that the uses for the ceramic powder metallurgy products are changing quickly, with a major emphasis on structural applications. These materials are becoming more complex, in terms of being strengthened and toughened by transformation processes, the addition of other ceramic or metallic materials. The successful use of the materials requires the development of equally advanced joining technologies. Advanced ceramic powder metallurgy products include the electronic ceramics, such as those used as substrates and encapsulants, ionic conductors, piezoelectric devices, and high-critical-temperature superconductors needing to be joined to metals with new joining technologies.

Conclusions

Ceramic powder metallurgy products/metal interfaces are used in a variety of applications. This has prompted a concerted effort to develop fundamental structure and property relationships for these structures. This study aimed at revealing the recent development of joining methods of ceramics to metals. Brazing, ultrasonic welding, diffusion bonding, laser welding and partial transition liquid phase bonding were the most common techniques used for this purpose. Characterization and successful developments were briefly discussed.

In summary, however, in spite of extensive research on joining of powder metallurgy products to metals, there still should be more attention on the mechanism involved during the bonding of the substrates. Importantly, as multiple examples discussed earlier illustrate, the combined application of several experimental and theoretical approaches to a particular interface system has led to significant breakthroughs in the understanding of the structure and chemistry of ceramic powder metallurgy products/metal interfaces. Nonetheless, experimentally, the use of metallic foam as a buffer layer between ceramic and metal could be an effective way of avoiding thermal expansion mismatch between the two materials when bonded together by brazing.

Reactive penetration technique used to produce an Al_2O_3 -Al composite interlayer between Al_2O_3 and Al_2O_3 , and between Al_2O_3 and Al- Al_2O_3 metal matrix composite, where Al_2O_3 /Al- Al_2O_3 / Al_2O_3 joints and Al-MMC/Al- Al_2O_3 / Al_2O_3 joints were produced are still needing more development to predict the diffusion barrier and joining mechanisms.

ZrO_2 ceramic/stainless steel joints can be produced by the pressureless brazing with Ag-Cu filler metal and TiH_2 powder. A double-layer structure including a reaction layer and a sublayer can be introduced at the ZrO_2 /filler interface, where Zr^{2+} , Ti^{4+} and Ti^{2+} were detected.

In the Al_2O_3 /Ti/filler metal/Ni system a number of intermetallics (TiCu , Ti_2Cu and Ti_2Cu_3) of the Ti-Cu system were observed. In most cases, the intermetallics had shown some solubility for Ag, which replaces Cu in the superlattices of the Ti-Cu binary system.

Finally, it should be pointed out that an atomic-scale understanding of the role of impurities on adhesion needs to be found and a strategy developed to effectively describe the mechanism of bonding in different techniques.

References

- [1] K. Katayama, et al: SAETech. Paper Series, 861128, (1986)
- [2] R. W. Rice: Advances in Joining, Brook Hill Publ. Co., Chestnut Hill, MA, (1976), 69.
- [3] Katsuaki Sugauma, Materials Science and Engineering, National Defense Academy, Hashirimizu, Yokosuka, Kanagawa-ken, 239, Japan (1990).
- [4] K. Sugauma. Bull. Jpn. Inst. Met., 29 (1990), No. 12
- [5] M. Schwartz. Brazing. UK: Chapman & Hall; 1995.
- [6] G.W. Liu, et al., Journal of Alloys and Compounds, 470 (2009) 163–167.
- [7] N. Decrestofaro, C. Henchel., Weld J, 1978; 57(7):33 – 8.

- [8] R. M. do Nascimento, A. E. Martinelli, A. J. A. Buschinelli, Recent advances in metal-ceramic brazing, *Cerâmica* 49 (2003) 178-198.
- [9] M. G. Nicholas, and S. D. Peteves., *Scripta Mater.*, 1994, 31(8), 1091–1096.
- [10] Jolanta Zimmerman, et al., *Journal of materials processing technology*, 20 9 (2009) 1644-1653.
- [11] T. W. Eager, et al., Pergamon press, Oxford, 1207, 1994.
- [12] R. P. Messner and Y. M. Chiang., *J. Am. Ceram. Soc.*, 1990, 73(5), 1193– 1200.
- [13] N.Yu. Taranets, H. Jones, *Mater. Sci. Eng.*, 2004, A379, 251–257.
- [14] N. Eustathopoulos, et al. Pergamon 1998.
- [15] R. Asthana, et al. *Metall. Mater. Trans.*, 2002, 33A, 2119– 2128.
- [16] E. Saiz and A. P. Tomsia., *J. Am. Ceram. Soc.*, 1988, 81(9), 2381–2393.
- [17] J. P. Martins and F. Margarido, *Mater. Chem. Phys.*, 1996, 44, 156–169.
- [18] Murray JL. Calculations of stable and metastable equilibrium diagram of the Ag–Cu and Cd–Zn system. *Metall Trans*, 1984;15(A):261–8.
- [19] Murray JL. In: Massalski TB, editor. *Binary alloy phase diagrams*, vol. 1. Materials Park (OH): ASM International; 1986. p. 874.
- [20] J. Q. Li, P. Xiao, *Journal of the European Ceramic Society* 22 (2002) 1225–1233.
- [21] Proceedings of the Conference on Cellular Metals and Metal Foaming Technology, Papers on Applications and Industrialisation, Verlag Metall Innovation Technologie MIT Publisher, Bremen, Germany, June, 2001, pp. 5–62.
- [22] M. Rohde, I. Südmeyer, A. Urbanek, M. Torge, *Ceramics International* 35 (2009) 333–337.
- [23] J. A. Howe, *Intern. Mater. Rev.* 38, 5 (1993) 257.
- [24] O. Toruna, et al., *Materials Characterization*, 59(2008) 852-856.
- [25] M.I. Barrena, et al. *Materials Letters*, 63 (2009) 2142–2145.
- [26] Zhand Yong, et al., *Journal of Iron and Steel Research, International* 2006, 13(2): 01-05.
- [27] A. A. Shirzadi, et al., *Materials Science and Engineering A* 496 (2008) 501–506.
- [28] *Journal of Advanced Engineering Materials*, Special Issue on Metal Foams, 2000, vol. 2, No. 4.
- [29] H. Choe, D.C. Dunand, *Acta Materialia* 52 (2004) 1283–1295.
- [30] M.G. Fillabi, A. Simchi, and A.H. Kokabi, *Materials & Design* volume 29, 2008, Pages 411-417.
- [31] R. Asthana a, M. Singh, *Journal of the European Ceramic Society*, 28 (2008) 617–631
- [32] S. Mandal., et al., *Mater. Sci. Eng.*, 2004, A383, 235–244.
- [33] C. G. Levi, et al., *Acta Mater.*, 1999, 47(14), 3927–3934.
- [34] N. Shinozaki, *Trans. Joining Weld. Res. Ins. Osaka Univ. WRI, Jpn*, 2001, 30, 161–166.
- [35] I. Chakravarty, S.P. Gupta, *Materials Characterization*, 51 (2003) 235– 241.
- [36] Murray JL. In: Massalski TB, editor. *Binary alloy phase diagrams*, vol. 1. Materials Park (OH): ASM International; 1986. p. 942.
- [37] M. C. A. Nono, et al., *Mater. Sci. Eng. A*, 2006, 435–436, 602–605.
- [38] Murray JL. In: Massalski TB, editor. *Binary alloy phase diagrams*, vol. 2. Materials Park (OH): ASM International; 1986. p. 1763–4.

- [39] Voytovich, et al., *Scripta Mater.*, 2004, 51, 431–435.
- [40] RT. Cassidy., Monsanto Research, U.S. Dept. Energy; 1987 Oct.
- [41] Dong-You Chung, Yeon-Hyuk Heo, Seung-Bok Lee, Tak-Hyoung Lim, Rak-Hyun Song, Dong-Ryul Shin, *International journal of hydrogen energy*, 2010, 1-7
- [42] Chunguang Zhang, Guanjun Qiao, and Zhihao Jina, *Journal of the European Ceramic Society*, Vol. 22, No.13, 2002, 2181-2186.