



Advances in Joining of Ceramics

Edited by
Charles A. Lewinsohn
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Advances in Joining of Ceramics

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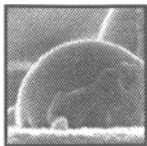
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Ceramic
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Volume 138

Advances in Joining of Ceramics

*Proceedings of the Joining of Ceramics symposium held at the
104th Annual Meeting of The American Ceramic Society,
April 28–May 1, 2002 in St. Louis, Missouri.*

Edited by
Charles A. Lewinsohn
Ceramatec, Inc.

Mrityunjay Singh
QSS Group Inc.
NASA Glenn Research Center

Ronald Loehman
Sandia National Laboratory

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Preface

Joining remains an enabling technology in several key areas related to the use of ceramics. Development of ceramic materials for electronic, biomedical, power generation, and many other fields continues at a rapid pace. The inherent cost of technical ceramics, as compared with metals, usually drives device designers to specify that ceramic components possessing desirable properties to be used only in critical locations in an assembly. Hence, joining of ceramics is a critical issue in the integration of ceramic components in engineering design.

The papers contained in this volume were a result of an international symposium on Joining of Ceramics held at the 104th Annual Meeting of The American Ceramic Society, April 28–May 1, 2002 in St. Louis, Missouri. Twenty speakers from North America, Europe, and Asia presented their papers during this symposium. Some of the papers reviewed the state-of-the-art of ceramic joining, whereas others presented results from recent studies concerned with developing new joining materials and methods. Several of the papers described innovative ways to model joint behavior and properties.

The papers in this volume were peer-reviewed and divided into four general categories: designing joints in ceramics, brazing, biomedical applications, and high temperature applications. The selection of materials for joints (Case and Greenhut), approaches to modeling joint properties (Takahashi and Stamille), and a review of the state-of-the-art of ceramic joining methods (Greenhut) are presented in the section on designing joints in ceramics. In the section on brazing there are two papers describing innovative brazing compositions and methods (Knowles and Weil). In the section on biomedical applications there is a review of the state-of-the-art (Agathopoulos) and two papers describing recent advances (Shin and Saiz). In the section on high temperature applications there are two papers on the development of joints for new applications of ceramic materials (Chou and Weil), a paper on new joining methods for silicon carbide-based ceramics (Lewinsohn) and a paper illustrating the use of ceramic joining to fabricate ceramic components (Kwon).

The editors thank all the contributors, speakers, and attendees at the symposium for making it such a rewarding forum for discussion of recent advances in joining. We also thank the staff of The American Ceramic Society for their support and assistance in publishing this volume.

Charles A. Lewinsohn

Mrityunjay Singh

Ronald Loehman

Designing Joints in Ceramics

SELECTION AND FUNCTION OF INTERLAYER MATERIALS IN CERAMIC/CERAMIC JOINING

Eldon D. Case

Chemical Engineering and Materials Science Department

Michigan State University

East Lansing, MI 48824

ABSTRACT

The ability to join ceramics with ceramics is important to both the design and function of ceramics. Ceramic/ceramic joining can allow ceramic components to be fabricated with a greater range of both external and internal morphologies (where "internal" morphology can refer to channels within the component, for example). Also, ceramic/ceramic joining can allow one to integrate ceramics of different properties and function into a single structure. A variety of interlayer materials and interlayer fabrication techniques have been used to successfully join ceramics. This paper reviews the function of interlayer materials and aspects of the choice of interlayer materials and/or interlayer application techniques in ceramic/ceramic joining.

INTRODUCTION

Ceramic/ceramic bonding has been of technological importance for a very long time. For example, joining stoneware cups and handles has long been performed by first "throwing" the cup from clay then allowing the cup to dry to "leather hard" consistency. A freshly pulled handle then can be joined to the cup by first abrading the intended joint region with a scribe and attaching the cup and handle using slip. This simple example highlights the use of joining to obtain morphologies that may not otherwise be practical or even feasible. More recently, many different ceramics have been joined, where the ceramics may be either in the "green state" (unfired) or densified prior to joining.

THE FUNCTION OF CERAMIC/CERAMIC BONDING

It can be difficult to machine or form intricate shapes in ceramic components. However, ceramic/ceramic joining can facilitate the fabrication of more complex shapes. It can be useful to consider the topic morphological complexity of

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ceramic components in terms of an external geometry or shape as well as an "internal geometry", which can consist, for example, of channels that penetrate the bulk of the component [1]. Such channels could serve as a conduit for a variety of fluids, including cooling fluids for electronic devices, medicine in bioceramics, fuel or coolant in engine components, and a variety of other uses.

Channels in ceramics can be formed by a number of different techniques. External channels in ceramics can be formed by cutting or by ultrasonic grinding [2,3], or by "stamping" or "pressing in" of channels into a powder compact [4]. Shallow channels (channels with submicron depths) can be formed by photolithographic techniques [5]. External channels with dimensions of up to several hundred microns or more can be formed by pressing fugitive phase elements into the surface of a powder compact, with the external channels formed when a fugitive phase is burned out of the powder compact [6,7]. Likewise, internal channels in ceramics can be formed by the burnout of fugitive phase elements that have been pressed into a powder compact, for example [6-8]. (In the case of the direct formation of channels using fugitive phase elements, it is important that the elements are "burned out" by a pre-sintering heat treatment that allows sufficient time for the volatile phase to escape from the powder compact. Otherwise, an over-rapid binder burnout can lead to bloating and cracking).

However, internal channels in ceramics also can be formed by joining ceramic components having external channels (Figure 1). In this technique, typically sintered ceramic specimens containing exterior channels are joined using an interlayer. This technique of forming internal channels by joining specimens with external channels has at least one inherent advantage over the method of directly forming internal channels by fugitive phase burnout; namely the external channels can be examined in detail prior to joining. Thus, the accessibility of external channels allows one to sort out defective channels or to make detailed measurements of channel dimension prior to joining the specimens.

When interlayers are used in ceramic/ceramic joining, there are a number of different methods of applying the interlayer, as will be discussed below. Also, the thickness of the interlayer can potentially be important in ceramic/ceramic joining. However, before we discuss the selection and function of interlayer materials in joining, we shall briefly discuss ceramic/ceramic joining for which no interlayer material is applied prior to joining.

SYSTEMS THAT JOIN WITHOUT THE USE OF AN INTERLAYER

The presence of an interlayer is not universally required for ceramic/ceramic joining. There are two significant types of ceramic/ceramic joining for which one does not apply an interlayer material prior to joining, namely:

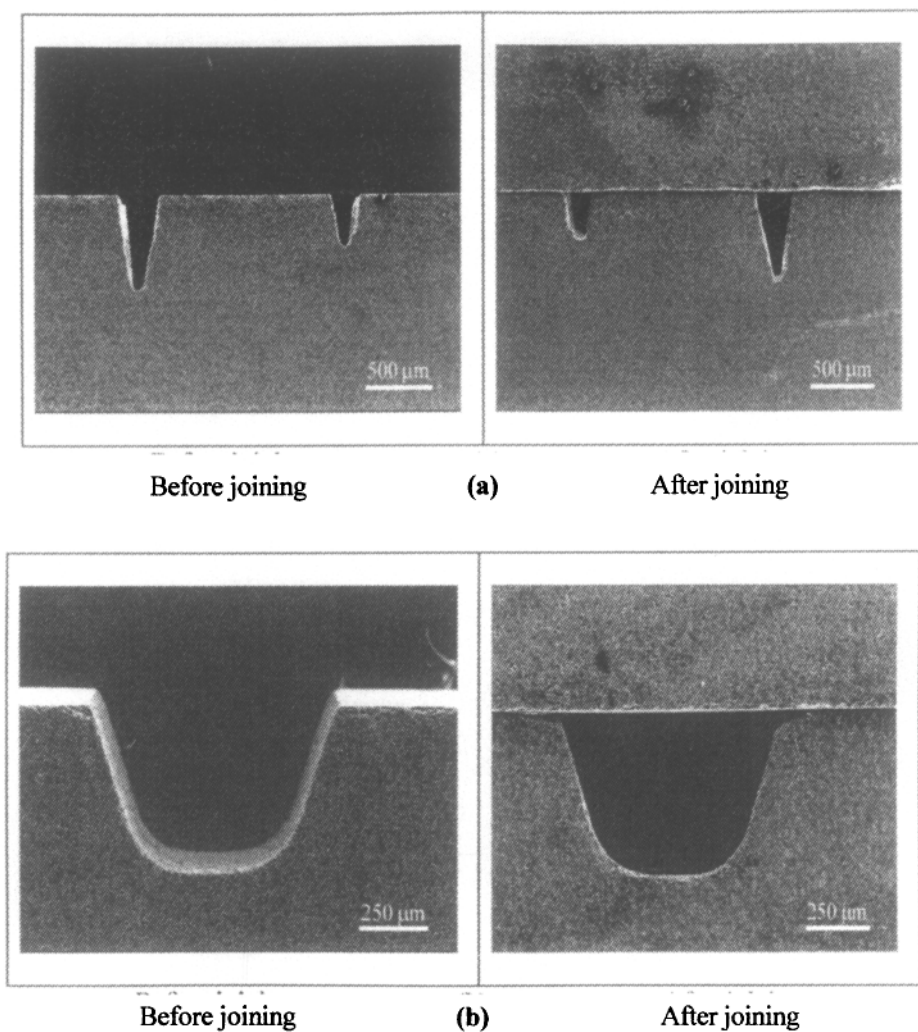


Figure 1. Microwave-joined specimens of (a) MaCorTM and (b) alumina (Reprinted with permission from [2], Copyright CRC Press, Boca Raton, Florida).

(a) "Freeform fabrication" of ceramics, in which multiple layers of ceramics can be joined by first pressing together sheets with significant loading of binders and plasticizers, then sintering the body, allowing for binder burnout. Thus, joining in the green state is facilitated by the binder phase. The specimen is then densified after the individual layers are joined in the green state.

(b) Diffusion bonding can be used to join densified specimens at high temperatures and applied pressures. Joining by superplastic deformation is a special case of diffusion bonding. Although interlayers or surface dopants are often used in diffusion bonding, diffusion bonding has been done without the presence of an interlayer.

Ceramic/ceramic joining via green tape (Freeform fabrication)

Ceramic/ceramic joining has been accomplished via the pressing together of "green tape" material [9,10], where green tape is a term used to describe ceramics that are impregnated with a binder phase and produced in flexible sheets. Green tapes typically incorporate binders such as polyvinyl butyral (PVB) [11,12] and plasticizers such as polyethylene glycol (PEG) [11] or dibutyl phthalate (DBP) [12], where the sheets are cut, stacked and pressed together to form a component.

For more than two decades, green tapes have been used in the fabrication of electronic ceramics, including multilayer ceramic capacitors and multilayer electronics packaging [13,14]. The binder and plasticizers must be removed prior to sintering the capacitors, but binder burnout can involve processing problems. For example, Tavernor et al. note that "the most common defects found in multilayer ceramic capacitors are derived from residual porosity formed when solvents and binders are released from a ceramic green body" [15]. Another difficulty with green tape materials is that (especially for components made of multiple layers) the time that is required for binder burnout can be very lengthy. For example, the binder burnout time for multilayer capacitors or multilayer ceramic piezoelectric components can range from tens of hours to more than one week [16,17].

Diffusion Bonding

Diffusion bonding can be used to join both metals and ceramics at high temperatures and pressures. In diffusion bonding, the mass transfer across the joined interface is due to solid-state diffusion, but additional interlayer materials that act as fluxes are sometimes also used in order to enhance the mass diffusion rates [18]. Diffusion bonding (and creep) can be a sensitive function of grain size. The tendency for both creep and successful diffusion bonding increases as the grain size decreases. For example, Elssner et al. showed that alumina plates with a mean grain size of 18 microns did not join, while similar alumina plates with a one micron average grain size joined when processed under nominally identical conditions [19].

Using superplastic joining at pressures of up to 35 MPa and a joining temperature of 1400°C, Dominguez-Rodriguez and co-workers [20, 21] were able to form bonds between partially stabilized zirconia specimens for which the interface could not be distinguished from the bulk material. Dominguez-Rodriguez et al. intentionally avoided the use of an interlayer material, terming it "deleterious" [20, 21]. The microstructure near the joint was apparently identical to the microstructure far from the joint. However, the superplastic flow induced plastic strains of up to 10 percent in the specimens [20, 21].

JOINING FACILITATED BY LOW MELTING POINT PHASES

Bonding can occur without an applied interlayer between ceramic components that include significant phase fractions of glassy or low melting temperature phases. In this case, the interlayer evolves in-situ during joining. More than a decade ago, Fukushima observed that microwave heating could induce joining between 92% and 96% purity alumina specimens, while 99% alumina specimens failed to join [23]. When Fukushima inserted billets of 92% alumina between the faces of 99% alumina specimens, the specimens did join. In addition, Fukushima was unable to join yttria-doped silicon nitride by microwave heating until a low-purity silicon nitride billet was inserted between the specimen faces [23]. The enhancement in microwave joining as the impurity level increases results from at least two factors. First, microwave heating is proportional to the dielectric loss of the material [37], and dielectric loss increases as the impurity level increases in a given material [4, 37, 42]. Second, the impurities often favor the formation of glassy grain boundary phases which can lead to a glassy interlayer being formed at the interface between the low purity specimen and another specimen.

More recently, Binner et al. [24,25] have shown that in both alumina and in silicon carbide, the presence of low melting point phases can affect joining behavior. For example, Binner et al. were able to microwave join RBSC (Reaction Bonded Silicon Carbide) specimens while more pure silicon carbide specimens did not join under similar conditions. In addition, in a series of micrographs for joints processed at a series of differing joining temperatures, Binner et al. observed a glassy phase that appeared to "bleed" out from the RBSC and form an interlayer that bonded the specimens together.

As demonstrated by the work of both Fukushima [23] and Binner et al. [24,25], low melting point phases can lead to the formation of interlayers that lead to joining. Thus, interlayers can in this circumstance lead to bonding, although interlayer materials were not externally applied to the specimens prior to heating.

HEATING TECHNIQUES FOR CERAMIC/CERAMIC JOINING

Ceramic/ceramic joining has been accomplished by conventional heating, microwave heating, capacitive discharge, laser heating, and frictional heating.

Microwave heating

Microwave heating has been used by a number of researchers to process ceramics [2,3,9,10,26-55]. The author and co-workers have used microwave energy in a variety of ceramic processing studies which has included sintering [9, 10,26-31], binder burnout [32,33], crack healing [34-36], modeling of refractory heating [37-41], thermal etching [42] and joining of similar ceramics, dissimilar ceramics and ceramic composites (Figures 2 - 6) both oxide ceramics [2,3,34,40, 43-48] and non-oxide ceramics [46, 49,50].

Laser heating

An example of a joining technique involving laser heating is gas-phase selective area laser deposition (SALD). In this technique, an interlayer is deposited in situ during the joining process [56]. Using the SALD technique for joining means that a laser beam is rastered over the joint region to induce the desired chemical reactions in the gas phase that is introduced into a vacuum chamber. For example, Harrison and Marcus [56] used SALD in which the laser heating induced thermal decomposition of tetramethylsilane and hydrogen gas mixture in a vacuum chamber to produce solid SiC that was used to join Hexoloy (Carborundum) tubes with an outer diameter of 0.95 cm.

Capacitive discharge joining

Alumina and PSZ (a partially stabilized 3 mol% yttria-zirconia) have been joined using a capacitor discharge system [57]. Specimens polished with 1 micron diamond paste prior to joining. Plate-shaped specimens were joined with specimen dimensions of 10 mm X 10 mm X 4 mm and 5 mm X 4 mm X 10 mm. The interlayer materials were Ti or Al foils or amorphous aluminum-rich $\text{Al}_a\text{Ni}_b\text{Y}_c$ foils, where the foil thickness was about 10 - 75 microns thick. The alumina joints were approximately 40 microns thick, with a significant porosity. However, the zirconia joint thickness were about 15 microns thick for the $\text{Al}_a\text{Ni}_b\text{Y}_c$ amorphous foil [57].

Frictional heating

Iijima and Watanabe [58] were able to join silicon nitride plates without an interlayer material using friction heating. Applying a 19kHz ultrasonic signal (with the direction of the vibration being parallel to the silicon nitride/ silicon nitride interface), Iijima and Watanabe achieved a silicon nitride/ silicon nitride joint within 10 seconds, where the highest joint strength achieved was approximately 33 MPa [58].

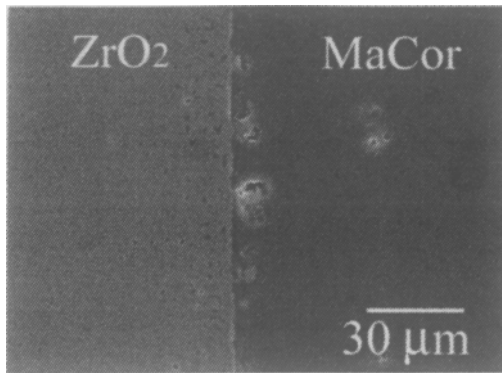


Figure 2. SEM micrographs of microwave-joined ZrO₂ with MaCorTM joined at 1020⁰C for 20 minutes, using a 20 gm dead weight (after [79]).

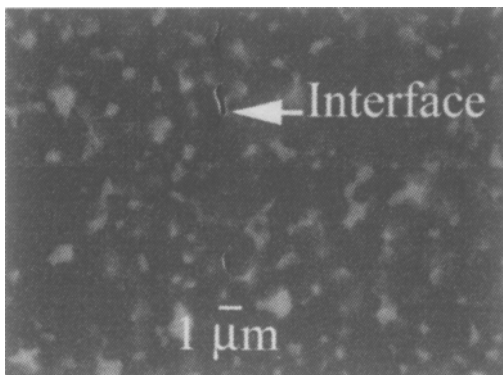
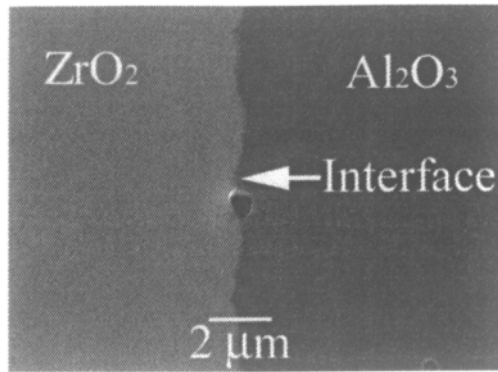
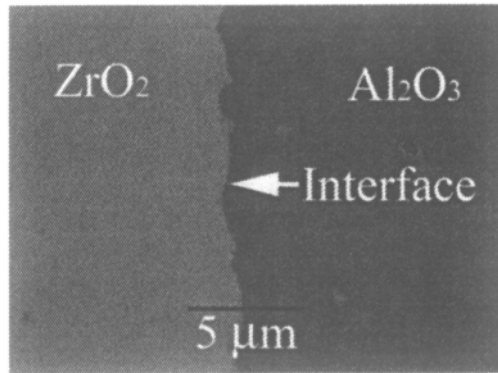


Figure 3. SEM micrographs of microwave-joined alumina/zirconia composites joined at 1450⁰C for 20 minutes (after [79]). Occasional porosity aids in locating the interface, but much of the interface was without porosity that could be resolved with the SEM.

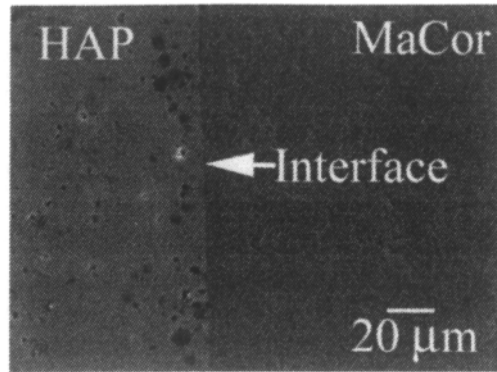


(a)

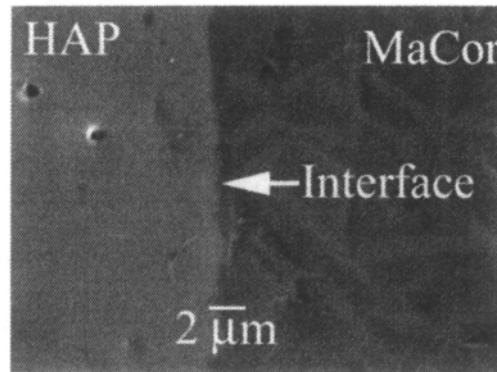


(b)

Figure 4. SEM micrographs of microwave-joined $\text{ZrO}_2/\text{Al}_2\text{O}_3$, using a 50 gm dead weight, joined at 1500°C for 20 minutes using a silica interlayer (Figure 4(a) is reprinted with permission from Journal of Advanced and Specialty Materials II, ASM International, Materials Park, OH, 44073-0002, Fig.3 pg 14, reference [45]). The same type of interlocking microstructure observed in the above two micrographs also were observed for conventionally joined $\text{ZrO}_2/\text{Al}_2\text{O}_3$ specimens [85].



(a)



(b)

Figure 5. SEM micrographs of microwave-joined MaCorTM and HAP, joined at 1020°C for 20 minutes using a 20 gm dead weight using a silica interlayer (after [79]). For the higher magnification micrograph (Fig. 5b), one can observe the randomly-oriented mica platelets in the MaCorTM microstructure.

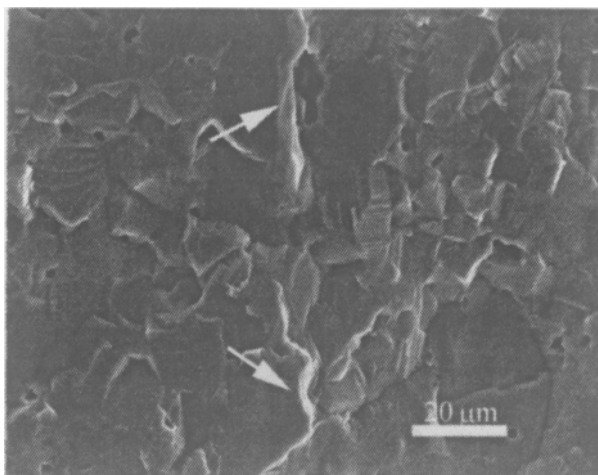


Figure 6a. The fracture surface of alumina specimens that were microwave-joined at 1625°C for 10 minutes. The arrows indicate the position of the joined interface (after [34]).

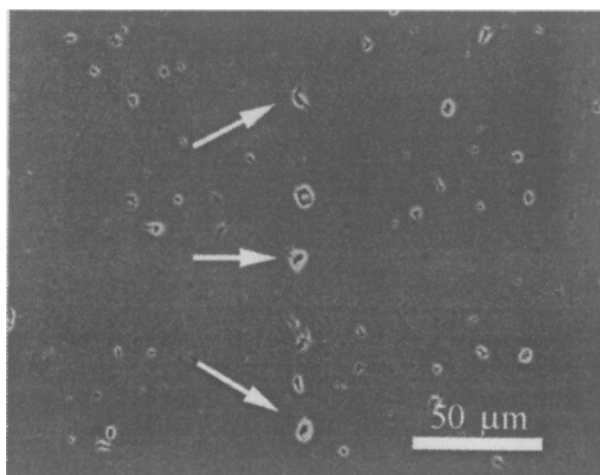


Figure 6b. A polished surface of the microwave-joined alumina specimen, joined at 1625°C for 10 minutes. The arrows indicate the position of the joined interface (after [34]).

APPLIED STRESS DURING JOINING

The applied stress during joining is a crucial processing variable. Many researchers apply high stresses during joining; especially those researchers that fabricate joints via diffusion bonding, superplastic deformation, or by hot pressing. For diffusion bonding and superplastic deformation, a high-applied stress is indispensable, since the mass transport that is involved is facilitated by creep conditions of high-applied stress at high temperature. Researchers that use metallic interlayers or other thick interlayers often employ hot pressing. If one is using initially thick interlayers, then the high applied pressures available via hot pressing can enable the foil or interlayer to conform to the gap between the materials to be joined.

A disadvantage of applying high stresses during bonding is the fixturing needed to apply the load to the specimen. In hot pressing (as well as in diffusion bonding and superplastic deformation) one needs to have sections of the load train adjacent to the specimen fabricated from materials that can perform under the temperatures and stress conditions

In addition to fixturing considerations, high stress at the joining temperature can induce creep in the specimens, depending on the times, temperatures, and the details of the creep mechanisms for the particular materials [20,59]. Of course, a problem that can be induced by creep is the loss of dimensional tolerances during joining. Joining by diffusion bonding does involve high-applied stress, and such stresses can lead to significant deformation of the specimens [20]. High-applied stresses sometimes are used even when interlayer materials are used. For example, many researchers have employed hot pressing to apply stresses in the range of 1 to 5 MPa in order to join materials for which an interlayer has been included.

CHEMICAL COMPOSITION OF THE INTERLAYER MATERIAL

A wide variety of materials have been used as an interlayer. For example, ceramic/ceramic joining has been successfully performed using metallic interlayers [57,60,61], ceramic interlayers [59,62], carbon-based interlayers [63] and glass interlayers [64-67].

Metallic interlayers

Metallic interlayers have been used to join a variety of oxide and non-oxide ceramics. A number of the metal interlayer joining processes involve reactive metal brazing, in which the metal reacts with the ceramic, forming a compound that is more readily wet by molten metal. For example, for the multilayer interlayers used by Young and Duh [68] to join AlN to AlN. AlN is not wet by Cu, Al, Ag, or Ni [56], but a Ni alloy that has been electroless-plated onto the AlN forms an AlNi compound that is more readily wet by molten metal than AlN.