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STRUCTURAL CERAMICS JOINING II

Arthur J. Moorhead • Ronald E. Loehman • Sylvia M. Johnson

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Edited by Arthur J. Moorhead
Oak Ridge National Laboratory
Ronald E. Loehman
Sandia National Laboratories
Sylvia M. Johnson
SRI International

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Preface

Ceramic materials have traditionally been used at relatively low temperatures, or under low tensile stress at high temperatures. Only in recent times have major efforts been made to use ceramics in structural applications that include a combination of conditions including significant tensile or flexural stresses, a range of temperatures, and corrosive environments. The ceramics that are being developed to meet these needs make up one segment of the broader class of materials that in the United States are referred to as advanced ceramics or high-performance ceramics, and in Japan are referred to as "fine ceramics." One of the main drivers for much of the interest in structural ceramics is the desire to improve the efficiency of transportation systems (utilizing advanced heat engines) and industrial processes (using ceramic heat exchangers to recover the waste heat from industrial furnaces).

In recent years it has become widely recognized that one of the key technologies that will enhance or restrict the use of ceramic materials in advanced structural applications such as in heat engines or heat exchangers is the technology to reliably join simple-shape components to form complex assemblies, join unit lengths of material to form large systems, or join ceramic components to metals. However, it has also become evident that the development and successful application of such joining technologies is difficult, costly, and only achieved by the combination of the thoughts and efforts of a worldwide body of experts in this field.

In order to facilitate the exchange of ideas on the science and technology of structural ceramics joining, the First International Forum on Structural Ceramics Joining was held in Pittsburgh, April 28-29, 1987, at the 89th Annual Meeting of the American Ceramic Society. Thirty-one peer-reviewed papers made up the proceedings of Structural Ceramics Joining I, which was published as the November–December 1989 issue of *Ceramic Engineering and Science Proceedings*.

However, given the rapid pace of research and development in this field, it is appropriate to periodically bring together the active participants for updates on their studies. Accordingly, researchers from around the world gathered on November 3-4, 1992, in San Francisco, for the Second International Forum on Structural Ceramics Joining, held at the Third International Ceramic Science and Technology Congress and Pacific Coast Regional Meeting of the American Ceramic Society.

A total of 37 papers were given, many of which are included in this volume. The forum also included invited presentations by two distinguished researchers in the field. Professor Nobuya Iwamoto of the Welding Research Institute, Osaka University, Osaka, Japan, gave an overview of the state of the art for joining ceramic materials in his country. Dr. Mike Nicholas of AEA Technology, Harwell Laboratory, United Kingdom, reviewed the material science aspects of the technology for joining ceramics using active metal brazing. All of the sessions were well attended, and the discussions were penetrating and informative.

This volume comprises the papers that were submitted for publication by the Forum participants and that were then peer-reviewed according to the normal procedures of the American Ceramic Society. We have arranged the proceedings into the same four sections as were used in the symposium sessions. Our goal, then and now, is that the papers present a logical progression of topics: Model Systems; Interfacial Reactions; Strength, Fracture, and Residual Stresses; and Novel Techniques. However, as is often the case, many papers could be placed equally well in different sections, and hence the sections divisions should be considered as general guidelines.

We would like to take this opportunity to thank all of speakers at the forum, and particularly those authors and coauthors who promptly provided us with manuscripts so that this volume could be published in a timely fashion. We would also like to thank those who chaired the sessions, and particularly those of you who reviewed the manuscripts. On behalf of the participants, we gratefully acknowledge the indispensable financial support provided by Robert B. Schulz, U.S. Department of Energy, and D. Ray Johnson, Oak Ridge National Laboratory, through the Ceramic Technology Project, U.S. DOE Office of Transportation Technologies.

Finally, we give special thanks to secretaries Billie Russell and Fay Christie, who cheerfully spent many hours assisting us in organizing the forum, sending out the many mailings, and managing the collection of drafts and final manuscripts.

Artie Moorhead
Ron Loehman
Sylvia Johnson

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Section I. Model Systems

REVIEW OF THE STATE OF ART IN JAPAN FOR JOINING CERAMIC MATERIALS

N. IWAMOTO

Welding Research Institute
Osaka University
Osaka, Japan

ABSTRACT

The state of art for joining ceramic materials in Japan is reviewed. In the application of ceramic turbocharger attached to a metal shaft the maximum usable temperature in thermal cycling was determined by acoustic emission. Furthermore the development of high temperature brazing filler metal is discussed based both on wettability and the standpoint of barriers to inhibit the diffusion of element to form intermetallic compounds at the interface. Potential applications for such joints include high efficiency heat-exchangers and high temperature fuel cells.

INTRODUCTION

Nitride ceramics, Si_3N_4 or AlN, are important materials for a variety of uses including turbochargers in automobile engines (Si_3N_4) and electronic uses (AlN). In general, Ag-Cu-Ti system brazing filler metal is being widely used for joining Si_3N_4 rotors to incoloy turbocharger shafts. It is understood that the effect of titanium in brazing filler metals is good wettability on Si_3N_4 as well as AlN. TiN is easily formed at the interface, however, the potentially serious problem of the formation of brittle Ni-Si intermetallic compounds is widely recognized.

Many investigators have studied only the kinds of intermetallic compounds formed when various transition metals are added to brazing filler metals. The phenomenon of segregation of the silver in solidified brazing alloy region was also confirmed. Such segregation means that the use temperature of metal-ceramic joints is limited by the melting temperature of segregated silver in the braze alloy.

To prevent the formation of brittle intermetallic compounds such as Ni_2Si or NiSi at the interface, the solution of using insert metals between Si_3N_4 and braze alloys has been proposed although it seems difficult to find an appropriate process for adding the insert so the possibility of many kinds of coating were examined.

The development of techniques for measurement of the residual stresses in joints has been accompanied through the application of fine-focused X-ray diffraction and ultrasonic microscopy. Also acoustic emission detection of cracks developing during thermal cycling and mechanical testing was established.

Research Project and Groups for the Joining Ceramic Materials in Japan

The Japanese Government via The Ministry of Education, Science and Culture funded research work titled "Advanced Materials Mechanical Properties " via Grants in Aid for scientific research from 1989 till 1993. Prof.T.Yokobori is head of this program. The research was conducted by four organizations with the author in charge of one group on the "Mechanical and Physical Properties of Joints and Advanced Composed Materials".

Our research aims were as follows:

1) Interfacial Structure Analysis
   Members: Prof. Y.Ishida (Univ. of Tokyo)
   Prof. T.Iseki(Tokyo Inst. Technology)
   Prof. T.Suga(Univ. of Tokyo)
   Mr. R.Maeda(Natl.Mechanical Eng. Inst.)
   Prof. N.Iwamoto(Osaka Univ.)
   Metal-Ceramic Systems: Nb/Al_2O_3, Si_3N_4/Ni,
   Si_3N_4/Braze Alloys, SiC/Braze Alloys
2) Morphological Structure Control of Interface
   Members: Prof. A.Okura(Natl.Inst. Space and Aeronautical Sci.)
   Prof. Y.Kagawa(Univ. of Tokyo)
   Prof. T.Nishizawa(Tohoku Univ.)
   Metal-Ceramic Systems, Metal-Metal Systems:
   SiC/Ti, Ni-base/ γ', γ/γ'
3) Wettability Problem of Braze Alloys
   Members :Prof. M.Naka(Osaka Univ.)
   Prof. K.Nogi(Osaka Univ.)
   Metal-Ceramic Systems:
   Al_2O_3/Braze Alloys, SiC/Cu, Si_3N_4/Molten Metals
4) Measurement of Static or Dynamical Fatigue and Destruction Behaviors
   Members :Prof. A.Otsuka(Nagoya Univ.)
   Mr. Y. Obata(Nihon Univ.)

Metal-Ceramic Systems:(Ceramic-Ceramic Systems)
Si_3N_4/Si_3N_4, SiC/SiC, Si_3N_4/Cu/Miled steel

5) Evaluation of Fracture Toughness, Finite Element Analysis of Thermal Stress and Stress-Strain Relation, Measurement of Dynamical Characteristics and the, Establishment of Fracture Model of Joints

Members :Prof. K.Ikegami(Tokyo Inst. Technology)
Prof. Y.Ichikawa(Tokyo Electrocommun. Univ.)
Prof. T.Suga(Univ. of Tokyo)
Prof. A.Okura(Natl. Inst. space and Aeronautical Sci.)
Prof. Y.Kagawa(Univ. of Tokyo)

The major results of these research tasks are summarized below.

Interfacial Structure Analysis

High resolution electron microscopic analysis of the interfaces of Nb:Al_2O_3 joints made using high vacuum solid state bonding at 1073K, showed the formation of compound CaO•6Al_2O_3 from the impure CaO in Nb and good epitaxial relationships. It was formed that if the interfacial orientations are so chosen that the ship plane of the metal is made parallel to the interface, the glide motion of the misfit dislocations may be preserved up to ambient temperature. Thermal stress relaxation in such an interface can be made even at ambient temperature and therefore metal-ceramic interfaces could be simple and strong.[1] Further, though Nb-Si_3N_4 joining was performed, the joint strength was very low. Such behavior was attributed to the formation of brittle Nb_3Si which precluded the relaxation of thermal stress and suggested the necessity for a diffusion barrier such as Al_2O_3.

In the joining of Si_3N_4 using Cu-Ti brazing filler metal, an increment of the bonding strength is considered due to homogeneous distribution of thin fine TiN particles without formation of bonding phases as well as stress relaxation effect of fine TiN particles between Si_3N_4 and the bulk crystals shown in Fig.1.[2]

The effect of friction in ultra high vacuum and surface preparation on joining strength was studied by Maeda. Application of sliding motion in ultra high vacuum was proved to be favorable to increase the joining strength.[3] Fig.2 shows the effect of the sputter deposition time on the bonding strength. Aluminum alloy was deposited on alumina surface by sputtering prior to loading bonding pressure. The strength increases up to sputtering time of 30 min and no further sputtering is effective to improve the

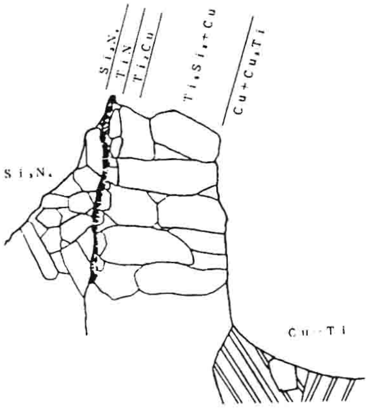

Fig. 1 Schematic representation of the interface between Si_3N_4 and Cu-Ti braze alloy.

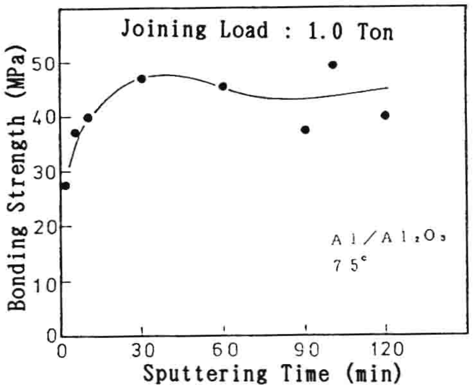

Fig. 2 Effect of sputtering of Ti/Ni bonding strength between Al and Al_2O_3.

strength, which suggest that etching or cleaning of the joining surface and full coverage of metal on ceramic surface are important. The effect of surface preparation was also studied using some surface polishing techniques such as electrolytic powder polishing and diamond polishing. The joining strength was greatly affected by ceramic surface conditions such as surface roughness and surface defects.

Electromobility Experiment

Minegishi has undertaken a study to clarify the interfacial segregation phenomenon of Ti in Cu-Ti braze alloys when joining AlN and ZrB_2 ceramics. He has found that if an electric circuit is constructed in the ceramic/reactive metal containing braze alloy /ceramic system, the ceramic could be polarized and behaves as cathode against the braze alloys. Accordingly, the reactive metal can migrate in the braze as a cation and segregate at the metal/ceramic interface. In Fig.3, the result of Ti segregation in a AlN/Cu-Ti/AlN sample held at 1373K for 600s with an electric current of 6.5mA is shown. It can be seen that remarkable segregation of Ti occurred at the interface between the Ti braze alloy and the AlN cathode.[4] He concluded that thickness of segregation at the interface of braze alloy and ceramic is in order of Ti/Zr/Hf, which is an inverse relation with the ease of compound formation based on the respective free energies.[5]

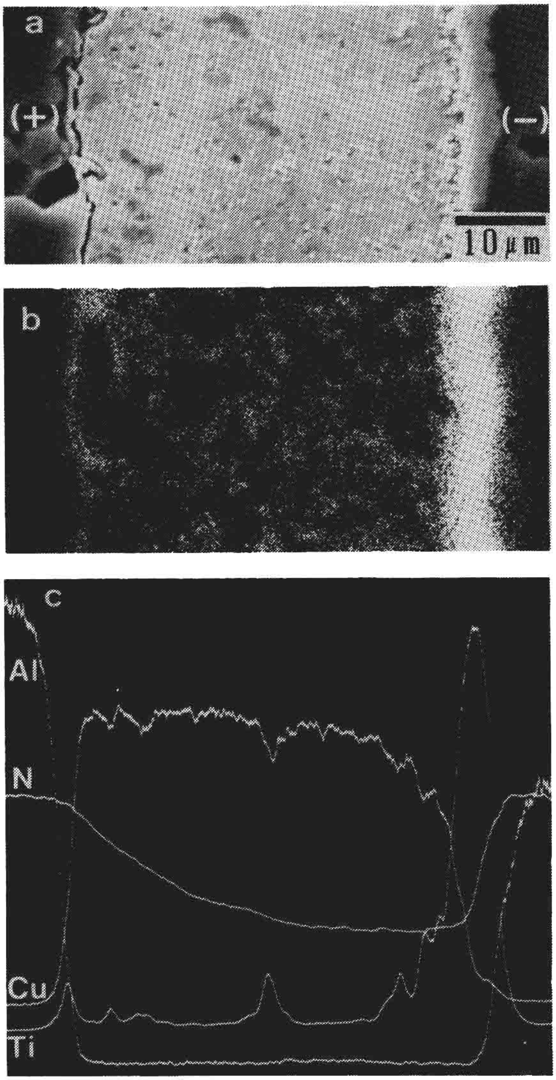

Fig. 3 Aspect of segregation of Ti at the braze-ceramic interface in the AlN/Cu-Ti/AlN system heat treated at 1373K for 600s under applide d.c. and examined by EPMA: (a)SEI, (b)Ti and (c)line profiles.

Wettability of Ceramics

Iseki studied the wetting behavior of Ag-Cu braze alloys on TiC, Ti_5Si_3 and Ti_3SiC_2. As shown in Fig.4, wetting angles on any of these compounds did not change with temperature variation. He considers that the additional effect of Ti in Ag-Cu braze alloys is not effective on wettability, but it behaves only as adsorbent at the interface.[6]

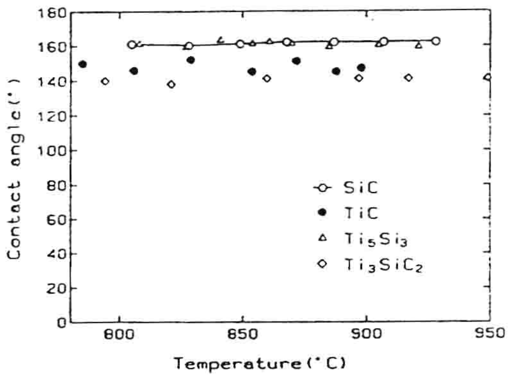

Fig. 4 Relationship between temperature and contact angle of molten Ag-Cu braze alloy on SiC, TiC, Ti_5Si_3 and Ti_3SiC_2.

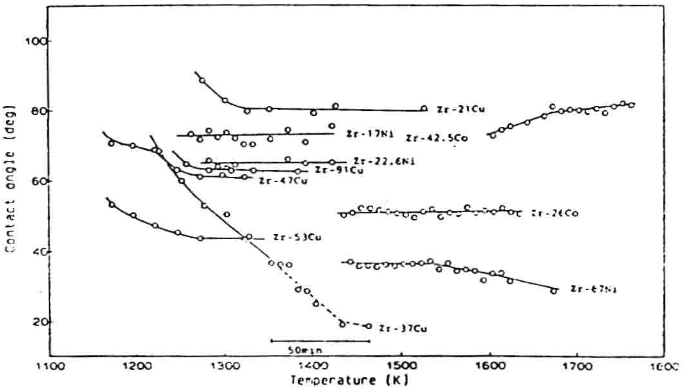

Fig. 5 Temperature dependence of the contact angle for Zr alloys against PSZ.

ZrO_2 ceramic is an important structural material and it is widely used for solid electrolytes and in fuel cells. In these hermetic seals, ZrO_2-Pt-Al_2O_3 solid state bonding was previously used. Bond strength was measured and also the interfacial reaction behavior was studied, however, a clear explanation for the reason for bonding is unknown.[7-10] Further it has been shown that molten metals do not readily wet ZrO_2.[11] At first, the author tried joining ZrO_2 with the use of oxide solder, but achieved only one case in which shear strength was over 100MPa.[12]

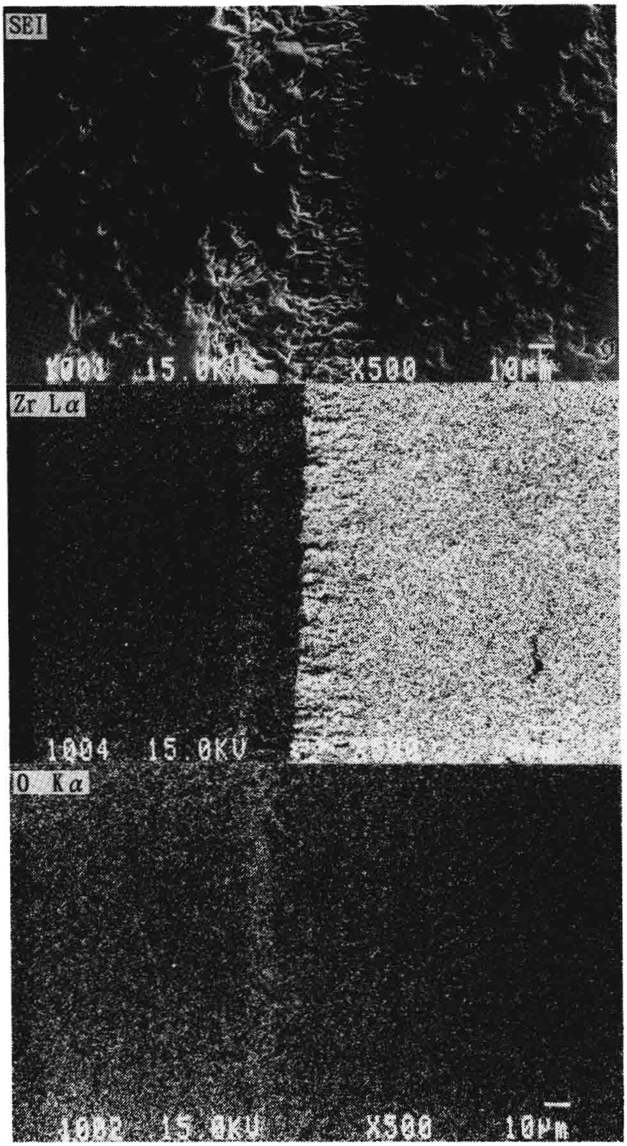

Fig. 6 Scanning electron micrograph and EPMA results of the interface of PSZ joined at 1273K for 1.8ks using Zr-17Ni alloy.