

Ceramic Joining

Edited by
Ivar E. Reimanis
Charles H. Henager Jr.
Antoni P. Tomsia





Geramic ransactions
Volume 77

Ceramic Joining

Edited by

Ivar E. Reimanis

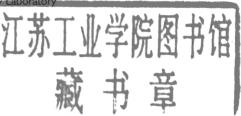
Colorado School of Mines

Charles H. Henager Jr.

Battelle Pacific Northwest Laboratories

Antoni P. Tomsia

Lawrence Berkeley Laboratory



Published by
The American Ceramic Society
735 Ceramic Place
Westerville, Ohio 43081

Proceedings of the Ceramic Joining symposium, held at the 98th Annual Meeting of the American Ceramic Society in Indianapolis, Indiana, April 14–17, 1996.

Copyright © 1997, The American Ceramic Society. All rights reserved.

No part of this book may be reproduced, stored in a retrieval system, or transmitted in any form or by any means, electronic, mechanical, photocopying, microfilming, recording, or otherwise, without written permission from the publisher.

Permission to photocopy for personal or internal use beyond the limits of Sections 107 and 108 of the U.S. Copyright Law is granted by the American Ceramic Society, provided that the base fee of US\$5.00 per copy, plus US\$.50 per page, is paid directly to the Copyright Clearance Center, 222 Rosewood Dr., Danvers, MA 01923, USA. The fee code for users of the Transactional Reporting Service for Ceramic Transactions Volume 77 is 1-57498-022-X/96 \$5.00+\$.50. This consent does not extend to other kinds of copying, such as copying for general distribution, for advertising or promotional purposes, or for creating new collective works. Requests for special photocopying permission and reprint requests should be directed to the Director of Publications, The American Ceramic Society, 735 Ceramic Place, Westerville OH 43081, USA.

For information on ordering titles published by the American Ceramic Society, or to request a publications catalog, please call 614-794-5890.

Printed in the United States of America.

1 2 3 4-01 00 99 98 97

ISSN 1042-1122 ISBN 1-57498-022-X

Related titles published by the American Ceramic Society:

Advances in Ceramic-Matrix Composites III Edited by Narottam P. Bansal and J.P. Singh © 1996, ISBN 1-57498-20-3

Handbook on Continuous Fiber—Reinforced Ceramic Matrix Composites Edited by Richard L. Lehman, Said K. El-Rahaiby, and John B. Wachtman Jr. © 1995, ISBN 0-931682-43-6

Handbook on Discontinuously Reinforced Ceramic Matrix Composites Edited by Keith J. Bowman, Said K. El-Rahaiby, and John B. Wachtman Jr. © 1995, ISBN 0-931682-42-8

Databook on Mechanical and Thermophysical Properties of Whisker-Reinforced Ceramic Matrix Composites Christian X. Campbell and Said K. El-Rahaiby

© 1995, ISBN 0-931682-56-8

Databook on Mechanical and Thermophysical Properties of Particulate-Reinforced Ceramic Matrix Composites Christian X. Campbell and Said K. El-Rahaiby

© 1995, ISBN 0-931682-57-6

Advanced Synthesis and Processing of Composites and Advanced Ceramics II (Ceramic Transactions Volume 79)

Edited by Kathryn V. Logan, Zuhair Munir, and Richard M. Spriggs © 1996, ISBN 1-57498-17-3

High-Temperature Ceramic-Matrix Composites II: Manufacturing and Materials Development (Ceramic Transactions Volume 58)

Edited by A.G. Evans and R. Naslain

© 1995, ISBN 0-944904-99-8

High-Temperature Ceramic-Matrix Composites I: Design, Durability, and Performance (Ceramic Transactions Volume 57)

Edited by A.G. Evans and R. Naslain

© 1995, ISBN 0-944904-98-X

Advanced Synthesis and Processing of Composites and Advanced Ceramics (Ceramic Transactions Volume 56)

Edited by Kathryn V. Logan © 1995, ISBN 1-57498-000-9

Advances in Ceramic-Matrix Composites II (Ceramic Transactions Volume 46) Edited by J.P. Singh and Narottam P. Bansal

© 1994, ISBN 0-944904-80-7

Advances in Ceramic-Matrix Composites (Ceramic Transactions Volume 38) Edited by Narottam P. Bansal

© 1993, ISBN 0-944904-69-6

For information on ordering titles published by the American Ceramic Society, or to request a publications catalog, please call 614-890-5890, or write to Customer Service Department, 735 Ceramic Place, Westerville, OH 43081.



The advent of new materials for engineering applications almost always brings a new challenge: how will these new materials be joined to a larger engineering structure? New ceramic materials are being developed for a wide variety of applications in areas such as power generation, energy conversion, automotive and aerospace, with specific applications including heat exchangers, fuel cells, turbocharger rotors, combustor liners, and for many other applications. Typically the new materials will be exposed to more hostile environments with respect to temperature, corrosion, and stress than materials in the past, and thus, many of the conventional joining techniques developed for less hostile environments do not work. Understanding fundamental issues in joining enables the development of new techniques to be able to utilize new materials.

A large part of the motivation for this symposium arose from a workshop organized through the Department of Energy's Center of Excellence in Joining, held on April 20–21, 1995, in Salt Lake City, Utah. The workshop defined fundamental and critical issues in ceramic joining and classified them into four general areas: joining techniques, joint failure, residual stress, and characterization and testing. The present international symposium is an effort to discuss some of these fundamental issues and to define areas for future research.

The large number of attendees at the symposium's oral presentations (the average number of attendees throughout the entire meeting was 32) is attributed to the high degree of interest in producing and understanding reliable joints. There were 40 technical papers in the Ceramic Joining Symposium, including seven papers that were part a joint session with the Fatigue and Reliability of Structural Ceramics and Composites Symposium and seven papers which were part of a joint session with the International Symposium on Manufacture Reliability Analysis and Applications of Functionally Graded Materials. Twelve out of the forty papers were given by speakers from either lapan, Korea, Europe, or South America.

The papers in this volume were peer-reviewed in accordance with the normal procedures of the American Ceramic Society. Three session topics cover all of the papers: Techniques and Overviews Chemistry and Reactions, and Mechanical Properties. The papers appear in the same order as they appeared in the symposium.

We thank all of the contributors to this volume.

Ivar E. Reimanis Charles H. Henager Jr. Antoni P. Tomsia



Techniques and Overviews
Joining Ceramic Materials
Ceramic Joining Issues in Advanced Technology Development
Development of a Compound for Low Temperature Joining of SiC Ceramics and CFCC Composites
Joining of Ceramics Using Flexible Interlayers
Recent Research Advances in Ceramic Joining in the Context of the Needs of the CFCC Systems
Chemistry and Reactions
Joining MoSi ₂ to 316L Stainless Steel
Brazing of Zirconia to Ti and Ti6AlV
Indentation and Oxidation Studies of Silicon Nitride Joints
Brazing of Si ₃ N ₄ with Au-Ni-V-Mo Filler Alloy
Brazing of Zirconia to Metal for Development of Oxygen and pH Sensors for High-Temperature, High-Pressure Aqueous Environments
Microstructure in Brazed Joints of Al ₂ O ₃ /SiC Composites 107 M.A. Zurbuchen and A.H. Carim

Joining SiC Ceramics Using Displacement Reactions
Interfacial Reactions and Diffusion Between Silicon Based Ceramics and Metals
Mechanical Properties
The Effect of Interlayer Properties on Residual Stresses in Ceramic-Metal Joining
Effects of the Limited Region of K-Dominance in the Mixed Mode Delaminating Beam Test Specimen
Effect of Plasticity on the Toughness and Fatigue Crack Propagation of Metal-Ceramic Interfaces
Advances in Friction Welding and Ultrasonic Welding of Ceramics to Metals
Development of an Interlayer System for High-Strength Ceramic/Metal Joints
Mechanical Properties and Microstructure of a Novel SiC/SiC Joint 185 Ö. Ünal, I.E. Anderson, M. Nostrati, S. Ijadi-Maghsoodi, T.J. Barton, and F.C. Laabs

Techniques and Overviews

JOINING CERAMIC MATERIALS

John A Fernie TWI Abington Cambridge CB1 6AL UK

Abstract

Joining is generally acknowledged as a key enabling technology if ceramics are to be accepted by industry. There are several different ways of joining ceramics to themselves and to metals, with considerable effort placed on understanding the relevant chemistry. This paper reviews current (and emerging) joining technologies, but concentrates on design issues associated with thermal expansion mismatch. Industrial examples are used to describe best practice and the role of interlayers in joint design and formation.

INTRODUCTION

The interest in using ceramic materials is high, with both monolithic and composite materials finding application in a number of industries, including automotive, aerospace, electronic and biomedical. Among the factors inhibiting more widespread use is the cost and difficulty in manufacturing complex components, either in one step or by joining simple shapes. In addition, ceramic components are rarely used in isolation and frequently need to be bonded to another ceramic or, more commonly, dissimilar material, most often metallic in nature. Hence joining technologies are required to join simple shapes to make complex components - and to join ceramic components to the outside world. Improved joining techniques should ensure that the joint is not the performance limiting "weak link" of the component.

To the extent authorized under the laws of the United States of America, all copyright interests in this publication are the property of The American Ceramic Society. Any duplication, reproduction, or republication of this publication or any part thereof, without the express written consent of The American Ceramic Society or fee paid to the Copyright Clearance Center, is prohibited.

JOINING TECHNIQUES

There are several possible methods of producing ceramic-ceramic and ceramic-metal joints. These may be generally categorised as either mechanical or chemical; there are advantages and disadvantages to both, depending on the final requirements of the component. Two major complications arise, particularly when joining dissimilar materials; in general metals and ceramics have very different co-efficients of thermal expansion (CTE), Fig 1., which can manifest as stress during thermal treatment or cycling, and secondly, the nature of the interatomic bond in ceramic and metallic systems is very different.

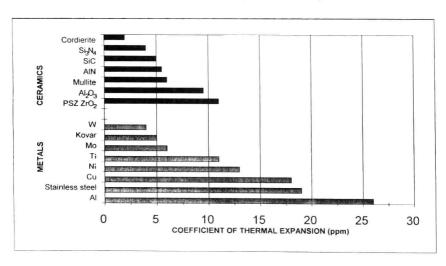


Fig 1. Co-efficients of thermal expansion for a range of metals and ceramics1

Mechanical joints can be used to incorporate ceramics into metallic or plastic structures. Designs tend to be relatively simple often using bolts, screw threads or shrink fitting; making assembly easy. However, machining of the ceramic can be expensive and care must be taken that the machined feature does not become a stress concentrator.

Chemical bonding is best achieved by producing intimate atomic contact at the interface and is frequently categorised as either solid or liquid phase. Solid phase bonding is achieved via atomic diffusion. In the absence of a liquid phase, the joint has to be made at sufficiently high temperature and pressure to produce intimate contact and provide sufficient thermal energy to cause diffusion and/or

chemical reaction. Surface flatness and cleanliness are therefore critical. Well known solid phase technologies include diffusion bonding and electrostatic bonding ^{2,3}.

Liquid phase bonding is achieved by introducing a material between the two surfaces to be joined, for example an adhesive or braze. Adhesive bonding is suitable for a number of applications below 150°C, and brazing when higher temperatures and hermeticity are important. Possibly the most famous adhesive bond is the space shuttle thermal protection system (TPS). The TPS consists of a low expansion/low thermal conductivity silica material (the heat shield) bonded to the aluminium structure of the shuttle. To overcome strains caused by thermal cycling (from the cold of space to the high temperatures on re-entry) and vibration during flight, a strain isolation pad (SIP) is used as an interlayer. The SIP is adhesively bonded between the silica tile and the shuttle structure. Adhesives could be used because the tiles had a protective coating (re-radiating 90% of the heat of re-entry), and any heat in the tile would take a significant period to diffuse to the bonded surface.

BRAZING

Brazing is an alternative to solid-state diffusion techniques and can be used at higher temperatures than adhesives, and is commonly used in industry. Brazing is a liquid phase joining process where a braze (a metal whose liquidus temperature is lower than the components to be joined) is placed between the two components, the entire assembly is then heated, usually in a controlled atmosphere or vacuum, to avoid formation of metallic oxides. The braze alloy is allowed to melt and flow between the components and thus produce a bond.

The problem most often encountered is that ceramics are relatively inert and are generally not wetted by liquid metals. This is a function of the relative surface energies of the materials concerned. Two methods are used to overcome this. The first is to "metallise" the ceramic component prior to conventional brazing. Metallisation treatments such as molybdenum-manganese (the most common), or titanium, are painted or sputtered on to the surface of the ceramic; this allows the braze to react and wet the ceramic. This approach is widely used, particularly in the electronics industry, for making gas tight seals and electrical isolation components. The second method uses 'active metal brazes' which contain deliberate additions of wetting agents, such as Ti and Zr, thus enhancing wetting.

ACTIVE METAL BRAZING

One of the simplest joining mechanisms for ceramic bonding is the use of ac metal brazing. There are a number of commercially available active braze all used in the joining of engineering ceramics such as alumina and silicon nitr An example is a silver-copper based braze which contains titanium as the ac metal. The Ag-Cu braze metal acts as a carrier for the titanium which diffinto, and reacts with, the surface of the ceramic. Indium can be added to red the eutectic temperature, if required.

The following sections outline case studies describing contrasting ways in what a ceramic-metal bond has been produced using active metal brazing a appropriate joint design.

Brazing Copper to Graphite

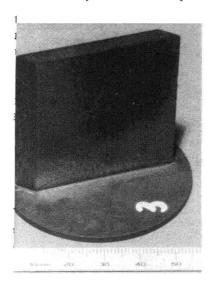
Copper-graphite assemblies can be used as targets for pulsed proton beams produce sub-atomic particles of interest when investigating atomic structure. T graphite targets, produced in a particular design for a number of years, had be brazed with a commercial Ni-Cr-P alloy. This braze was selected since it w known to wet onto graphite, with a required brazing temperature of 940°C.

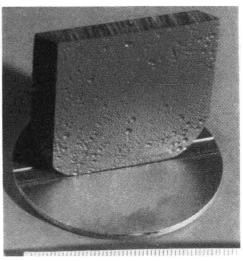
Successful shapes had previously been produced using a Ni-Cr-P braze describe earlier; however, when the brazing of "new" shapes was attempted, to alter the yield of particles, the graphite sections all failed as a result of cracking at the sharp edged corners, Fig 2a. Examination of the failed bonds showed classical residual stress cracks emanating from each corner of the graphite. This was due to both the large difference in (CTE) between graphite and copper (0.1x10⁻⁶/° and 18x10⁻⁶/°C respectively) and also the presence of sharp corners which a as stress concentrators. The ways used to resolve this problem were:

- i) a change in braze alloy to reduce the brazing temperature and hence reduce the thermal stresses induced during the heat treatment, a lowe temperature active metal braze based on Ag-Cu-Ti was used. Ag-Cu-Ti alloys are brazed at temperatures typically 100°C lower than Ni base alloys.
- ii) modification to the copper heat sink a change of dimensions to reducthe cross sectional thickness. This reduced thickness allows the copper to distort during cooling and so absorb stress more easily.

modification to the design of the graphite - removal of the sharp edges which act as local stress concentrators⁴.

successfully modified component is shown in Fig 2b.





 ξ 2. Showing a) a poor design with cracking at the edges and b) a successful nd produced using all three of the above described techniques 4 .

azing Ceramics to Metals Using Interlayers

e conventional way of overcoming the problem of thermal expansion smatch is to interpose a compliant layer between the two materials, such that e CTE of the layer is intermediate to those of the ceramic and metal, or to use layer of suitably graded composition, Fig 3. The depth of a solid compliant yer must be a significant fraction of its diameter. If not, the stresses built up ll either cause the bond to fail on cooling from the bonding temperature, or ll contribute to premature failure. The simplest interlayer is a single piece of aterial. Fig. 4 shows a shows a reaction bonded silicon carbide (SiSiC) burner zzle brazed to a stainless steel fixing⁴. A low expansion metal, in the form of ring, has been used along with a Ag-Cu-Ti active metal braze. In service, hough the hot end will reach temperatures of ~1400°C, the bond will only perience ~200°C, making this an acceptable design.

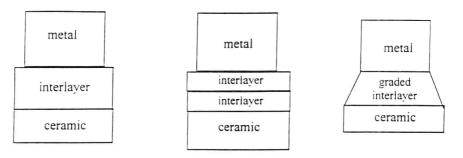


Fig 3. Designs for interlayers 1,4

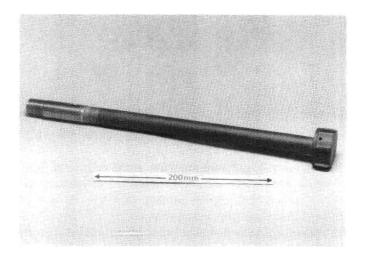
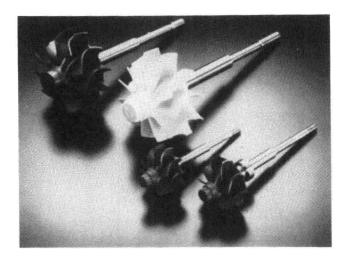


Fig. 4 A SiSiC burner nozzle (courtesy British Gas plc)⁴

A more complex design based on two interlayers has been used in the development of the ceramic turbocharger rotor ⁵ (Fig. 5). Again active metal brazing has been used to achieve the bond with low expansion metallics to absorb thermal expansion strains. The natural extension is the continuously graded interlayer. Such functionally graded interlayers would provide an excellent solution, but at present are economically not viable due to the high cost of production.



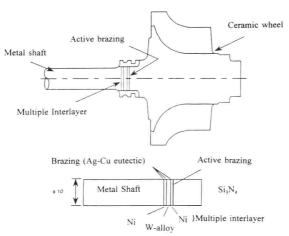


Fig. 5 Ceramic turbocharger rotors and bond design schematic

Mechanically Flexible Interlayers

In cases where weight (or volume) are critical then a heavy, solid interlayer may not be acceptable. In this case a non-solid, flexible, interlayer geometry may be used. A number of such flexible interlayer designs are being investigated including corrugated washers, honeycombs and dimples. These interlayers are produced from soft, ductile metals and alloys such as iron or stainless steel, and can also be manufactured in higher temperature materials such as nickel alloys.

They are then brazed into position. The important property of the interlayer is that it is ductile and able to absorb strain, caused by differing CTEs, during cooling from the joining temperature. This interlayer design is particularly versatile and has been demonstrated in a number of applications, such as ceramic faced tappets (Fig 6) and gas turbine components ^{6,7}.

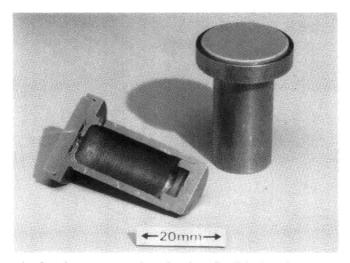


Fig. 6 Ceramic faced tappets produced using flexible interlayers

EMERGING TECHNOLOGIES

Brazing is only one of numerous techniques which can be used to produce ceramic-ceramic and ceramic-metal bonds. New techniques are constantly under development and among the leading contenders likely to play an increasingly strong role in joining are: friction welding, microwave bonding and glass-ceramics bonding.

Friction Welding

In the friction welding process, the two surfaces to be joined are aligned and made to rub together. The most common arrangement used in friction welding is where one of the components is held stationary while the other is rotated. The rotation provides friction between the two components when they are brought into contact, under uniaxial load. This not only generates heat at the interface, but also allows the break up of surface contaminants. Friction welding of aluminium to ceramics forms a soft/hard material combination such that the only