RESEARCH ON MICROWAVE JOINING OF SiC

LA-SUB--94-73

Final Report of Subcontract No. 9-XG1-U0554-1

Submitted to:

Materials Management Division
P.O. Box 990 Mail Stop P274
Los Alamos National Laboratory
Los Alamos, NM 87545

Submitted by:

Technology Assessment and Transfer, Inc.
133 Defense Highway, Suite 212
Annapolis, MD 21401

June 30, 1993

DISCLAIMER

ABSTRACT

Prior to this work, microwave joining had been demonstrated in oxide and nitride ceramics. These joints were as strong as the as received materials, were not detectable by microscopic observation, and the homogeneity in the vicinity of the joints was retained with little difference in the microstructure before and after joining. Preliminary work on microwave joining of sintered SiC (SSiC) as an alternative means to fabricate complex components and large assemblies from smaller and simpler shapes demonstrated that small samples could be joined using an interlayer of silicon, which was applied as a powder pressed between the specimens to be joined. Scanning Electron Micrographs (SEMs) of sections taken from these joints showed a smooth and homogeneous interlayer 50 μm in width. The objective of this contract was to optimize the properties of these joints through efforts to reduce the width of the interlayer and to match as closely as possible its thermal and mechanical properties to those of the silicon carbide.

The work performed under this contract demonstrated that the interlayer could be reduced to 10-20 μm using an oil-based slurry made from the Si powder, and to less than 5 μm by plasma spraying the Si on one of the SiC surfaces to be joined. Direct joints were then made in reaction bonded SiC (RBSC), using the residual Si present in the material. This included RBSC-RBSC and RBSC-SSiC. These results demonstrated that the bulk of the microwave joining results achieved earlier for ceramic oxides and nitrides could be extended to SiC. Excellent joints with good mechanical properties were obtained in both small specimens and in small scale tube assemblies that are prototypical of heat exchanger and radiant burner tubes. In situ reaction synthesis from powders to produce a SiC-TiC-SiC joint was demonstrated, as well as the feasibility of production of SiC from microwave-assisted decomposition of polymer precursors. Finally, new applicator designs, including a compound adjustable iris and a mitered bend single mode cavity, were demonstrated to provide improved heating of larger and longer specimens. This work provides the foundation for the scale-up of the microwave joining technique to SiC components needed for industrial applications.

1.0 INTRODUCTION AND BACKGROUND

Prior to this work, microwave joining had been demonstrated in oxide and nitride ceramics. Meek and Blake at Los Alamos had used a home microwave oven to join aluminum oxide to itself and to metal using a borosilicate glass interlayer (US Patents Nos. 4,529,856 and 4,529,857, July 16, 1985). Groups at QuesTech, Inc. [Palaith and Silberglitt, Mat. Res. Soc. Symp. Proc. **124**, 255 (1988)] and Toyota R&D Center (Fukushima, Yamanaka and Matsui, ibid., p. 267) used single mode cavity applicators to join mullite, and alumina and silicon nitride, respectively, to themselves. The samples were rods and tubes 0.32 cm and 0.95 cm in diameter, which were joined in minutes, as compared to hours using conventional techniques. These joints were as strong as the as received materials, were not detectable by microscopic observation, and the homogeneity in the vicinity of the joints was retained with little difference in the microstructure before and after joining. The power and compression requirements were modest (a few hundred watts and a few MPa) and the only surface preparation was that necessary for good contact and alignment.

The combination of high thermal conductivity, excellent thermal shock resistance and good corrosion resistance makes silicon carbide (SiC) an excellent material for heat exchangers, radiant burner tubes and advanced heat engine and pump components. The use of this material has been limited by the difficulty of sintering net shapes and the concomitant expense of machining complex components or fabricating large assemblies. Preliminary work on microwave joining of SiC as an alternative means to fabricate complex components and large assemblies from smaller and simpler shapes demonstrated that small samples could be joined using the same single mode applicator that was used to join mullite (Palaith, Silberglitt and Katz, Paper L3.11, MRS Spring Meeting, San Francisco, CA, April 16-21, 1990). An interlayer of silicon was used, which was applied as a powder pressed between the specimens to be joined. The specimens were disks of Carborundum Hexoloy™ approximately 0.95 cm in diameter and 0.64 cm in height. A Scanning Electron Micrograph (SEM) of a section taken from one of these joints is shown in Figure 1. It shows a smooth and homogeneous interlayer 50 μm in width. The objective of this contract was to optimize the properties of these joints through efforts to reduce the width of the interlayer and to match as closely as possible its thermal and mechanical properties to those of the silicon carbide.

2.0 SUMMARY OF WORK PERFORMED

The Statement of Work for this contract identified four tasks: Task 1 - Thinner Interlayer; Task 2 - Non-metal Compounds; Task 3 - Glasses & Glass Ceramics; and Task 4 - Experiments. Work performed on each task is summarized below.

2.1 Task 1 - Thinner Interlayer

The objective of this task was to reduce the width of the the 50 μm interlayer obtained using directly applied Si powder. As in the previous work, the specimens

were 0.95 cm diameter, 0.64 cm height sintered SiC disks. Two approaches were attempted and both were successful. First, the Si powder was mixed into an oil-based slurry using Nye watch oil. Joints were made by pressing together the two SiC disks with this slurry in between and heating the assembly to 1450°C in the single mode microwave joining apparatus for 5-10 minutes at approximately 250 watts of applied power. SEMs of several sectioned joined specimens showed joint interlayers of 10-20 μm. A typical SEM is shown in Figure 2. Second, a coating of Si was plasma-sprayed (at LANL) onto one SiC disk and this disk was sent to George Mason University and used in combination with an untreated SiC disk. A joint was made with these two SiC disks pressed together in the single mode microwave joining apparatus under similar conditions to those described above. SEM evaluation of this joint showed an interlayer width between 5 and 10 μm. (A joint was also made at LANL by placing a Si-coated SiC disk on top of an uncoated disk and heating the assembly in a multi-mode microwave cavity. SEM evaluations of this joint showed an interlayer width less than 5 μm.) Figure 3 is an SEM of a sectioned specimen joined with the plasma-sprayed Si method.

2.2 Task 2 - Non-metal Compounds

The objective of this task was to form non-metallic compounds in situ that can effect the joint. The work focused on attempts to react powder mixtures of Si, C and Ti. It was demonstrated at LANL that a compact of 2% Ti powder with a stoichiometric powder mixture of Si and C could be reacted in the multi-mode microwave cavity to form SiC. However, when similar powder mixtures containing up to 10% Ti were placed at the interface between two SiC disks and heated in the single mode microwave joining apparatus, the joints obtained looked very similar to those obtained in Task 1 and showed no evidence of the presence of carbides. Since the reaction $Ti + C \Rightarrow TiC$ is much more energetically favorable than $Si + C \Rightarrow SiC$, a cold-pressed disk made from Ti and C powders mixed in stoichiometric proportions was placed in between the SiC disks and heated in the single mode microwave joining apparatus. A strong reaction was observed, after which the SiC samples attained a temperature of 1500°C. This was held for several minutes. When the samples were cooled and sectioned, SEM and X-ray diffraction evaluation showed an interlayer of titanium oxides near the outer edge of the joined specimens and TiC over most of the central region of the joint. Figures 4 and 5 are SEM micrographs of sectioned joined specimens taken at the outer edge and interior of the specimen, respectively.

Following this initial success at forming a carbide interlayer through in situ reaction, the work focused on reacting powder mixtures of Si and C, with the objective of obtaining a homogeneous SiC interlayer that would bond to the SiC disks. Two SiC disks were pressed together with a mixture of Si and C powders in stoichiometric ratio placed in between. The assembly was then heated to a maximum temperature of 1540°C in the single mode microwave joining apparatus. The applied power was approximately 400 watts, and the sample was held at a temperature above 1500°C for 7 minutes. The applied pressure was modest--just enough to hold the assembly together. As shown in

Figure 6, SEM evaluation of the sectioned joint indicated that reaction occurred in the interior of the joint region. X-ray evaluation at LANL did indicate the presence of some SiC in the joint region. However, additional experiments using this powder approach were unsuccessful in producing the desired dense and homogeneous interlayer, and effort was shifted to the production of SiC in situ from the decomposition of chemical precursors, as described under Task 4 below.

2.3 Task 3 - Glasses & Glass Ceramics

The objective of this task was to investigate glass and glass ceramic materials as joining aids. Work was limited to literature review and discussions of potential compositions. In light of the success of the approaches described in Tasks 1 and 2 above, and the absence of glasses in the SiC material to be joined, this approach was not pursued further.

2.4 Task 4 - Experiments

This task comprised a major portion of the work performed under this contract, including interlayer material preparation and characterization, sample preparation, apparatus design and modification, and joining experiments and protocol development. SiC materials with different consolidation aides were investigated, in particular, reaction bonded SiC (RBSC), which is formed by infiltration of a preshaped preform of carbon and SiC with molten silicon. RBSC is nearly fully dense, with 5-15% residual silicon. It provides a less expensive and more formable alternative to sintered SiC (SSiC) at a slight sacrifice in service temperature and strength. The following text summarizes the experiments performed in three categories: applicator design; joining experiments; and synthesis of SiC.

2.4.1 Applicator Design

There were two improvements made in the LANL-furnished single mode microwave joining apparatus during this contract period of performance. The first was accomplished with support from the State of Virginia Center for Innovative Technology (CIT). A compound adjustable iris having both inductive and capacitive slits was designed, constructed and installed on the single mode microwave joining apparatus by Mr. Hussamaldin S. Sa'adaldin, a graduate student in Electrical and Computer Engineering at George Mason University. With this adjustable iris, improved coupling of microwave energy to SiC samples was obtained, allowing heating to 1650°C of rods of SiC filling the single mode cavity. When the sample was enclosed in insulation and oriented perpendicular to the electric field, the compound iris was demonstrated to provide a wide range of aperture settings to achieve critical coupling. For 600 watts of input power, reflected power was typically restricted to a few watts.

The second joining apparatus improvement was the design, fabrication and operation of a single mode mitered bend cavity operating in the TE_{103} mode. The cavity, which was constructed of WR-284 waveguide, is shown in Figure 7. As

indicated in the figure, choking structures were extended from the cavity walls to allow heating of long ceramic tubes without microwave leakage. Figure 8 shows a 3-D computer simulation of the electric field pattern in the mitered bend cavity. The simulation shows an extended area of maximum electric field, near the center of the cavity and in the vicinity of the bend. The mitered bend cavity is thus a good candidate for joining of long tubes extending through the walls with the joint region at the center of the cavity near the bend.

To test the mitered bend cavity approach, a sintered SiC rod of 0.9 cm diameter and 7.2 cm length was inserted along the axis of the bend and heated using the 0-1 kW 2.45 GHz microwave source. For these experiments, the compound adjustable iris was used and the mitered bend cavity was connected to an adjustable short. Input power was in the range of 400-550 watts and very good coupling was achieved (reflected power of 10 watts). The sample was heated to a temperature of 1660°C with 550 watts of input power within 4-5 minutes. Figure 9 illustrates the increased heating rate and higher temperature achieved for the same input power for the mitered bend cavity as compared to a rectangular single mode cavity.

2.4.2 Joining Experiments

Direct Joining of RBSC Rod and Disk Sections

Specimens of RBSC were joined to each other using both the single mode microwave joining apparatus supplied by LANL and a multi-mode microwave oven supplied by TA&T. The specimens joined in the single mode apparatus were sections of rods provided by Coors Ceramics of 0.95 cm diameter and 0.5 cm height. Joining was performed at temperatures of 1400-1450°C in 10-15 minutes. The compound adjustable iris was used in these experiments.

Several different RBSC specimen shapes and sizes were joined using the multi-mode microwave oven and an enclosure consisting of an alumina insulation lined on the inside with a thin layer of silicon carbide. This enclosure provided a combination of microwave and radiant heating. This hybrid heating technique provides very uniform heating and microstructure as demonstrated in microwave sintering experiments [De, Ahmad, Whitney and Clark, Ceram. Trans. **21**, 319 (1991)]. Specimens joined using this hybrid heating approach included rod sections similar to those joined in the single mode apparatus, as well as various configurations of whole and sectioned faucet disks provided by Laing Ceramics Corporation. Typical heating schedules were about 15 minutes to reach 1450-1550°C and a holding time of 20-30 minutes at this temperature.

Scanning electron micrographs (SEMs) of as cut sections of these RBSC-RBSC joints demonstrated excellent homogeneity across the joint interface, which was barely visible in SEMs taken at a magnification of 800. These sectioned specimens were then polished to further examine the interfacial region. Figure 10 shows an example of a micrograph of one of the polished sections. The interfacial region is identical to the bulk of the specimen, except for a silicon rich band of about 5 μm in width. Since

similar patches of silicon are evident throughout the RBSC material, the presence of this band should not substantially reduce the mechanical strength of the joint. In fact, micrographs of identical appearance were obtained for RBSC-RBSC joints that are as strong as the as-received material [Yiin et al, Ceram. Trans., **21**, 507 (1991)].

Direct Joining of RBSC Tubes

Short lengths (2.5 cm) of small scale (5 cm diameter) RBSC tubes intended for radiant burner tube applications were supplied by Coors. These tube sections were joined directly (butt joints with no interlayer material) in the 900 watt multi-mode microwave oven with hybrid heating. During joining, the tube sections were held at a temperature of 1450°C for 40 minutes. The joined tube section (shown in Figure 11) was installed on a vacuum line at a pressure of $12x10^{-3}$ torr, and held this level of vacuum as well as an as received tube. Joined tubes were cut into test bend bars and tested under four point flexure by Coors. Average mechanical strength was found to be 190 MPa, as shown in Table 1. The leak tightness and mechanical strength of these non-optimized joints are in an acceptable range for the radiant burner tube application.

Direct Joining of RBSC to Sintered SiC

The RBSC rod sections described above were butt joined to sections of Hexoloy™ SSiC using the single mode microwave joining apparatus. Typical time to reach 1450-1500°C was 5-8 minutes with 400-600 watts of power. The specimens were held in this temperature range for 10-15 minutes and then cooled by turning off the microwave power. Figure 12 shows a micrograph of a sectioned and polished joined specimen. A very homogeneous interlayer of about 5 μm in width is observed, similar in appearance to that observed in the RBSC-RBSC joints.

The joining of RBSC-SSiC would be especially useful in high temperature, high pressure heat exchangers, where the SSiC would be used only in the highest temperature sections and the low cost, easily fabricable RBSC would provide the transition to the metal sections. As a feasibility demonstration for RBSC-SSiC tube assemblies, a SSiC tube of 1" outer diameter was joined to a RBSC socket of 1" inner diameter and 1.5" outer diameter. To obtain leak-tight behavior, a butt joint was provided along the gas flow direction with a RBSC end cap of 0.5" inner diamter. This tube assembly, which is illustrated schematically in Figure 13, was joined in the 900 watt multi-mode microwave oven using hybrid heating. Leak-tight behavior was demonstrated before and after cycling between ambient temperature and 1100°C. As shown in Figures 14 and 15, SEMs of the butt joints are identical to those of the joined RBSC-RBSC tubes that had average mechanical strength of 190 MPa (Table 1). The joining of the RBSC-SSiC tube assembly was supported by a DOE SBIR contract; the evaluation of the joints was performed under subcontract 9-XG1-U0554-1.

2.4.3 Synthesis of SiC

An investigation of the effect of microwave heating on the production of SiC from polymer precursors was begun under this contract. Two different types of precursors were used: commercial polycarbosilane precursor material was purchased from Dow Corning, and precursors were prepared in-house via sodium reduction of trichloromethylsilane. Thermal decomposition of both precursor materials was performed first to provide a baseline for microwave processing. Infrared spectra of the products demonstrated the presence of SiC for T>1100°C. The commercial precursor was then subjected to microwave heating. The polycarbosilane precursor material was placed inside a quartz crucible, which was then placed at the (unloaded cavity) maximum electric field position of the TE_{103} single mode rectangular cavity. Alumina insulation was placed around the crucible, and the temperature was monitored through a hole in the insulation with a two-color infrared pyrometer. The temperature of the precursor was brought up to 1500°C and a product similar to that obtained by conventional heating was obtained. X-ray analysis demonstrated that the product was SiC. The microwave-assisted decomposition process was observed to provide a yield comparable to the conventional process. These results demonstrate considerable potential for the use of a polymer precursor as an in situ source of SiC during microwave joining.

3.0 SUMMARY AND CONCLUSIONS

The work performed under this contract demonstrated that the bulk of the microwave joining results achieved earlier for ceramic oxides and nitrides could be extended to SiC, first using Si-based interlayers, and then without the need for any interlayer material, using the residual Si present in RBSC. Excellent joints with good mechanical properties were obtained in both small specimens and in small scale tube assemblies that are prototypical of heat exchanger and radiant burner tubes. In situ reaction synthesis from powders to produce a SiC-TiC-SiC joint was demonstrated, as well as the feasibility of production of SiC from microwave-assisted decomposition of polymer precursors. Finally, new applicator designs, including a compound adjustable iris and a mitered bend single mode cavity, were demonstrated to provide improved heating of larger and longer specimens. This work provides the foundation for the scale-up of the microwave joining technique to SiC components needed for industrial applications.

4.0 PUBLICATIONS

1. I. Ahmad, R. Silberglitt, W.M. Black, H.S. Sa'adaldin, Y.L. Tian and J.D. Katz, "Microwave Joining of Dissimilar SiC Ceramics," presented at the 1993 ACerS Annual Meeting, Ceramic Transactions (in press).

2. H.S. Sa'adaldin, W.M. Black, I. Ahmad and R. Silberglitt, "Heating of SiC Rods in a Mitered Bend Single Mode Cavity," presented at the 1993 ACerS Annual Meeting, Ceramic Transactions (in press).

3. I. Ahmad, W. M. Black and R. Silberglitt, "Direct Microwave Joining of Reaction Bonded Silicon Carbide,", Ceramic Engineering and Science Proceedings, Vol. 13, No. 7-8, pp. 520-527 (1992).

4. H.S. Sa'adaldin, W.M. Black, I. Ahmad and R. Silberglitt, "Efficient Coupling with an Adjustable Compound Iris for Joining of SiC in a Single Mode Applicator," Materials Research Society Symposium Proceedings, Vol. 269, pp. 91-96 (1992).

5. I. Ahmad, R. Silberglitt, W.M. Black, H.S. Sa'adaldin and J.D. Katz, "Microwave Joining of SiC Using Several Different Approaches," Materials Research Society Symposium Proceedings, Vol. 269, pp. 271-276 (1992).

6. R. Silberglitt, D. Palaith, W. M. Black, H. S. Sa'adaldin, J. D. Katz and R. D. Blake "Investigation of Interlayer Materials for the Microwave Joining of SiC," Ceramic Transactions, Vol. 21, pp. 487-495 (1991).

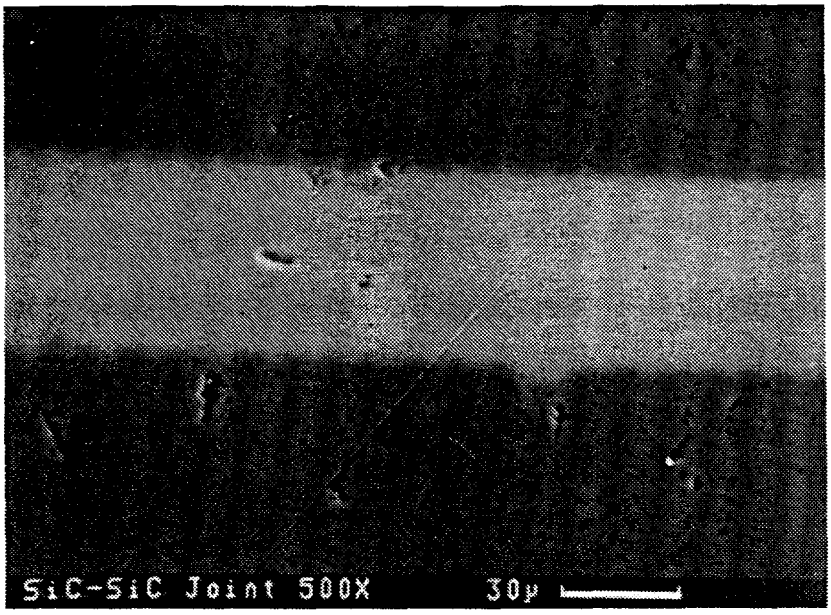

Figure 1 SEM micrograph of SiC joint made with Si powder.

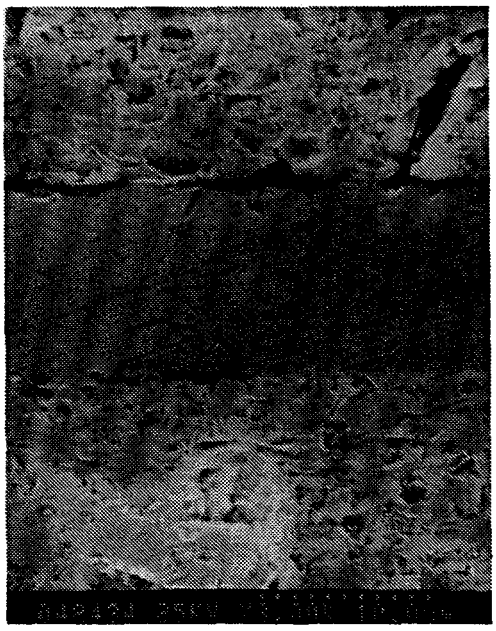

Figure 2 SEM micrograph of SiC joint made with oil-based slurry of Si powder.

Figure 3 SEM micrograph of SiC joint made with plasma-sprayed Si coating.

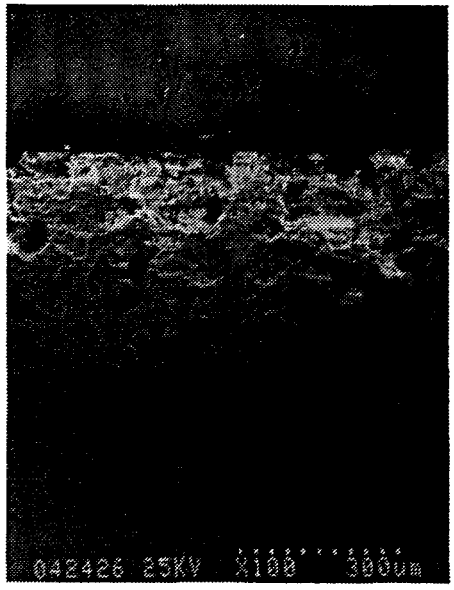

Figure 4 SEM micrograph of SiC joint made with cold pressed disk of Ti:C (outer edge).

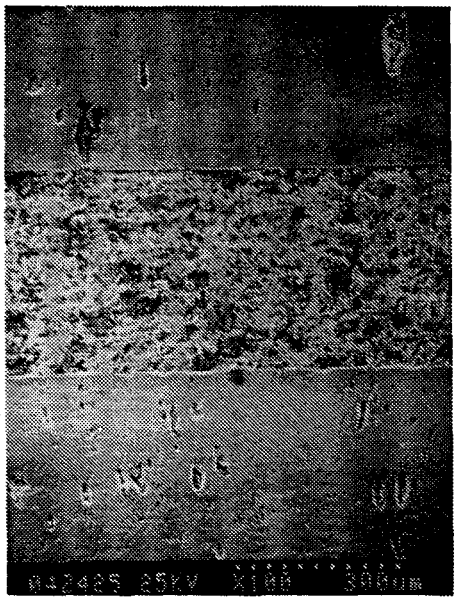

Figure 5 SEM micrograph of SiC joint made with cold pressed disk of Ti:C (interior of sectioned specimen).

Figure 6 SEM of SiC-SiC Joint Made By Combustion Synthesis of Si and C Powders (Interior of Specimen)

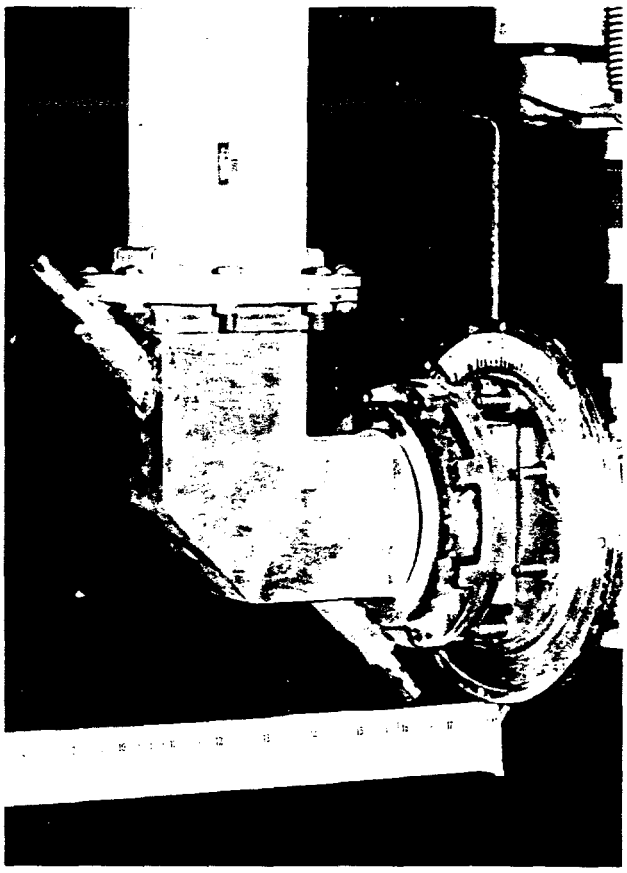

Figure 7 Mitered Bend Cavity Applicator

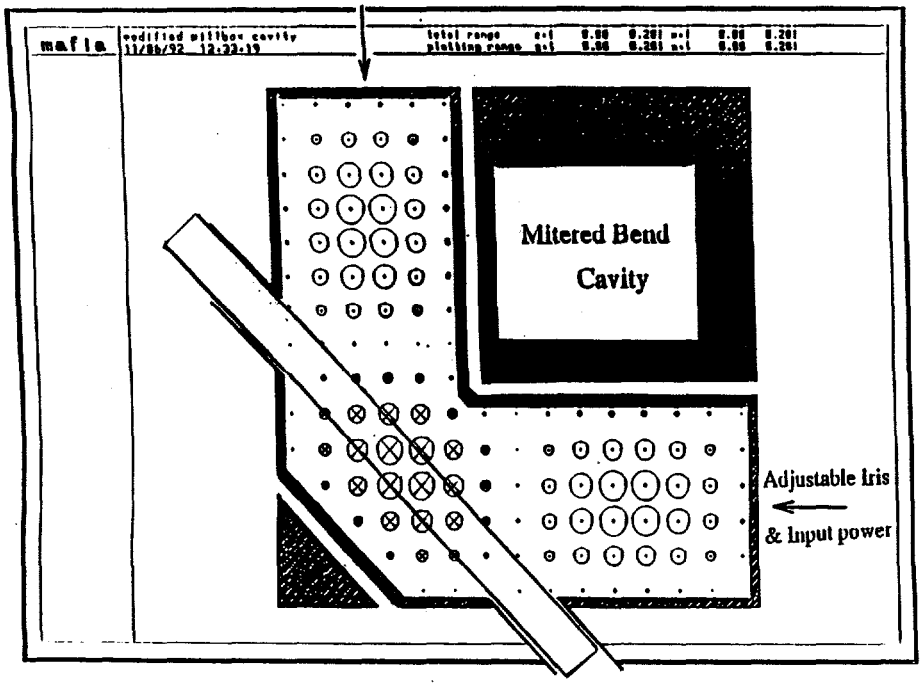

Figure 8 Electric Field Pattern in Mitered Bend Cavity

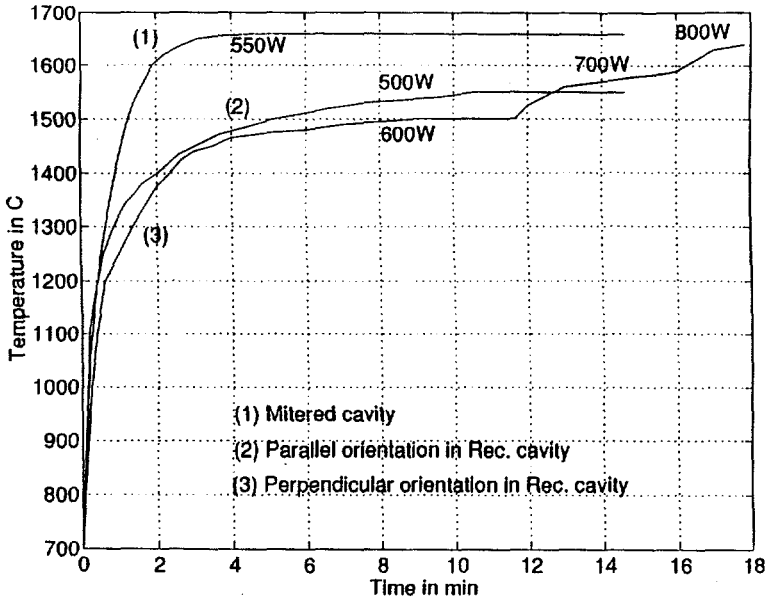

Figure 9 Comparison of Heating in Single Mode Applicators

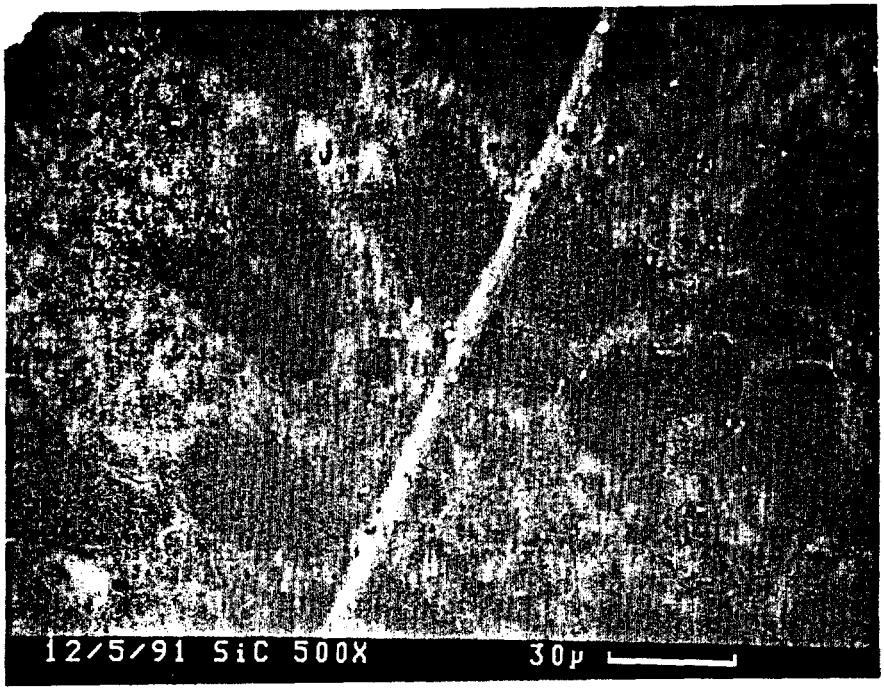

Figure 10 SEM of RBSC-RBSC Joint

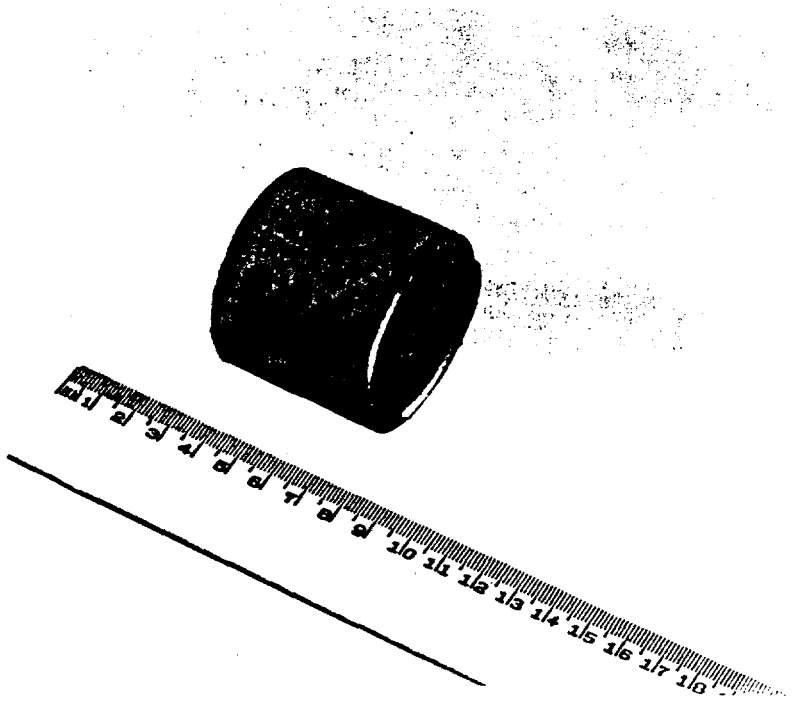

Figure 11 Photograph of Joined RBSC Tube Section

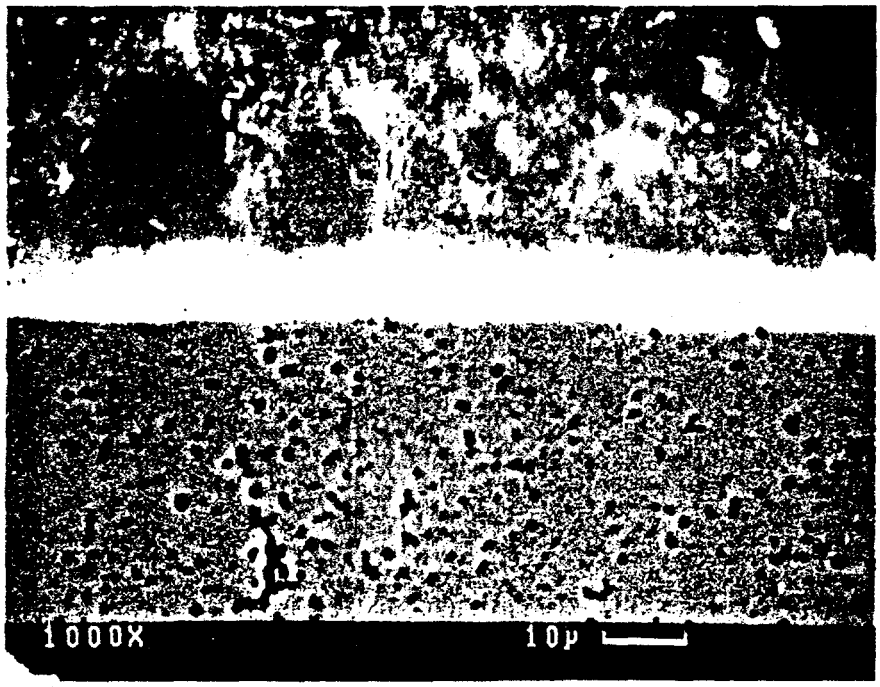

Figure 12 SEM of RBSC-SSiC Joint

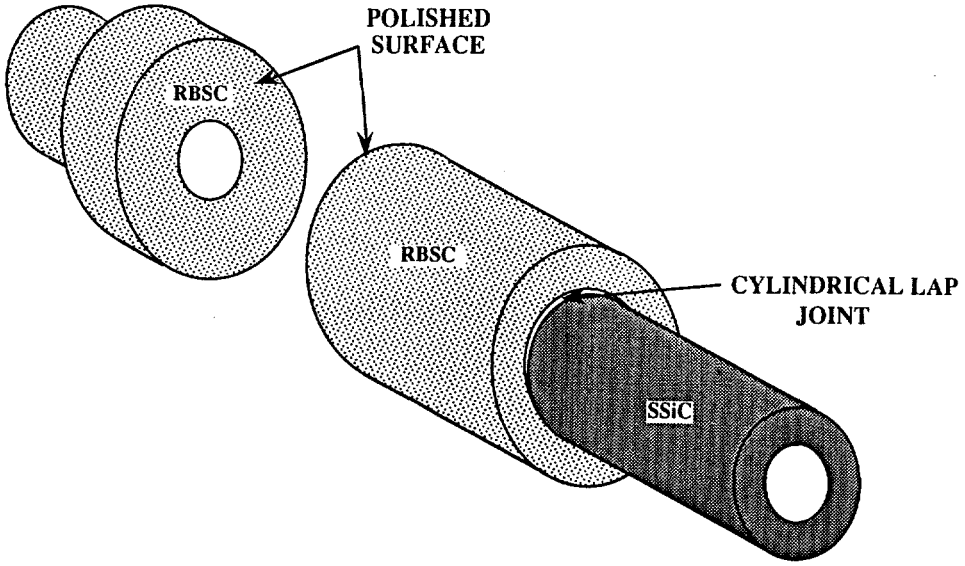

Figure 13 Schematic Illustration of RBSC-SSiC Tube Assembly

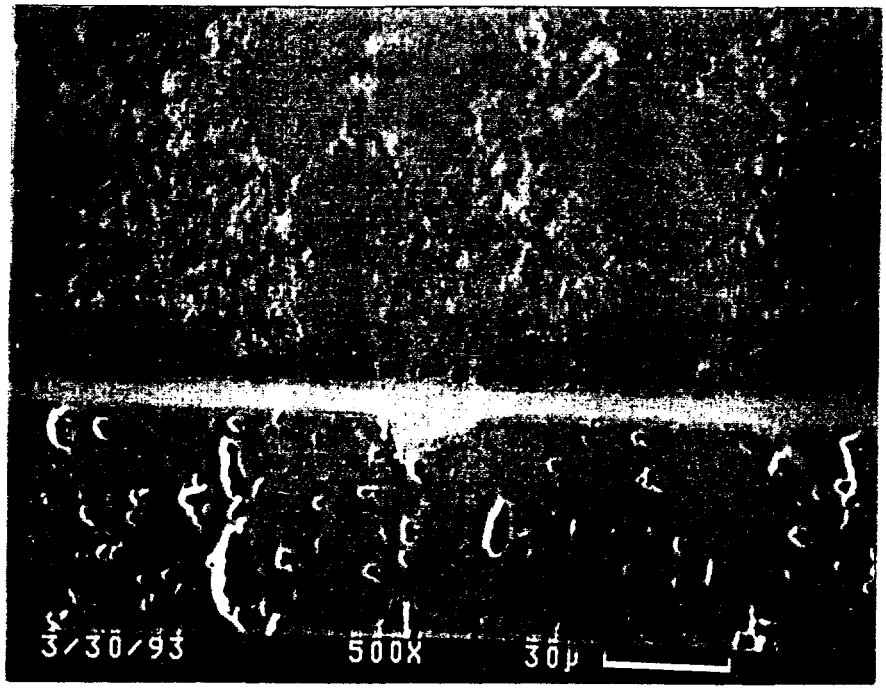

Figure 14 SEM of RBSC-SSiC Joint From Tube Assembly

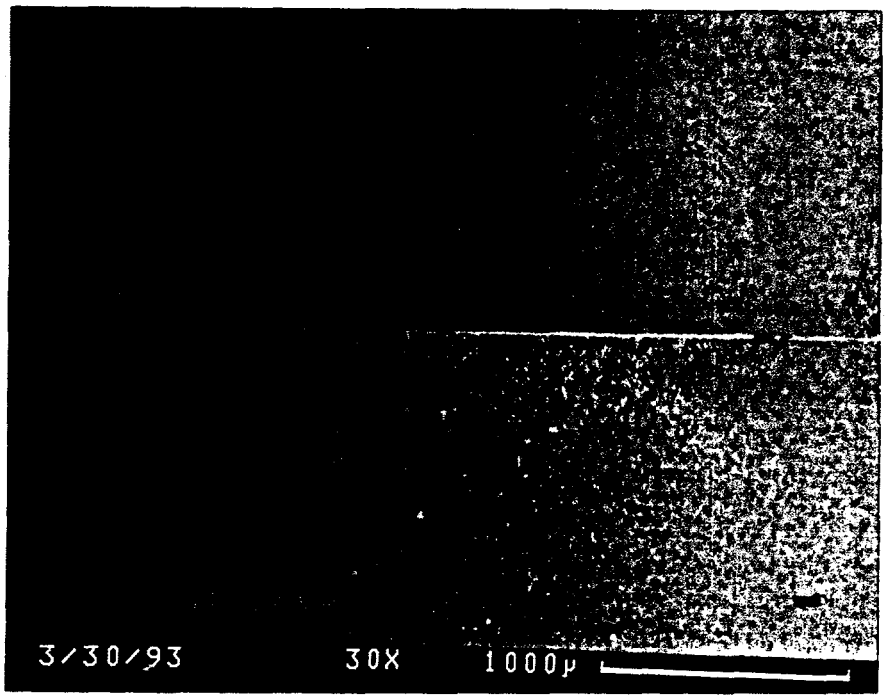

Figure 15 SEM of RBSC-RBSC Joint From Tube Assembly

MECHANICAL STRENGTH OF RBSC-RBSC JOINT

COORS ANALYTICAL LABORATORY (COORS CERAMICS)

WIDTH IN.	THICKNESS IN.	LOAD LBS	STRENGTH psi	STRENGTH MPa
0.1586	0.1116	60.5	36205	250
0.1580	0.1121	52.4	31180	215
0.1593	0.1121	37.5	22101	152
0.1602	0.1127	48.0	27874	192
0.1570	0.1116	57.7	34870	240
AVERAGE			27528	190

Technology Assessment & Transfer, Inc.
133 Defense Highway, Suite 212, Annapolis, MD 21401

020193MWF

Table 1 Mechanical Strength of RBSC-RBSC Joints From Tube Section