CONF-970568--

ULTRASONIC CHARACTERIZATION OF MICROWAVE JOINED
SILICON CARBIDE/ SILICON CARBIDE

M. B. House, P. S. Day

May 1997

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

MASTER

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States, nor the United States Department of Energy, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

KAPL ATOMIC POWER LABORATORY

SCHENECTADY, NEW YORK 10701

Operated for the U. S. Department of Energy
by KAPL, Inc. a Lockheed Martin company

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

ULTRASONIC CHARACTERIZATION OF MICROWAVE JOINED SILICON CARBIDE/SILICON CARBIDE

Michael B. House* and Paul S. Day
Lockheed Martin Corporation
P.O. Box 1072
Schenectady, NY 12301

ABSTRACT

High frequency (50-150 MHz), ultrasonic immersion testing has been used to characterize the surface and interfacial joint conditions of microwave bonded, monolithic silicon carbide (SiC) materials. The high resolution ultrasonic C-scan images point to damage accumulation after thermal cycling. Image processing was used to study the effects of the thermal cycling on waveform shape, amplitude and distribution. Such information is useful for concurrently engineering material fabrication processes and suitable nondestructive test procedures.

BACKGROUND

Ceramic materials are attractive due to their resistance to corrosion in high temperature gaseous environments. Unfortunately, current ceramic processing techniques require high temperatures and are not well suited to components of complex shape. Microwave joining offers the potential to rapidly join ceramic components into a complex shape at relatively low temperatures. Researchers at Los Alamos National Laboratory demonstrated the use of microwaves to join alumina to a metal using a glass interlayer [1]. Later investigators were able to fabricate joints between alumina and reaction bonded silicon nitride [2-4]. Under ideal conditions, diffusion occurs across the interface, providing a strong bond in microwave joined ceramics.

Pre-ceramic polymers may be used to form a joint material during microwave processing. With the proper heat treatment, these polymers transform into a ceramic material which may form a bond between the two surfaces to be joined. Pre-ceramic polymers, including allylhydridopolycarbosilane (AHPCS) may also be used to form the matrix phase in ceramic matrix composites [5]. Test methods are then sought to characterize the extent of material formation, presence of voids or degree of interfacial bonding.

Characterization of ceramics with ultrasound is not new [6-7], and many of the techniques developed for looking at these materials may be applied to microwave joints in ceramics. In-situ ultrasonic testing has been shown to be a good indicator of proper bond formation [8]. The work by Palaith *et al.* showed the in-situ decay of an ultrasonic waveform as bond formation proceeded. The work described herein centers on the use of microwave processing to form bonds between SiC materials using the pre-ceramic polymer AHPCS. High frequency ultrasonic testing is then used to determine joint integrity and the effects of thermal cycling. Fracture surface characterization is also used to verify the results of the ultrasonic tests.

MATERIALS AND TEST PROCEDURE

The specimens consisted of monolithic silicon carbide (Hexoloy SA™) disk pairs (7.62 mm diameter, 3.16 mm high) joined using cellulose nitrate adhesive (CNA) and through microwave processing. The CNA joined samples were used as a calibration to rapidly optimize the inspection and to account for the effects of specimen alignment, surface roughness and material attenuation. Prior to microwave processing of AHPCS, the disk pairs were polished to a finish of 30 μm and etched in a hydrofluoric acid solution to remove residual silicon dioxide. Next, a thin layer of AHPCS was applied to surfaces to be joined. The specimens were pressed together lightly and installed between quartz pushrods in an 0.457 m diameter, 0.559 m long microwave cavity equipped with a mode stirrer. The cavity was then evacuated and backfilled with Argon. After backfill, the cavity was opened and a small amount of AHPCS was applied to the joint region on the disk pair. Each specimen was infiltrated with the AHPCS using a total of three evacuation-backfill cycles prior to initiating the temperature cycle.

The temperature cycle consisted of a low and a high temperature segment; specific times and temperatures are reported in Table I below. A heating rate of $\pm 10^\circ\text{C min}^{-1}$ was maintained for all temperature ramps. After the first and second temperature cycles, Specimen 1 was infiltrated with the AHPCS using the procedure described above.

After infiltration, the disk pairs were examined by ultrasonic microscopy. A 50 MHz (2.54 cm focal length, 6.35 mm diameter) spherically focused quartz delay line transducer was used to determine the integrity of the joint between the disk pair. Spectroscopic techniques were used to assess the effects of interfacial porosity on the reflected ultrasonic signals. A five degree-of-freedom positioning head ensured the transducer was aligned with respect to the specimen.

Table I. Applied heat treatment to SiC disk pairs

Specimen	$T_{\text{dwell},1}$ (°C)	$t_{\text{dwell},1}$ (h)	$T_{\text{dwell},1}$ (°C)	$t_{\text{dwell},1}$ (h)	No. of Cycles
1	200	1	1000	2	3
2	400	5	1100	6	1

To determine how the joint was affected by temperature transients, both specimens were subjected to a high frequency thermal cycle test. The thermal cycle test was conducted between 500°C and 1000°C with a cycle time of approximately 3 minutes, an average cooling rate of 20°C sec⁻¹, and an average heating rate of 3.5°C sec⁻¹. A total of 10,000 such cycles were performed, after which the specimens were re-examined by ultrasonic microscopy.

Upon completion of the thermal cycling test, specimen 1 was fractured and characterized using optical microscopy, electron backscatter, and high frequency ultrasound. A 150 MHz (2 mm focal length, 6.35 mm diameter) spherically focused transducer was used to characterize the specimen fracture surface. Adequate sampling was ensured by using a 250 MHz bandwidth receiver and a 1 GHz analog-to-digital converter (no equivalent time sampling was required). The images were then statistically analyzed and correlated to the nondestructive test both pre- and post-thermal cycling. Post thermal cycle ultrasonic test data on specimen 2 has been obtained but not compared to destructive evaluation.

RESULTS

Figure 1 shows a typical A- and B-scan trace of an ultrasonic waveform through the total thickness of specimen 2 that was joined by the microwave process. As shown in Figure 1, two interfacial signals and the specimen backsurface are evident. Similar time traces were used to calculate the speed of sound (11.76 mm/μsec) from reference, unjoined SiC disks of equal thickness and composition. Figure 2 shows the C-scan (top view) of the first signal reflected from the interface (back surface of first disk) in specimens 1 and 2 before and after high frequency thermal cycling. Intuitively, a higher quality bond would allow the sound energy to pass through the interface without being reflected, thus the higher amplitude regions indicate relatively less bonding compared to the lower amplitude regions. Finally, Figure 3 shows optical, acoustic and electron backscatter images of the fracture surface as well as the C-scan for specimen 1 before thermal cycling.

DISCUSSION

The two microwave joined samples showed non-uniform reflectance indicating variable quality bonding with regions of lower bond quality (light regions of Fig. 2) near the specimen edges. The information contained in the C-scan images is confirmed by the acoustic and optical fractographs. Electron backscatter microscopy further confirmed regions that were successfully microwave bonded from other non-bonded regions based on magnified microstructural features. While the ultrasonic test could not estimate the degree or strength of the bond, it accurately imaged those regions that were unbonded.

Frequency domain information was calculated to investigate pre- and post-thermal cycle test conditions. A frequency downshift of the 50 MHz waveform was calculated (including the 95% confidence limits) point-by-point from the radio frequency waveform data over the entire scanned surface and is given in Table II for specimens 1 and 2.

Table II. Frequency analysis of reflected interface signals

Specimen Condition	Interface Signal	Reflected Peak Frequency	
		Specimen 1	Specimen 2
Before Thermal Cycle	Top	36.3±5.1 MHz	33.9±5.78 MHz
	Bottom	27.9±5.5 MHz	26.5±4.46 MHz
After Thermal Cycle	Top	36.7±4.3 MHz	28.7±7.72 MHz
	Bottom	25.7±4.7 MHz	26.0±26.2 MHz

A downshift of 2.2 MHz in mean frequency from the bottom interface signal and a 0.4 MHz upshift in mean frequency from the top interface were observed in specimen 1 after thermal cycling. The selective attenuation of higher frequency ultrasound at the bottom interface surface suggests (not conclusively) the deterioration of the bond quality during thermal cycling. Such a condition was not seen in specimen 2 due to large scatter in the peak frequency data upon thermal cycling. The scatter itself suggests the presence of damage.

CONCLUSIONS

The ultrasonic test was successful in indicating regions of non-uniform bonding and provided information on the capability of high frequency ultrasonic imaging for future material characterization efforts. The test was sensitive to damage induced in the SiC-SiC joint by thermal cycling, as demonstrated by observable changes in the frequency spectrum data. The ultrasonic tests were

Figure 1. Ultrasonic B- and A- scans of a microwaved SiC-SiC joint.

Figure 2. Ultrasonic C-scan images (taken at 50 MHz) of specimens 1 and 2 before and after high frequency thermal cycle testing.

Figure 3. (a) Optical fractograph, (b) 150 MHz acoustic surface scan of fracture surface, (c) electron backscatter images of fracture surface and (d) C-scan of joint interface prior to high frequency thermal cycle testing. All images are of specimen 1.

confirmed by acoustic, optical, and electron backscatter characterization of the fracture surfaces. Such concurrent engineering of nondestructive tests and material fabrication processing offers to aid in the development of future production inspections while also improving bond formation processes.

REFERENCES

1. T.T. Meek and R.D. Blake, U.S. Patent Nos. 4,529,856 and 4,529,857 (July 16, 1985).
2. D. Palaith, R. Silberglitt, C.C.M. Wu, R. Kleiner, and E.L. Libelo, in *Microwave Processing of Materials*, edited by W.H. Sutton, M.H. Brooks, and I.J. Chabinsky (Mater. Res. Soc. Symp. Proc. 124, Pittsburg, PA, 1988) p. 255.
3. H. Fukushima, T. Yamanaka, and M. Matsui, in *Microwave Processing of Materials*, edited by W.H. Sutton, M.H. Brooks, and I.J. Chabinsky (Mater. Res. Soc. Symp. Proc. 124, Pittsburg, PA, 1988) p. 267.
4. I. Ahmad, W.M. Black, and R. Silberglitt, *Ceram. Eng. Sci. Proc.* 13 (7,8) (1992) p. 250.
5. L. Kolaya, G. Borges and N. Lewis, "The influence of carbon on the structure of pre-ceramic polymer derived SiC" 21st Annual Coca Beach Conference and Exposition, Jan. 12-16, 1997.
6. B.T. Khuri-Yakub et al., "Nondestructive evaluation of ceramics" Review of Progress in QNDE, v 1 New York, NY: Plenum Press (1982).
7. N. Kulkarni et al., "Ultrasonic characterization of green and sintered ceramics: ii frequency domain" *American Ceramic Society Bulletin* v 73 n 7 July 1994, p. 83.
8. D. Palaith et al., "Microwave joining of ceramics" in *Microwave Processing of Materials*, edited by W.H. Sutton, M.H. Brooks, and I.J. Chabinsky (Mater. Res. Soc. Symp. Proc. 124, Pittsburg, PA, 1988) p. 255.