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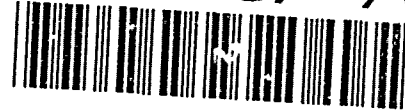
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DIELECTRIC PROPERTIES OF CERAMICS AT MICROWAVE FREQUENCIES

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The dielectric properties of alumina and silicon nitride have been determined using modified rectangular microwave waveguide techniques from 22° to 900°C over 8-12 GHz and modified coax techniques from 25° to 500°C over 2-18 GHz. The coax techniques were used to determine the temperature and frequency dependence of the dielectric properties of silicon nitride and of phosphate bonded alumina reinforced with silicon carbide whiskers. The coax techniques provide broadband dielectric property data that can be applied to the understanding and enhancement of the coupling behavior of ceramics, especially in the initial heat-up period of microwave processing.

INTRODUCTION

In a recent review of the state-of-the art in microwave processing of ceramics by Sutton, it was pointed out that one of the major obstacles to its widespread application in the ceramics industry is the lack of dielectric properties data for many ceramic materials [1]. The objective of our study was to develop a method for high temperature dielectric property measurement, over 2 to 18 GHz at elevated temperatures and to perform measurements on silicon nitride and a silicon carbide reinforced alumina composite. Measurement of the dielectric properties becomes particularly important when considering the effect of impurities or the addition of sintering aids on the microwave coupling of these materials as a function of temperature [2,3].

In Table 1 the advantages and disadvantages of three measurement techniques are compared. The resonant cavity is best for low loss tangent materials but is limited to a very narrow band, is very dimension sensitive and requires multiple fixtures and samples to cover the 2 to 18 GHz frequency range.

The waveguide method has the advantages of a convenient rectangular sample shape and is the least sensitive to the sample dimensions. However, a disadvantage of the waveguide method is that it requires five sets of waveguides, samples, calibrations, and measurements to cover the 2 to 18 GHz range. In addition the size of sample required for the lower frequency waveguides becomes very large.

In contrast the coaxial fixture covers the full bandwidth with only one small (7 mm outside diameter) sample and one measurement. More importantly the correction for the air gap is much less complex than for the rectangular waveguide due to the radial symmetry of the electric field.

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Table 1. Transmission Line Measurement Approach Comparison

1. RESONANT CAVITY	BEST FOR LOW LOSS TANGENT (<0.1)	VERY NARROW BAND REQUIRES MULTIPLE FREQUENCY SAMPLE POINTS VERY DIMENSION SENSITIVE
2. WAVEGUIDE FIXTURE	SIMPLE SAMPLE SHAPE LEAST SENSITIVE TO DIMENSIONS	MODERATE NUMBER OF BANDS - REQUIRES 5 OF EACH TO COVER 2 TO 20 GHz WAVEGUIDES SAMPLES CALIBRATIONS, MEASUREMENTS
3. COAXIAL FIXTURE	COVERS FULL BAND REQUIRES ONLY ONE SAMPLE EASIEST TO CALIBRATE	COMPLEX SAMPLE SHAPE VERY DIMENSION SENSITIVE SOME COMPLEXITY WITH CENTER CONDUCTOR

The disadvantages of the coaxial fixture include the sample shape complexity, the dimensional sensitivity and some complexity with the center conductor.

Measurements were based on a modification of standard transmission methods [4] in specially prepared high temperature waveguide and coax fixtures at Hughes. The Hughes coaxial fixture differs from other high temperature coax fixtures in that it involves a dual port transmission measurement which inherently provides greater sensitivity than the commonly used single port techniques.

The free space method used by W. Ho is readily adaptable to the use of a furnace to obtain dielectric properties measurements at elevated temperatures [5]. However the free space method is limited to frequencies greater than 30 GHz. Since most commercial microwave heating equipment operates at 0.915 and 2.45 GHz, some at 5.8 GHz and a very few at 28 GHz, the dielectric properties obtainable by the 7 mm coax in the 2 to 18 GHz range would be quite useful in developing the data base necessary for widespread utilization of microwave processing in ceramics.

While the Hughes coax is limited to ultimately 900°C, by its current materials of construction, it has only been used in dielectric measurements to 500°C. The temperature range of ambient to 500°C is of interest in studying the behavior of ceramic materials during the initial heating period in the sintering process. It is expected that the Hughes coax fixture will be useable to 900°C with further refinements.

EXPERIMENTAL PROCEDURE

The elevated temperature transmission line microwave measurements system schematic diagram is shown in Figure 1. The transmission line measurement utilizes an HP8510B network analyzer, an

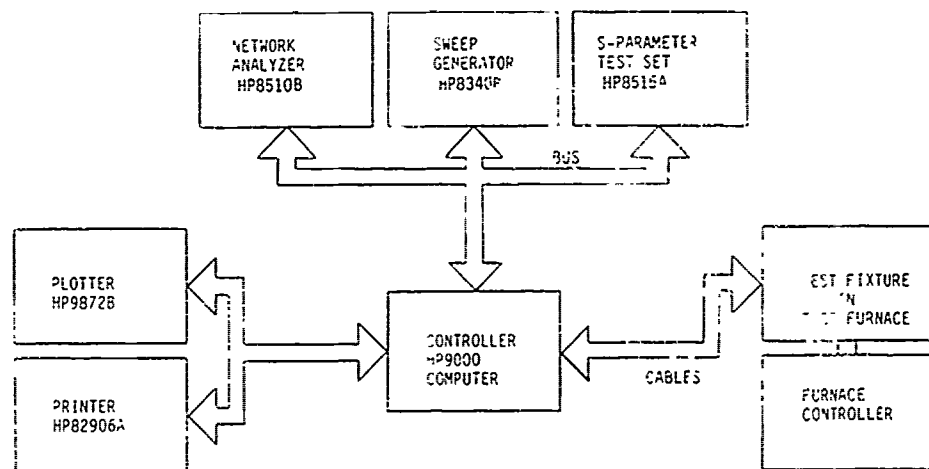


Figure 1. Elevated Temperature Transmission Line Microwave Measurement System.

HP8340B sweep generator and other associated components controlled by an HP9000 computer.* The primary source of error which must be taken into consideration is that of the air gap. The air gap error was not corrected in the waveguide but was corrected in the coax fixture based upon the radial symmetry of the electric field. A diagram representing the cross-section through the coax fixture and the test specimen is shown in Figure 2. The relation for the effective dielectric constant, ϵ_{eff} , relative to the magnitude of the air gaps is given as follows:

$$\epsilon_{\text{eff}} = \frac{\ln(d/a)}{[\ln(d/e) + (1/\epsilon_a)\ln(c/b) + \ln(b/a)]} \quad (1)$$

where a is the outside diameter of inner conductor, b is the inside diameter of the sample, $b - a$ is the inner air gap, d is the inside diameter of the 7 mm coax fixture, c is the outside diameter of the sample and $d - c$ is the outer air gap. These dimensions are shown in Figure 2. The usefulness of this relation in providing an air gap correction was demonstrated first for the outside diameter (OD) correction in Figure 3 and for the inside diameter (ID) correction in Figure 4 using a series of specially machined Rexolite** specimens. The correction in the measurement software controlling the network analyzer totally eliminated the air gap effect for up to a 5.5% OD gap $[(d - c)/c]$ and for as much as a 13.5% ID gap $[(b - a)/a]$. The magnitude of the gap correction can be seen by comparison of the corrected and uncorrected data in Table 2 and in Figures 3 and 4. The program was able to correct for air gap dimensions larger than the anticipated gaps resulting from thermal expansion mismatch between the metal fixture and the ceramic specimen.

* HP denotes products of Hewlett-Packard Co., Palo Alto, CA.

**Rexolite is a polymer product of Union Carbide Co., Danbury, CT.

Table 2 Gap Dimensions of Rexolite in Coax Fixture at 22°C.

NUMBER	LENGTH(mm)	OUTSIDE DIAMETER(mm)	INSIDE DIAMETER(mm)
RX-01	4.059	7.264	3.200
RX-02	4.084	7.264	3.272
RX-03	4.064	7.264	3.594
RX-04	4.069	7.264	4.572
RX-05	4.072	7.209	3.175
RX-06	4.069	6.858	3.175
RX-07	4.072	5.842	3.175

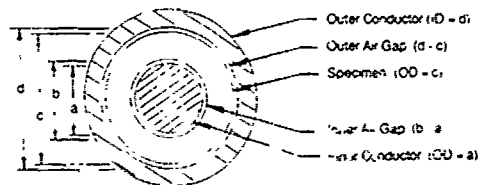


Figure 2. Coax Gap Error Considerations (Coax Gap Error Relationship).

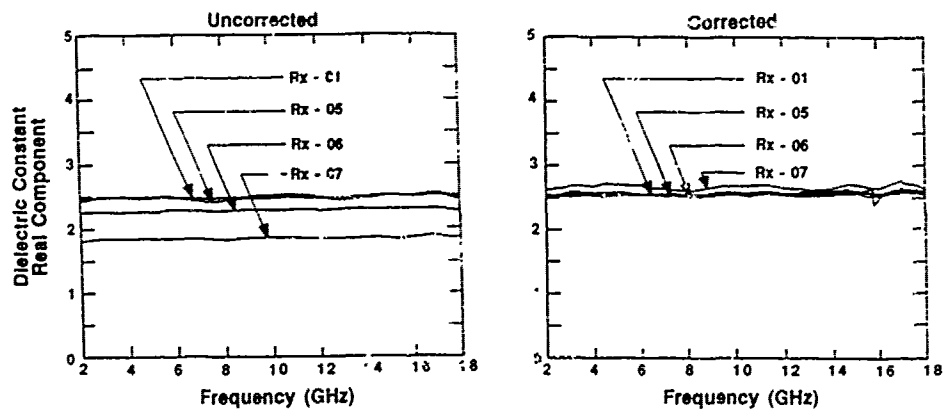


Figure 3. Coax Gap Error Considerations (Outside Diameter Correction).

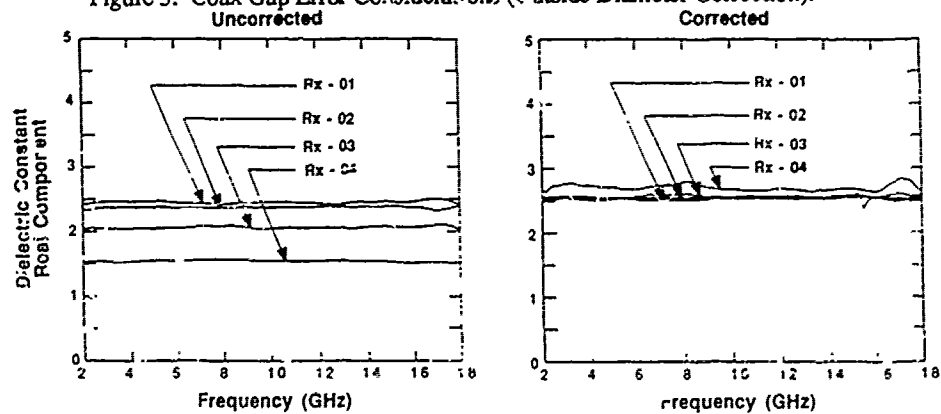


Figure 4. Coax Gap Error Considerations (Inside Diameter Correction).

In the present study specimens were machined from disks prepared from submicron alumina*** mixed with 10% phosphoric acid**** and from 0 to 5 weight percent silicon carbide whiskers*****. The materials were mixed, screened -200 mesh, and then cold pressed with a uniaxial pressure of 10,000 psi. The disks were then cured at 750°C for 4 hours. After machining, the specimens were dried in a vacuum oven at 150°C for more than 24 hours. The specimens were stored in a desiccator until the dielectric measurements were performed to prevent misleading results due to the absorption of moisture from the atmosphere.

The dielectric properties of silicon nitride* were determined from 25° to 900°C in the X-band waveguide. The same silicon nitride* was measured in the coax fixture along with a silicon nitride** and mixtures of phosphate bonded alumina with 0, 1.0, 2.5, and 5.0% (by weight) SiC whiskers. The gap sensitivity measurements for the coax fixture dielectric properties were performed on a series of Rexolite specimens at 22°C. The air gaps in the Rexolite specimens were machine'd so that they accounted for the anticipated gaps resulting from the difference in thermal expansion coefficients of the metal coax fixture and various ceramic test specimens.

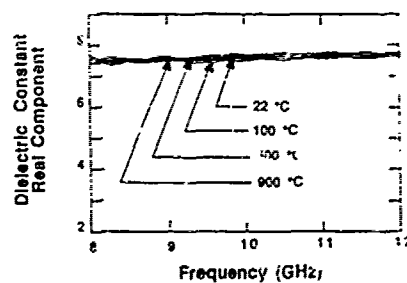


Figure 5. Dielectric constant (real component) of silicon nitride (Kyocera 220) over 8-12 GHz from 22° to 900°C.

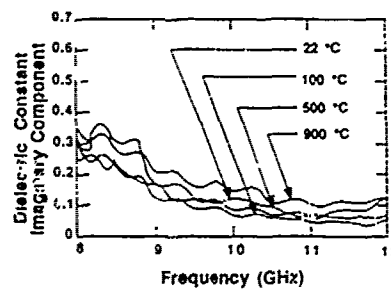


Figure 6. Dielectric constant (imaginary component) of silicon nitride (Kyocera 220) over 8-12 GHz from 22° to 900°C.

- * Silicon nitride 220M, slip cast, Kyocera, Nagoya, Japan.
- ** Silicon nitride, Toshiba Ceramics, Tokyo, Japan.
- *** Alumina, Grade A-16-SG, ALCOA, Bauxite, AR.
- **** Phosphoric acid, reagent grade, J. T. Baker Co., Phillipsburg, NJ.
- ***** Silicon nitride whiskers, ARCO Specialties Co., NC.

RESULTS AND DISCUSSION

The results of the dielectric constant determination (real and imaginary components respectively) are shown in Figures 5 and 6 for the silicon nitride* in the X-band waveguide with temperatures from 25° to 900°C. These results are uncorrected for air gaps. The air gap between the test sample and the waveguide increases with higher temperatures due to the higher thermal expansion coefficient of the metal waveguide compared with the ceramic test specimens.

The same silicon nitride* was measured in the coax fixture with the results shown in Figure 7 for both the real and imaginary components of the dielectric constant for 25°, 200° and 500°C each over the 2 to 18 GHz frequency range. The coax results for another silicon nitride** are shown at 25° and 500°C over the 2 to 18 GHz frequency range in Figure 8. The values obtained were 7.5 +/- 0.5 for the real and 0.2 +/- 0.1 for the imaginary component of the dielectric constant which compare favorably with the values reported by W. Ho at 35 GHz ($\epsilon_r = 7.5 - 8.8$ at 23°C), [5]. The

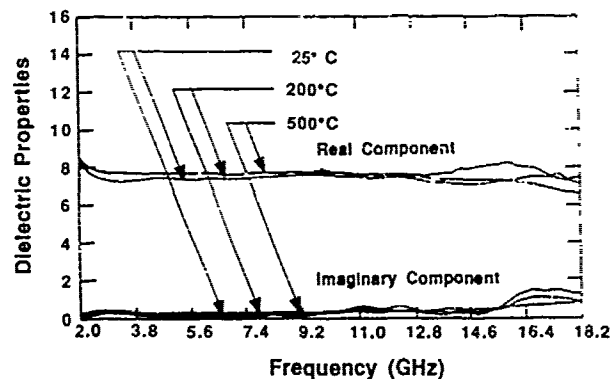


Figure 7. Dielectric constant vs. frequency (2-18 GHz) of silicon nitride (Kyocera 220) at 25°, 200°, and 500°C.

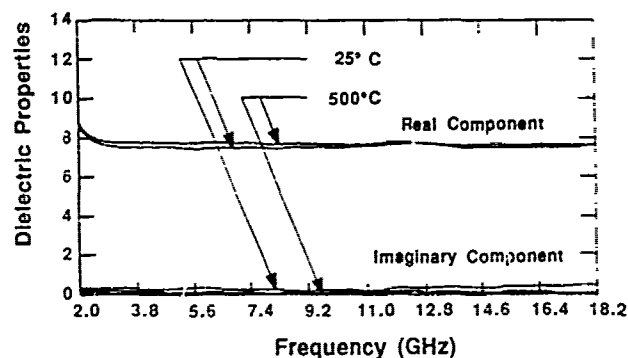


Figure 8. Dielectric constant vs. frequency (2-18 GHz) of silicon nitride (Toshiba) at 25° and 500°C.

* 220M, slip cast silicon nitride, Kyocera, Nagoya, Japan.

** Silicon nitride, Toshiba Ceramics, Tokyo, Japan

broad spectral results of the coax method points out that the narrow band waveguide results may be in question. As seen in Figure 6 the imaginary component of the dielectric constant as measured in a waveguide increases anomalously at lower frequencies but remains relatively constant when measured in a coax as shown in Figure 7. It has been determined that this effect is caused by the controller software and that it can be resolved with program modification.

The dielectric constant real and imaginary components were determined for a series of phosphate bonded alumina samples with silicon carbide reinforcing from 0 to 5.0% by weight. The real components are shown at 25°C and 500°C in Figure 9. The imaginary components are shown at 25° and 500°C in Figure 10. It can be seen that at 25°C the effect of the SiC whisker additions is

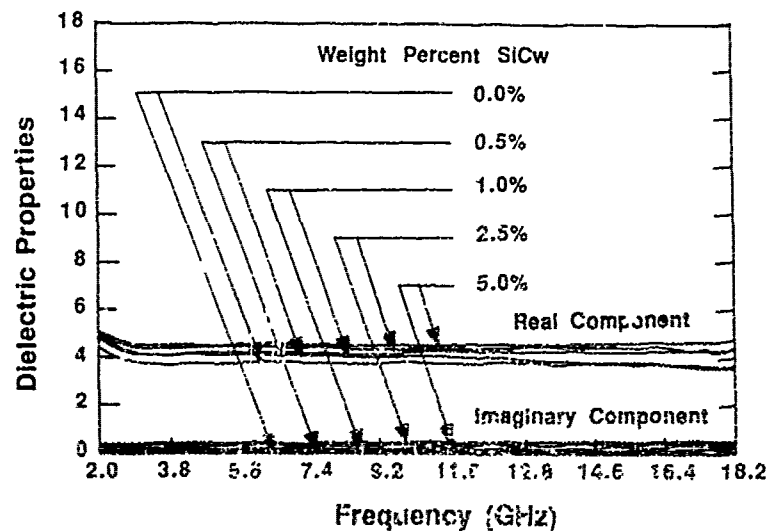


Figure 9. Dielectric constant (real component) vs. frequency (2-18 GHz) at 25°C for 0-5% SiCw/Al₂O₃ + 10% H₃PO₄. (Gap error correction and 10% curve smoothing).

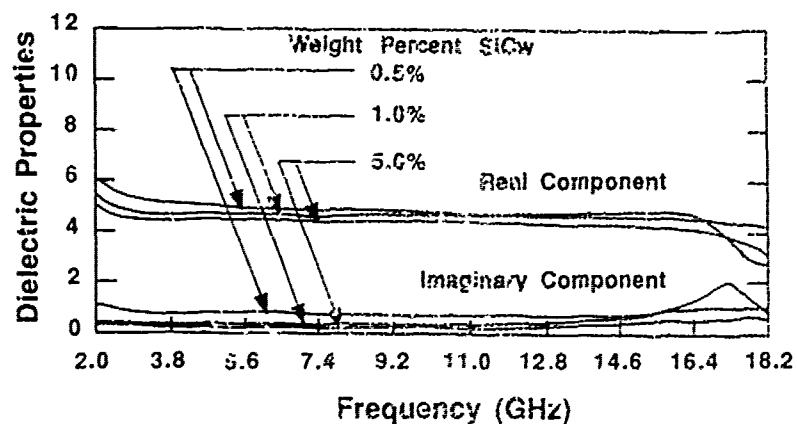


Figure 10. Dielectric constant (imaginary component) vs. frequency (2-18 GHz) at 500°C for 0-5% SiCw/Al₂O₃ + 10% H₃PO₄. (Gap error correction and 10% curve smoothing).

negligible. However at 500°C the increasing SiC content resulted in a clearly discernable increase in both the real and imaginary components of the dielectric constant. The higher imaginary component values for the dielectric constant would be expected to enhance the coupling of the composite with microwave radiation so that faster heating rates would result. It is apparent that modification of the composition of ceramic materials is a critical part of optimizing a material system for microwave processing.

Microwave processing of ceramic composites is a complex process. However it is possible, with semi-empirical shape factors, to develop methods for predicting the dielectric properties of discontinuous fiber, whisker and particulate reinforced composite systems. The Hughes coax measurement technique can be used to determine the shape factors necessary to design composites optimized for microwave processing.

CONCLUSION

A high temperature broad frequency band dielectric properties measurement capability has been achieved with a Hughes coax fixture design. A method for correcting coax air gap errors was developed and incorporated into the measurements software. The coax fixture demonstrated sensitive broad band capability to 500°C for silicon nitride with dielectric constant values which compared favorably with published data. Increasing concentrations of SiC whiskers in a phosphate bonded alumina composite resulted in an increase in both the real and imaginary dielectric constant values at 500°C. The Hughes coax design was shown to be a useful method for determining the dielectric properties of ceramic materials up to 500°C in the frequency range of 2 to 18 GHz. The dielectric properties data obtained with the Hughes coax system will be useful in predicting the response of ceramic materials during the initial heat up period of microwave processing.

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