A system to measure complex permittivity of low loss ceramics at microwave frequencies and over large temperature ranges

Binshen Meng
Department of Electrical and Computer Engineering, University of Wisconsin, Madison, Wisconsin 53706

John Booske
Department of Electrical and Computer Engineering and Materials Science Program, University of Wisconsin, Madison, Wisconsin 53706

Reid Cooper
Department of Materials Science and Engineering and Materials Science Program, University of Wisconsin, Madison, Wisconsin 53706

(Received 9 September 1994; accepted for publication 3 October 1994)

A system has been developed for measuring the complex permittivities of low loss ceramic materials at frequencies from 2 to 20 GHz and over a temperature range 20–1000 °C. The measurement technique involves a modified version of the conventional cavity perturbation method. Details of the design and fabrication of the circular cylindrical cavity and the input and output coupling transmission lines are discussed. Particular features related to high-temperature operation and temperature cycling are described. Data are presented for an illustrative measurement of the complex microwave dielectric properties of NaCl single crystals between 20 and 400 °C. The experimental results are in excellent agreement with theoretical models. © *1995 American Institute of Physics.*

I. INTRODUCTION

An understanding of the fundamental physical mechanisms of interaction between microwave-frequency electromagnetic radiation and crystalline solids is crucial to the development of predictive capabilities having an impact on several materials applications of growing interest and importance, namely (1) microwave processing of ceramics (e.g., sintering, bonding), (2) complex dielectric properties of ceramic substrate materials for microwave and high-speed digital circuits (microwave circuit substrates and integrated circuit packaging), and (3) materials selection and fabrication for window and insulator structures in high power coherent microwave sources employed in conventional power tube markets as well as research enterprises such as high-energy physics accelerators and fusion plasma experiments. To accomplish these goals, it is necessary to develop a measurement system to accurately determine the microwave dielectric properties of low loss ceramic materials over a broad range of temperatures and frequencies.

In this paper, we describe the design and performance of a system for measuring the complex dielectric constant of low loss ceramics in 2–20 GHz microwave regime. The specific demonstrated capabilities include a minimum measurable loss tangent tan $\delta \sim 10^{-5}$ and a temperature range of 20–400 °C (minor straightforward modifications described later extend the maximum temperature up to 1000 °C).

II. THEORY OF THE MEASUREMENTS

The system described in this paper determines the complex dielectric constant of low loss samples and is based on the cavity perturbation method. This technique has been extensively employed for studying the dielectric properties of materials in the microwave-frequency regime.[1–4] In our system, a circular cylindrical cavity is used as the resonator into which cylindrical rod-shaped samples are inserted along the cavity axis. The intensity of the electromagnetic field inside the cavity is determined by the input power and the cavity Q factor. The real part of the complex dielectric constant ϵ' of an inserted sample is determined from the shift of the resonant frequency of the cavity due to the presence of the sample, while the imaginary part ϵ'' (representing microwave energy absorption by the sample) is obtained by measuring the change of the Q factor of the cavity induced by insertion of the sample. In conventional cavity perturbation analysis, ϵ' and ϵ'' are calculated from the resonant frequency shift and the change in cavity Q, respectively, using the formulas

$$\epsilon' \simeq 1 - C\,\frac{\Delta f}{f_0} \tag{1}$$

and

$$\epsilon'' \simeq C\left(\frac{1}{Q} - \frac{1}{Q_0}\right). \tag{2}$$

In Eqs. (1) and (2), Q and Q_0 are the quality factors of the cavity with and without the sample, respectively, and C is a constant depending on the cavity configuration, mode structure, and sample shape. For a specific experimental configuration C can be either calculated analytically assuming several approximations[5,6] or determined experimentally by calibration with a known material as the sample.

The validity of the conventional method embodied by Eqs. (1) and (2) and the approximate calculation of constant C[5,6] assumes the sample is sufficiently small to have negligible effect on the cavity fields.

For many of the applications cited earlier, the materials of interest include crystalline solids that may be difficult to fabricate into sufficiently small samples to satisfy the size requirements of the conventional cavity perturbation method.

 0034-6748/95/66(2)/1068/4/$6.00

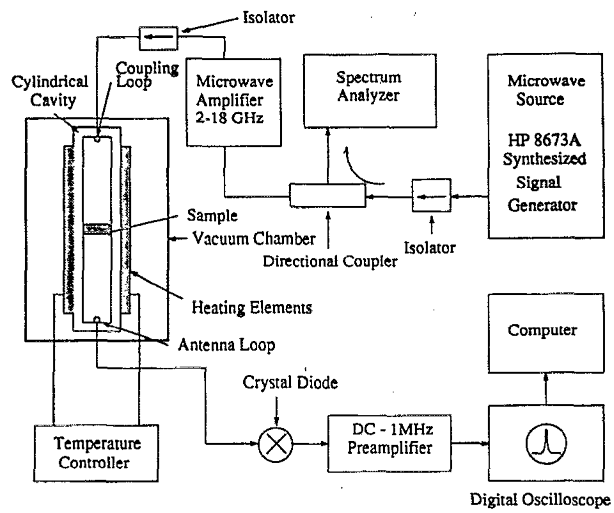

FIG. 1. Schematic of the experimental system.

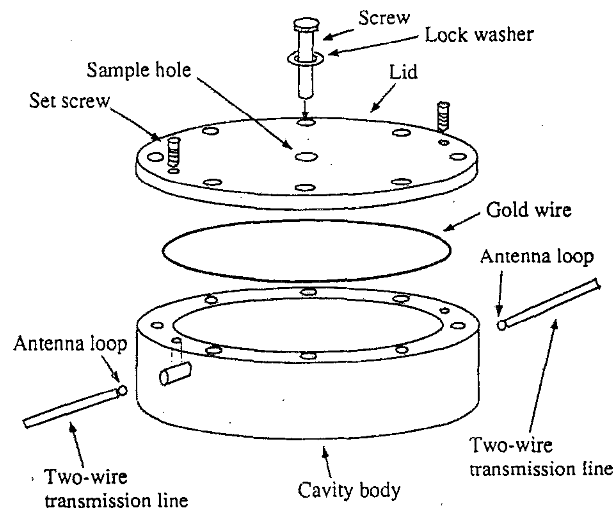

FIG. 2. Schematic of the cavity assembly.

To solve this problem, an extended version of the cavity perturbation method has been developed and tested for circular cylindrical cavities resonating in TM_{0n0} modes. This extended version of cavity perturbation allows for thicker samples for which the conventional analysis would yield considerable errors, especially for ϵ''. In the improved method, ϵ' is determined from the resonant mode frequency shift data and an exact solution of the dispersion relation for the sample-filled cavity [instead of Eq. (1)]. Equation (2) is still used to obtain ϵ'' from the measured values of Q and Q_0 (the sample-inserted and empty cavity quality factors, respectively). However, the calculation of C is done in a more exact form than in previous approaches, and hence a more accurate value for ϵ'' is obtained. A more detailed discussion of the theoretical details are in preparation and will be submitted as a future publication.

III. DESCRIPTION OF THE MEASUREMENT SYSTEM

A schematic of the system configuration is illustrated in Fig. 1. A Hewlett–Packard 8637A synthesized signal generator is used as the microwave source. It provides 10 mW of power over a frequency range of 2–26 GHz that can be covered in discrete increments as small as 1 kHz. Two low-noise, high gain microwave amplifiers (one covering 2–6 and the other covering 6–18 GHz) increase the microwave power of 10 mW up to 1 W.

To span a frequency range of 2–18 GHz, two circular cavities resonating in several TM_{0n0} modes are employed. Two cavities are required since it was discovered that only the first three modes (TM_{010}, TM_{020}, and TM_{030}) were usable in each cavity. Higher-order modes (i.e., TM_{0n0}, $n \geqslant 4$) were found to overlap with other nearby, densely spaced modes, rendering it difficult to clearly ascertain their resonance line-shape properties. Each cavity is a right circular cylinder, one with an internal radius of 5.22 cm and a height of 2 cm the other having an internal radius of 2.3 cm and a height of 1 cm. The larger cavity's first three eigenfrequencies are approximately 2, 5, and 8 GHz, while the smaller cavity's first three eigenfrequencies are approximately 5, 11, and 18 GHz. The redundancy at 5 GHz was intentional to cross-check that both cavities yield identical data at 5 GHz. For initial measurements, prototype cavities were fabricated from copper.

Cavity Q is determined from the linewidth (full width half maximum) of the two-port transmission coefficient between two coupling loop antennas. The two antennas are inserted through diametrically opposed small holes in the sides of the cavity and weakly couple the cavity to two shielded, two-wire transmission lines, as indicated in Fig. 2. The diameter of each transmission line is 3.175 mm and the diameters of the loops are approximately 2 mm for the larger cavity and 1.5 mm for the smaller cavity. Internal access to the prototype cavities was provided by having removable lids, as shown in Fig. 2.

In working with the prototype cavities, it was determined that two design features were critical for reliable operation. First, an excellent and unchanging ohmic contact is required between the lid and the cavity body. One technique that was found to significantly improve this electrical contact was to compress a piece of thin gold wire (0.1 mm diameter) between the lid and the cavity body. Second, a precise positioning of the loop antennas for extremely weak (but nonzero) coupling is necessary to ensure that the measured cavity Q is as close to the unloaded Q as practical. Even very small eigenmode field perturbations caused by insertion of a dielectric sample (through a small hole in the lid) can unacceptably distort the eigenfrequency line shapes if the antenna–cavity coupling is not sufficiently weak. The combination of the 30 dB (1 W maximum output) microwave amplifier(s) and a low-noise preamplifier (10^5 gain, dc-1 MHz) after the crystal diode detector were found adequate to compensate for the weak signals associated with the very weak cavity coupling. The set screws indicated in Fig. 2 serve the function of fixing the positions of the antenna loops, once an acceptably weak coupling is empirically determined.

Observing variations in ϵ' and ϵ'' over large dynamic temperature ranges represents an important tool for determining the physical mechanisms that are responsible for the complex dielectric behavior of solid materials.[7] Consequently, considerable attention was focused on system design features that ensured repeatable, reliable operation at high temperatures and under conditions of regular thermal cycling. For example, except for the piece of gold wire, all cavity components were fabricated from either copper or stainless steel, due to the similar coefficients of thermal expansion between these two materials. The rigid two-wire transmission lines feeding the coupling loops were custom fabricated for high-temperature operation from copper wire for the transmission line, double-bore alumina tubing for the dielectric, and thin copper tubing for electromagnetic shielding. Stainless-steel lock washers were placed under the screw heads (Fig. 2) to act as compression springs, thereby alleviating hysteresis in the lid–body mechanical spacing due to differential expansion, contraction, and stress relief during thermal cycling. Placing the set screws for the rigid transmission lines directly on the cavity body (rather than on an external mechanical support) was found necessary to avoid relative positional shifting of the coupling loops (causing shifts in cavity coupling) during cycling between 20 and 400 °C.

To ensure that the specimen is isothermal, the entire cavity is resistively heated by ohmic heating rings on the top and bottom (Fig. 1), and the entire assembly is enclosed in pressed ceramic fiber board insulation. To avoid oxidation of the hot cavity, the assembly is also placed in controlled gas conditions (typically, a 10 mTorr vacuum is found adequate). It was also found necessary to prebake all insulation materials in order to burn-out organic binders that would otherwise leave carbon deposits on exposed cavity and dielectric sample surfaces as the system was heated to high temperatures. The heater element power supply was converted to a dc supply after it was discovered that ac current was a source for unwanted 60 Hz noise. Control of the cavity and sample temperature was accomplished by a commercial autotuned temperature controller reading a K-type thermocouple. This feature was found to be necessary for precise, repeatable measurements of temperature-dependent dielectric properties in low loss ceramics.

A good test of whether the cavity design is free from mechanical expansion and contraction effects during thermal cycling is to see whether the temperature dependence of the empty cavity quality factor Q_0 can be solely accounted for by the temperature dependence of the wall material's resistivity. Undesired thermal expansion and contraction effects on the cavity Q would include variations in the ohmic contact between the cavity lid and body, as well as excessive relative shifting between the coupling loop positions and the cavity body. In the absence of such effects, the empty cavity quality factor should vary with temperature T as

$$Q_0(T)=\frac{1}{2}\sqrt{\frac{1}{\pi f_n \epsilon_0 \rho(T)}}\frac{x_{0n}}{a/d+1} \tag{3}$$

or

$$\rho(T)\propto\frac{f_n}{[Q_0(T)]^2}, \tag{4}$$

where ρ is the resistivity of the cavity material, x_{0n} the nth zero of the zero-order Bessel function J_0, f_n the eigenfrequency of the TM_{0n0} mode, and a and d the cavity radius and height, respectively. In the case of copper above 100 K, the resistivity is expected to vary in an approximately linear fashion with temperature:[8]

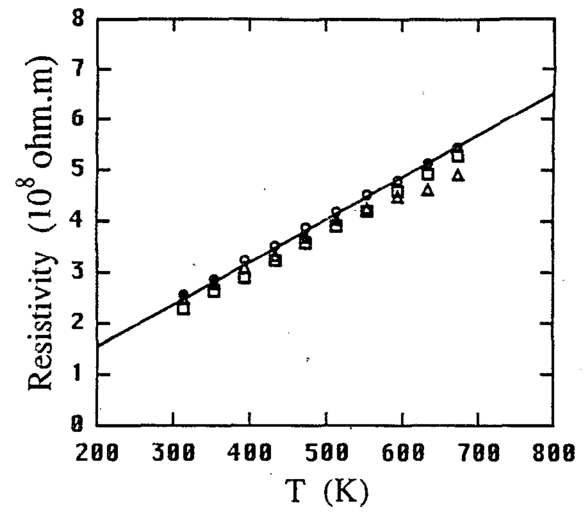

FIG. 3. Comparison of experimental measurements of $f_n/[Q_0(T)]^2$ (data points) with the expected scaling (solid line) assuming that the variation was entirely due to changes in the cavity wall resistivity [Eq. (5)]. ○: 2 (n=1), □: 5 (n=2), and △: 8 GHz (n=3).

$$\rho=\rho_0+\alpha T, \tag{5}$$

where $\alpha=8.3\times10^{-11}$, ρ_0 depends on the impurity concentration ($\rho_0\approx-0.315\times10^{-8}$ for "pure" copper), and T is the absolute temperature. To evaluate whether the configuration was free of undesired thermal contraction and expansion effects, repeated measurements were made of the empty cavity quality factor Q_0 as the cavity temperature was varied between 300 and 700 K for the first three TM_{0n0} eigenmodes (corresponding to resonant frequencies of approximately 2, 5, and 8 GHz). Measurement precision was determined to be within 0.5%. In Fig. 3 it is apparent that the measured (relative) variations of $f_n/[Q_0(T)]^2$ with temperature are in excellent agreement with the temperature scaling of copper's resistivity based on Eq. (5). This demonstration of a stable and repeatable variation of empty cavity Q with temperature cycling was considered positive proof that to within very small error limits the cavity behavior was free from anomalous thermal cycling effects. Accurate and predictable knowledge of the exact dependence of $Q_0(T)$ was critical for an accurate determination of temperature-dependent complex dielectric properties based on Eq. (2).

IV. TYPICAL EXPERIMENT RESULTS

To illustrate the capabilities of this new microwave dielectric spectrometer, we have measured the complex dielectric permittivity of nearly pure and impurity-doped NaCl single-crystal specimens. Comparing experimental measurements of the imaginary part ϵ'' with theoretical models provides an excellent method for determining the impact of point defects on microwave absorption in ionic crystalline solids.[7] In Fig. 4 we provide such a comparison for the mi-

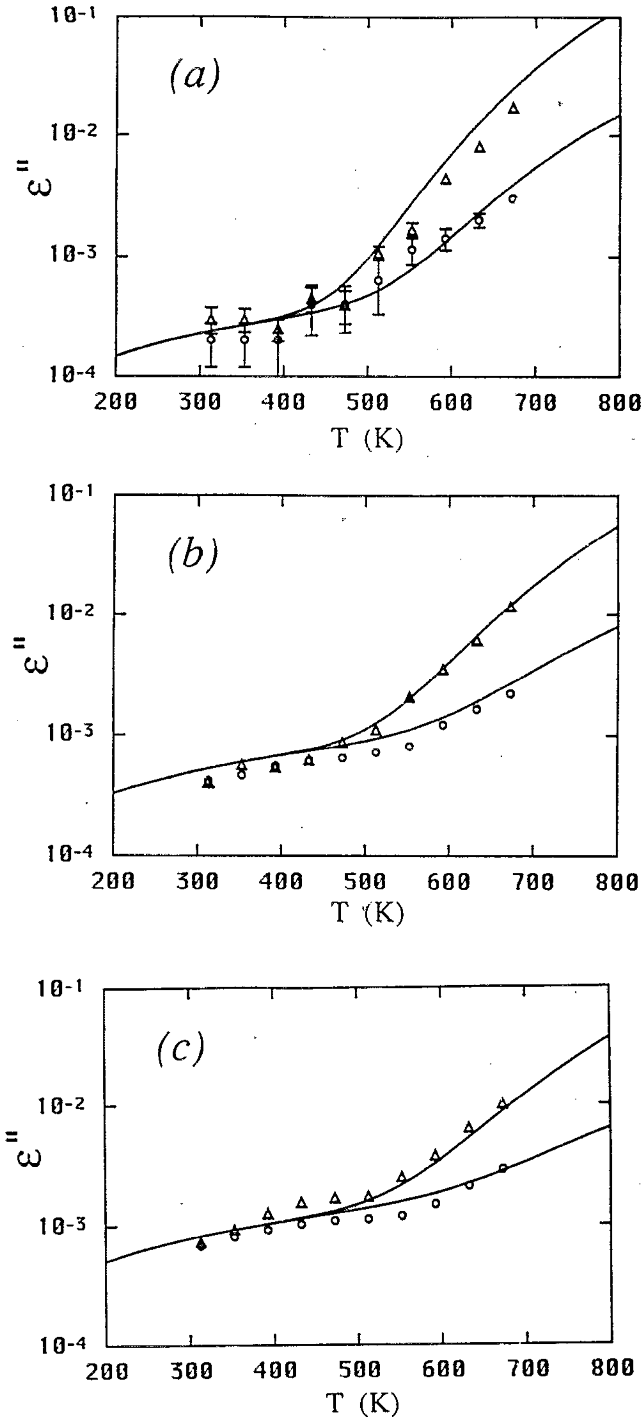

FIG. 4. The experimentally measured ϵ'' and theoretically predicted ϵ'' in NaCl single crystals at frequencies of (a) 2.2, (b) 4.8, and (c) 7.4 GHz vs temperature. In the all plots, O: undoped crystal (pure crystal), △: doped crystal,—: theoretical prediction.

crowave absorption in both a nearly pure NaCl single-crystal sample and a second sample doped with 250 ppm Ca^{++} cation impurity concentration. Data corresponding to the first three TM_{0n0} resonances from one of our two cavities are shown. Results from the second (higher frequency) cavity are similar to the data shown here. Note that the theory and experimental measurements are in excellent agreement, even at values of ϵ'' as small as $\sim 10^{-4}$ (corresponding to a loss tangent tan $\delta \sim 10^{-5}$). A detailed discussion of the theory of microwave absorption in ionic crystalline solids is beyond the scope of this paper but will be provided in a future publication. An outline of the basic physics can be found in Ref. 7.

V. DISCUSSION

A system has been developed to measure the complex dielectric properties of low loss ceramic materials over large temperature ranges. A prototype system has demonstrated reliable operation for a temperature range of 20–400 °C and at five frequencies spanning the frequency range of 2–20 GHz. An extended version of the cavity perturbation method allows the use of higher-order mode measurements (TM_{0n0}, $n \geqslant 2$) with dielectric samples that are difficult to fabricate to the small dimensions that would be required by the conventional cavity perturbation approach. Extension of the maximum operating temperature to 1000 °C can be accomplished by straightforward modifications of the prototype system described here. In particular, the use of molybdenum for the cavities provides the high-temperature capability with an electrical conductivity only 3× less than that of copper. Hence the cost of modifying for higher temperature operation is only a modest reduction of measurement sensitivity by a factor of 1.7 (square root of the reduction in conductivity). The only other design modification required for higher temperature operation is to similarly select refractory metals (such as molybdenum or stainless steel) for fabrication of the rigid transmission lines feeding the coupling loops. Finally, illustrative measurements of the complex permittivity of NaCl crystals with different levels of cation impurity concentrations are in excellent agreement with theoretical predictions over a large range of temperature, at several different frequencies, and at very small loss tangent values, tan $\delta \sim 10^{-5}$.

ACKNOWLEDGMENTS

The authors gratefully acknowledge the support of the Electric Power Research Institute, the Wisconsin Alumni Research Foundation, the National Science Foundation through a Presidential Young Investigator Award, and Nicolet Corp. for donation of the digital oscilloscope used in the experiments. Sam Freeman and Tom Wadzinski contributed valuable assistance to the conduct of this research.

[1] B. Birnbaum and J. Franeau, J. Appl. Phys. **20**, 817 (1949).
[2] K. S. Champlin and R. R. Krongard, IRE Trans. Microwave Theory Tech. **MTT-9**, 545 (1961).
[3] J. Verweel, Philips Res. Rep. **20**, 404 (1965).
[4] A. Parkash, J. K. Vaid, and A. Mansingh, IEEE Trans. Microwave. Theory Tech. **MTT-27**, 791 (1979).
[5] R. A. Waldron, Proc. Inst. Electr. Eng. **107C**, 272 (1960).
[6] R. F. Harrington, *Time-Harmonic Electromagnetic Fields* (McGraw-Hill, New York, 1961), p. 317.
[7] B. Meng, J. H. Booske, R. F. Cooper, and S. A. Freeman, MRS Symp. Proc., Vol. 347: Microwave Processing of Materials IV, 467 (1994).
[8] C. A. Wert and R. M. Thomson, *Physics of Solids* (McGraw-Hill, New York, 1964), p. 205.