Electrical properties of films at microwave frequencies

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Abstract. A technique suitable for the measurement of electrical properties of films/sheets at microwave frequencies has been described. Groups of materials are identified for which the absence of any kind of electrodes in these microwave measurements can be utilised with advantage. Results are presented for a variety of materials such as metal films, superionic conductors, semiconductors and insulating polymers.

Keywords. Microwaves; waveguide; dielectric constant; a.c. conductivity.

1. Introduction

Knowledge of the electrical properties of films at microwave frequencies is helpful in material study and characterization for device fabrication. Thin metal and insulating films, for example, are extensively used in microwave integrated circuits. There, it is of interest to know the attenuation and phase produced by a particular film and variation of these quantities with parameters such as film thickness, temperature etc. Thin semi-metal and metal films were studied by several workers (Gunn 1967; D'Aiello and Freedman 1969; Duggal and Rup 1969; Ramey *et al* 1970). The microwave applications of thin metallic films have also gained importance as exemplified by the use of films as microwave mirrors and as large size film reflectors (Korolev and Gridnev 1963). Microwave measurements are essential in the above cases. But in many situations advantage can be taken of the fact that at microwave frequencies the measurements basically are electrodeless and therefore these are recommended to study materials which require special type of electrodes. Groups of materials may be identified where contacts do pose problems, such as *e.g.* (i) semi-conductors (ii) fast ion conducting solids (iii) membranes and other biological systems. Such materials can be conveniently studied at microwave frequencies.

Section 2 describes few techniques including the one developed earlier (Dube and Natarajan 1973), for the measurement of dielectric constant, loss and conductivity of films and foils. Finally the results are presented for a variety of films.

2. Measurement techniques

The most widely used method for microwave measurements of dielectric parameters of solids is the short-circuited line technique of Von Hippel (Von Hippel 1961). The technique involves the measurement of vswr (voltage standing wave ratio $= E_{max}/E_{min}$) on the standing waves set-up when the microwaves are totally reflected from a short circuit. A dielectric specimen in the form of a small block which completely
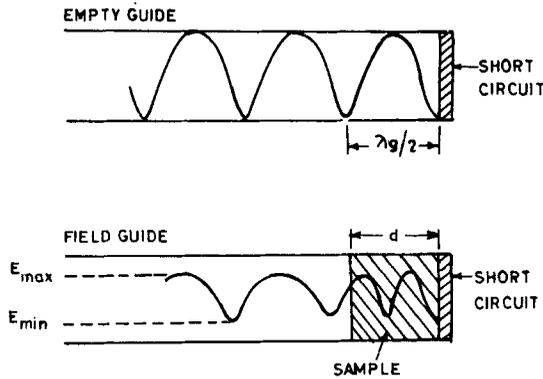
EMPTY GUIDE

SHORT CIRCUIT

$\lambda g/2$

FIELD GUIDE

d

E_{max}

E_{min}

SHORT CIRCUIT

SAMPLE

Figure 1. Standing wave pattern in empty and field guide.

fills the section of waveguide is placed next to the short circuit. This alters both the position of standing wave pattern and the size of the voltage ratio by amounts which depend on the dielectric constant and loss of the material and on the length of the 'filled guide' (figure 1).

Von Hippel's method when applied to a very thin specimen does not give accurate results, the reason being the lack of strong interaction between the e.m. wave and the thin specimen. Therefore the presence of the specimen shifts the voltage minima only slightly (often insignificantly) and the change in standing wave ratio is also small (often not measurable accurately).

Recently, Von Hippel's technique has been modified to suit thin samples by measuring impedance not only with short circuit (figure 1) but also using an open circuit and matched terminations (Das Gupta and Pramanic 1982). The method is effective only for high dielectric constant films.

Cavity methods which measure the change in resonance frequency and broadening of resonance response on introduction of the specimen are also not very successful for the same reasons of lack of strong interaction when the specimen is very thin. However, attempts have been made to study thin samples using specific forms of cavities (Conklin 1965; Sobol and Hughes 1967) and other forms of sample holders (Pilipenko *et al* 1970). Cavity perturbation technique has also been used (Rzepecka and Hamid 1972).

3. An accurate and convenient technique

In the above techniques such as that of Von Hippel, the accuracy of measurement is poor due to weak interaction of e.m. wave with the specimen. And the reason for the poor interaction is the fact that in normally used TE_{10} mode of excitation the electric field is maximum at the centre of the waveguide and diminishes to zero at the side walls (figure 2); most of the portion of the specimen lies in the weak field regions. Keeping this in view, a suitable technique was developed (Dube and Natarajan 1973) to study thin film specimens. In this technique the thin specimen is placed longitudinally at the centre of the broad side of a hollow rectangular waveguide excited in the TE_{10} mode so that the whole specimen remains in maximum electric field as shown in figure 3. In this geometry Maxwell's equations can be solved for the TE_{10} mode by taking Z variations of the form $\exp(-i\beta Z)$ (axis of waveguide is along the z-direction). Then by usual
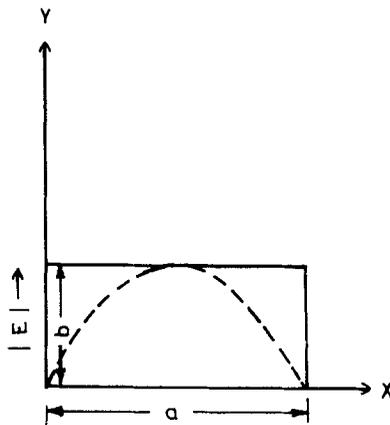
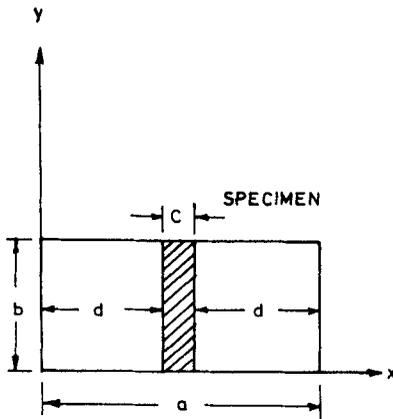**Figure 2.** Electric field distribution in a rectangular waveguide in the TE_{10} mode.

Figure 3. Cross-section of a rectangular waveguide with specimen.

methods (Collin 1960), the following dispersion relation is obtained

$$\tan\left[d(k^2 - \beta^2)^{1/2}\right]\tan\left[\tfrac{1}{2}c(\varepsilon k^2 - \beta^2)^{1/2}\right] = \frac{(k^2 - \beta^2)^{1/2}}{(\varepsilon k^2 - \beta^2)^{1/2}} \tag{1}$$

where $k = \omega/C_0$ is the free space propagation vector and ω is the angular frequency. Equation 1 is a transcendental equation, and its solution can be obtained only by graphical methods. For complex dielectric constants, the solution of (1) is very difficult even graphically. Therefore, we simplify (1) under the approximation $\tfrac{1}{2}c(\varepsilon k^2 - \beta^2)^{1/2} < 1$, which holds very well in the case of thin films; hence using $\varepsilon = \varepsilon_r - i\varepsilon_i$ and $\beta = \beta_r - i\beta_i$, we get the following expressions for the real and imaginary parts of the complex dielectric constant:

$$\varepsilon_r = \frac{1}{k^2}\left((\beta_r^2 - \beta_i^2) + \frac{a_1 a_3 + a_2 a_4}{a_3^2 + a_4^2} \cdot \frac{1}{\tfrac{1}{2}cd}\right), \tag{2}$$

$$\varepsilon_i = \frac{1}{k^2}\left(2\beta_r\beta_i - \frac{a_2 a_3 - a_1 a_4}{a_3^2 + a_4^2} \cdot \frac{1}{\tfrac{1}{2}cd}\right), \tag{3}$$

where

$$a_1 = d \operatorname{Re} (k^2 - \beta^2)^{1/2},$$

$$a_2 = d \operatorname{Im} (k^2 - \beta^2)^{1/2},$$

$$a_3 = \frac{2 \sin 2a_1}{\exp(2a_2) + \exp(-2a_2) + 2 \cos 2a_1},$$

and

$$a_4 = \frac{\exp(2a_2) - \exp(-2a_2)}{\exp(2a_2) + \exp(-2a_2) + 2 \cos 2a_1},$$

and

β_i = attenuation introduced by unit length of the specimen (measured in nepers m^{-1})

$\beta_r = \beta_p + \beta_0$ (measured in radians m^{-1})

where

β_p = phase shift produced by the unit length of the specimen

and

$\beta_0 = 2\pi/\lambda g$, where λg is the guide wavelength in the empty guide,

c = thickness of specimen, and

d = $\frac{1}{2}(a - c)$, a being the width of the waveguide (figure 3).

Thus, in complex dielectrics, the value of ε_r and ε_i can be obtained from (2) and (3). For loss-free dielectrics the solution of (1) can be obtained without any approximations. The two main quantities required to be measured experimentally are the phase and attenuation introduced by the specimen. Two experimental arrangements have been proposed (Dube and Natarajan 1973, 1974). Either may be used for these measurements. Both make use of rectangular waveguide system excited in the TE$_{10}$ mode. One arrangement has been shown in figure 4.

Microwave power is obtained from a Klystron source. The power is divided into two arms at the 10-dB directional coupler. The lower arm contains the standard impedance, *i.e.*, a standard variable attenuator and a standard variable phase shifter (0° → 360°). The upper arm contains the sample holder which is an approximately 10 cm long waveguide having a sharply cut longitudinal slot at the middle. The sample is introduced through this slot. The powers from the upper and the lower arms are again combined at a 3-dB directional coupler. The power output from the directional coupler is fed to the detecting system.

To start with, a null point is obtained in the detector by properly adjusting the variable attenuator and the phase shifter. The introduction of the specimen disturbs the balance. The null conditions are restored by varying the attenuator and phase shifter. Thus the phase shift and attenuation introduced by the sample can be accurately measured. Large sample lengths should be used for producing large phase shifts and attenuations.

Reflections from the edge of the sample are normally extremely small. But these can be detected by a separate experiment and if required, tuner no. 8 in figure 4 may be used for matching purposes.

The minimum detectable phase shift and attenuation in the measuring system were 0·25° and 0·02 dB, respectively.
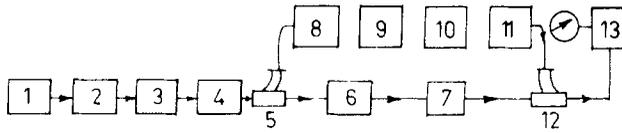
Figure 4. Schematic diagram of the experimental set up: 1. Klystron power source; 2. Frequency meter; 3. Padder attenuator: 4. Ferrite isolator; 5. and 12, Directional couplers; 6. Calibrated variable phase shifter; 8 and 11 tuners; 9. Variable attenuator; 10. sample holder waveguide; 13. Amplifier and meter.
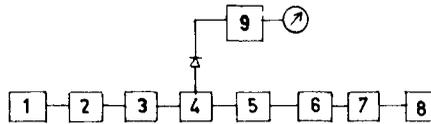

Figure 5. Schematic diagram of the experimental set-up; 1. Klystron power source; 2. Frequency meter; 3. Ferrite isolator; 4. Waveguide travelling detector; 5. variable attenuator; 6. E-H tuner; 7. sample holder waveguide; 8. movable short; 9. Amplifier and meter.

To study dielectric parameters as a function of temperature, the guide section containing the specimen may be cooled by a low temperature bath around the section or may be heated by a cylindrical furnace.

The other experimental set-up (Dube and Natarajan 1974) is shown in figure 5. This is a simpler arrangement and does not require calibrated variable phase shifter and attenuator and is as accurate as the first set-up.

Here the travelling wave detector is used to measure the shift of the voltage minimum when the specimen is introduced. Let the specimen length be L cm and the phase shift produced be X cm, then phase introduced by the specimen per unit length is

$$\beta_s = \beta_0 \frac{X}{L}$$

Thus the total phase constant β_r is given by

$$\beta_r = \beta_0 + \beta_s$$

or $$\beta_r = \beta_0 \left(1 + \frac{X}{L}\right)$$

For measuring the dielectric loss, the VSWR is taken with and without the specimen. Let these be ρ_1 and ρ_2; then

$$r = \frac{\rho_1 - 1}{\rho_1 + 1} = \exp(-\alpha), \tag{4}$$

and

$$r' = \frac{\rho_2 - 1}{\rho_2 + 1} = \exp - (\alpha + 2\beta_i L), \tag{5}$$

where r and r' are the reflection coefficients of the system with and without the sample and α is the attenuation constant. Hence β_i can be calculated from (4) and (5).

4. Results and discussion

The accuracy of the proposed technique for measuring the dielectric parameters of films was checked by studying two materials namely, myler and teflon whose dielectric properties are well known over a wide frequency range including microwaves. Results are summarised in table 1.

In the recent past the technique has been used to study films/sheets/wafers of varying nature and differing widely in electrical properties. Some of the results are given below.

Complex permittivity and conductivity have been studied for silicon semiconductor (Natarajan *et al* 1978). ε_r and ε_i values for one sample of *p*-silicon for various thicknesses are given in table 2. The thickness of the starting sample was reduced by etching and lapping.

The d.c. conductivities measured by the four-probe technique and the microwave conductivities obtained from the relation $\sigma_{a.c.} = \omega\varepsilon_0\varepsilon_i$ are compared in table 3 for three samples of *p*-silicon.

The microwave conductivity is slightly less than the d.c. values. This is somewhat

Table 1. Dielectric parameters of teflon and mylar films at 9·375 GHz at room temperature (310°K)

Sample	Sample thickness (μm)	Phase shift per unit length (deg.)	Dielectric constant	
			Present work	Literature
Teflon	90	0·61	2·03	2·08[a]
	130	0·96	2·03	2·07 ± 0·03[b]
	186	1·28	2·02	
Mylar	75	1·15	3·13	
	130	1·91	3·10	3·145[c]
	250	3·74	3·18	

[a] Von Hippel (1961); [b] Pilipenko *et al* (1970); [c] Conklin (1965).

Table 2. Complex permittivity for a *p*-type silicon (5·45 Ω cm) at room temperature (310°K) and at a frequency of 9·410 GHz

Thickness (microns)	Phase shift per unit length (degrees)	Attenuation per unit length (decibles)	ε_r	ε_i
272	15·0	8·6	11·61	30·58
226	13.0	7·5	11·65	32·12
200	12·0	6·9	11·81	33·13
180	10·8	6·1	11·58	32·68
140	8·6	5·1	11·62	34·61
94	5·8	3·4	11·41	34·76
70	4·5	2·4	11·63	32·69

Table 3. Comparison of d.c. and a.c. conductivities for three samples of p-silicon

d.c. conductivity by four-probe technique $(\Omega m)^{-1}$	Microwave conductivity from $\sigma_{a.c.} = \omega \varepsilon_0 \varepsilon_i$ $(\Omega m)^{-1}$
10.26	9.79
15.63	15.17
18.35	17.97

consistant with the usual expressions for a.c. and d.c. conductivities.

$$\sigma_{\text{a.c.}} = \frac{ne^2 v}{m[v^2 + \omega^2]},$$

and

$$\sigma_{\text{d.c.}} = \frac{ne^2}{mv},$$

where m, e, n and v are the mass, charge, concentration and collision frequency of the holes in p-silicon.

The microwave data may also be used to determine other transport parameters of silicon. The dielectric constant and conductivity in terms of effective mass m^* and relaxation time τ may be written as

$$\varepsilon_r = \varepsilon_L - \frac{ne^2 \tau^2}{\varepsilon_0 m^* [1 + (\omega\tau)^2]},$$

$$\sigma = \frac{ne^2 \tau}{m^* [1 + (\omega\tau)^2]} = \omega \varepsilon_0 \varepsilon_i,$$

where ε_L is the lattice dielectric constant and ε_0 is the permittivity of free space. From the above two equations, we get

$$\tau = \frac{\varepsilon_L - \varepsilon_r}{\omega \varepsilon_i}.$$

With the present results the relaxation time in silicon comes out to be around 10^{-13} sec, which agrees well with the data ($\simeq 10^{-13}$ sec) produced by Blatt (1968).

The conductivity variations with temperature for p-silicon samples (d.c. resistivities 10·5, 40 and 170 ohm cm) in the temperature range 190–525°K are plotted in figure 6. The curves for the three silicon samples exhibit broadly similar characteristics. *viz* the conductivity initially decreases with increase of temperature and later increases very rapidly with temperature. This is, of course, expected, considering the relative predominance of extrinsic and intrinsic conduction mechanisms. The conductivity of the samples (having resistivities 10·5 and 40 ohm-cm) decreases from room temperature to 440°K. In this temperature region lattice scattering by acoustical phonons plays the dominant role (Blackmore 1969). As the temperature is raised further, the concentration of free electrons and holes increases and the intrinsic conduction predominates thereafter. In the case of the third sample (170 ohm cm, the highest resistivity of all the
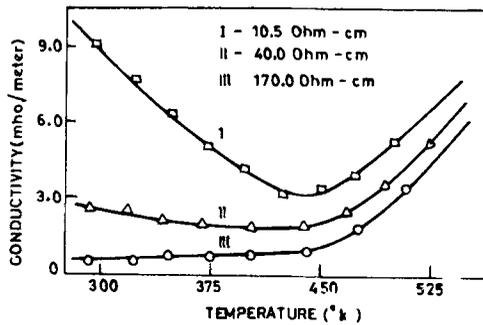
Figure 6. Microwave conductivity variation of silicon wafers in the temperature range 293–523°K.

three) the conductivity rises slowly from room temperature up to 440 K and increases very rapidly subsequently thereby showing its intrinsic behaviour right from room temperature. Bhar (1963) made similar measurements on temperature variations of microwave conductivity for bulk p-silicon samples having conductivities 0·7 and 0·1 mho m^{-1} in the temperature range 273–480°K. His results also resemble the curves of figure 6.

Electrical parameters have been measured for metal and semimetal films of thicknesses less than the skin depths deposited on glass substrates by vacuum thermal evaporation technique (Dube *et al* 1978).

Figures 7 and 8 show the phase shift and attenuation constants as a function of film thickness for the semimetal bismuth. Table 4 contains the results for ε_r and ε_i for bismuth films of different thicknesses. The high values of ε_r and ε_i are attributed to the presence of free carriers and are consistent with the results of Rahneberg (1971) for bismuth films measured by a different technique at 14·50 GHz. Gold and aluminium films also show high dielectric parameters of the order of 10^5 (Dube *et al* 1978).

Dielectric behaviour of pure polyvinyl chloride (PVC) and iodine-doped PVC has been studied at microwave frequencies using the proposed technique (Natarajan and Dube 1981). Pure PVC films were obtained by dissolving the polymer powder in tetrahydrofuran and permitting the evaporation of the solvent at room temperature. Iodine-doped PVC samples were prepared from the PVC solution in which a requisite amount of iodine was dissolved. The concentration of iodine in the samples varied from 4–40 % by weight.

Figure 9 depicts the variation of ε_r with temperature for all PVC samples. It is seen that at any fixed temperature, the dielectric constant increases as the iodine content in the sample increases. This effect is more pronounced for iodine concentrations above ~ 5 % (by weight). For temperatures above 90°C the dielectric constant decreases for all samples containing iodine.

The rise in ε_r with iodine concentration may be attributed to the presence of highly polarizable molecular iodine in the specimen. The presence of free iodine was detected in the absorption spectrum in the visible region.

The volume expansion due to transition from glassy to rubbery state seems to account for the decrease of ε_r with temperature in the vicinity of 85°C as the glass transition temperature (T_g) of PVC falls in the region 70–105°C (Brandrup and Immergut 1975). Variation of loss-tangent (tan δ) with temperature for different iodine
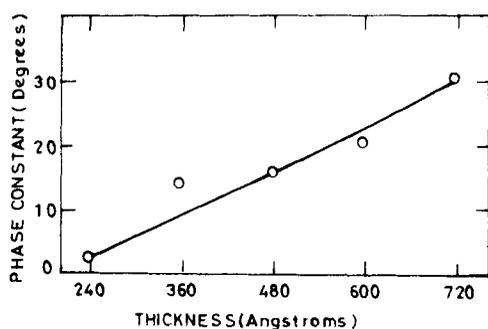
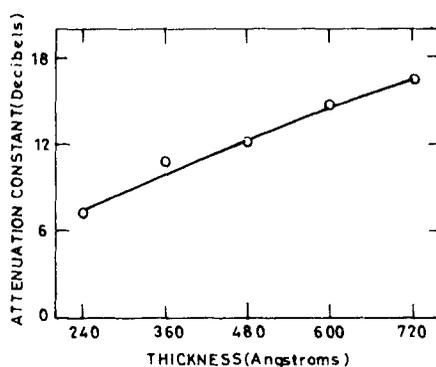**Figure 7.** Variation of phase constant as a function of bismuth film thickness.

Figure 8. Variation of attenuation constant as a function of bismuth film thickness.

Table 4. Complex permittivity for thin bismuth films for various thicknesses at 310°K and at a frequency of 9·410 GHz.

Thickness (angstroms)	Phase shift/ length (degrees)	Attenuation/ length (decibels)	Dielectric parameters ($\times 10^5$)	
			ε_r	ε_i
240	2·5	7·5	0·36	3·12
360	12·0	10·8	0·91	2·91
480	15·5	12·2	0·89	2·38
600	20·0	13·7	0·83	1·83
720	30·0	16·3	1·05	1·86

dopings has been shown in figure 10. For iodine content of 10% and above, well-defined loss-peak appears at around 90°C in each case. With increasing iodine content, the position of loss peak does not change; however, the peak height changes considerably. The fact that loss tangent maxima does not change in position but only in magnitude with iodine concentration shows that on increasing the doping the number of dipoles participating in the orientation process increases, while the frequency of orientation remains unchanged.
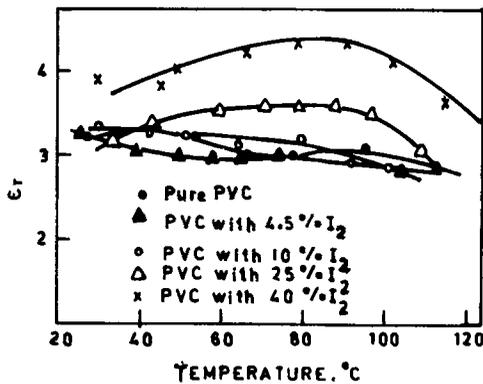
Figure 9. Variation of ε_r with temperature for pure and iodine doped PVC.
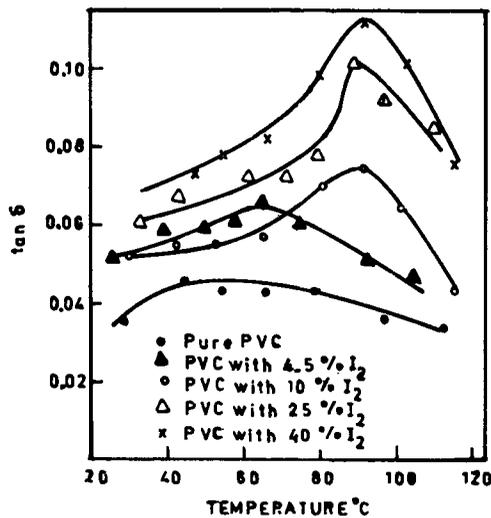

Figure 10. Variation of tan δ with temperature for pure and iodine doped PVC.

Fast ion conducting solids (also called superionic conductors) have become very important in the recent past due to their applications in many electrochemical devices. Electrical measurements on these materials require special attention due to the variety of electrodes (blocking, reversible and their combinations) (Bottelberghs 1978) used making the analysis more complex. The ionic conduction has been studied recently at microwave frequencies (Dube and Saraswat 1982) in silver iodide in the high conductivity α-phase. Thin pellets of AgI of varying packing density were used and dielectric and conductivity data obtained. This data can be transformed to 100 % dense bulk using correlation formulae (Landau and Lifshitz 1960; Leoyenga 1965; Dube 1970).

$$\varepsilon_p'^{1/3} - 1 = \delta(\varepsilon_b'^{1/3} - 1),$$

$$\varepsilon_b'' = \frac{\varepsilon_p''}{\delta}\,(\varepsilon_b'/\varepsilon_p')^{2/3},$$

Table 5. Conductivity and dielectric constant for α-AgI ($T = 160°C$) at 10 kHz

Packing fraction %	Observed parameters for pellets		Values converted to 100% density by correlation formulae		Values reported in literature in microwave region (Gebhardt et al 1980)	
	Dielectric constant	Conductivity $(\Omega\,cm)^{-1}$	Dielectric constant	Conductivity $(\Omega\,cm)^{-1}$	Dielectric constant	Conductivity $(\Omega\,cm)^{-1}$
72	12·0	0·044	21·74	0·092		
82	15·4	0·076	22·30	0·119		
86	16·9	0·084	22·45	0·110	22·00	1·22
92	19·4	0·088	22·75	0·116		

and

$$\sigma_b = \omega \varepsilon_0 \varepsilon_b'',$$

where ε_p' and ε_p'' are the dielectric constant and loss factor for the pellet of packing fraction δ and ε_b', ε_b'' are the corresponding parameters for the bulk. σ_b is the conductivity at angular frequency ω. ε_0 is permittivity of free space ($= 8\cdot85 \times 10^{-12}$ Fm^{-1}). Various results have been summarised in table 5. It is seen that while the deduced dielectric constant data match with experimentally reported value of 22, the conductivity results are found reduced by an order of magnitude. This reduction in conductivity is attributed to the presence of grain boundaries in the unsintered samples used in the above studies.

In conclusion, it may be said that the microwave technique described is very convenient and accurate and can be used to study electrical transport behaviour of materials differing widely in characteristics.

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