AN INVESTIGATION ON DIELECTRIC PROPERTIES OF MIXTURES AT RADIO FREQUENCIES

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ABSTRACT

The major formulae suggested for the dielectric constant of a mixture of two components, at least one of which has the form of particles containing many molecules, are related to one another. Assuming this very recently Pradhan and Gupta have derived three different formulae for determining dielectric constant for three different shaped particles. The validity of these equations have been tested in the present paper. Theoretical and experimental results have been compared. It is found that these results tally quite satisfactorily.

INTRODUCTION

Considerable theoretical effort has been expended on a problem that has no direct relation to atomic and molecular processes, but occurs in connection with dielectric constants of solids. The problem is to calculate the dielectric constant of a mixture of two materials, for instance, particles of the material suspended in a matrix of the other, from the dielectric constants of the component materials, or conversely from the measurements of the dielectric constant of mixture and from the knowledge of the dielectric of one component, to calculate the dielectric constant of the other component. The problem is encountered when, for instance, the material whose dielectric constant is to be measured is available only in the form of powder, with spaces between the particles.

Very little work has been carried out by different workers from time to time in an endeavour to clarify the behaviour of solid mixture material under alternating electric stress. When a solid mixture is subjected to high frequency electric stress, various factors come into play within the material of the solid mixture, depending on the physical and chemical complexities. This makes an understanding of the dielectric behaviour of solid insulating material under high frequency stress, a difficult matter.

Recently, it has been shown by Reynold and Hough (1957) that the formulae which are suggested in literature for mixture of two components can be derived from the fundamental formulae given by them. Smith (1956) has shown that the effective dielectric constant of a mixture not only depends on the dielectric constants of the components and their volume fractions but also on the densities of the particles.

According to Reynold and Hough, the dielectric constant of a mixture is a function of the volume fraction, shape and dielectric constant of each component. Theoretical calculations are most easily made for systems where the components are arranged in a regular array but normal mixtures show a random arrangement of the components. For the last fifty or sixty years many formulae have been suggested by many workers (Fricke, 1924; Polder and Van Santen, 1946; Niesel, 1952) for such mixtures, the method of derivation has varied greatly and it is not always obvious what relation exists between the formulae.

However, most recently Pradhan and Gupta (1959) have derived three different formulae for determining dielectric constants of mixtures of two components, having different shaped particles, i.e. spherical, lamellar, and rod type of one component embedded in a continuous medium of the other component, using very simple assumption. In this paper the authors have tested the validity of these formulae experimentally at three different radio frequencies.

In order to avoid difficulties the authors have taken the simplest, that is, the case of only two components, one of them being randomly dispersed in the other and the latter being continuous and homogeneous. The extension to more complicated cases has been discussed in detail in the previous work of Bruggeman (1935).

Theory

Reynold and Hough have derived equations for the cases (1) particles dispersed in a continuous medium and (2) where the mixture is a statistical one. These equations are written in the following forms respectively.

$$\epsilon = \epsilon_2 + (\epsilon_1 - \epsilon_2) v_1 f_1 \quad .. \quad .. \quad .. \quad .. \quad (1)$$

$$(\epsilon - \epsilon_1) v_1 f_1 + (\epsilon - \epsilon_2) v_2 f_2 = 0 \quad .. \quad .. \quad .. \quad (2)$$

where ϵ, ϵ_1 and ϵ_2 are dielectric constants of mixture and two components of mixtures respectively, v_1 and v_2 are fractional volumes of components, f_1 and f_2 are the field ratios.

The equations (1) and (2) represent the general forms of the formulae for the dielectric constants of mixtures. Theoretically it appears that both the equations are equivalent but as soon as we make certain approximation in the value of f_1 they are no longer so.

In this investigation equation (1) has been used since one of the components is continuous and homogeneous, i.e. (Rosin).

The determination of values of f_1 for ellipsoidal particles has already been dealt with in the previous paper (Pradhan and Gupta, 1959) and is given by equations (3).

$$\left.\begin{aligned} f_1 &= \frac{3\epsilon_1}{\epsilon_1 + 2\epsilon} && \text{(for Spherical)} \\ &= \frac{\epsilon + 2\epsilon_1}{3\epsilon_1} && \text{(for Lamellae)} \\ &= \frac{\epsilon_1 + 5\epsilon}{3(\epsilon + \epsilon_1)} && \text{(for Rod like)} \end{aligned}\right\} \quad .. \quad .. \quad .. \quad (3)$$

On putting these values in equation (1) and knowing that ϵ_1, ϵ_2 and ϵ are complex, we have, on simplifying, the three equations for different types of particles as derived earlier (Pradhan and Gupta, 1959).

Case I. (For Spherical)

$$4\epsilon' = \epsilon_2'(2-3v_1) - \epsilon_1'(1-3v_1) + R^{\frac{1}{2}} \cos\left(\tfrac{1}{2}\tan^{-1}\frac{Y}{X}\right) \quad .. \quad .. \quad (4)$$

$$4\epsilon'' = \epsilon_2''(2-3v_1) - \epsilon_1''(1-3v_1) + R^{\frac{1}{2}} \sin\left(\tfrac{1}{2}\tan^{-1}\frac{Y}{X}\right) \quad .. \quad .. \quad (5)$$

where

$$X = (\epsilon_1'^2 - \epsilon_1''^2)(1-3v_1)^2 + (\epsilon_2'^2 - \epsilon_2''^2)(2-3v_1)^2 + 2(\epsilon_1'\epsilon_2' - \epsilon_1''\epsilon_2'')(2+9v_1-9v_1^2)$$

$$Y = 2\epsilon_1'\epsilon_1''(1-3v_1)^2 + 2\epsilon_2'\epsilon_2''(2-3v_1)^2 + 2(\epsilon_1''\epsilon_2' + \epsilon_1'\epsilon_2'')(2+9v_1-9v_1^2)$$

and

$$R^2 = X^2 + Y^2$$

Case II. (For Lamellae)

$$\epsilon' = \frac{a_1 + a_2 v_1 + a_3 v_1^2}{c_1 + c_2 v_1 + c_3 v_1^2} \quad .. \quad .. \quad .. \quad .. \quad (6)$$

$$\epsilon'' = \frac{b_1 + b_2 v_1 + b_3 v_1^2}{c_1 + c_2 v_1 + c_3 v_1^2} \quad .. \quad .. \quad .. \quad .. \quad (7)$$

where

$$a_1 = 9\epsilon_2'(\epsilon_1'^2 + \epsilon_1''^2)$$

$$a_2 = 3(\epsilon_1'\epsilon_2'^2 - 3\epsilon_1'^2\epsilon_2' + 2\epsilon_1'^3 + \epsilon_1'\epsilon_2''^2 + 2\epsilon_1'\epsilon_1''^2 - 3\epsilon_1''^2\epsilon_2')$$

$$a_3 = 2(2\epsilon_1'^2\epsilon_2' - \epsilon_1''^2\epsilon_2' - \epsilon_1'\epsilon_2'^2 - \epsilon_1'^3 + 2\epsilon_1'\epsilon_1''\epsilon_2'' - \epsilon_1'\epsilon_2''^2 - \epsilon_1'\epsilon_1''^2 + \epsilon_1''^2\epsilon_2')$$

$$b_1 = 9\epsilon_2''(\epsilon_1'^2 + \epsilon_1''^2)$$

$$b_2 = -3[3\epsilon_2''(\epsilon_1'^2 + \epsilon_1''^2) - \epsilon_1''(\epsilon_2''^2 + \epsilon_2'^2) + 2\epsilon_1'^2\epsilon_1'' - 2\epsilon_1''^2]$$

$$b_3 = 2[\epsilon_1''^3 + \epsilon_1'^2\epsilon_1'' + \epsilon_1''\epsilon_2'^2 + \epsilon_1''\epsilon_2''^2 - 2\epsilon_1''^2\epsilon_2'' - 2\epsilon_1'\epsilon_2'\epsilon_1'']$$

and

$$c_1 = 9(\epsilon_1'^2 + \epsilon_1''^2)$$

$$c_2 = 6[(\epsilon_2' - \epsilon_1')\epsilon_1' + \epsilon_1''(\epsilon_2'' - \epsilon_1'')]$$

$$c_3 = [(\epsilon_2' - \epsilon_1')^2 + (\epsilon_2'' - \epsilon_1'')^2]$$

Case III. (For Rod like)

$$6\epsilon' = (\epsilon_2' - \epsilon_1')(3 - 5v_1) + R^{\frac{1}{2}} \cos\left(\tfrac{1}{2}\tan^{-1}\frac{Y}{X}\right) \quad .. \quad .. \quad (8)$$

$$6\epsilon'' = (\epsilon_2'' - \epsilon_1')(3 - 5v_1) + R^{\frac{1}{2}} \sin\left(\tfrac{1}{2}\tan^{-1}\frac{Y}{X}\right) \quad .. \quad .. \quad (9)$$

where

$$X = (\epsilon_1'^2 - \epsilon_1''^2)(9 - 18v_1 + 25v_1^2) + (\epsilon_2'^2 - \epsilon_2''^2)(9 - 30v_1 + 25v_1^2) + 2(\epsilon_1'\epsilon_2' - \epsilon_1''\epsilon_2'')(9 + 24v_1 - 25v_1^2)$$

$$Y = 2\epsilon_1'\epsilon_1''(9 - 18v_1 + 25v_1^2) + 2\epsilon_2'\epsilon_2''(9 - 30v_1 + 25v_1^2) + 2(\epsilon_1'\epsilon_2'' + \epsilon_1''\epsilon_2')(9 + 24v_1 - 25v_1^2)$$

Experimental Technique

Hartshorn's (Hartshorn and Ward, 1936) method for the measurement of dielectric constant and loss tangent at radio frequencies has been used. The sample is placed between two metal plates forming a condenser of capacitance c_1. A coil and a thermionic voltmeter are connected to it. The circuit so formed is loosely coupled to the coil of a variable frequency oscillator. The experimental arrangement has already been discussed earlier by Sharma and Prakash (1958). The distance between the plates is adjusted by means of a vertical micrometer head attached to the upper plate, the lower plate remaining in a fixed position.

Samples in the form of a disc between 51 and 54 mm. in diameter preferably 53 mm. and about 2 to 3 mm. thick are required for measurement on this apparatus. The disc should be flat and of uniform thickness.

The size and shape of the samples are of utmost importance for obtaining accurate results and, therefore, the samples were carefully prepared. Powders were mixed in known percentages and were melted in a vessel at constant temperature, with air bubbles removed (Gupta and Pradhan, 1958). The molten mixture was stirred well until the mixture became so viscous that it did not allow the heavy particles to settle down in liquid rosin. Molten mixture was poured in a brass cast and the samples obtained from the cast were rubbed and polished. Samples were desiccated for 36 hours before experiments (Surber and Crouch, 1948).

The samples thus prepared were placed between two copper electrodes, and the main condenser screwed down till the sample was held. Now the apparatus was tuned to the oscillator frequency and the maximum deflection (θ_1) noted. The side micrometer condenser was varied first in one direction and then in the other until the deflection became $\frac{\theta_1}{2}$. The capacitance change, ΔC_1, between these two readings was thus obtained.

With the circuit in response the sample was removed and the resonance again restored by the adjustment of the main condenser only. Reading of the maximum deflection θ_0 and the capacitance change ΔC_0 corresponding to deflection $\frac{1}{2}\theta_0$ were obtained as before. In addition the reading of the main condenser, Cm, was noted. The capacitance of the specimen C_s is equal to the capacitance Cm less the edge capacitance.

The dielectric constants K of rosin samples and mixtures are given by

$$K = \frac{3{\cdot}6C_s t}{r^2} \quad .. \quad .. \quad .. \quad .. \quad .. \quad (10)$$

where C_s is the capacitance of the sample in $\mu\mu f$, t is the thickness of the sample in cms. and r is the radius of the electrode (2·5 cms.). The tangent loss is given by

$$\tan \delta = \frac{\Delta C_1 - \Delta C_0}{2C_s} \quad .. \quad .. \quad .. \quad .. \quad (11)$$

Results and Discussion

Table I gives the dielectric constant of mixtures along with that of rosin at three different frequencies. Table II gives the loss tan δ along with dielectric conductivity of these samples. Dielectric constants of rosin thus obtained were used in calculating the theoretical values for mixtures. Values of dielectric constant for SiO_2, MgO and ZnO were taken from the measurements made by Smith (1956) and are given in Table III.

The values of dielectric constants of mixtures ($= \epsilon'\epsilon_0$) and loss tan δ ($= \epsilon''/\epsilon'$) have been calculated theoretically by equations (4–9) for three different shapes of particles embedded in rosin with different percentages of the different particles. Dielectric conductivities were calculated from the following formula (Sharma, 1960) for conductivity

$$\sigma = 0{\cdot}0053024 \left(\frac{\pi k \tan \delta}{\lambda}\right) \text{ mhos/metre} \quad .. \quad .. \quad (12)$$

where wavelength λ (is in metre).

In the present experiment SiO_2 (Spherical), MgO (Lamellae) and ZnO (Rod type) were used. Their shapes and axial ratios had already been tested and verified with electron and optical microscopes by Smith (1956). It is well known that ZnO crystals naturally grow as cylinders. When these particles are pressed against one another, they coalesce, and flatten against one another forming oblong

TABLE I

Experimental Values for Dielectric Constants of Mixtures and Rosin

Substance	Percentage	Frequencies		
		61 Kc/s.	7·25 Mc/s.	15·6 Mc/s.
Rosin	100	2·85	2·68	2·62
SiO_2	10	3·01	2·86	3·79
,,	20	3·18	3·04	2·97
,,	30	3·28	3·22	3·17
,,	40	3·53	3·44	3·40
,,	50	3·75	3·65	3·59
MgO	10	3·48	3·30	3·27
,,	20	4·12	3·98	3·93
,,	30	4·85	4·69	4·65
,,	40	5·58	5·44	5·42
,,	50	6·32	6·25	6·19
ZnO	10	3·25	3·08	3·01
,,	20	3·68	3·46	3·39
,,	30	4·10	3·39	3·85
,,	40	4·60	4·44	4·41
,,	50	5·11	4·99	4·91

TABLE II

Loss Tangent δ and Dielectric Conductivity of Mixtures and Rosin

Substance	Per-centage	Frequencies					
		61 Kc/s.		7·25 Mc/s.		15·6 Mc/s.	
		tan δ $\times 10^{+4}$	σ (in μ mhos/metre)	tan δ $\times 10^{+4}$	σ (in μ mhos/metre)	tan δ $\times 10^{+4}$	σ (in μ mhos/metre)
Rosin	100	43·0	$41{\cdot}5\times10^{-3}$	52·0	$56{\cdot}1\times10^{-1}$	58·0	132×10^{-1}
SiO_2	10	38·9	$39{\cdot}2\times10^{-3}$	46·3	$53{\cdot}2\times10^{-1}$	52·0	125×10^{-1}
,,	20	34·0	36·5	41·0	49·3	45·7	117
,,	30	29·8	33·1	35·5	45·7	39·8	108
,,	40	24·1	28·1	29·7	39·9	33·0	95
,,	50	20·1	25·3	24·5	35·3	27·3	83
MgO	10	34·2	$39{\cdot}1\times10^{-3}$	40·2	$53{\cdot}0\times10^{-1}$	45·0	125×10^{-1}
,,	20	27·1	36·7	32·2	49·6	35·0	118
,,	30	21·8	33·8	25·0	45·9	27·5	109
,,	40	17·0	30·6	19·8	41·4	22·1′	98
,,	50	13·2	26·8	15·1	36·7	17·1	86
ZnO	10	37·0	$40{\cdot}3\times10^{-3}$	45·5	$55{\cdot}8\times10^{-1}$	50·2	132×10^{-1}
,,	20	31·5	38·3	38·1	53·1	42·0	124
,,	30	26·4	35·6	31·9	49·5	35·0	116
,,	40	21·6	32·7	26·0	44·8	28·9	105
,,	50	17·1	28·6	20·5	39·4	22·9	94

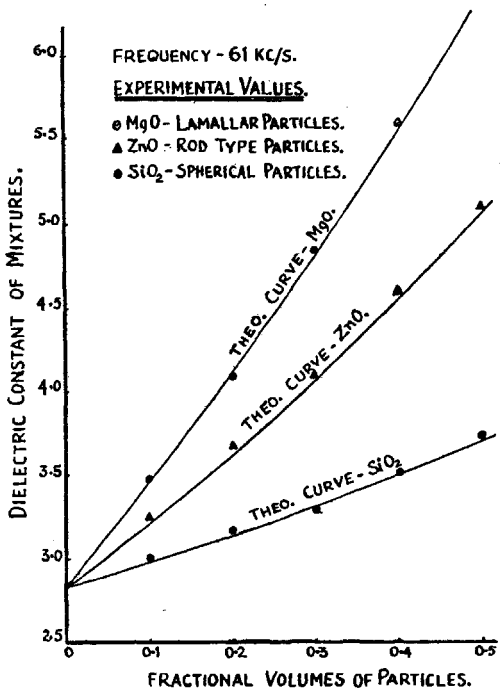

FIG. 1. Comparison of Experimental and Theoretical Values of Dielectric Constants at 61 Kc/s.

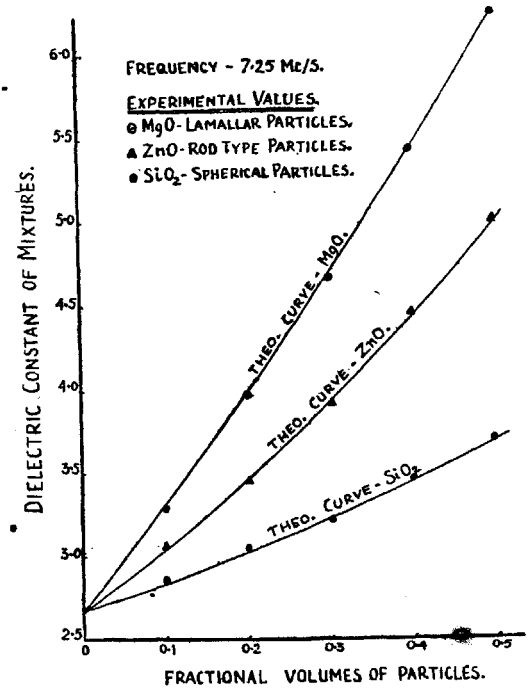

FIG. 2. Comparison of Experimental and Theoretical Values of Dielectric Constants at 7·25 Mc/s.

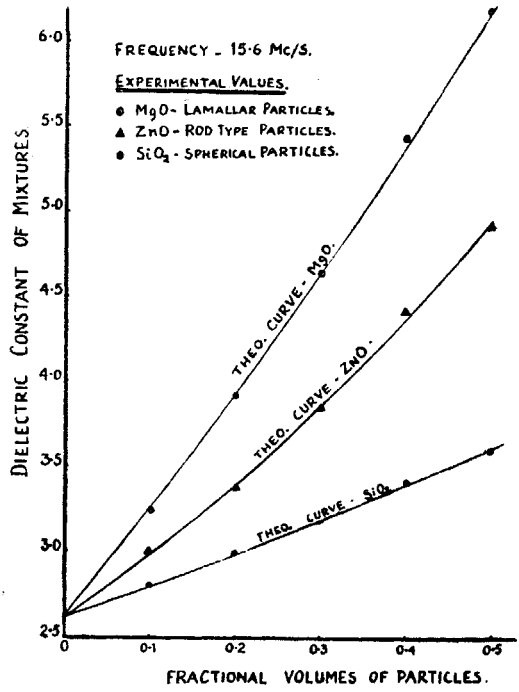

FIG. 3. Comparison of Experimental and Theoretical Values of Dielectric Constants at 15·6 Mc/s.

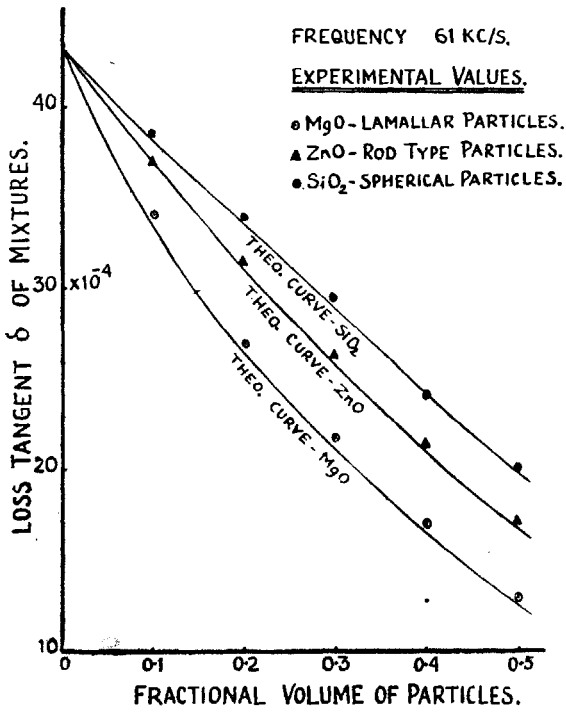

FIG. 4. Comparison of Experimental and Theoretical Values of Loss Tangent at 61 Kc/s.

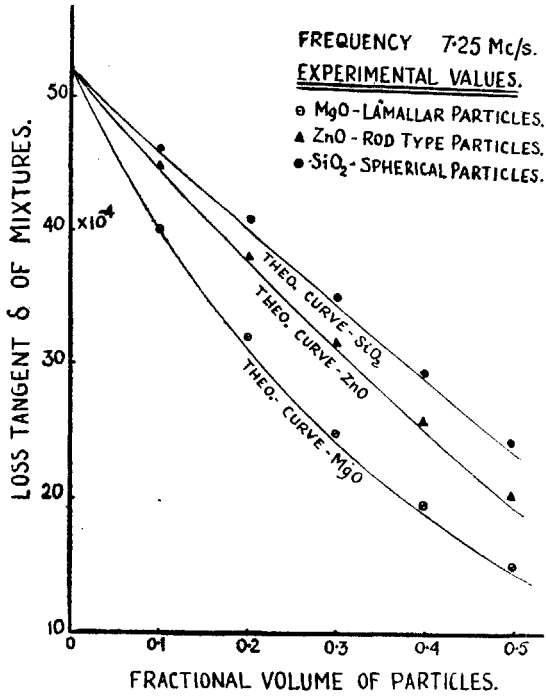

FIG. 5. Comparison of Experimental and Theoretical Values of Loss Tangent at 7·25 Mc/s.

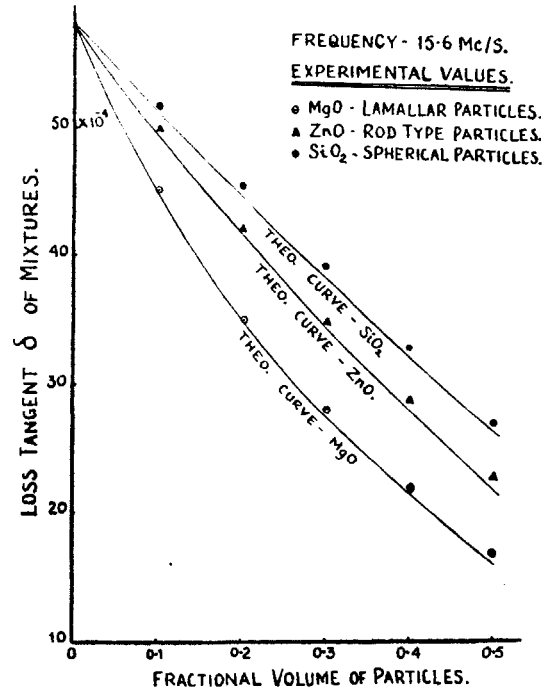

FIG. 6. Comparison of Experimental and Theoretical Values of Loss Tangent at 15·6 Mc/s.

TABLE III

Dielectric Constants of Particles

Substance	Dielectric Constant
ZnO	8·13
SiO_2	4·80
MgO	1·1·00

particles. The silica particles consist of fairly hard spherical kinds. The experimental data confirm the fact that the particles act as hard spheres.

Curves (Figs. 1 to 6) show that there is good agreement between theoretical and experimentally obtained values. Graphs show that in each case the dielectric constant increases as the fraction volume increases and loss tangent also increases with v_1. But as the frequency increases dielectric constant decreases, while loss tangents show an increase with frequency. These experimental data verify and test the validity of the formulae derived earlier (Pradhan and Gupta, 1959).

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