

Dielectric study of particulate material in e.m. field at microwave frequencies

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Abstract : The present investigation aims at finding out the extent upto which microwaves can be used as a probe to estimate the presence of conducting impurity in some powdered materials. Dielectric properties of pure polyvinyl chloride (PVC) powder and mixtures of pure PVC with different percentages of copper, aluminium and magnesium powders as impurity varying from five to eighty and fifty percent by weight are studied at microwave frequency 10.17 GHz and temperature 34.5°C. Presence of metal modifies dielectric behaviour of pure PVC powder to a great extent. These studies may be useful for estimating quantitatively and qualitatively the percentage of metal in natural ore samples and mineral rocks. This work is the extension of our previous work [1].

Keywords : Dielectric parameters, dipolar motion, dipolar hinderance, conducting centres, energy dissipation, relaxation time.

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1. Introduction

PVC has a wide range of applications as insulators, isolators and passivation devices, dielectrics for the capacitors *etc*, it is therefore very useful in micro electronics and optical waveguide systems. The simplicity of fabrication makes PVC insulation pre-eminent in the wire building field. It is therefore, used as a base material in powder form to prepare various mixtures with metal impurities. The corresponding change in permittivity ϵ' and dielectric loss ϵ'' of the mixtures of PVC with known percentages of copper, aluminium and magnesium powder is observed. Such measurements may lead to understand the behaviour of particulate materials in an electromagnetic field at microwave frequencies. The selection of Cu, Al and Mg metal powder is made because of appreciable difference in their conductivities and these are easily available in powder form.

2. Experimental details

The experimental set up, theory and procedure used for the present work is the same as used earlier by Yadav and Gandhi [2]. Microwave power obtained from the source forms standing waves inside the slotted section due to reflections from shorting plunger situated in the dielectric cell. With the insertion of powder in the cell, a shift of standing wave pattern is observed. The experimental details and determination of dielectric parameters are given elsewhere [2]. The PVC, copper, aluminium and magnesium powders used for the present investigation have grain sizes ranging from 125–180 microns. Nine different samples with copper powder varying from 5 to 80% six different samples with aluminium powder varying from 5 to 50% and six different samples with magnesium powder varying from 5 to 50% by weight in pure PVC powder varying are prepared. To ensure uniform mixing of metal powder in PVC powder, the mixtures are shaken well vigorously for a long time before subjecting them for the experimental measurements. The same process is repeated for at least fifty times with each sample and the most probable values of λ_d and α_d are obtained for each sample. All measurements are carried out at a constant microwave frequency 10.17 GHz and at room temperature 34.5°C. The set up is so arranged that the equal pressure is being exerted on the sample inside the measurement cell during the course of measurements. A sample leveling piece is used to level the sample inside the cell to obtain uniform sample thickness from corner to corner of the cell. The temperature is maintained constant within $\pm 0.5^\circ\text{C}$.

The dielectric constant (ϵ') and dielectric loss (ϵ'') for pure PVC powder as well as for different mixture compositions (PVC + metal) are determined by using eqs. (1) and (2) respectively which are given below :

$$\epsilon' = (\lambda_0/\lambda_c)^2 + (\lambda_0/\lambda_d)^2 [1 - (\alpha_d/\beta_d)^2] \quad (1)$$

$$\epsilon'' = 2(\lambda_0/\lambda_d)^2(\alpha_d/\beta_d) \quad (2)$$

where α_d is the attenuation and $\beta_d = 2\pi/\lambda_d$ is the phase shift introduced per unit length of the sample. λ_0 , λ_c and λ_d are the free space wave length, cutt-off wave length and the wave length in the dielectric medium respectively.

The other dielectric parameters like a.c. conductivity ($\sigma = \omega\epsilon_0\epsilon''$), relaxation time ($\tau = \epsilon''/\omega\epsilon'$) and loss tangent ($\tan\delta = \epsilon''/\epsilon'$) are determined using the values of ϵ' and ϵ'' determined as above.

Saraswat and Dube [3] have reported earlier that for powdered compacts of $\beta\text{-AgI}$, the ϵ' and ϵ'' values remain constant within three percent over a frequency range of 9 to 26 GHz indicating the complete absence of structural effects in the sample. It is therefore, assumed here that no structural effects are present in the powdered samples under investigation. Hence, all measurements are carried out at a single microwave frequency.

The experimental values of dielectric constant (ϵ'_{exp}) for all mixture compositions containing metal as impurity are compared with those values (ϵ'_{cal}) calculated theoretically by using three established relations namely Maxwell-Wagner [4–6] absorption expression (3), Bruggeman's [7] formula (4), modified Bruggeman's formula [7] eq. (5) and our empirically modified Maxwell-Wagner absorption relation (6). The three theoretical relations are as under :

The Maxwell-Wagner absorption relation is

$$\epsilon' = \epsilon'_\gamma \left[1 + S / (1 + \omega^2 T^2) \right] \quad (3)$$

where $S = 9\gamma\epsilon'_1 / (2\epsilon'_1 + \epsilon'_2)$

$$\epsilon'_\gamma = \epsilon'_1 \left[1 + 3\gamma(\epsilon'_2 - \epsilon'_1) / (2\epsilon'_1 + \epsilon'_2) \right]$$

and

$$T = (2\epsilon'_1 + \epsilon'_2) / (4\pi \times 90 \times 10^{10} \sigma_2)$$

With γ = volume fraction of phase 2 material (metal) ϵ'_1 - dielectric constant of phase 1 material, ϵ'_2 - dielectric constant of phase 2 material (metal), σ_2 - conductivity of phase 2 material (metal).

The Bruggeman's formula is

$$\epsilon' = \epsilon'_1 / (1 - \gamma)^3 \quad (4)$$

and modified Bruggeman's formula is

$$\epsilon' = \epsilon'_1 (1 + 3\gamma) \quad (5)$$

Here symbols have their usual meanings.

We have modified Maxwell-Wagner absorption relation (3) empirically by replacing S by S' . Our empirically modified Maxwell-Wagner absorption relation is

$$\epsilon' = \epsilon'_\gamma \left[1 + (S - \gamma) / (1 + \omega^2 T^2) \right] \quad (6)$$

where

$$S' = S - \gamma = (7\epsilon'_1 - \epsilon'_2) / (2\epsilon'_1 + \epsilon'_2) \quad (7)$$

where symbols have their usual meanings.

The research grade PVC powder used for the present investigations is obtained from Apex Chemicals, Bombay. The research grade metal powders copper, aluminium and magnesium used for the study are 99% pure and are obtained from Metal Powder Company Ltd., Madurai.

3. Results and discussion

Experimental dielectric parameters like ϵ' , ϵ'' , τ and $\tan\delta$ for pure PVC powder as well

as for different mixture compositions of (PVC + Cu), (PVC + Al) and (PVC + Mg) systems are obtained at a single microwave frequency 10.17 GHz and at a single temperature 34.5°C. Variations of dielectric constant ϵ' , dielectric loss ϵ'' and relaxation time τ with varying concentration of metal impurities in PVC powder for each metal are compared in Figures 1, 2 and 3 respectively. Experimentally determined values of

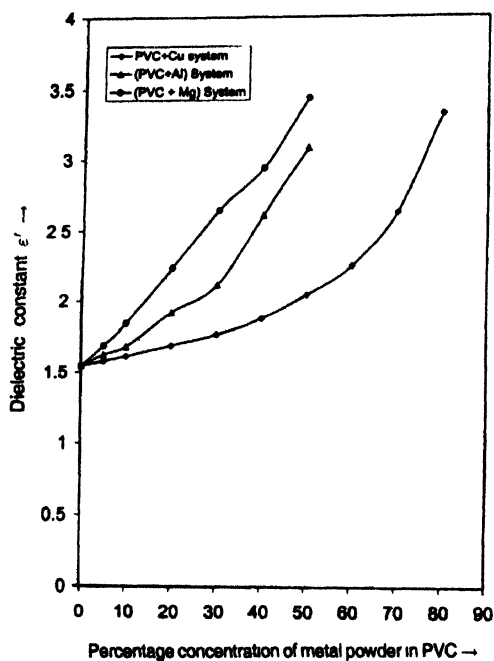


Figure 1. Comparison curves of variations of dielectric constant ϵ' with percentage concentration of metal powder in PVC powder.

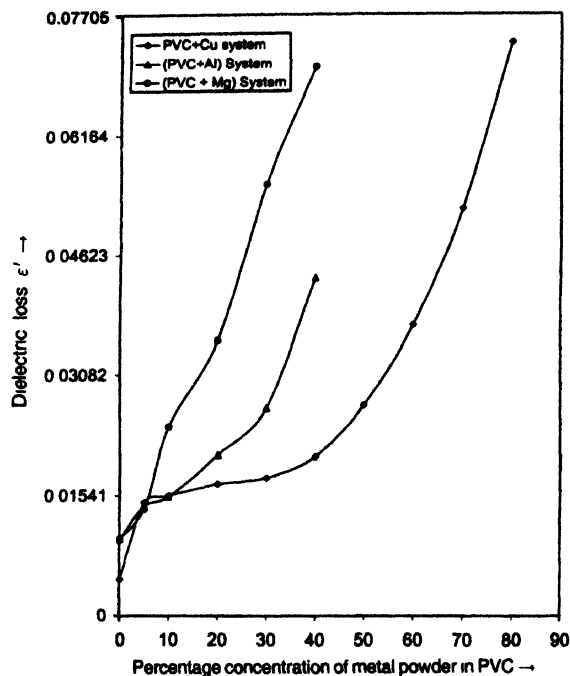


Figure 2. Comparison curves of variations of dielectric loss ϵ'' with percentage concentration of metal powder in PVC powder.

ϵ' are compared with the values calculated using various theoretical relations (3), (4), (5) and (6) in Figure 4. The literature [8] value of dielectric constant ϵ' for solid PVC is 2.62 which is much higher than the experimental value 1.548 for pure PVC powder of grain size 125–180 microns. The lower value of ϵ' for PVC powder than its solid sample is because of the reason that it contains air and hence, it is lower in density. Thus, the hinderance offered to the dipolar motion of a material in an electromagnetic field at microwave frequency for compact medium will be much higher than that of the material constituted by less bounded particles, which causes lower value of ϵ' for PVC powder than that of its solid sample. The similar conclusion was already been reported by other workers [3,9].

It is evident from Figure 1 that ϵ' values increases with increasing metal impurity in PVC powder. Further examination of Figure 1 reveals that the samples with conducting centres of higher electrical conductivity has lower values of ϵ' than that of

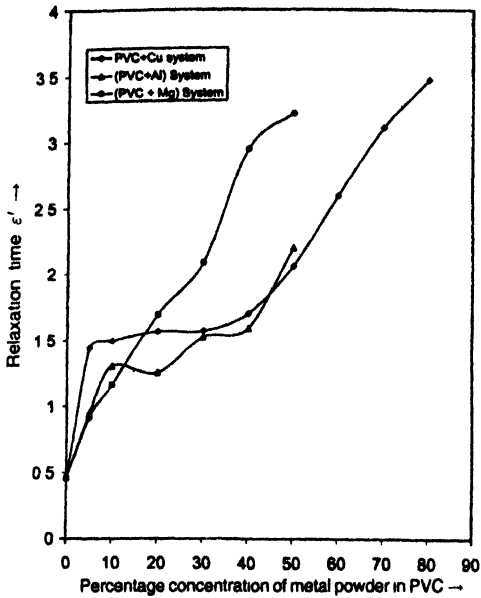


Figure 3. Comparison curves of variations of relaxation time (t) with percentage concentration of metal powder in PVC powder.

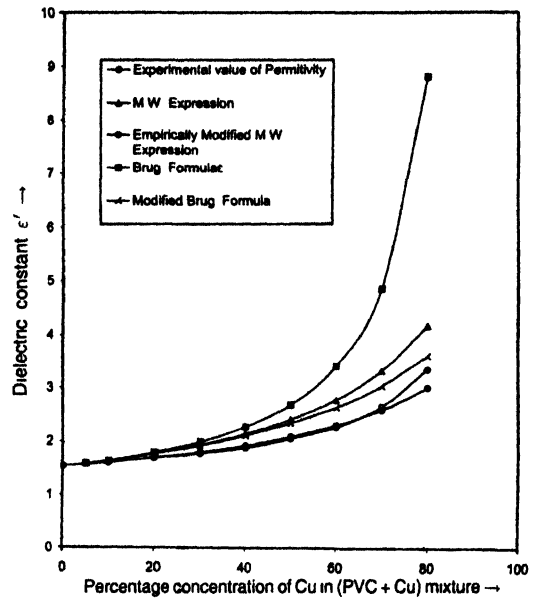


Figure 4. Comparison curves of variations of experimental and theoretically calculated values of ϵ' with percentage concentration of copper in (PVC + Cu) mixture compositions

the samples with conducting centres of lower electrical conductivity. This can be explained as under :

The dielectric behaviour of powdered sample with conducting centres is governed by different factors namely :

- (1) Firstly, when particles of second phase are dispersed within the principal one, energy dissipation will occur by scattering of radiation at interfacial boundaries in a non homogeneous medium which can be explained on the basis of Maxwell-Wagner-Sillars effect [3,4,10].
- (2) Secondly, the hinderance offered to the dipolar motion is a function of the basic nature of the particles of the dielectric powder, their grain size, packing fraction etc. The presence of conducting centres may modify this behaviour to some extent.
- (3) Thirdly, the amplitude of the field available to cause dipolar motion controls the degree of dipolar motion. With lower amplitude of the field the degree of dipolar motion causing lower effective hinderance in the dielectric medium with conducting centres. The incident energy gets absorbed due to electrical conduction and hence the amplitude of the field available to cause dipolar motion reduces, resulting into a total low effective hinderance.
- (4) Another possible factor, may be lower density of Mg powder in comparison to

Al and Cu. Copper has the highest density among these metals. Therefore, the magnesium powder occupies more volume in the mixture resulting into higher volume fraction γ than that of the copper and aluminium powders of the same mass.

These factors are responsible for low ϵ' values for mixture compositions having conducting centres of higher electrical conductivity in comparison to the mixture compositions with conducting centres of lower electrical conductivity under similar conditions as shown in Figure 1. This is in excellent agreement with the results obtained from three theoretical relations (3), (4) and (5).

It is evident from Figure 1 that the percentage deviations between experimental and calculated values of dielectric constant ϵ' increases with increasing the percentage concentration of metal powder in PVC for most of the mixture compositions. It can be observed that at lower percentage of metal impurity in the sample the percentage deviations between experimental and calculated values of ϵ' are very small. However, at higher percentages of metal impurity such deviations are quite large. It indicates that at lower percentages of conducting impurity the experimental values of ϵ' are in good agreement with the theoretical values of ϵ' obtained. The similar behaviour was reported by Naiki and Matsumura [7].

Higher deviations at higher percentage of metal impurity similar to those that of Naiki *et al* [7], can not be explained only on the basis of Maxwell-Wagner [4,10] absorption. The evaluation of ϵ' of a mixture by Maxwell-Wagner relation is largely controlled by the Sillars factor S . Because this factor relates ϵ'_1 and ϵ'_2 with the volume fraction γ of phase 2 material and any variation in this factor will show a sizable variation in the final evaluated values of ϵ' .

In our experimental results, we have found that if we replace S by S' in eq. (3) with $S' = S - \gamma$ given by eq. (7) then eq. (3) reduces to our empirically modified Maxwell-Wagner absorption relation (6). It is found that the evaluated values of ϵ' using our relation (6) exhibit a striking agreement with the experimental values of ϵ' . An inter comparison between Maxwell-Wagner absorption relation (3) and our empirically modified Maxwell-Wagner relation (6) is made for (PVC + Cu) system in Figure 4.

The replacement of S by S' may be explained that the theoretical expression (3) of Maxwell-Wagner does not include the deviation of grains from the spherical shapes as well as the effect of actual adjustment of actual grains of powder with each other and the metal grains.

Variation of ϵ' with percentage concentration of metal in (PVC + Cu) mixture compositions using all four theoretical relations (3), (4), (5) and (6) is presented in Figures 4. It is evident from the Figure 4 that the experimental values of ϵ' show a fair agreement with ϵ' values obtained by our empirically modified Maxwell-Wagner relation (6). Average deviations between experimental and calculated ϵ' values obtained

using Maxwell-Wagner relation (3), Bruggeman's relation (4) and modified Bruggeman relation (5) are 6.17%, 11.70% and 4.46% respectively. However, it is observed that the average deviation between experimental ϵ' values and those obtained from our empirically modified relation (6) is 1.45%. Which is much smaller than that of the average deviation obtained earlier using established relations (3), (4) and (5). Available literature value of experimental results and Figure 4 show that the deviation between experimental and calculated values of ϵ' increases tremendously at higher percentages of metal impurity. However, it is found that our experimental results are in excellent agreement with the results obtained using our empirically modified Maxwell-Wagner relation (6) even at higher percentages of metallic impurities.

Further Figure 2 indicates that the dielectric loss ϵ'' increases with the increase in metal impurity in PVC. As the electromagnetic energy passes through the medium, absorption of energy by conducting centres takes place. Presence of more conducting centres in the medium increases the loss of e.m. energy in the medium. The variation of ϵ'' with metal impurity for all three systems are compared in Figure 2 also. It is evident from this figure that at small percentages of metal impurity, the losses in the samples containing conducting centres of lower electrical conductivity are lower than that of samples containing conducting centres of higher electrical conductivity while at higher percentage of metal impurity, the losses in the samples containing conducting centres of lower electrical conductivity are higher than that of the samples containing conducting centres of higher electrical conductivity. This can be explained as :

The losses in the powdered dielectric medium containing conducting centres are controlled mainly by two factors :

- (1) Firstly, the absorption of energy by conducting centres in the dielectric medium termed as conduction losses.
- (2) Secondly, the dissipation of energy used in the dipolar motion.

At small percentages of metallic impurity in a dielectric medium, the losses due to dipolar motion are dominant over conducting losses. This is probably due to the heating of conducting centres because the amplitude of the electromagnetic field available to cause dipolar motion reduces with an increase in metallic impurity. The losses due to dipolar motion increases with the increase in metal powder in a dielectric sample. Hence, at lower concentrations of metallic impurity, the losses due to dipolar motion in mixture compositions with conducting centres of higher electrical conductivity are higher than that of the mixture compositions with conducting centres of lower electrical conductivity under similar conditions. On increasing the concentration of metallic impurity in a dielectric sample, the losses due to dipolar motion becomes equal to the conduction losses at certain percentages of metallic impurity. On further increasing the concentration of metallic impurity, dielectric losses due to conduction losses starts dominating over losses due to dipolar motion. Because of more and

more conducting centres available in the dielectric medium to face the same e.m. field resulting into an increase of heating effect in conducting centres in the medium and hence, increases the dissipation of energy in the dielectric sample having conducting centres of higher electrical conductivity. This is the reason that at higher concentration of metallic impurity, dielectric loss ϵ'' in PVC mixture compositions with copper are less than that of the losses in mixtures with aluminium or magnesium.

It is obvious from Figure 3 that the relaxation time (τ) values of pure PVC and its mixture compositions obtained during the present investigations are very much comparable to the values (10^{-13} sec) reported earlier in the literature for silicon [9,11], marble and soap stone samples [2]. An examination of Figures 2 and 3 reveals that the variations of relaxation time (τ) values with metallic concentrations in PVC powder follow almost the similar trend as followed by variations of dielectric losses ϵ'' with metallic impurity. This can be explained as :

The relaxation time (τ) is determined using the relation ($\tau = \epsilon''/\omega\epsilon'$) and the variations of ϵ' with metallic impurity in PVC are almost linear as shown in Figure 1. Therefore the relaxation time variations follow the same trend as followed by dielectric loss ϵ'' variations. This also indicates that the relaxation time increases as the dipolar hinderance increases due to increase in metallic impurity in the dielectric medium.

The a.c. conductivity ($\sigma = \omega\epsilon_0\epsilon''$), relaxation time ($\tau = \epsilon''/\omega\epsilon'$) and loss tangent ($\tan\delta = \epsilon''/\epsilon'$) variations with metallic impurity follow the similar trend as followed by ϵ'' variations which can be explained on the same lines mentioned above for (τ) variations.

Though, enough care has been taken during the course of measurements still accuracy of measurements is limited due to the possibility of higher order mode generations and power leakage through the slotted line section. Still it can be concluded from the present investigations that dielectric measurements of powdered samples with conducting impurities are interesting. Such results can be used for estimating the presence of metallic impurities and their nature in dielectric powders. Similar measurements can be employed to estimate the concentration of metals present in natural ores and rocks.

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