Dielectric Behavior and Functionality of PVC / Cellulose acetate Composite

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Abstract

Cellulose acetate powder which is extracted from the cigarette (insulator part as a filler) has been used with polymer to produce PVC Cement/cellulose acetate composite. The dielectric behavior of this composite is analyzed as a function of weight ratio of cellulose acetate powder (0.1, 0.2, 0.3, 0.4, and 0.5 wt%), temperature range (30 - 120) $^{\circ}$C and frequency (120Hz - 2MHz). Impedance and   AC conductivity ($\sigma_{ac.}$) of the composites behaviors as function of frequency and temperature have also studied. The results show that, the real permittivity, dielectric loss and loss tangent for all composites increase the with increasing of cellulose acetate filler content.

1. Introduction

Composites which are made of polymer with inorganic filler have been successfully used in electrical applications. These systems are considered heterogeneous and their electrical characteristics depend on many of factors such as size, shape, volume ratio, conductivity of the filler, the adhesion between the polymer-matrix and the filler, and the method of processing. The advantage of such composites is that it can be produced to exhibit enhanced and compatible properties that the constituent materials may not exhibit [1- 4].

Filler can improve the properties of

mechanical, thermal and electrical. It can lower the shrinking in addition to the price reduction consideration[1- 2]. In order to achieve both electrically insulating and thermally conducting polymer composites, fillers such as (barium titanium, boron nitride, silicon carbide, alumina, aluminum nitride and beryllium oxide), are used [5-8]. Metallic fillers, graphite and carbon black are used to enhance both thermal and electrical properties [9,10]. Non conductive fillers increase the dielectric permittivity due to interfacial polarization (Maxwell-Wagner-Sillars polarization). For con fillers, dielectric permittivity and electrical conductivity increase with increasing the filler ratio until drastic changes in these properties reach a critical range of filler concentration called percolation threshold. The effective use of composites strongly depends on the ability to disperse the fillers homogeneously throughout the material [11].PVC cement composite which consists of an PVC cement resin and conductive or nonconductive filler. It has been reported to possess interesting properties and it is used in a verity of applications such as encapsulating, thin film coating, packing of electronic circuits protective coatings, electromagnetic frequency interference shields, anti- static devices and thermistors [12-14]. The raw material for the manufacture of cigarette filters is cellulose acetate (obtained from wood).We can dispose and use of cigarette butts (waste) by mixing them polymer matrix to produce new composite polymer.

PVC cement is an adhesive-like substance used to join (glue) two pieces of PVC together. PVCs cement are widely used as suitable matrices, besides their other applications in modern technology, because they offer versatility, low shrinking, chemical resistance, relativity low dielectric constant and outstanding adhesion [7]. While referred to as cementing, it is more similar to welding, as it fuses the two pieces of PVC into one piece in much the same way a welding does. PVC cement based composite are popular insulators in voltage engineering, such as PVC cement/Carbon composite which is used for the dielectric application [8].The Predication of the dielectric of blend materials are very important in technological applications [9, 10]. Research in PVC cement based composite dielectric systems is gathering momentum for their preferred electrical properties [15,16].Since interesting properties of polymer attributable to complex motion within their molecular matrix, therefore, the study of dielectric properties as a function of frequency and temperature is one of the most convenient and sensitive methods of studying polymer

structure.

In this study, (PVC cement / cellulose acetate) composites were prepared and their dielectric properties were investigated as a function of frequency in the range (120Hz-2MHz) filler weight fraction (0.1, 0.2, 0.3, 0.4, and 0.5 wt%), and temperature in the range (30 - 120)˚C.

2. Experiment

2.1. Materials

A commercial LICHIDE PVC cement 717-21 heavy duty-clear, with Glass transition Temperature (Tg=75°), Dielectric Constant ε' =3.19, molecular weight Mw = 620 gm/mol and density = 1.42gm/cm^{3} supplied by SWAN TRADING (L.L.C) was used as polymer matrices for the composites.

2.2. Sample Preparation

In order to insure a excellent dispersion of the filler and to provide a homogenous composite, the cellulose acetate powders were added to the PVC cement (as received) in different weight ratio (0.1, 0.2, 0.3, 0.4, and 0.5 wt%) , It is suitably mixed at about 65˚C for 3 minutes. The mixture then was casted as a film on Al substrates. The initial treatment was carried out at room temperature for 24 hours, followed the samples were placed in a vacuum oven (post curing treatment) at 120˚C for 2 hours, and then cooled down to room temperature to eliminate thermal history Circular disk shaped thin film Aluminum electrodes (1 mm thick, 9 mm in diameter) were vacuum deposited on the upper side of the casted composites. The film samples were placed between two Al-coated electrodes which were finally made.

2.3. Characterization and Measurements

Dielectric behavior of the composite amples were measured by using a digital RCL bridge type Megger B131 and RCL bridge type (Metrapoint-RLC2 and ME 1634 Function Generator). The capacitance of the film samples and loss tangent were measured in the frequency range between 120Hz and 2MHz. The relative complex permittivity (ε^{*}) can be expressed as follows:

$$\varepsilon^{*} = \varepsilon' - i\varepsilon'' \qquad (1)$$

where, $i = \sqrt{-1}$. The real part (ε') and the imaginary part (ε'') of relative permittivity. Dielectric loss of permittivity can be calculated from the measured capacitance and loss tangent [17, 18];

$$\varepsilon' = C\, d\, /\varepsilon_O\, A \qquad (2)$$

$$\sigma_{a.c} = \varepsilon_O \omega\, \varepsilon'' \qquad (4)$$

$$\varepsilon'' = \varepsilon' \tan\delta \qquad (3)$$

where ω is the angular frequency. The complex impedance (Z^*) can be expressed as

where, d is a separation distance between two electrodes, A is electrodes area, ε_O is the permittivity of the free space, $(\varepsilon_O = 8.85 \times 10^{-12}$ F/m$)$

$$Z^* = Z + iZ' \qquad (5)$$

w parts of impedance, respectively. The real part of impedance Z at different frequencies up to 500 kHz was measured by impedance analyzer (HeweltpacardA4800).

a.c. conductivity $(\sigma_{a.c})$ was calculated according to the relation [19, 20].

follows

3. Results and Discussion

Figure (1) shows the variation real permittivity part (ε') of PVC cement composite as a function of frequency range between 120Hz and 2MHz for different cellulose acetate filler content in the room temperature. It show see that, there is slightly decrease in real permittivity as the frequency increases. High values real permittivity in the low frequency range, are due to the process of interfacial polarization effect that appears in hetero-phase systems (Maxwell Wagner Sillars process) and the polarization induced by segmental mobility in the polymer matrix which appears more effective at low frequency [19]. The dipoles responsible of these two polarizations have less time to orient themselves in the direction of the alternating field at high frequencies [21].

The effect of temperature on real part permittivity (ε') of cellulose acetate filled PVC cement composite versus temperature range between 30˚C and 120˚C for cellulose acetate filler content at fixed frequency is depicted in fig.(2). Here, since there is no significant change in the filler permittivity with increasing temperature due to, the disruption of contacts between filler particles caused by the differential thermal expansion of resin and filler; this mechanism should decrease real permittivity, as well as, the increased mobility of segments of polymer molecules which increases by increasing temperature; this mechanism should increase the real permittivity due to

greater freedom of movement of the dipole molecular chains within the polymer at high temperature [18].
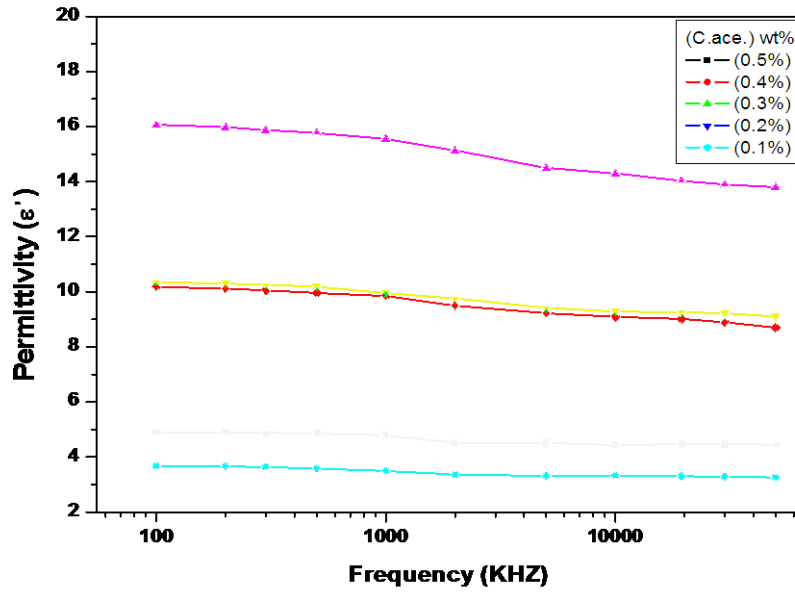
Fig. (1) Permittivity as a function of frequency of PVC Cement/cellulose acetate composite .
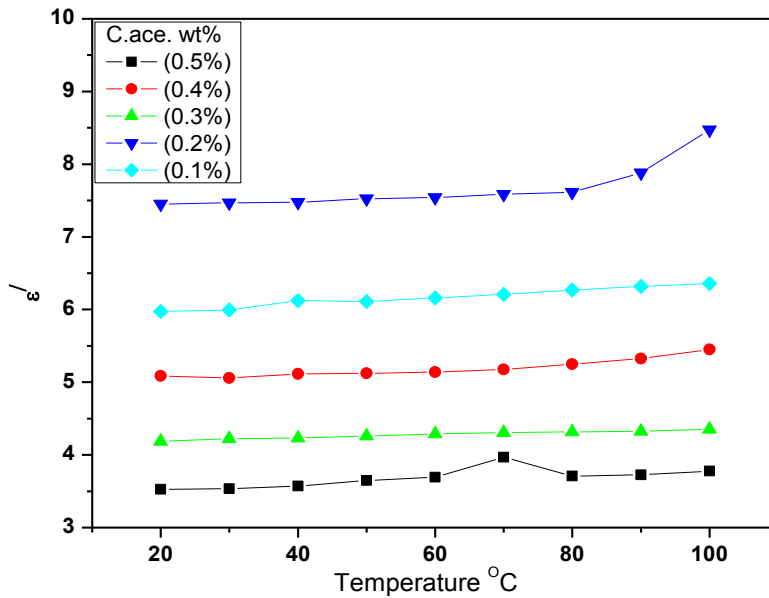
Fig. (2) Permittivity as a function of temperature of PVC Cement/cellulose acetate composite.

Figure (3) shows the variation of dielectric loss ε'' as a function of frequency in the range (120Hz - 2MHz) at room temperatures with different filler cellulose acetate content. This kind of frequency dependency is called dielectric relaxation, being characterized by a relaxation time τ or relaxation frequency f_o corresponding to $\Delta\varepsilon/2$. This relaxations shift to lower frequencies with the increasing of cellulose acetate filler content. It can be observed that, the values of dielectric loss increases gradually with increasing the frequency to reach the highest value (maximum value). The increases in dielectric loss may be related to a.c conductivity (σ_{ac}) which depends on the number of charge carriers [21,22].

Figure (4) shows the variation of dielectric loss ε'' as a function of temperature in the range 30°C - 120°C at a constant frequency (1 kHz) with different cellulose acetate filler content. It is observed that ε'' is increased in general as the filler content or temperature increases. The increase in ε'' by increasing of filler contents is related to the interfacial polarization, while that caused by increasing temperature may be related to the increase of segmental mobility. Since the rise in temperature (and the consequence drop in viscosity) exerts an effect on the amount of the losses due to the friction of the rotating dipoles, the degree of dipole orientation increases and ionic conduction increases, due to the thermal

dissociation of molecule [18, 23, 24]. It can be seen that the a.c. conductivity increases by increasing filler content.

Figure (5) Shows the impedance (Z) real component frequency dependence PVC cement/cellulose acetate composites for cellulose acetate filler content. Impedance values decrease by increasing frequency. The observed decrease in impedance with cellulose acetate content is due to protonic migration transporting the impurities existing in the (cellulose acetate) filler. This motion leads to higher electrical conduction in the filled composites. As can be seen, there is an exponential decrease in the impedance with the increase in frequency for all filler cellulose acetate filler content, and this decrease is greater for high filler contents composites.

Figure (6) Shows the impedance Z (real component) temperature for PVC cement/cellulose acetate composites (different wt%). There is an obvious decrease in impedance by the increasing filler content due to the increased interfacial polarization, and with the rising in temperature. The effect of adding cellulose acetate filler to the PVC cement on σ_{ac} conductivity as a function of temperature is shown in Fig. (7).

It can be seen that the increase in σ_{ac} conductivity with cellulose acetate content and temperature is due to contributed 1st, by the charge carriers weakly bound ions in PVC cement resin, and polymer segmental chains,

that would increase dramatically with increasing temperature [26]. 2nd by considering that the conduction mechanisms are usually enhanced by thermal fluctuation of local field [25].
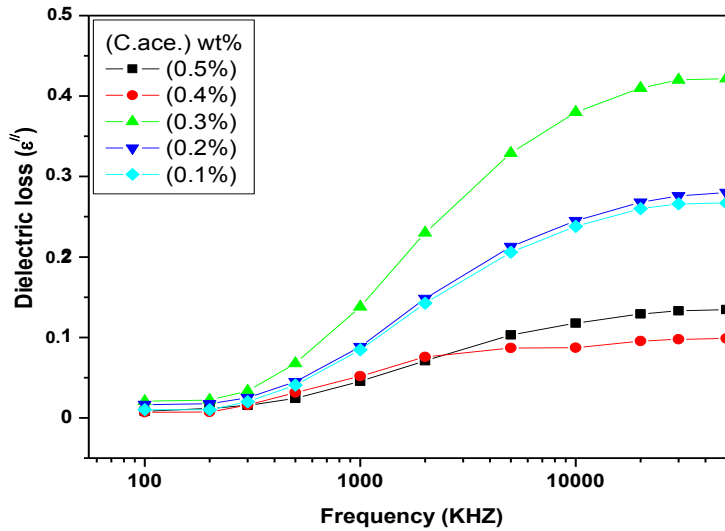
Fig. (3) PVC Cement/cellulose acetate composite dielectric loss as a function of frequency.
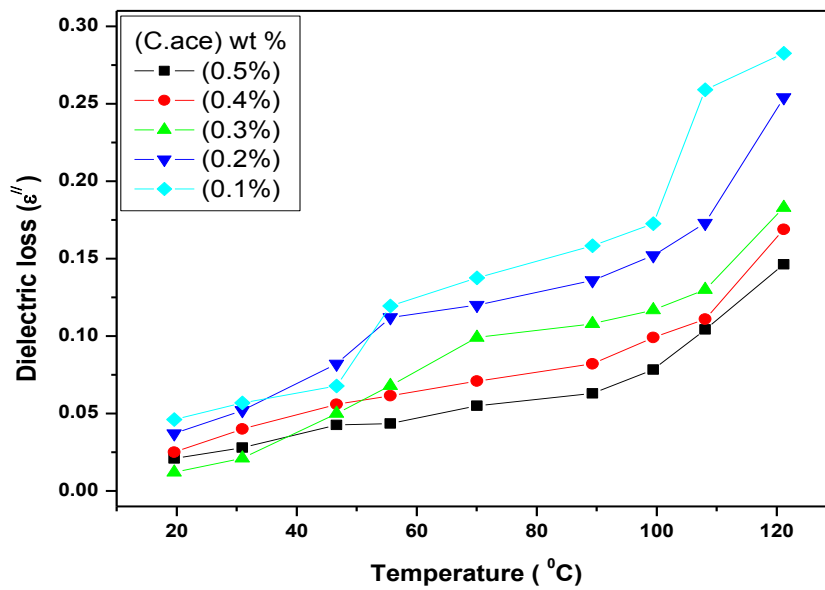
Fig. (4) PVC Cement/cellulose acetate composite dielectric loss as a function of temperature.
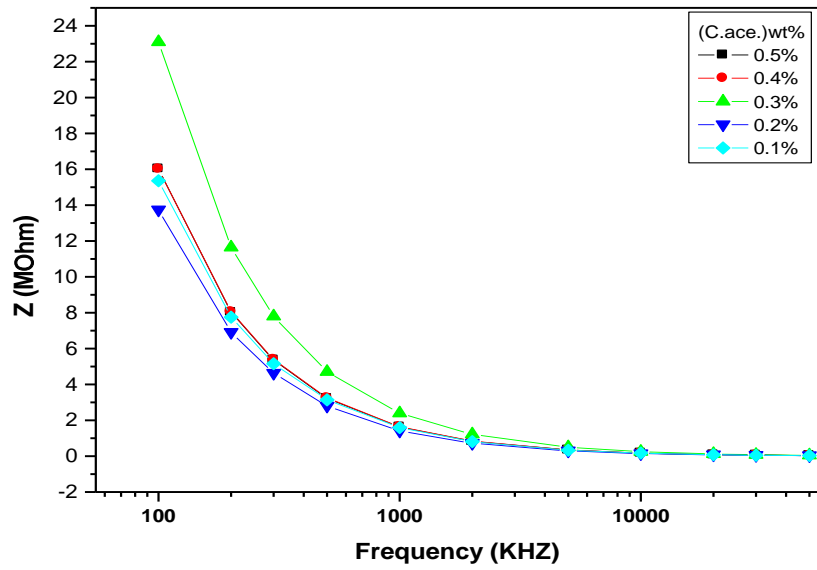
Fig. (5) PVC Cement/cellulose acetate composite impedance as a function of frequency.
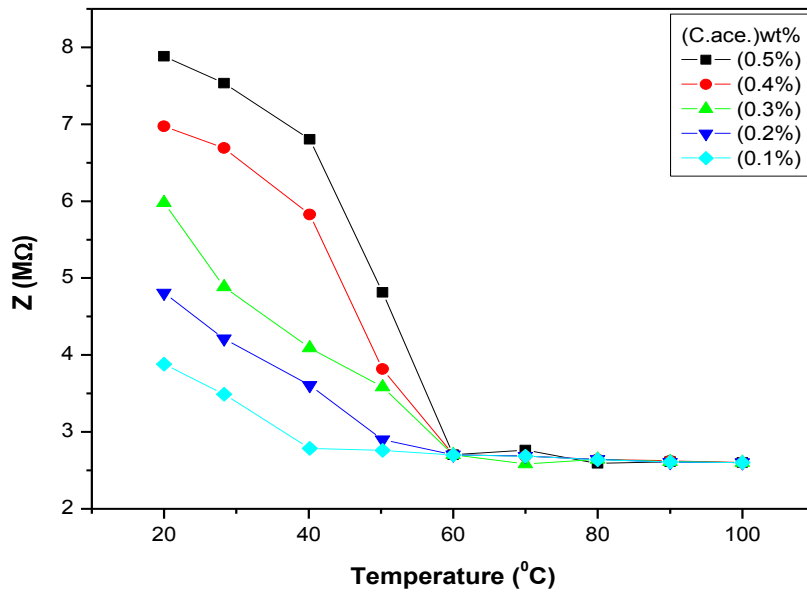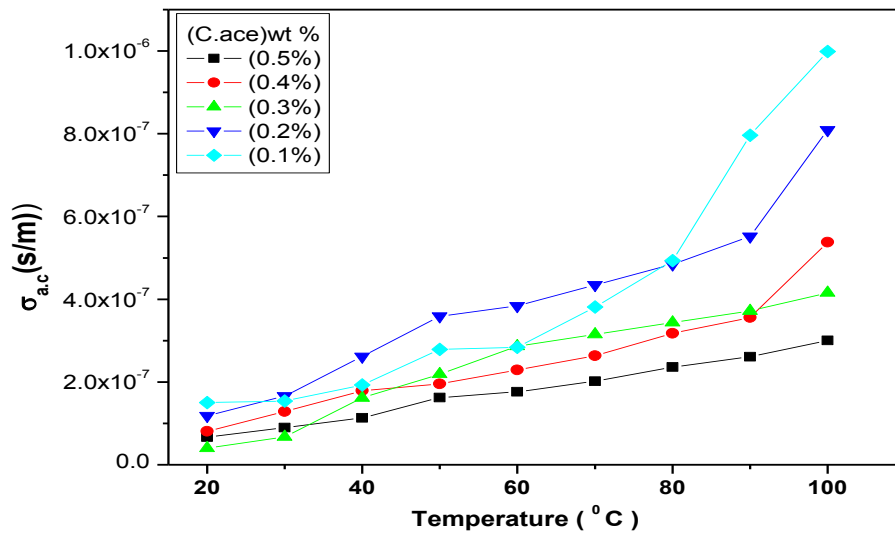

Fig. (6) PVC Cement/cellulose acetate composite impedance as a function of temperature.

Fig. (7) PVC Cement/cellulose acetate composite impedance as a function of temperature.

The dependence of the calculated a.c conductivity on frequency range (120Hz - 2MHz) for different weight ratio of cellulose acetate filler at room temperature is shown in figure (8) It increased when the filler content increased for all frequency range studied. This increase in σ_{ac} conductivity is expected, since at higher applied field frequency more ions and impurities are moved. The observed enhancement in the σ_{ac} conductivity is attributed to ionic interactions and impurity motion taken in the bulk of electrolyte polymer composite [26]. The relation between the dielectric loss ε'' as a function of the dielectric permittivity ε' for (PVC cement/ cellulose acetate) Composite at room temperature is shown in figure (9). From the Cole-Cole plot, the generalized relaxation time, the parameter relating to the distribution of relaxation times is determined.
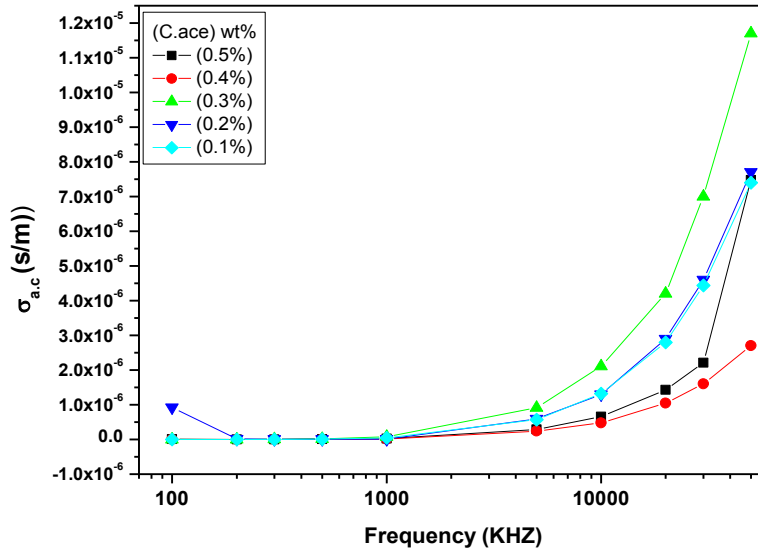
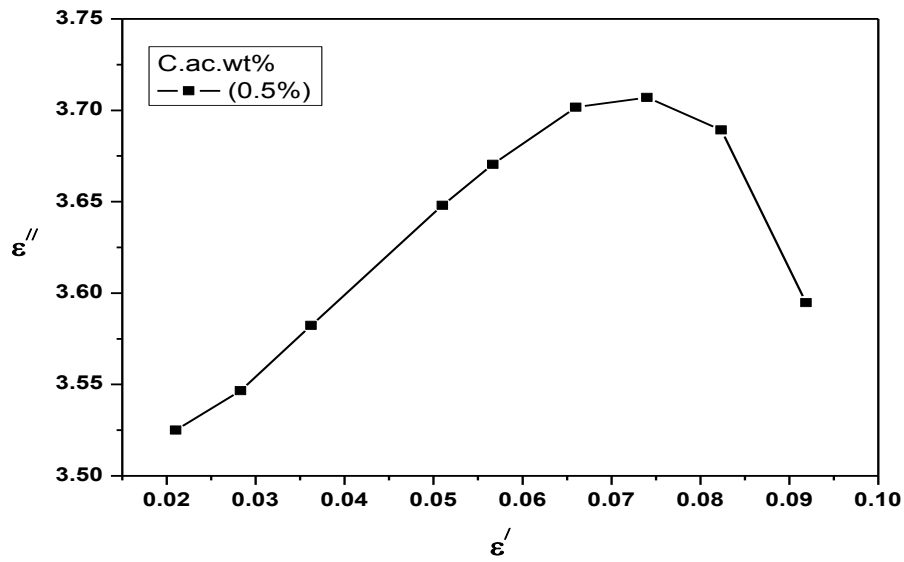**Fig. (8)** PVC Cement/cellulose acetate composite impedance as a function of frequency.

Fig. (9) The Cole-Cole plot for PVC Cement/cellulose acetate composite.

4. Conclusion

It is found that the permittivity, dielectric loss for all composites increase with increasing the cellulose acetate filler content, or temperature which has been attributed to interfacial polarization and segmental mobility of the polymer molecules, respectively. The permittivity decreases with the increasing of frequency because interfacial and segmental mobility polarizations cannot keep up orientation in the direction of the alternating field. The impedance of the composite decreases with the increase of filler content, frequency and temperature.

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الخلاصة:

تم في هذا البحث استخلاص مسحوق خلات السليلوز وخلطة بوليمر بولي كلوريدالفينيل لتكوين متراكب جديد. درست الخواص الوزنية العازلية كدالة للنسب (0.1, 0.2, 0.3,0.4,0.5) بدرجة حرارة ($30\text{-}120\ ^{O}C$) والتردد ($120\text{-}2MHz$) . تم دراسة الممانعة والتوصيلية الكهربائية المتناوبة للمتراكبات المحضرة كدالة للتردد ودرجة الحرارة . أظهرت النتائج زيادة في السماحية الحقيقية وخسائر العزل وعامل الفقد لكل النسب المحضرة وكدالة لمحتوى الحشوة .