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The Conductivity of AC, Loss Tangent, and Relative Permittivity for Composites of PVC Paste/Graphite Electrode Waste

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Abstract

The behavior of AC conductivity (σ_{ac}), loss tangent (tan δ), and relative permittivity (ε′) for composites of PVC-P/graphite electrode waste (GEW) was investigated, and a qualitative explanation was provided as a function of PVC-P weight fractions (0, 5, 10, 15, 20, and 25) wt. percent, temperature (30-90) °C, and frequency (100Hz-2MHz). The behaviors of the composites' ac. conductivity and impedance as a frequency function and temperature have been examined. The permittivity was shown to rise with increasing temperature (Tg). The relative permittivity increased as the GEW filler concentration increased and was highest in the low-frequency range; nevertheless decreased as the frequency increased.

Keywords: Polyvinyl chloride paste, AC conductivity, graphite electrode waste, permittivity, composite.

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1- Introduction

Polymer and organic filler composites have been effective in the electrical and electronic industries. The electrical properties of these heterogeneous systems are influenced by the size, volume fraction, conductivity of the filler, shape, the filler's adherence to the polymer, and the processing step.

Composites like this have the advantage of being able to be built to have improved and compatible qualities that separate materials might not have [1-4].

Fillers, for instance, diamond, silicon carbide, alumina, boron nitride, aluminum nitride, beryllium oxide, and fused SiO_2, are utilized to make both electrically insulating and thermally conducting composites of polymer-matrix [5-8]. To increase electrical and thermal properties, carbon black, metallic fillers, and graphite are utilized [8-10].

The dielectric permittivity of non-conductive fillers is increased by polarization between the two surfaces (Maxwell-Wagner-Sillars polarization).

Due to interfacial polarization, nonconductive fillers increase dielectric permittivity (Maxwell-Wagner-Sillars polarization). To properly use composites, it is necessary to be able to evenly distribute fillers throughout the material [11].

Exfoliating graphite intercalation compounds results in expanded graphite with a high aspect ratio and decent electrical conductivity by rapidly heating them in a microwave or furnace.

Graphite electrode waste from electric arc furnaces can be used in place of manufactured graphite to reinforce carbon composites.

The industry has made extensive use of antistatic, electromagnetic shielding, and electronic technology materials [12, 13]. Composite materials have been employed in electronic technology to create a device with superior energy storage systems and electromagnetic [14] or energy storage technologies based on electrochemistry [15].

In this work, the dielectric characteristics of (PVC-P as a matrix and GEW powder as a filler) the properties of composites were investigated as a function of the filler weight percentage (5, 10, 15, 20, and 25 wt. percent), temperature (30-120 °C) and frequency (100 Hz-2 MHz). Impedance and AC conductivity were also investigated.

2- Experiment Method and Materials

2.1. Materials

As polymer matrix for the composites, Commercial (LICHIDE) PVC paste 717-21 heavy duty-clear was supplied by SWAN TRADING-CHINA (L.L.C.) (as received from the market), with permittivity (ε'= 3.2), volume resistivity ~ 5.5×10^{15} ohm/cm, and density=1.43 gm/cm^3 [15].

The iron and steel factory's GEW powder, with a particle size of ~ 45nm, is utilized as an additive.

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2.2. Preparation of Samples

The GEW powders were incorporated into the PVC-P in varying % of weight (5, 10, 15, 20, and 25 wt. percent) and adequately mixed at approximately 65°C for 7 minutes to provide decent filler dispersion and a homogeneous composite. Then, the mixture was decanted over clean Al substrates as a thick layer. The primary curing occurred 24 hours at room temperature, after that a 2-hour post-cure at 120°C. On the casted composites' upper surface, thin-film aluminum electrodes in the shape of a circular disk with a diameter of 6 mm were vacuum deposited and 10^{-6} torr. Finally, an Al/PVC-P: GEW/Al sandwich was investigated as depicted in Fig. **1**

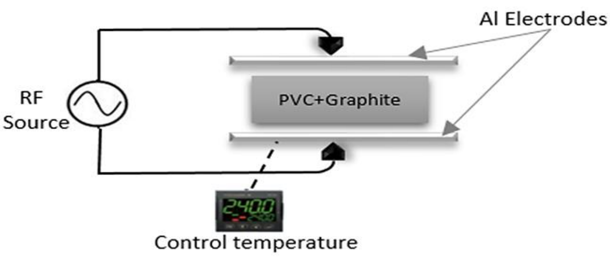

Fig. 1. Schematic diagram of the fabrication dielectric device

2.3. Characterization and Measurement

At 120Hz and 1 kHz, the sample capacitance and loss tangent (tan δ) of composites were determined using a Fluke PM6303A-Kelvin.

3- Results and Discussion

Dielectric relaxation strength, energy loss, and electric permittivity are essential factors in material science because to the fact that they are utilized to determine whether a material is suitable for a certain application.

This experiment was aimed to investigate the PVC-P/GEW composite's behavior and dielectric characteristics. The frequency dependence of dielectric characteristics is shown in Fig. **2**, at room temperature, the fluctuation of tan of GEW filled PVC-P composites is a function of the range's frequency (120 Hz - 2 MHz).

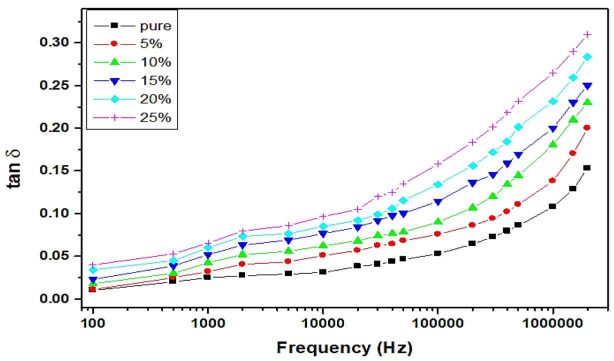

Fig. 2. The loss tangent of the composite PVC-P/GEW as a function of frequency

For all frequency ranges studied, tan δ rises as filler content increases. The increase in tan δ with increased filler content could be due to induced polarization caused by the applied electric field at the polar ends of the particle. The charge carriers are then transported efficiently around the interface and converted the conducting [16].

This will result in a permittivity greater than that of the particle. The presence of a relaxation peak at roughly 100 kHz, indicating changes in the polymer molecular structure, is intriguing.

The relaxing of the polymer happens when GEW interrupts the original crystallization process of PVC-P, resulting in more free volume and more flaws due to the poor interface [17].

The fluctuation of tan δ as a function of temperature in the range 30-90 C for pure PVC-P and PVC-P/GEW composites at a constant frequency (1kHz) is illustrated in Fig. **3** (with different filler concentrations).

Tan δ is known to rise in general, when the filler amount or temperature rises. Increased filler content causes an increase in tan, which is connected to interfacial polarization, whereas increased temperature causes an increase in segmental mobility and ionic conductivity.

Due to the molecule's thermal dissociation, the degree of dipole orientation increases and ionic conduction increases, since temperature (and thus viscosity) affect the number of losses due to dipole friction [17-21].

The following formula was utilized to quantify the AC electrical characteristics of a composite material sample. $\varepsilon^*(\omega)$ is the frequency dependence complex dielectric permittivity [19-21].

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

Where: $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$ are the real and imaginary components of complex dielectric permittivity.

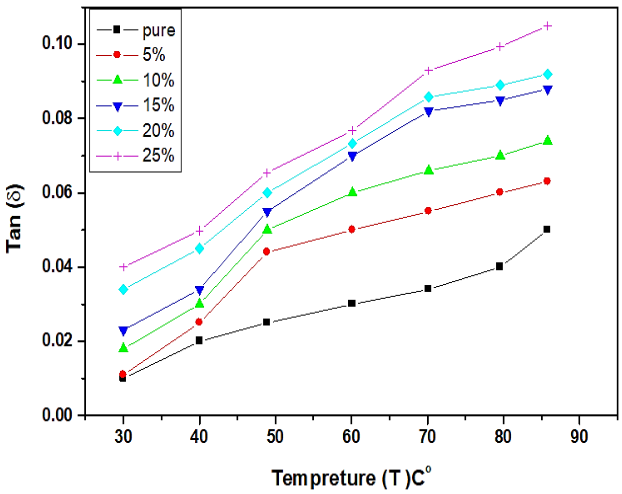

Fig. 3. The loss tangent of the PVC-P/GEW composite as a function of temperature-dependent

Fig. **4** shows PVC-P/GEW at room temperature, as a function of frequency. Electrical conductivity (σ) is inversely proportional to dielectric constant variation with frequency, as expected.

The ε' has a value that is large at low frequencies and diminishes exponentially as the frequency increases. The interfacial polarization process and the polarization caused by segmental mobility in the polymer, which is more effective at low frequency and high temperature, respectively, are attributed to high values ε' in the low-frequency range [18-22].

At high frequencies, the dipoles that cause these two polarizations have less time to align themselves with the alternating field's direction.

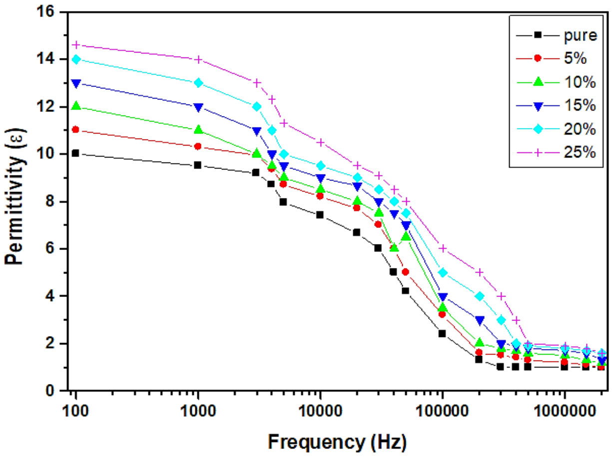

Fig. 4. Frequency-dependence of the dielectric permittivity of PVC-P/GEW composite

Fig. **5** illustrates the dielectric permittivity (ε') of the PVC-P/GEW composite for varied filler concentrations as a function of temperature in the range of 30-90°C. For comparison, pure PVC-P was presented in the illustration.

It is demonstrable that ε' grows with rising temperature in all circumstances, up to a point where a further rise in temperature causes the ε' value to decrease.

Because the filler permittivity does not change significantly with temperature, the composites' dielectric response may be attributed to two factors: first, the polymer's segmental mobility, which rises with temperature; this mechanism should enhance the dielectric constant at high temperatures due to the increased freedom of movement of the dipole molecular chains inside the polymer [23], and second, the dielectric response of the composites.

Second, the heat expansion of the polymer and filer causes the breakage of connections between filler particles; this mechanism should lower the dielectric constant [23-26].

The structure's orientation within the material also affects dielectric characteristics. From the measured loss tangent and capacitance, permittivity dielectric loss can be calculated as follows [20-23]:

$$V_\theta = V_T \sin(\pi - \varphi) \quad (2)$$

$$\tan\delta = (V_T^2 - V_\theta^2)^{1/2} / V_\theta \quad (3)$$

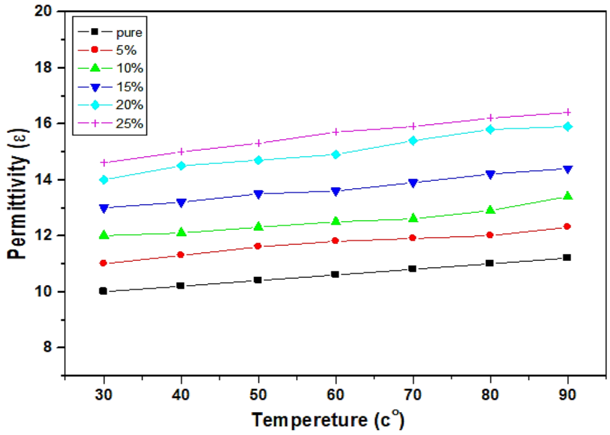

Fig. 5. The real dielectric permittivity of the PVC-P/GEW composite as a function of temperature-dependent

Fig. **6** shows the fluctuation of AC conductivity (σ_{ac}) at room temperature with a frequency from 120Hz to 2MHz for pure PVC-P and PVC-P/GEW of composites. At room temperature, it is clear that σ_{ac} increases as the frequency increase. This is typical behavior for dielectric material, which may be attributed to the insulating phase of (PVC-P), which forms the boundaries between the conductive particles of GEW filler. These boundaries are more active to prevent hopping and tunneling conduction between the insulated conductive particles. Temperature dependence of AC conductivity (σ_{ac}) in the range (30 – 90 °C) at constant electric field frequency at 1kHz of the pure PVC-P and PVC-P/GEW of composites shown in Fig. **7**.

It was noticed that increase in AC conductivity with temperature is due to contributed by free charges available in the composite system. It was also found that, at a higher temperature, high mobility of free charges make them more frequency independent conductivity[18,19,26].

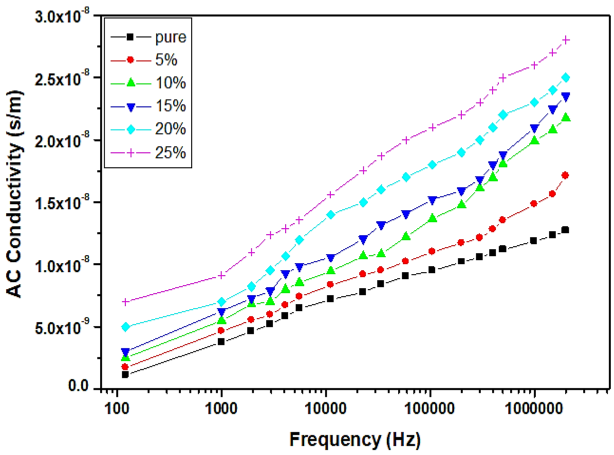

Fig. 6. AC conductivity of PVC-P/GEW composite as a function of frequency-dependent

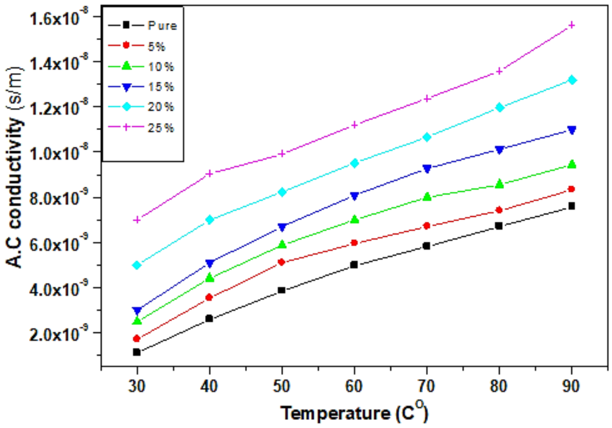

Fig. 7. AC conductivity of the PVC-P/GEW composite as a function of temperature-dependent

The AC conductivity (a.c) was computed using the following formula:

$$\varepsilon'(\omega) = C\, d / \varepsilon_o A \qquad (4)$$

$$\varepsilon''(\omega) = \varepsilon' \tan\delta \qquad (5)$$

Where: d is the distance between two electrodes when they are separated, *A* is the electrode's area, ε_o is the free space's permittivity, (ε_o=8.85×10^{-12} F/m).

$$\sigma_{ac} = \varepsilon_o\, \omega\, \varepsilon'' \qquad (6)$$

Where: $\omega = 2\pi\upsilon$ is the angular frequency.
The following formula uses to calculate the complex impedance (Z^*)

$$Z^* = Z + iZ \qquad (7)$$

Where: Z is the real and iZ is the imaginary impedance portions, correspondingly. The fluctuation of impedance Z with frequency for PVC-P: GEW composites is shown in Fig. **8**.

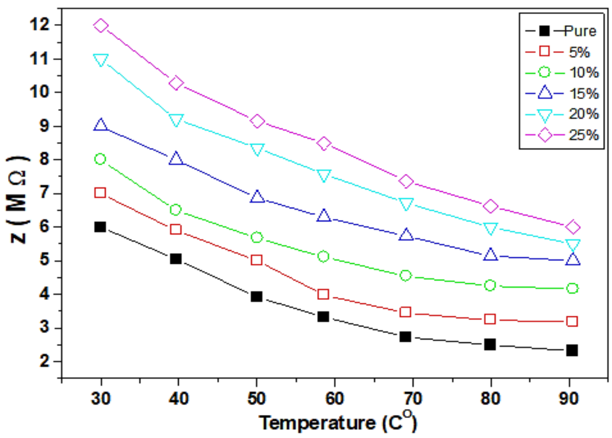

Fig. 8. Temperature dependence of the frequency for PVC-P/GEW composite

The phase angle was always negative, indicating that the composites were capacitive and could be modeled as a series of parallel RC networks (lumped circuits). With the rising frequency and GEW concentration, impedance values drop. The process of protonic migration and contaminants contained in GEW filler is responsible for the observed decrease in impedance as GEW concentration increases. This movement results in increased electrical conductivity in filled composites [21]. The impedance decreases exponentially with increasing frequency for all filler volume fractions, but the decline is larger for composites with a high filler content [22]. The temperature dependence of impedance Z of PVC-P: GEW composites are shown in Fig. **9**.

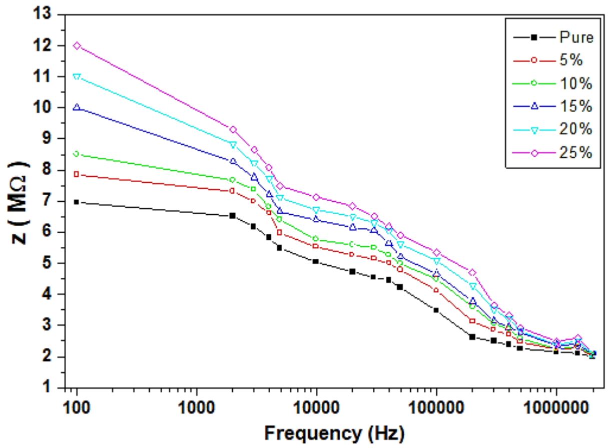

Fig. 9. PVC-P/GEW composite impedance frequency dependency

As the filler volume fraction increases due to greater interfacial polarization and as the temperature rises, there is a noticeable drop in Z. For all situations in which filler was added, the impedance Z reduces as the temperature rises, with clear dips near (90^{o}C) temperature. As the temperature increased, the mobility of segmental molecules increased as well. could explain the drop in Z. The dips could be related to the temperature zone of the glass transition where segmental mobility is higher. As a result, these dips may refer to the glass transition temperatures of all composite materials.

4- Conclusions

Electric permittivity, dielectric loss, and loss tangent all raise as the Graphite electrode waste filler concentration or temperature is increased, which has been attributed to interfacial polarization and segmental mobility of the polymer molecules, respectively.
Electric permittivity decreases with increasing frequency due to the difficulty of interfacial and segmental mobility polarizations to retain orientation in the alternating field's direction.
The composite's impedance Z decreases as the frequency, volume of filler, and temperature increase.

Acknowledgements

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دراسة التوصيلية الكهربائية المتناوبة وزاوية الفقد والسماحية الكهربائية لمتراكبات البولي فينيل كلورايد اللاصق / مخلفات اقطاب الكرافيت

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الخلاصة

تم فحص سلوك التوصيلية المتناوبة (**AC**) ، وزاوية الفقد (**tan δ**) ، والسماحية النسبية (**ε′**) لمتراكبات بوليمر **PVC–P** / مخلفات اقطاب الجرافيت (**GEW**) ، وتم تقديم تفسير نوعي كدالة لـ (**PVC–P**) بحسب النسب الوزنية (0, 5, 10, 15, 20 و25) ودرجة الحرارة (30–90) درجة مئوية والتردد (100 هرتز –2 ميجاهرتز).تم فحص سلوك المركبات 'AC التوصيلية المتناوبة والمقاومة كدالة للتردد ودرجة الحرارة. لوحظ ازدياد السماحية مع زيادة درجة الحرارة (**Tg**). زادت السماحية النسبية مع زيادة تركيز حشو **GEW** وكان أعلى في نطاق التردد المنخفض ؛ ومع ذلك انخفض مع زيادة التردد.

الكلمات الدالة: معجون بولي فينيل كلوريد ، موصلية AC ، بقايا قطب الجرافيت ، السماحية ، المركب