Research Article

Inspection of the Spark Plug Effect NGK Nanoparticle on the Dielectric Properties of Filled Polyvinyl Chloride Paste Resin

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Abstract

In this study, the investigation of the use of polyvinyl chloride-paste (PVC-P) composites which contained spark plug aluminum silicate nanoparticles (NGK-NPs) for different insulation applications was carried out. The spark plug NGK-NPs acted as a matrix for the aluminum silicate nanoparticle powder. We analyzed the dielectric properties of the PVC-P / NGK-NPs composite material for the function of its filler content, temperature and frequency. We also studied the AC conductivity and impedance of the composite. Results indicated that the dielectric loss, dielectric constant and the loss tangent of the composites increased with an increase in the NGK-NPs filler content.

Keywords: Dielectric properties; Polyvinyl; NGK-NPs; Nanoparticle filler

Introduction

Polymeric composites containing inorganic fillers were used in many electronic and electrical applications. These systems were characterized to be heterogeneous and their different electrical characteristics were dependent on factors like size, shape, volume fraction, the conductivity of the filler, processing technique and the adhesion between filler and polymer materials. The main advantage of using these composites was that they exhibited compatible and better properties which were could not be displayed by the constituent materials [1-4].

Fillers tend to improve the thermal, mechanical and electrical (i.e., permittivity and conductivity) properties of the composites. They decrease the shrinking and are cost-effective. Furthermore, for developing polymer-matrix composites that displayed better electrical insulation and thermal conductivity, the researchers used fillers (such as boron nitride, diamond, aluminum nitride, alumina, fused SiO_2, silicon carbide and beryllium oxide) [5-8]. Some studies used metallic fillers, graphite and carbon black for enhancing the thermal and electrical properties of the fillers [8-10]. Nonconductive fillers were noted to improve the dielectric permittivity owing to interfacial polarization (i.e., Maxwell-Wagner-Sillars polarization). The dielectric permittivity and electrical conductivity of the composite fillers increased with an increase in the filler volume fractions. This continued

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until any drastic changes in the properties reached a critical filler concentration known as a percolation threshold. The use of the composites was dependent on their capability to disperse the filler molecules homogeneously uniformly in the material [11]. The PVC-P composites contain the poly vinyl chloride-paste resin along with a conductive/non-conductive filler. These composites display very interesting properties and are used for several applications like thin-film coating, encapsulating, electromagnetic frequency interference shields, packaging of electronic circuits, antistatic devices, protective coatings and thermistors [12-14].

In the past few years, the research in the PVC-P resin-based composite dielectric systems has garnered a lot of interest owing to their interesting electrical properties [15, 16]. These properties have been attributed to the complex motion of the molecules in the molecular matrix. Hence, investigating the dielectric loss, A.C. conductivity and dielectric constant, as the function of frequency and temperature was regarded as a sensitive technique for understanding the polymer structure. Here, we used PVC-P resin as the matrix and used the alumina silicate NGK-NPs (SiO_2 and Al_2O_3) powder as the filler composite. We investigated the dielectric properties of these composites as the function of the filler weight function, frequency and temperature. We also studied the impedance and A.C. conductivity.

Experimental
Materials and methods

In this study, we selected a commercial PVC-P resin (717-heavy duty-clear) due to its effective dielectric properties ($\varepsilon' = 3.19$, $\tan\theta = 0.002$), dielectric strength 544 volts/mil, glass transition temperature (T_g) = 75 °C, and density = 1.42 gm/cm^3. This resin was purchased from Swan Trading Company (L.L.C). We also acquired the solvent-borne PVC-P resin-based cement which was best suited for PVC-P pipe joining. This cement could be used for developing natural gas pipes, portable water irrigation, conduit, drain, etc. We also purchased the alumina silicate nanoparticle (NGK-NPs) ceramic powder (grain size of ≈ 80 nm; density of 5.8 gm/cm^3) from the local market and used it as the filler.

Sample preparation

To ensure that the filler was uniformly dispersed,

and a homogeneous composite was obtained, different concentrations of the NGK-NPs powder (5, 10, 15, 20 and 25% wt.%) to the PVC-P resin was added. This mixture was blended with the help of using the Petri dish mixer for 10 min at 70 °C. This mixture was cast as a thick film on the clean Al substrates. Thereafter, they carried out the initial curing for 24 h at room temperature, followed by the post-curing for 2 h at 120 °C. Then, circular, thick disk-shaped film aluminum electrodes (diameter of 6 mm and 2.5 cm sheet) were vacuum deposited on the upper regions of the casted composites. Finally, a sandwich containing Al/ thermosetting sheets made of composite/Al was developed. These films were placed between 2 stainless steel gold-coated electrodes. The measurements of the dielectric constant and the tangent loss were conducted in a frequency range of 120 Hz and 1 kHz at 25 °C.

Characterization and measurements

The measurement of the dielectric values of the above-mentioned composite samples was carried out using the Hewlett Packard 4284 A-Impedance Analyzer. All film samples (20 mm diameter and 1 mm thickness) were rested between the steel electrodes. Thereafter, the loss tangent and permittivity values were measured for the frequency values ranging between 100 and 106 Hz (fixed temperature of 25 °C) and temperature values ranging between 30 and 120 °C (fixed frequency of 1 kHz). The following formula was used for calculating the relative complex permittivity (ε^*):

$$\varepsilon^* = \varepsilon' - i\varepsilon''. \tag{1}$$

Here, $I = \sqrt{-1}$. ε' and ε'' denoted the real and the imaginary components of the relative permittivity, respectively. The dielectric loss for the permittivity was calculated using the values of the loss tangent and Capacitance [17, 18]. The actual correlation between the Capacitance, C and real permittivity, (ε'), values of a ferrite sample was estimated as follows:

$$C = \varepsilon_o \varepsilon^* A/d, \tag{2}$$

and

$$\varepsilon'' = \varepsilon' \tan \delta. \tag{3}$$

Wherein, d = separation distance between the electrodes; A = area of the electrodes, and ε_o is the permittivity of free space that equals to 8.85×10^{-12} F/m. The A.C. conductivity ($\sigma_{a.c}$) was estimated using the following formula [19, 20]:

$$\sigma_{a.c} = \varepsilon_o \omega \varepsilon''. \tag{4}$$

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Wherein; ω = angular frequency. The following formula was used for determining the complex impedance (Z^*) value:

$$Z^* = Z + iZ'. \tag{5}$$

Z and Z′ were the real and imaginary components of impedance, respectively. The real impedance values, Z, at differing frequencies (up to 500 kHz) were measured using the impedance analyzer (Hewlett Packard A4800).

Results and Discussion

Fig. 1 shows the dielectric permittivity (ε′) for the PVC-P/NGK-NPs composites as the function of the NGK-NPs powder for 2 fixed frequency values, i.e., 120 Hz and 1 kHz at room temperature. It was noted that the addition of NGK-NPs filler to the polymers increased the value of permittivity of the composite, which increased with an increase in the NGK-NPs filler content. This direct increase in the permittivity value was based on an increase in the filler content, and was related to the interfacial polarization or the Maxwell-Wagner Sillars (MWS) value as noted to have occurred between the PVC-P matrix and NGK-NPs particles [15-17].
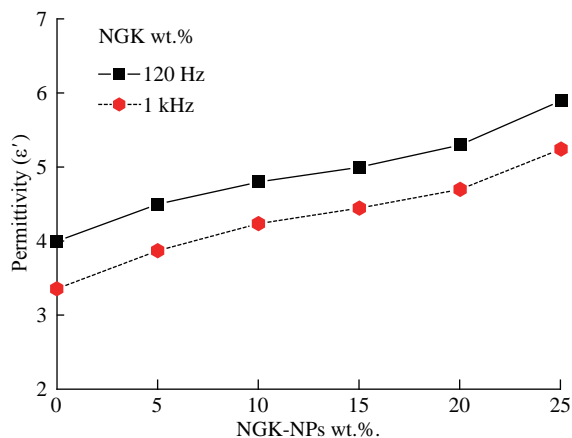

Fig. 1 The permittivity as a function of filler content for 2 different frequencies.

Fig. 2 shows the temperature dependence of the dielectric permittivity (ε′) value for different temperature values ranging between 30 and 110 °C at a fixed electric field frequency value of 1 kHz for the PVC-P/NGK-NPs composites. The results indicated that the permittivity increased with an increasing temperature value.

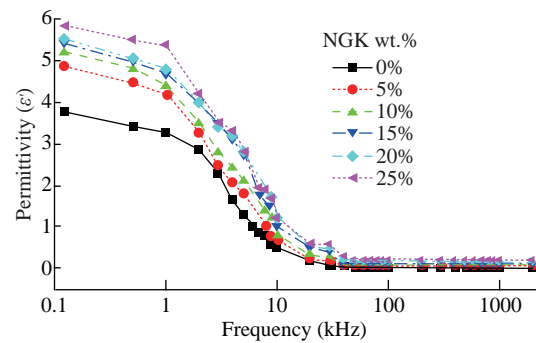This was attributed to the increase in the segmental

Fig. 2 The variation of dielectric permittivity of PVC-P/NGK-NPs composites with temperature and filler content.

mobility of the dipoles within the matrix (the dipole molecular chains of the polymers had greater freedom of movement at a higher temperature) and the changes occurring in the dielectric response of filler particles as a function of temperature [18]. As depicted in Fig. 3, the dependence of the dielectric permittivity (ε′) values of the PVC-P/NGK-NPs composites on the different frequency values, ranging between 120 Hz and 2 MHz for varying weight fractions of NGK-NPs at room temperature. The dielectric permittivity value decreased with an increase in the frequency values for all filler contents. Furthermore, it was seen that a reduction in the dipolar polarization of this matrix and an accumulation of the charges at the interface of the filler particles and the polymer led to a large-scale field distortion [19]. This change decreased at frequency values above 100 kHz.
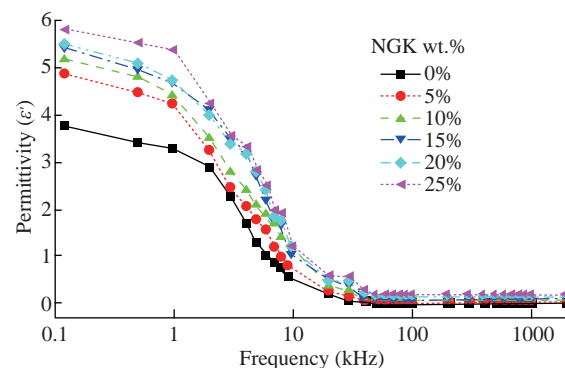

Fig. 3 The variation of dielectric permittivity of PVC-P/NGK-NPs composites with frequency and filler content.

In Fig. 4, the changing dielectric loss factor (ε″) was noted for the PVC-P/NGK-NPs composites, as the function of the filler weight fractions and temperatures, at the constant frequency of 1 kHz that was applied to the field. It was seen that the dielectric loss factor increased with increasing temperature and filler content. The increasing dielectric loss that was
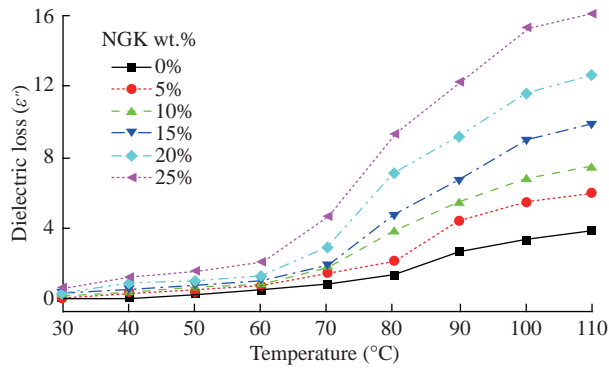
Fig. 4 The variation of dielectric loss factor of PVC-P/NGK-NPs composites with temperature and filler content.

noted with increasing temperatures was related to the tendency of the dipole molecules to rotate in the electric field and overcome the internal frictional forces between the molecular frictions or intermolecular interactions, which led to an expenditure of some electrical energy or its conversion to thermal energy [20].

The variations occurred in the dielectric loss values (ε'') at different frequency values ranging between 120 Hz and 2 MHz at room temperature for different samples (Fig. 5). This frequency dependency is known as dielectric relaxation as it is characterized by relaxation time, τ, or the relaxation frequency, f_o, which corresponds to $\Delta\varepsilon/2$. This relaxation shifts to the lower frequencies when the NGK-NPs content increases. A gradual increase in the dielectric loss value was noted when the frequency value increased until it reached a maximal value. This increase in the dielectric loss was related to the A.C. conductivity ($\sigma_{a.c}$) that was dependent on the number of charge carriers, frequency domain and relaxation time. The dielectric loss value showed a gradual decrease when the temperature value
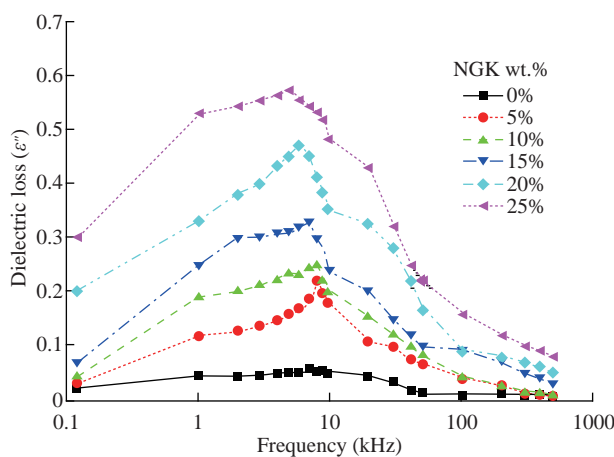

Fig. 5 The variation of dielectric loss factor of PVC-P/NGK-NPs composites with frequency and filler content.

was maintained as constant. The relaxation process was affected by the interfacial polarization [21, 22].

Fig. 6 presents the plots for the relaxation time (τ) versus NGK-NPs powder content. The results presented in the figure indicated that the relaxation time increased with an increase in the filler content. This was related to the increase in the A.C. conductivity and was attributed to the interfacial or MWS polarization seen in a complex system which exhibited electrical heterogeneity, because of the accumulation of the charges at the system interface [23].
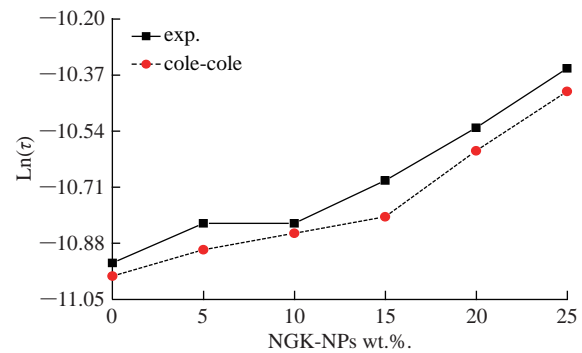

Fig. 6 Relaxation time of the filler content on PVC-P/NGK-NPs composites.

The relationship between the dielectric loss (ε'') as the function of dielectric permittivity (ε') for the PVC-P/NGK-NPs composites at room temperature is presented in Fig. 7. We used the Cole-Cole plot for estimating the generalized relaxation time, which was related to the distribution of all relaxation times. All the values for the relaxation time are collected in Table 1.
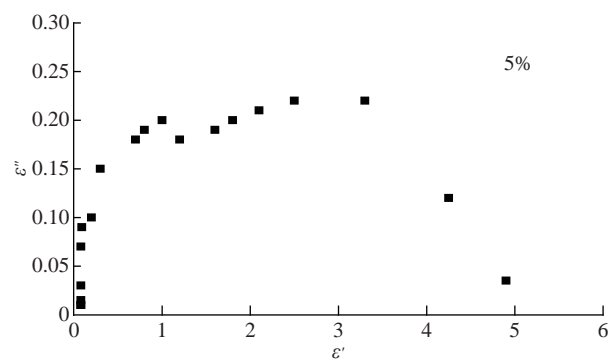

Fig. 7 Relaxation time of the filler content on PVC-P/NGK-NPs composites.

Table 1 The relaxation time for PVC-P/NGK-NPs composites

wt. %	0	5	10	15	20	25
t (sec.* 10^{-5})	1.7	1.88	2.14	2.61	3	3.7

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Fig. 8 describes the temperature dependence of the A.C. conductivity for varying temperatures (ranging from 30 to 110 °C) at the constant electrical field frequency value of 1 kHz for the PVC-P/NGK-NPs composites (differing concentrations). The results attributed the direct relationship between the A.C. conductivity and the temperatures and NGK-NPs concentration to the free charges which were available in this composite system. Furthermore, it was seen that at high temperatures, the mobility of the free charges was responsible for the frequency-dependent conductivity value [25, 26].
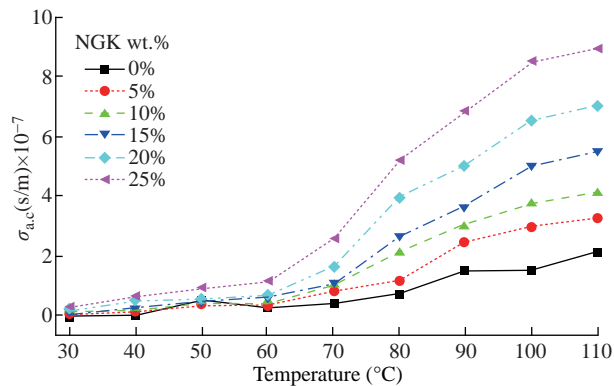

Fig. 8 The variation of $\sigma_{a.c}$ of PVC-P/NGK-NPs composites with temperature and filler content.

The study of the dependence of the A.C. conductivity value on the varying frequency values (ranging from 120 Hz to 2 MHz) for the NGK-NPs weight fractions at the room temperature is depicted in Fig. 9.
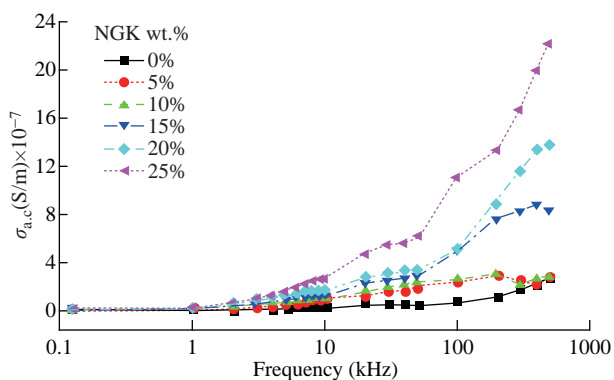

Fig. 9 The variation of $\sigma_{a.c}$ of PVC-P/NGK-NPs composites with frequency and filler content.

The graph shown in Fig. 9 indicated that this A.C. value increased with an increase in the filler content for all the frequency values that were studied. The increase in the A.C. conductivity was expected at a higher field frequency, with more impurities and when

ions could be moved. This increased A.C. conductivity was attributed to the ionic interactions and the impurity movements in the electrolyte polymer composites.

In Fig. 10, we plotted the impedance (Z) for the PVC-P/NGK-NPs composites for varying temperatures (ranging between 30 and 110 °C) for the different NGK-NPs weight fractions at the constant electric field frequency value of 1 kHz. It was seen that after the addition of the NGK-NPs filler powder, the impedance value could be decreased by increasing the temperature value. A decreasing impedance value was attributed to the higher motility of the segmental molecules with an increase in temperature.

Finally, the illustration in Fig. 11 presents the measured impedance of the composites with different NGK-NPs filler content (5, 10, 15, 20 and 25% w/w) at room temperature and for different frequency values (ranging between 120 Hz and 2 MHz). The results indicated that for all the cases, the impedance value exponentially decreased with an increasing frequency and NGK-NPs concentration. This decrease was higher when the filler content was maximal. This
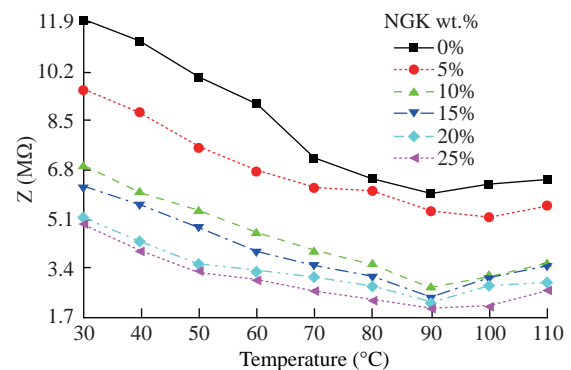

Fig. 10 The variation of impedance of PVC-P/NGK-NPs composites with temperature and filler content.
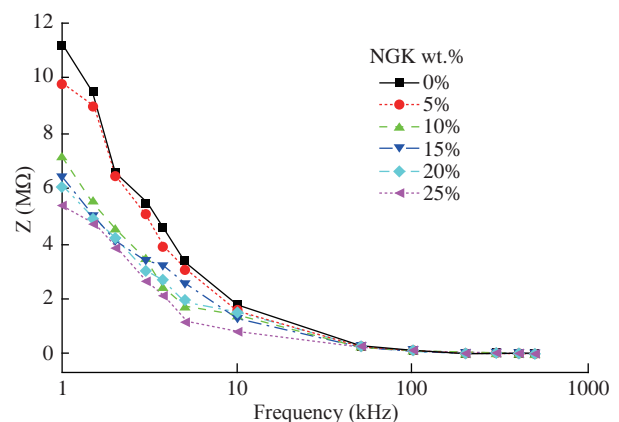

Fig. 11 The variation of impedance of PVC-P/NGK-NPs composites with frequency and filler content.

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decreasing impedance was attributed to increasing A.C. conductivity.

Conclusions

In this study, we investigated the frequency and the temperature dependence of the dielectric constant (ε') and the dielectric loss factor (ε") for the PVC-P/NGK-NPs composites for different frequency values (ranging between 120 Hz and 2 MHz) and differing temperatures (between 30 and 110 °C). Experimental results indicated that the values of the ε' and ε" increased after NGK-NPs were added to the PVC-P resin. At the low-frequency value, the permittivity decreased with an increasing frequency value; however, after the frequency value was increased beyond 100 kHz, these changes were ignored. Thereafter, an increase in the ε' at increasing temperatures was attributed to the higher freedom of the movement of dipoles of the polymeric molecular chains, which occurred at high temperatures. The dielectric loss noted in the composite materials at higher temperatures was ascribed to the glass transition temperature of the PVC-P materials. Lastly, the A.C. conductivity and the impedance value were seen to be temperature and frequency dependent.

Acknowledgements

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