

An Experimental Study of the Dielectric Parameters of PVC Nano-Composites under Corona Conditions

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ABSTRACT

Poly-Vinyl Chloride (PVC) is a commonly used material used in cable insulation sheaths, but its dielectric properties can be negatively impacted by electric aging. This study investigates the use of nano-fillers, specifically alumina (Al_2O_3), titanium dioxide (TiO_2), calcium carbonate ($CaCO_3$), and barium titanate ($BaTiO_3$), in order to improve the dielectric properties of PVC. Films of PVC were doped with nano-fillers and were then exposed to an alternating voltage of 15kV for various time periods (1, 2, and 3 hours). The dielectric properties of PVC were measured using an impedance analyzer, and the results indicated that the use of these nano-fillers had a positive effect on the dielectric characteristics of PVC.

Keywords-PVC; polymer composite; dielectric parameters; corona discharge

I. INTRODUCTION

Polymeric electrical insulation is subjected to electric fields and thermal stresses in distribution power lines, making it crucial to develop new insulation coordination procedures to improve electric insulation parts in substations. In recent years, nanotechnology has been increasingly utilized in the synthesis of polymer-nano-composites, which have shown improved mechanical, electrical, and chemical properties [1-4]. Dielectric spectroscopies in time and frequency domains are useful tools for evaluating and diagnosing electrical insulation and understanding the dynamics of complex solid polymer systems [5-7]. Metal oxide nanoparticles in polymers have been studied as alternative materials for electrical applications [8-11]. Studies have demonstrated that the incorporation of nanoparticles in similar systems can have both positive and negative effects on breakdown strength [12-14]. Furthermore, it has been established that the production of artificial reinforcement fillers can lead to either an increase or decrease in the dielectric strength of polymeric composite insulation, which is utilized in high voltage outdoor applications [15]. Polyvinyl chloride (PVC) is a commonly used polymer in

industrial and communication applications. It is preferred due to its ease of production and processing, good adhesion with reinforcing elements, and resistance to corrosive environments. In addition, PVC has ductile electrical performance and excellent dielectric strength, low dielectric permittivity, low loss factor, and favorable thermo-mechanical behavior. PVC has been utilized as electrical insulators in distribution for a long time due to these properties [16].

A study of PVC's compression behavior was conducted in which its dielectric reaction was evaluated within the frequency range of 20Hz-1GHz [17]. Various research efforts have focused on developing high-performance materials with exceptional dielectric properties [18-23]. Modern polymer nano-composites have gained significant attention for their expanded applicability. There has been a growing interest in utilizing nanotechnology to improve the dielectric properties of materials. In this regard, research has been conducted to investigate the impact of different nanoparticles, such as clay, fumed silica, zinc oxide, and titanium dioxide, on the electric and dielectric loss performance of PVC in experimental settings. As the field of polymer nano-composites continues to

advance, this study examines how the type and concentration of nanoparticles at various voltages and frequencies (10Hz-10KHz) affect the electric and dielectric characteristics of PVC [24-32].

Corona discharge is a significant factor that can have a notable impact on the properties of polymeric insulating materials like PVC. Several studies have investigated the effect of corona discharge on the surface properties of RTV and HTV silicone rubber filled with micro-sized alumina trihydrate (ATH) fillers, utilizing various corona generation techniques [34, 35]. Authors in [36, 37] reported improved corona resistance performance of silicone rubber composites. The focus of the current paper is to examine the impact of different types of nano-fillers and their loading on the dielectric characteristics of PVC composites when subjected to AC corona discharge.

II. EXPERIMENTAL SETUP

Table I provides information on the composition of the samples analyzed in this study. The main component of the samples was PVC resin of the 4000M type, which is commonly used in the Algerian industry. An antioxidant (Barium-Zinc) was added to ensure thermal stability, and dicumyl peroxide was used as a linking agent, along with lubricant and Plasticizer DOP (Di-Octyle Phthalate) from Zhengzhou P&B Chemical Co., Ltd (99.5%). According to [38], two types of samples were prepared: standard PVC and 4 ceramic-doped PVC samples with 10% of BaTiO₃, Al₂O₃, TiO₂, and CaCO₃, respectively. The formulation was mixed in a two-roll mixer at a temperature of 160°C for 10 minutes. The resulting mixture was then formed into sheets of predetermined thickness, which were subsequently cut into 10cm×10cm squares with a thickness of 0.3mm. Some of the composite samples were subjected to electric discharge at 15KV for 1, 2, and 3 hours, as shown in Figure 1.

TABLE I. SAMPLES USED

1	2	3	4	5
Pure PVC	PVC + 10% BaTiO ₃	PVC + 10% Al ₂ O ₃	PVC + 10% TiO ₂	PVC + 10% CaCO ₃



Fig. 1. Samples before and after electric aging.

The electrical aging of the specimens was performed in the high voltage test chamber of the HV laboratory at the Mouloud Mammeri University of Tizi-Ouzou. The experimental setup for electrical aging is shown in Figure 2(a), and the test protocol involved the following steps [39]:

- The test protocol for electrical aging involved placing the specimen between 2 flat-tip electrodes, with the upper

electrode connected to the high voltage and the lower electrode connected to the ground. The electrodes were carried by a bakelite support designed to be perfectly opposite and perpendicular to the plane, and to facilitate adjustment of the inter-electrode distance and ensure good contact with the sample. An alternating voltage of 15kV was applied to the specimen for 1, 2 and 3 hours.

- To avoid the phenomenon of contentment, a green plastic plate or other material with a surface area greater than that of the sample was placed between the contact electrode (lower) and the test specimen, as shown in Figure 2(b).
- After placing the specimen between the electrodes and the plastic plate, the chamber door was closed, and the main circuit breaker for the faraday chamber was powered on.
- Next, the manual circuit breaker of the control console was powered on.
- After powering on the circuit breaker of the control console, the green button was pressed to power the system. The voltage was varied and monitored on a voltmeter until it reached the desired voltage of 15kV, which was obtained by using the autotransformer located in the control panel. This process was repeated for 1, 2, and 3 hours of electrical aging.

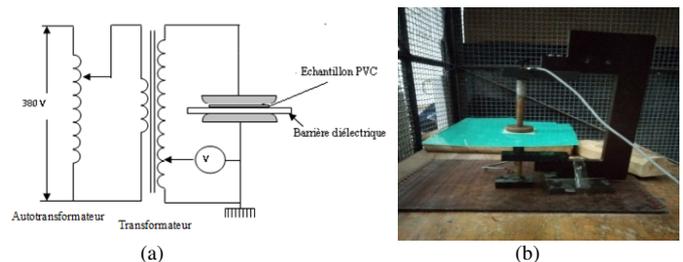


Fig. 2. Experimental setup for electrical aging

Impedance analyzers are commonly used to measure the electrical properties of materials, such as capacitance and resistance, as a function of frequency. In this case, the RLC 817 meter was used to measure the dielectric response of the samples up to 10MHz. The samples were placed between two plane electrodes made of copper, and an alternating electric field of 1V/m was applied. From these measurements, tgδ (dissipation factor), ε_r (relative permittivity), and loss index were calculated. The measurement error was to be ±0.05%.



Fig. 3. Samples in electric aging.

III. RESULTS AND DISCUSSION

In this research, the dielectric parameters $\text{tg}\delta$, ϵ_r , and loss index were measured at frequencies ranging from 10Hz to 10KHz for all the manufactured samples, including the standard PVC.

A. Relative Permittivity

Figure 4 shows that for all types of PVC samples, the relative permittivity initially increases with frequency up to a certain point, and then decreases with further increase in frequency. Additionally, the magnitude of the relative permittivity peaks varies with aging time and type of nanoparticle filler. It is worth noting that the observed behavior in the low frequency range (12Hz - 500Hz) may be related to the presence of electrode polarization effects, which can lead to inaccurate measurements of the material's dielectric properties in this frequency range.

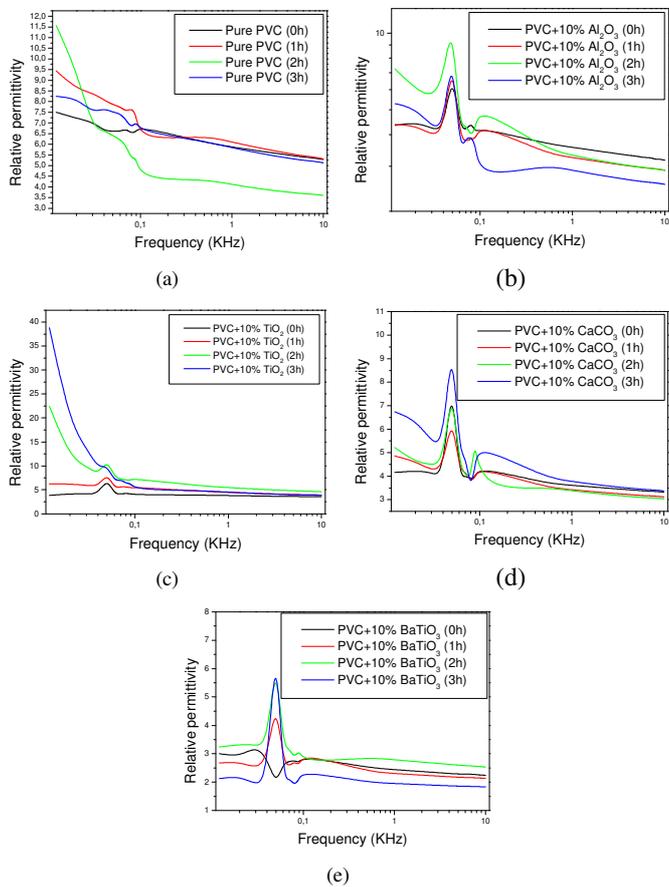


Fig. 4. Variation of the relative permittivity as a function of frequency and aging time. (a) Pure PVC, (b) PVC+10% Al_2O_3 , (c) PVC+10% TiO_2 , (d) PVC+10% CaCO_3 , (f) PVC+10% BaTiO_3 .

B. Dissipation Factor

The values of $\text{tg}\delta$ come directly from the RLC meter. Figure 5 represents the variation of the dissipation factor as a function of frequency and aging time.

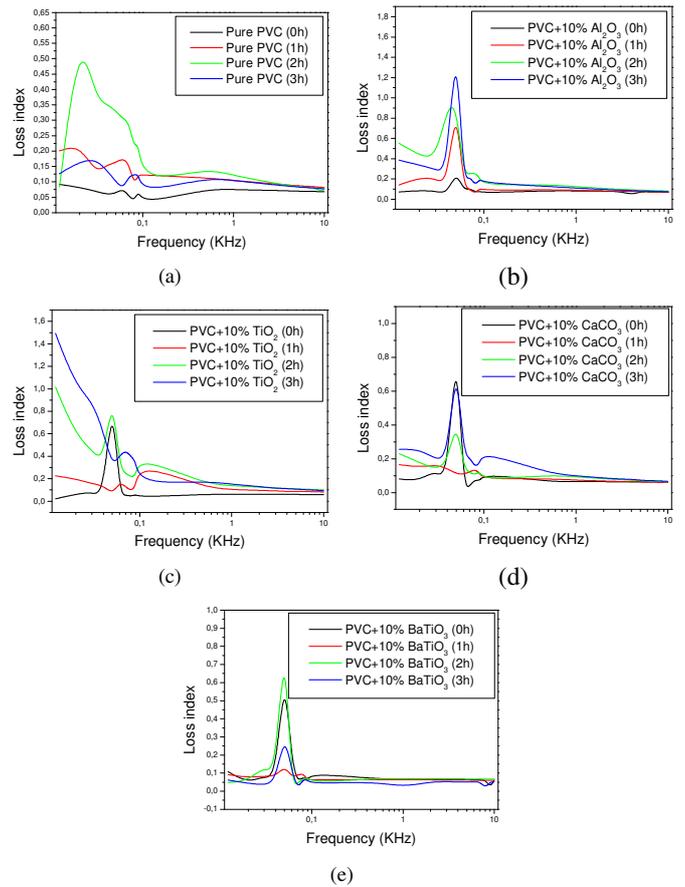


Fig. 5. Variation of the dissipation factor $\text{tg}\delta$ as a function of frequency and aging time. (a) Pure PVC, (b) PVC+10% Al_2O_3 , (c) PVC+10% TiO_2 , (d) PVC+10% CaCO_3 , (f) PVC+10% BaTiO_3 .

The analysis of the results of the evolution of the dissipation factor allowed us to note that it decreases with the increase in frequency (500hz-10Khz) and increases with the increase in the duration of application of the stress (electric corona discharge).

C. Dielectric Losses

The variations of dielectric losses as a function of frequency and aging time for the 5 considered types of PVC are shown in Figure 6. We notice that all the curves show almost the same pace, with a peak at low frequencies (between 50-80Hz) and then stabilize for the other frequencies. Losses increase with increasing corona discharge time but with a slight difference (especially for high frequencies > 1KHz). Figure 7 shows a comparison between the results of relative permittivity, dissipation factor and dielectric losses of all nanocomposites after electric aging for 3 hours. It is observed that the curves have the same pace (increase in the low frequencies and consequently decrease until they stabilize in high frequencies). It seems that BaTiO_3 is the most effective dopant among the 5 tested, with the smallest values for dielectric losses. Al_2O_3 comes second, followed by CaCO_3 . This information can be useful for selecting the most appropriate dopant for improving the electrical properties of PVC.

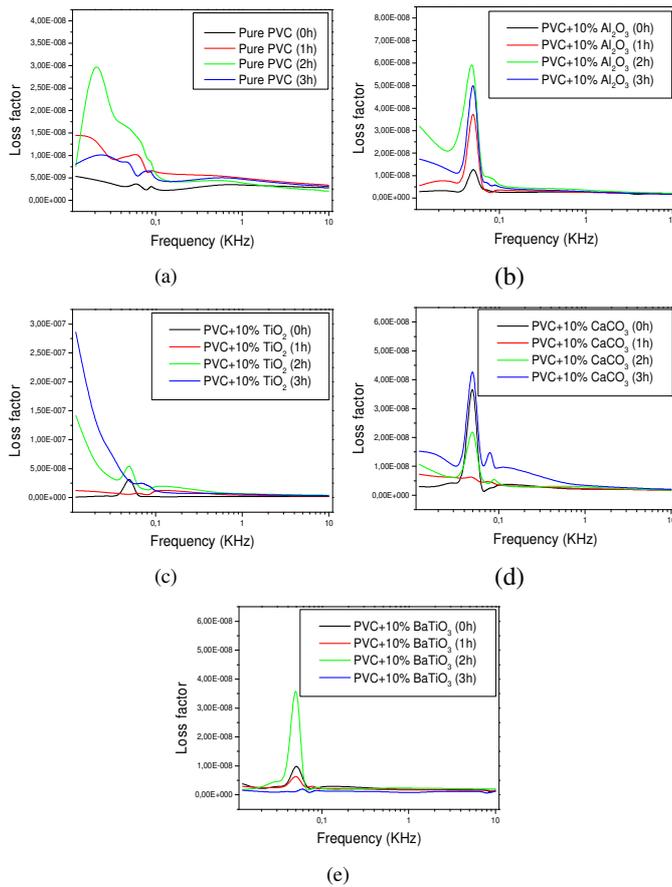


Fig. 6. Variation of dielectric losses as a function of frequency and aging time. (a) Pure PVC, (b) PVC+10% Al₂O₃, (c) PVC+10% TiO₂, (d) PVC+10% CaCO₃, (e) PVC+10% BaTiO₃.

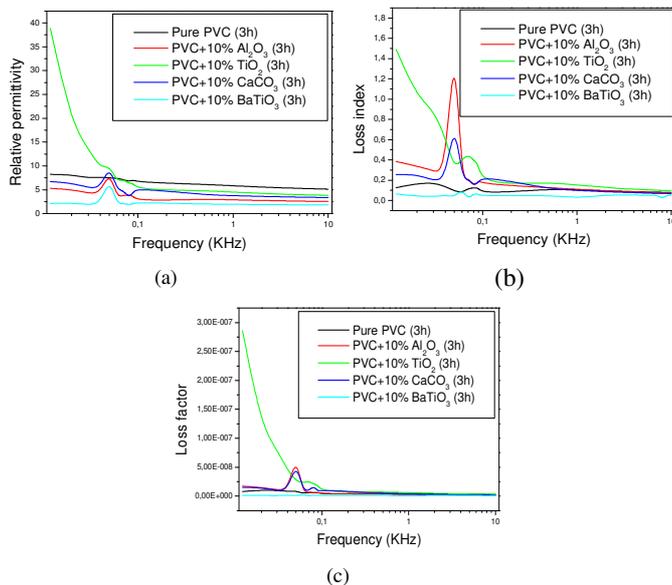


Fig. 7. Comparisons between nanocomposites after electric aging for 3 hours of: (a) Relative permittivity, (b) dissipation factor, (c) Dielectric losses.

IV. DISCUSSION

In general, the existing literature indicates that the dielectric constants and loss tangents of insulators are influenced by various physical, chemical, and structural changes that occur during their use [8, 19-22]. In order to enhance the performance of insulators, new polymeric composites have been developed that incorporate ceramic powders. To assess the impact of different nano-fillers such as BaTiO₃, CaCO₃, Al₂O₃, and TiO₂, as well as the aging under corona discharge conditions, the dielectric properties of PVC were analyzed.

Incorporating nano fillers into the PVC matrix allows for the modification of the composite's dielectric parameters. These values are dependent on the type of filler, the integration rate, and the dielectric parameters of the fillers themselves [1, 21, 35]. Specifically, in our study, the dielectric constant (ϵ_r) increased and the loss tangent ($\text{tg}\delta$) decreased as the nano-filler content increased, as shown in Figures 4 and 5. As demonstrated in Figure 4, for a frequency of 100Hz, the values of ϵ_r for BaTiO₃, TiO₂, CaCO₃, and Al₂O₃ decrease by approximately 57%, 50%, 35%, and 37%, respectively, compared to the value of ϵ_r for pure PVC prior to aging. These findings are in agreement with the findings of [13, 25, 33], and can be validated through (1) [1]:

$$\frac{\epsilon_1 - \epsilon_c}{\epsilon_1 - \epsilon_b} \left(\frac{\epsilon_b}{\epsilon_c} \right) = 1 - P_{\text{Par}} \tag{1}$$

where ϵ_b , ϵ_1 , and ϵ_c represent the relative permittivity's of PEI, BaTiO₃ nano-particles, and the nano-composite, respectively, P_{par} is the volume fraction of the particles, and μ is a parameter that depends on subtleties of the microstructure such as particle clustering and surface roughness.

The doped samples exhibit greater resistance than pure PVC to the aging process. Specifically, at a frequency of 100Hz, the values of ϵ_r for BaTiO₃, TiO₂, CaCO₃, and Al₂O₃ increase by 45%, 14%, 18%, and 16%, respectively. The observed reduction in space charge in the PVC matrix, as evidenced by the capturing behavior of ceramics, supports the findings of [16, 40]. The primary current carriers were attributed to free ions generated from ingredients used in the polymerization reaction of PVC, such as the stabilizer, as well as ingredients in the plastifier itself. Additionally, the similarity in the shape of the curves of all the sample types of PVC/doped PVC can be attributed to interfacial polarization and rearrangement of the molecular structure. This relaxation is reflected in the peaks observed in the ϵ_r curves, which are consistent with those reported in [5, 29-31, 41].

The dielectric parameters of the composites are affected by aging, as demonstrated by the decrease in ϵ_r values and the increase in $\text{tg}\delta$ values over time for all composites. However, the incorporation of the load helps to slow down the electrical aging effect and alter the dielectric parameter values. Composite aging is characterized by a broadening of the peak in the low-frequency region, as shown in Figures 4 and 5. This result is consistent with previous research on BaTiO₃ doped PVC [28], where a peak in the dielectric constant curve was observed around 50Hz, and a second peak was detected at 500Hz. The same relaxation phenomena were also observed in our previous work [1, 28] and in [29]. Specifically, the first and

second relaxation phenomena occurred at low frequencies around 50 and 500Hz, respectively, as shown in Figures 4 and 5.

Based on the analysis of Figure 7 and a comparison of the various doped PVC samples, it is observed that an increase in nano filler content resulted in an increase in ϵ_r values and a decrease in $\tan\delta$ values. Among the different nano fillers studied, BaTiO₃ exhibited better resistance to aging. This can be attributed to the reduction of space charge, which is a result of the capturing behavior of ceramics in the PVC matrix [25, 43]. The main carriers present in the samples were attributed to free ions from the ingredients used in the polymerization reaction of PVC, such as the stabilizer, as well as ingredients in the plasticizer itself.

In addition, the breakdown strength of the samples, as shown in Figure 7, was found to be dependent on the ceramic content, with values decreasing as the ceramic content increased up to a certain practical limit, as reported in [27-28]. The formation of void defects and the resulting electromechanical stress led to the formation of cracks, which played a crucial role in the injection of electrons from the electrodes, providing them with enough energy to increase the probability of macromolecule ionization and the initiation of electron avalanche. This process accelerated the development of conducting channels and ultimately led to breakdown, as reported in [29-31]. This phenomenon can be explained by the reduction of space charge due to the capturing behavior of ceramics in the PVC matrix [26, 42-44].

V. CONCLUSION

It is important to note that the choice of nanoparticles and their concentration should be carefully considered to avoid any negative impact on other properties of the insulation material, such as mechanical strength, thermal stability, and moisture resistance. Additionally, the production process of the nanocomposites should be optimized to ensure homogeneous dispersion of the nanoparticles in the polymer matrix, which is crucial for achieving consistent and reliable electrical insulation performance.

Overall, the use of nanoparticles in electrical insulation materials shows a great potential for improving the electrical properties and performance of the materials, and can lead to more efficient and reliable electrical power systems.

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