



Experimental Study of Space Charge Characteristics in Thin Films of Polyvinyl Chloride Nanocomposites

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Abstract-- This paper aims to provide a view of a lively space charge accumulation in new polyvinyl chloride (PVC) nanocomposite thin films. Space charges in thin films of polyvinyl chloride nanocomposite insulation materials are done to distort the local electric fields and affect high-field conduction and breakdown phenomena, whatever, adding nanofillers to polyvinyl chloride changes their electrical ages. Thus, space charge characteristics have been investigated by pulsed electroacoustic (PEA) measurement system for thin thickness of polyvinyl chloride nanocomposite materials which filled with different separately nanoparticles like ZnO, TiO₂, and Al₂O₃. This paper depicts that space charge accumulation in polyvinyl chloride nanocomposites differs with respect to type and concentration of nanoparticles.

Keywords: Space charge, Nanocomposite, PEA system, Polymers, Polyvinyl Chloride, Insulation, Nanoparticles

1. Introduction

Space charge accumulation is known to play a very important role in the aging and breakdown of HVDC solid polymeric insulation, due to local electric field enhancement. If the applied field exceeds the threshold for space charge accumulation, charge injected from the electrodes can accumulate in traps located at the interface with electrodes and in the insulation bulk [1-5]. The existence of such a threshold would have considerable impact on the design of insulation systems. Designing an insulation system for an electrical stress below the threshold would assure very long life and high reliability, provided that there are no other significant stresses acting on the insulation. In order to obtain a better understanding of the role of the nanoparticles in the process of dielectric breakdown, lifetime, and space charge behavior, the interfacial region needs to be investigated [6, 7]. A number of mechanisms have been inferred for the electro thermal aging process occurring in insulating polymers with the aim of deriving usable life expressions. These mechanisms have followed two prevailing directions: a macroscopic approach, based on an overall description of degradation processes, and a microscopic approach, based on the presumption that the prevailing cause of electrical and mechanical aging in practical insulation systems is accelerated localized degradation triggered by micro defects [8, 9]. For the enhancement of electrical insulation reliability and compact design in electric power apparatus, the electrical properties of polymer composites filled with metal oxide particles will play an important role. Recently, it has been recognized that the introduction of metal oxide nanoparticles to polymer has unique dielectric behavior and advantages when compared to the base polymer and the polymer filled with conventional micrometric particles [10-16]. The effects of nanosized particle additives on the electrical properties of common dielectrics are currently of considerable interest [17-22]. The trapped space charge density is obtained from the depolarization current characteristics according to the following expression [23],

$$q(t) = \int_0^t i(t) dt \quad (1)$$

Where, $i(t)$ is the depolarization current, $q(t)$ the trapped charge density and t is time.

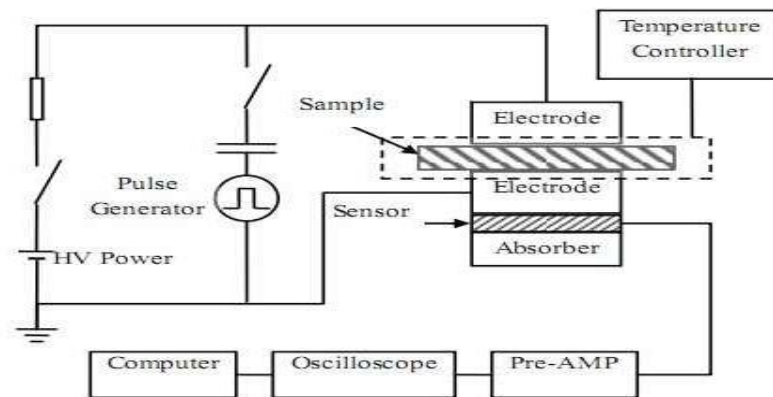
Polyvinyl Chloride is stronger and more rigid than other general purpose thermoplastic materials like ac and dc cable insulations, etc. It has a high tensile strength and modulus of elasticity. Nowadays, the research goes towards for concerning about the effect of nanotechnology on space charge distribution in polymeric insulations. With a continual progress in polymer nanocomposites, the current research depicts the effects of types and concentration of nanoparticles in new nanocomposite industrial polymer material. Space Charge Dynamics in polyvinyl chloride Nano-composite materials which use ZnO or TiO₂ or Al₂O₃, nanoparticles separately under DC Stress have been investigated and discussed experimentally.

2. Experimental Setup

Acoustic wave propagation in the thick samples is significantly attenuated and dispersed due to the loss and dispersive properties of the polymer and principle of space charge measurement using the PEA technique. The specifications of each module in PEA system is being as follows: The oscilloscope (LECROY, LC334AM, 500MHz), impulse generator (TECHMP, 10nsec), VHF fixed attenuator (30W, 40dB) and stabilized voltage supply (Type L30). Measurements are performed in silicon oil to avoid partial discharges, and to keep the temperature constant. The applied voltage was monitored simultaneously by using a resistance voltage divider. In order to observe the effect of pre-stressing on impulse breakdown, it existed that an impulse generator, and a coupling capacitor (2000 pF). Figure 1 shows Life photo and Schematic diagram of the PEA system which used in measuring.



(a) Life photo



(b) Schematic diagram

Figure 1. PEA system testing equipment's

3. Material Preparation and Characterization

Although nanoparticles are generally considered a discovery of modern science, they actually have a very long history. With respect to electrical and mechanical importance of zinc oxide, titanium dioxide, and aluminum oxide in industrial applications, they are suggested to embed as inclusions in Polyvinyl Chloride. Additives of clay, zinc oxide, and fumed silica nanoparticles to the base industrial polymers (Polyvinyl Chloride) has been fabricated by using mixing, ultrasonic, and heating processes in nanotechnology Research Centre, Aswan - Egypt. Preparations of studied polyvinyl chloride nanocomposites have been used SOL-GEL method. The sol-gel processing of the nanoparticles inside the polymer dissolved in non-aqueous or aqueous solution is the ideal procedure for the formation of interpenetrating networks between inorganic and organic moieties at the milder temperature in improving good compatibility and building strong interfacial interaction between two phases. This process has been used successfully to prepare nanocomposites with nanoparticles in a range of polymer matrices. Several strategies for the sol-gel process are applied for formation of the hybrid materials. One method involves the polymerization of organic functional groups from a preformed sol-gel network. The sol- gel process is a rich chemistry which has been reviewed elsewhere on the processing of materials from glass to polymers. The organic-inorganic hybrid nanocomposites comprising of polymer, and nanoparticles were synthesized through sol-gel technique at ambient temperature. The inorganic phase was generated in situ by hydrolysis-condensation of tetraethoxysilane (TEOS) in different concentrations, under acid catalysis, in presence of the organic phase, polymer, dissolved in formic acid [24]. Penetration of nanofillers into polyvinyl chloride has been shown by SEM measurements in Figure 2.

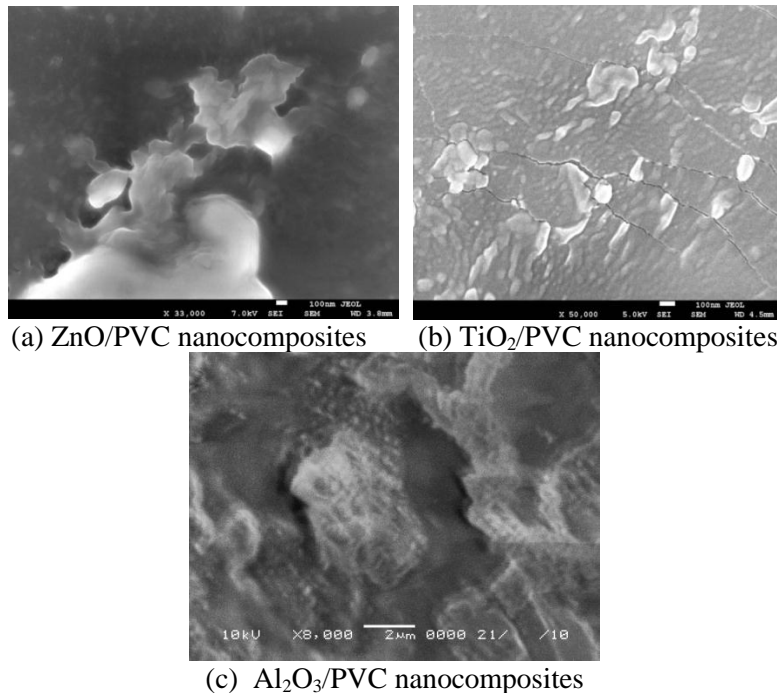


Figure 2. SEM measurements for PVC nanocomposites

HIOKI 3522-50 LCR Hi-tester device is measuring characterization of nanocomposite insulation industrial materials. Thus, the industrial materials studied here is Polyvinyl Chloride, has been formulated utilizing nanoparticles of ZnO, TiO₂, and Al₂O₃. The base of all these polymer materials is a commercially available material already in use in the manufacturing of high-voltage (HV) industrial products and their properties detailed in table 1.

TABLE 1
DIELECTRIC PROPERTIES OF PURE AND NANO-COMPOSITE MATERIALS

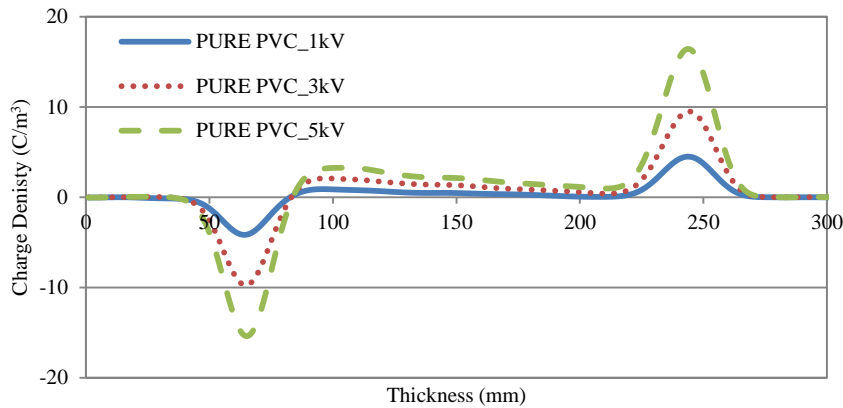
MATERIALS	DIELECTRIC CONSTANT AT 1KHZ	RESISTIVITY ($\Omega.M$)
Pure PVC	3.3	10^{13}
PVC + 1% wt ZnO	3.24	10^{14}
PVC + 5% wt ZnO	3.19	$10^{14}-10^{15}$
PVC + 10% wt ZnO	3.04	$10^{15}-10^{16}$
PVC + 1% wt TiO ₂	3.94	10^{12}
PVC + 5% wt TiO ₂	4.64	$10^{12}-10^{11}$
PVC + 10% wt TiO ₂	5.17	$10^{11}-10^{10}$
PVC + 1% wt Al ₂ O ₃	3.39	10^{13}
PVC + 5% wt Al ₂ O ₃	3.59	$10^{12}-10^{11}$
PVC + 10% wt Al ₂ O ₃	3.99	$10^{12}-10^{11}$

4. Results and Discussion

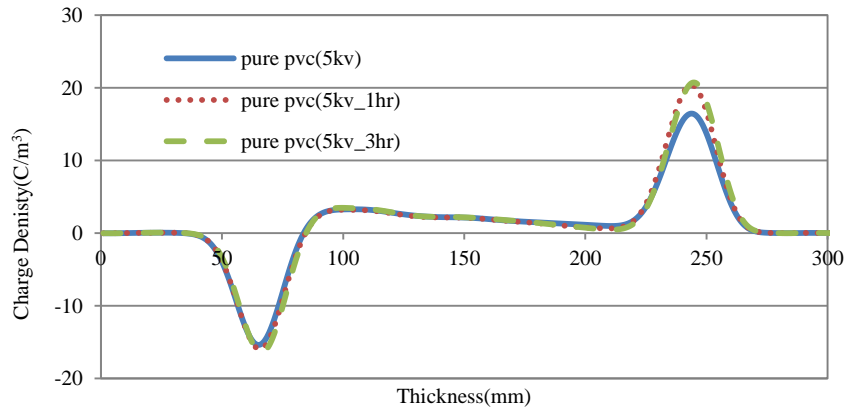
PEA system measurements were carried out using two semiconductor cylindrical stainless steel electrodes with diameter 50 mm, and placing specimens in a mineral oil vessel kept at 20 °C. Tests were realized applying an increasing dc breakdown voltage up to 7.5 kV. Space charge distribution at various dc voltages stressed and period times can be seen the positive and negative space charge peaks related to the cathode and anode electrodes that found subsequently using variant applied voltages and times. The measurements imply the injection of impulse high voltages on the charge density of Pure PVC dielectric materials; the cathode is at the left and the anode is at the right.

4.1 Space Charge Accumulation in Pure Polyvinyl Chloride

Figure 3.a shows space charge distribution in pure polyvinyl chloride under various DC electric fields; the charge density increases with increasing impulse voltages and hetero-charge accumulated near the cathode.



(a) Pure PVC with varying DC voltages



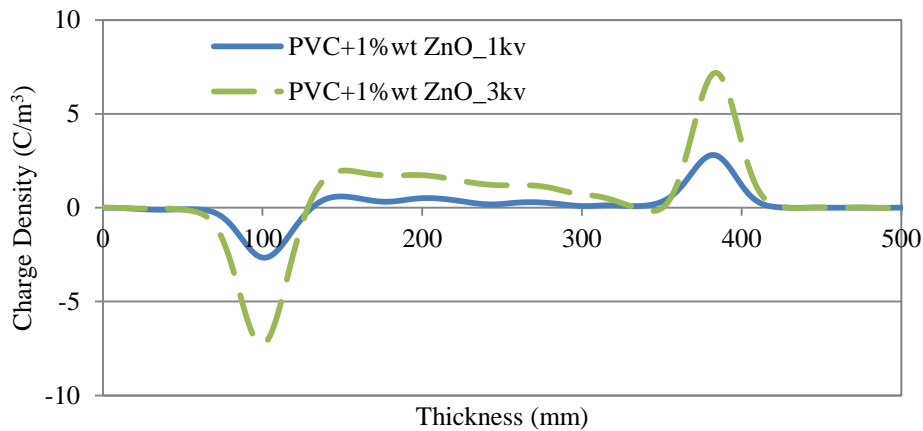
(b) Pure PVC with varying time

Figure 3. Space charge measurement profiles for pure polyvinyl chloride

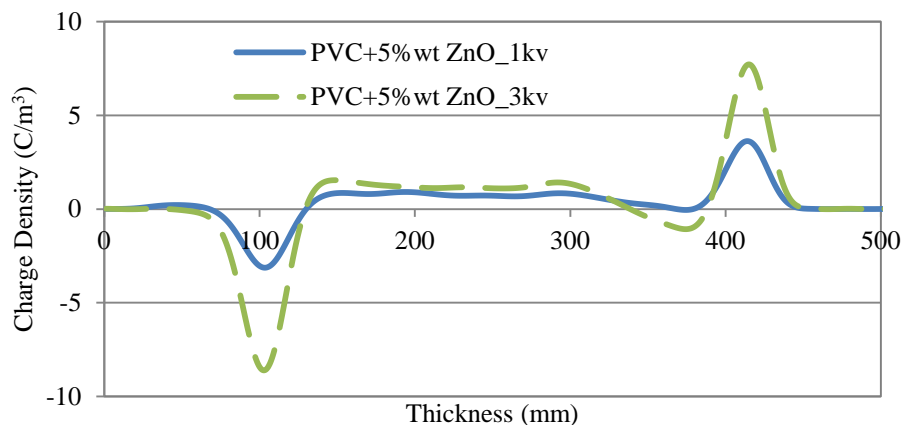
On the other wise; Figure 3.b contrasts on the space charge distribution, and its decay happened in remaining 5kV, observed in the samples after three hour space charge electrodes. Charge density increases with remaining impulse voltages at 5kV especially at the Anode but the hetero-charge accumulated occurs near the cathode. The measurements therefore imply the injection of negative charge from the cathode, which remains trapped close to the injecting electrode when the duration is three hours.

4.2 Space Charge Accumulation in ZnO/ Polyvinyl Chloride Nanocomposites

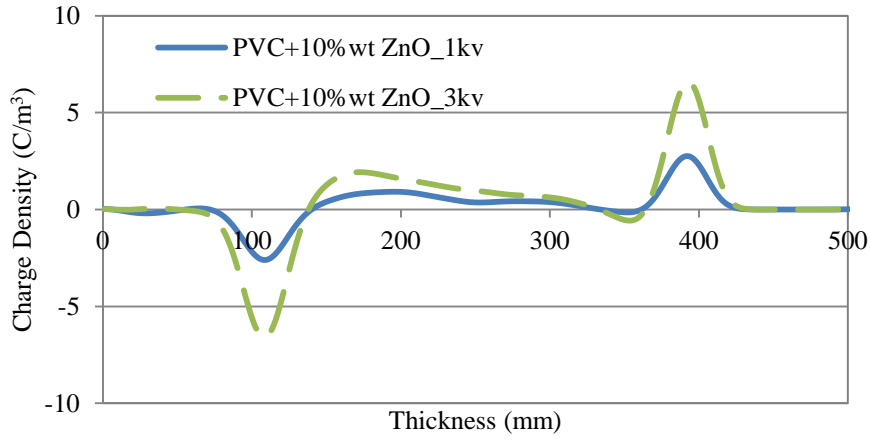
Figure 4.a and Figure 4.b show space charge distributions relevant to ZnO/PVC nanocomposite specimens 1%wt, and 5%wt of ZnO nanoparticles respectively under various DC electric fields. The measurements imply the injection of impulse high voltages on the charge density of ZnO/PVC nanocomposites; charging density at the cathode is more than charging density at the anode with increasing ZnO nanoparticles percentage and impulse applied voltages; especially through 5%wt specimens and there is hetero-charge accumulated near anode and cathode electrodes. Figure 4.c shows space charge distributions relevant to ZnO/PVC nanocomposites with 10%wt ZnO nanoparticles percentage under various DC electric fields. The measurements imply the injection of impulse high voltages on the charge density of ZnO/PVC nanocomposites; charging density at the cathode is more than charging density at the anode with increasing impulse applied voltages and there is hetero-charge accumulated near the anode. It is noticed that increasing ZnO nanoparticles from 5%wt up to 10%wt decreasing charge density in the nanocomposite molecules at electrodes. On the other wise, Figure 5 contrasts on the space charge distribution in ZnO/PVC nanocomposites with 10%wt nanoparticles percentage, and its decay that happened in the samples through three hour remaining voltage. Charge density increases with increasing remaining time up to three hours, especially at the Anode.



(a) 1% wt ZnO / PVC nanocomposite



(b) 5% wt ZnO / PVC nanocomposite



(c) 10% wt ZnO / PVC nanocomposite

Figure 4. Space charge profiles for ZnO/PVC nanocomposite with varying DC electric fields

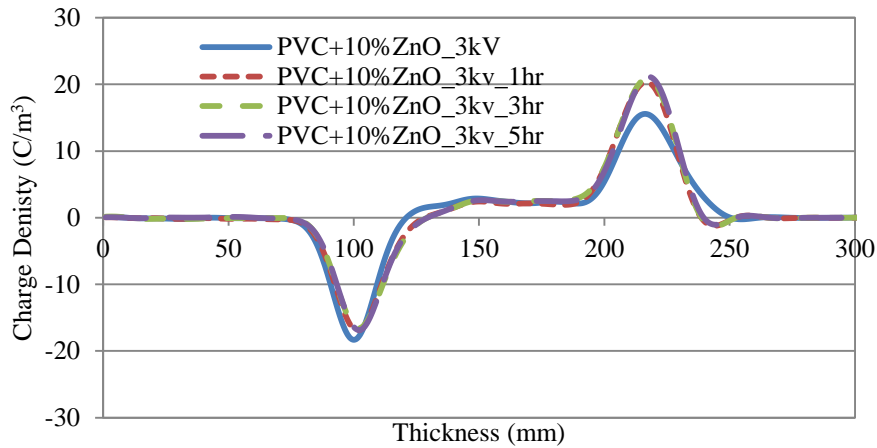
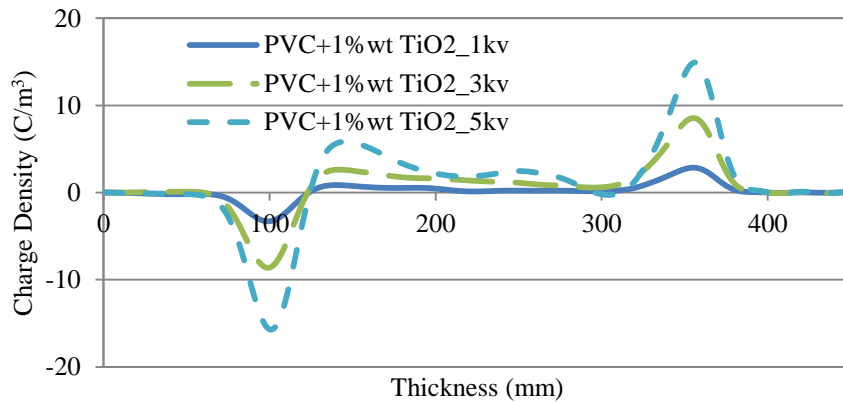


Figure 5. Space charge profiles for 10% wt ZnO/PVC nanocomposite with varying time

4.3 Space Charge Accumulation in TiO₂/ Polyvinyl Chloride Nanocomposites

Figure 6.a and Figure 6.b show space charge distributions relevant to TiO₂/PVC nanocomposite specimens 1%wt, and 5%wt of TiO₂ nanoparticles respectively under various DC electric fields. The measurements imply the injection of impulse high voltages on the charge density of TiO₂/PVC nanocomposites; charging density between electrodes increases with increasing TiO₂ nanoparticles percentage and impulse applied voltages; especially through 5% wt specimens and there is hetero-charge accumulated between electrodes.

(a) 1% wt TiO₂ /PVC nanocomposite

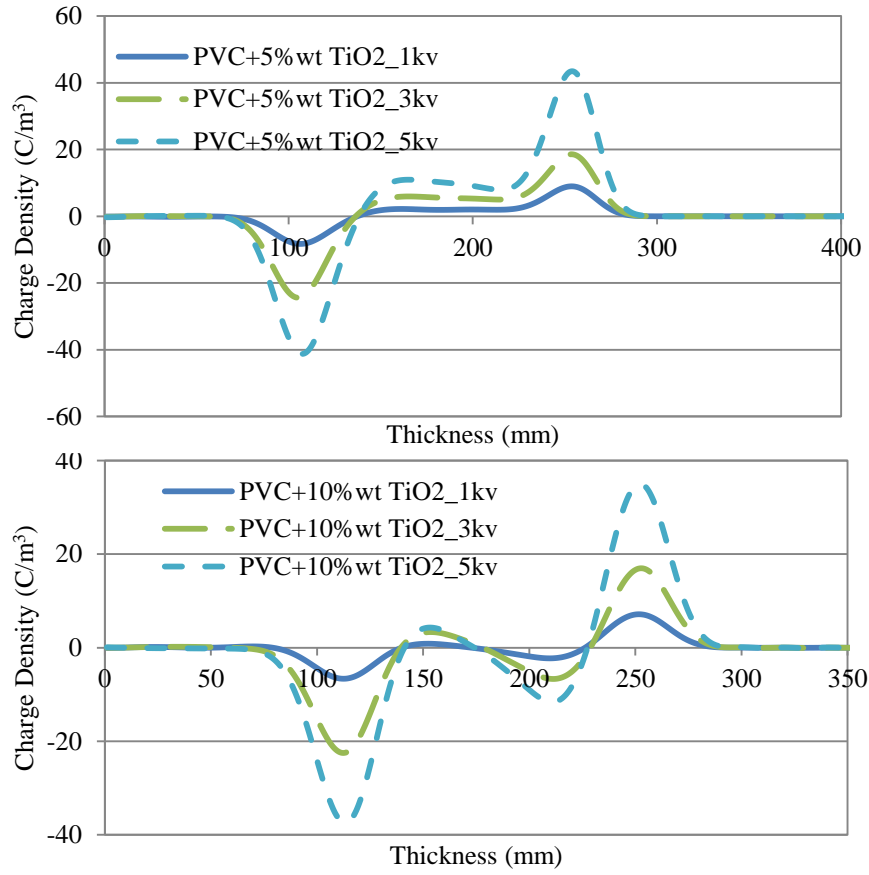
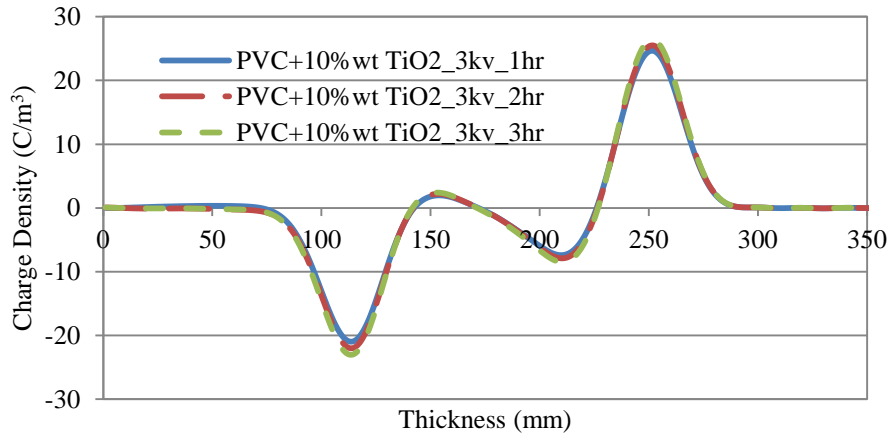
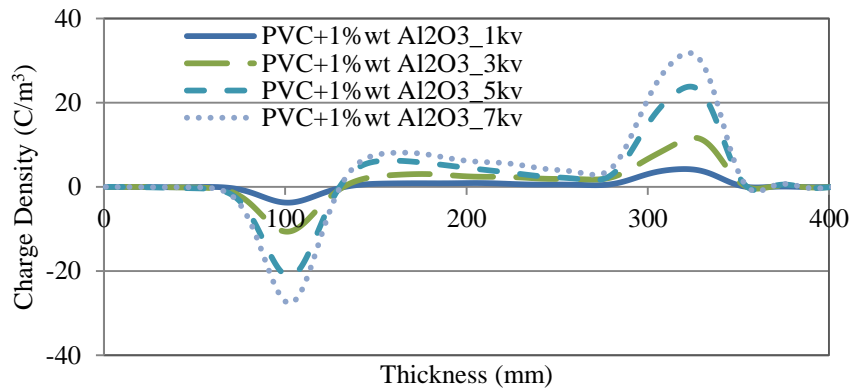
(c) 10% wt TiO₂/PVC nanocompositeFigure 6. Space charge profiles for TiO₂/PVC nanocomposite with varying DC electric fieldsFigure 7. Space charge profiles for 10% wt TiO₂/PVC nanocomposite with varying time

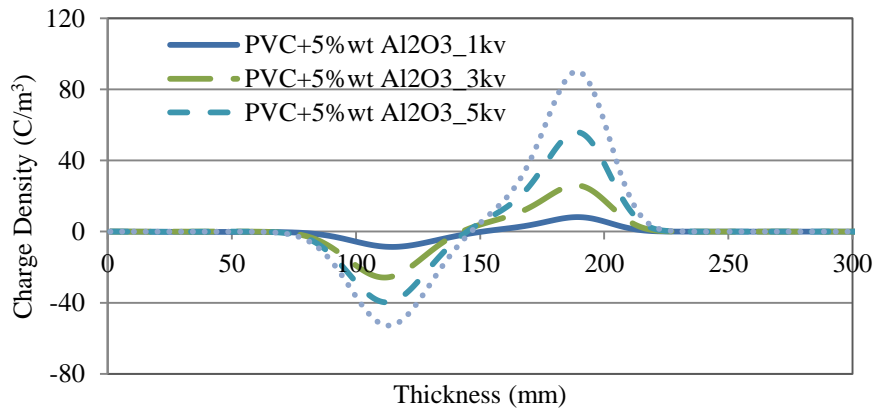
Figure 6.c shows space charge distribution relevant TiO₂/PVC nanocomposites with 10%wt nanoparticles percentage with varying DC electric fields. The measurements imply the injection of impulse high voltages on the charge density of relevant TiO₂/PVC nanocomposites with 10%wt nanoparticles percentage; the charge density at both electrodes increases with increasing impulse applied voltages and there is high hetero-charge accumulated between anode and cathode electrodes. It is noticed that increasing TiO₂ nanoparticles from 5%wt up to 10%wt decreasing charge density in the nanocomposite molecules at electrodes. Also, Figure 7 contrasts the space charge distribution with remaining 3 kV over TiO₂/PVC nanocomposites with 10%wt nanoparticles percentage through three hours. There is no changing in Charge density with remaining impulse voltages between both anode and cathode electrodes.

4.4 Space Charge Accumulation in Al₂O₃/ Polyvinyl Chloride Nanocomposites

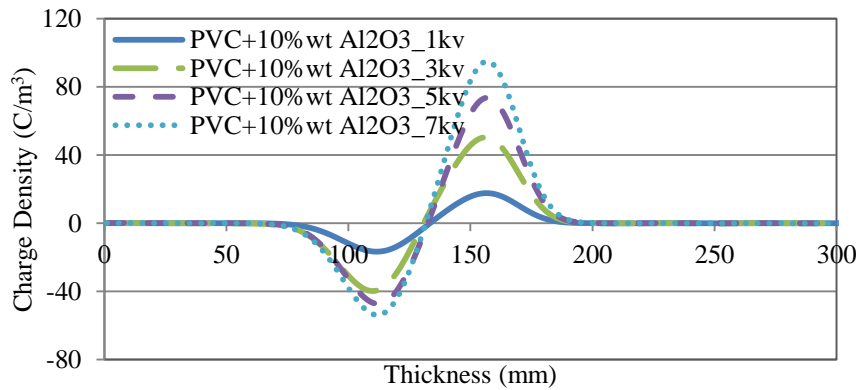
Figure 8 shows space charge distribution relevant Al₂O₃/Polyvinyl Chloride nanocomposites aluminum oxide nanoparticles percentages (1%wt, 5%wt and 10%wt) under varying DC electric field 70kV/mm. The measurements imply the injection of impulse high voltages on the charge density of relevant Al₂O₃/Polyvinyl Chloride nanocomposites with variant nanoparticles percentage; the charge density at both electrodes increases with increasing impulse applied voltages up to 5kV and there is high hetero-charge accumulated between anode and cathode, specially, in case adding 1%wt percentage. Noting that, Figure 8.b and Figure 8.c contrast the space charge distribution with remaining 3 kV over Al₂O₃/Polyvinyl Chloride nanocomposites with 10%wt nanoparticles under varying DC electric field. There is no changing in Charge density with remaining impulse voltages at cathode but there is a very small rising at the Anode.



(a) 1% wt Al₂O₃/ PVC nanocomposite



(b) 5% wt Al₂O₃/PVC nanocomposite



(c) 10% wt Al₂O₃/PVC nanocomposite

Figure 8. Space charge profiles for Al₂O₃/ PVC nanocomposite with varying DC electric fields

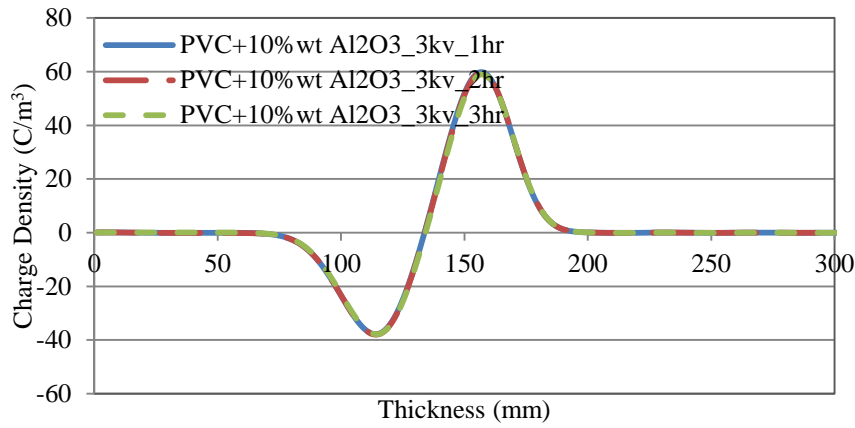


Figure 9. Space charge profiles for 10% wt Al_2O_3 / PVC nanocomposite under constant DC electric field

Figure 9 shows space charge distribution relevant Al_2O_3 /Polyvinyl Chloride nanocomposites with 10%wt nanoparticles percentage with remaining 3 kV over specimens. The measurements imply the injection of impulse high voltages on the charge density of relevant Al_2O_3 /Polyvinyl Chloride nanocomposites with 10%wt nanoparticles percentage; there is no increasing in charge density at both electrodes with remaining impulse voltages at cathode and there is no hetero-charge accumulated between anode and cathode

5. Effects of Nanoparticles on Space Charge of Polyvinyl Chloride

Space charge accumulation in pure polyvinyl chloride is shown in Figure 3 the charge density increases with increasing impulse voltages and hetero-charge accumulated near the cathode. Also, charge density increases with remaining impulse voltages at the Anode and the hetero-charge accumulated near the cathode. The effect of raising concentration of ZnO nanofillers in polyvinyl chloride decreases its dielectric constant as shown in table I, whatever, Figure's (4, 5) depicts charging density at the cathode is more than charging density at the anode with increasing ZnO nanoparticles percentage and impulse applied voltages; especially through 5%wt specimens and there is hetero-charge accumulated near the cathode. Increasing ZnO nanoparticles from 5%wt up to 10%wt decreases charge density in the nanocomposite molecules at electrodes. Although, The effect of raising concentration of TiO_2 nanofillers in polyvinyl chloride increases its dielectric constant as shown in table I, the charging density at the cathode is more than charging density at the anode with increasing TiO_2 nanoparticles percentage and impulse applied voltages; especially through 5%wt specimens and there is hetero-charge accumulated between electrodes as shown in Figure's (6, 7). Also, it is noticed that increasing TiO_2 nanoparticles from 5%wt up to 10%wt decreasing charge density in the nanocomposite molecules at electrodes. On the other wise, Figure's (8, 9) show the effect of raising concentration of Al_2O_3 nanofillers in polyvinyl chloride increases its dielectric constant as shown in table I, the charging density at the cathode is more than charging density at the anode with increasing Al_2O_3 nanoparticles percentage and impulse applied voltages; especially through 1%wt specimens and there is no hetero-charge accumulated between electrodes through 5%wt and 10%wt of aluminum oxide. Also, it is noticed that there is no increasing in charge density at both electrodes with remaining impulse voltages at cathode and there is no hetero-charge accumulated between anode and cathode.

6. Conclusion

- Adding ZnO nanofillers in polyvinyl chloride decreases its dielectric constant but, the charge density at both electrodes increases with increasing ZnO percentages up to 5%wt and impulse applied voltages, then; it increases hetero-charge accumulated at cathode. Whatever, adding TiO₂ nanofillers in polyvinyl chloride increases its dielectric constant and the charge density at both electrodes increases with increasing TiO₂ percentages up to 5%wt and impulse applied voltages, then; it increases hetero-charge accumulated between electrodes. On the otherwise, adding Al₂O₃ nanofillers in polyvinyl chloride increases its dielectric constant and the charging density at the cathode is more than charging density at the anode with increasing Al₂O₃ nanoparticles percentage and impulse applied voltages; especially through 1%wt specimens and there is no hetero-charge accumulated between electrodes through 5%wt and 10%wt of aluminum oxide. So, there is no increasing in charge density at both electrodes with remaining impulse voltages at cathode and there is no hetero-charge accumulated between anode and cathode.
- Type and percentage of nanoparticles are vital parameters in specifying electrical aging and the distribution of space charge density accumulation in the nanocomposite molecules between electrodes at DC electric fields.

7. Acknowledgements

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