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A study on space charge distribution in LDPE nanocomposites for future electric power applications

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ABSTRACT. *Nanotechnology techniques are used for changing properties of polymers to be more reliability in the future of electrical power applications. This paper presents the effect of small amounts of nanometer size fillers (Clay, or ZnO or* Al_2O_3*) that are homogeneously dispersed by only several weight percentages in low density polyethylene (LDPE) on space charge characterization. Pulsed electroacoustic (PEA) system used to study the changing in the accumulation conduction phenomena, the amount of stored charges, and the trap density distribution in new low density polyethylene nanocomposite insulating materials with depolarization current. It has been controlled in electric and dielectric characterization of the new low density polyethylene nanocomposite insulating materials for enhancing electrical power applications.*

RÉSUMÉ. *Les nanotechnologies sont utilisées pour changer les propriétés de polymères en vue d'augmenter leur fiabilité pour des nouvelles applications dans le domaine de la puissance électrique. Cet article présente l'effet sur les charges d'espace des petites quantités (quelques pourcentages en poids) de nanoparticules (Argile, ou ZnO ou Al2O3) dispersées d'une façon homogène dans une matrice de polyéthylène basse densité (LDPE). La méthode électro-acoustique pulsée (PEA) est utilisée pour étudier le changement dans les phénomènes de conduction, la quantité de charges d'espace, la distribution de la densité de pièges avec le courant de dépolarisation dans un nouveau isolant nano-composite à matrice polyéthylène basse densité. Cet isolant est caractérisé en vue d'améliorer les applications dans le domaine de la puissance électrique.*

KEYWORDS: *space charge, nanocomposite, PEA system, polymers, low density polyethylene, nanoparticles.*

MOTS-CLÉS: *charge d'espace, nano-composite, PEA, polymères, polyéthylène basse densité, nanoparticules.*

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1. Introduction

A nanocomposite polymer is composed of nanofillers which diameter is as small as a few tens to a few hundreds of nanometers, and a polymer as a matrix. As the sectional area of the interface between the filler and the matrix is enormously large compared to that of conventional microcomposite materials, some improvement of insulating performance is expected to be realized by using such technology as shown in researches (Matsui et al., 2005; Murakami *et al.,* 2008; Takada *et al*., 2008; Wang *et al*., 2008). Also, There may be mentioned the researchers of (Tanaka *et al.,* 2004; Cai *et al.,* 2008; Ishimoto *et al.,* 2009; Jiandong *et al.,* 2010; Tian *et al.,* 2012; Thabet, 2013a; 2013b; Gouda *et al.,* 2014a; 2014b; Thabet, 2015a) to be compared with traditional materials, nanocomposite materials have a set of special characteristics, such as surface effect, small size effect, quantum size effect, macroscopic quantum tunneling effect and dielectric confinement effect. Attracted by these outstanding characteristics, a large number of researchers in the field of dielectrics and electric insulation have attempted to add nanofillers into polymers in order to improve dielectric and insulating characteristics of polymers. It has already been found that nanoparticles can improve some key properties of polymers to a certain extent.

The effects of nanoparticle additives on the electrical properties of common dielectrics are currently considerable interest. It is recognized that the interfaces between the host dielectric and the nanoparticles can strongly influence the dielectric properties of the composite material as a whole. This is not surprising when one compares the volume of the interface between a nanoparticle and the host with the combined (interface + particle) volume (See Fleming *et al.,* 2005; Lewis, 2004; 2005; Nelson *et al.,* 2004; 2005). It is recognized that the interfaces between the host dielectric and the nanoparticles can strongly influence the dielectric properties of the composite material as a whole. Thus, a given physical property may have an effective value for the composite differing considerably from that of either component, or not necessarily bounded by the component values as shown in researches of (Thabet, 2015b; Fleming *et al.,* 2008). Based on the traditional PEA space charge measurement system (See Wang *et al*., 2008; Li *et al*., 1994), Space charge distribution in samples was measured by the pulsed electro-acoustic (PEA) method at room temperature, (DC source 0-7 kV). The sample was sandwiched between aluminum electrodes. Silicone oil was used as an acoustic coupling agent in order to ensure good acoustic contact between the sample and the electrodes. The sample thickness was about 0.15- 0.4 mm.

The trapped space charge density is obtained from the depolarization current characteristics according to the following expression (See Das *et al.,* 2010),

$$\mathrm{q(t)} = \int_0^t \mathrm{i\,(t)dt} \tag{1}$$

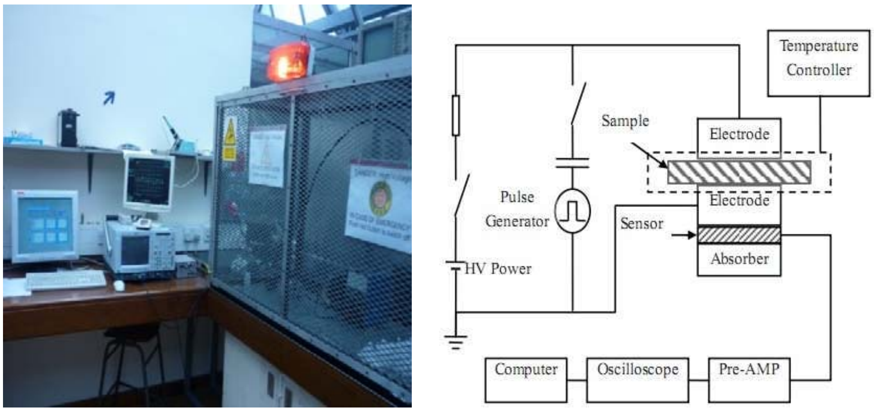
Where, i(t) is the depolarization current, q(t) the trapped charge density and t is time.

Polymer nanocomposites are defined as polymers in which small amounts (1-10%wt.) of nanometer-size fillers are homogeneously dispersed. The properties of space charge accumulation in low density polyethylene (LDPE)/ (Clay, or ZnO or Al_2O_3) nanocomposite film under high dc electric stress (up to 70kV/mm) have been experimentally investigated. The nanocomposite film contains dispersed (Clay, or ZnO or Al_2O_3) nanoparticles about 50 to 100 nm in diameter and 1 to 10%wt. in content. Thus, this research goes towards for concerning the effect of nanoparticles on space charge distribution in polymeric insulations. With a continual progress in Low density polyethylene polymer nanocomposites, the current research depicts that the effects of types and concentration of nanoparticles in new nanocomposite industrial polymer material. Space Charge Dynamics have been investigated and discussed experimentally for Low density polyethylene nanocomposites that use clay or aluminum oxide or zinc oxide nanoparticles separately under DC Stress.

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, 50Hz, 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. Nanoparticles are generally considered a discovery of modern science; they actually have a very long history. So that, Zinc oxide (ZnO) is an inorganic compound; it is widely used as an additive in numerous materials and products including plastics, ceramics, glass, cement, lubricants. Spherical clay nanoparticle shape is the most important characteristic of nano-clay for polymer applications. Cost less of clay catalyst to be the best filler among nanofillers industrial materials. Aluminum oxide (Al_2O_3) is commonly referred to as alumina; Alumina catalysis a variety of reactions that are useful industrially. Also, it serves as a catalyst support for many industrial catalysts.

Low-density polyethylene is a thermoplastic made from petroleum. Molecules of LDPE are less tightly packed and less crystalline because of the side branches, its density is lower. Low-density polyethylene contains the chemical elements carbon and hydrogen. Thus, SEM images illustrate penetration of nanoparticles in low-density polyethylene nanocomposite films as shown in Figure 2.

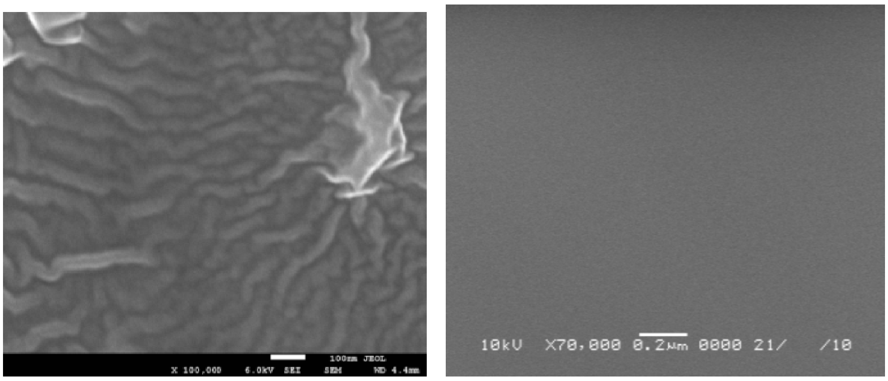


(a) Life photo for PEA system testing equipment's

(b) Schematic diagram of PEA system

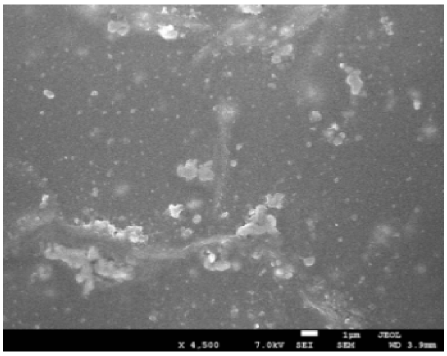
Figure 1. PEA system testing equipment's

HIOKI 3522-50 LCR Hi-tester device has been measured electrical parameters of nano-metric solid dielectric insulation specimens at various frequencies, therefore; it can be measured all dielectric properties for pure and nanocomposite industrial materials. The studied industrial materials here is low-density polyethylene nanocomposite films that have been formulated by using nanotechnology techniques and utilizing nanoparticles (Dia.: 10nm) of clay (montmorillonite-type) and ZnO, and Al_2O_3. The base of all these 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.



(a) Clay/LDPE nanocomposite film

(b) Al_2O_3/LDPE nanocomposite film



(c) ZnO/LDPE nanocomposite film

Figure 2. SEM images for low density polyethylene nanocomposite films

Table 1. Electric and Dielectric Properties for LDPE Nanocomposite films

Materials	Dielectric Constant at 1kHz	Resistivity (Ω.m)
Pure LDPE	2.3	10^{14}
LDPE + 1%wt Clay	2.23	10^{15}
LDPE + 5%wt Clay	1.99	10^{15}-10^{18}
LDPE + 10%wt Clay	1.76	10^{18}-10^{20}
LDPE + 1%wt ZnO	2.28	10^{14}
LDPE + 5%wt ZnO	2.27	10^{14} -10^{15}
LDPE + 10%wt ZnO	2.25	10^{15}
LDPE + 1%wt Al_2O_3	2.42	10^{13}
LDPE + 5%wt Al_2O_3	2.59	10^{13}-10^{11}
LDPE + 10%wt Al_2O_3	2.97	10^{11}-10^{9}

3. 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. All measurements show space charge distribution at various dc voltage stressed and variant period times. It 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.

3.1. Space charge distribution in pure low density polyethylene

Figure 3 shows the measurements that 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 that the charge density increases increasing impulse voltages and hetero-charge accumulated near the cathode.

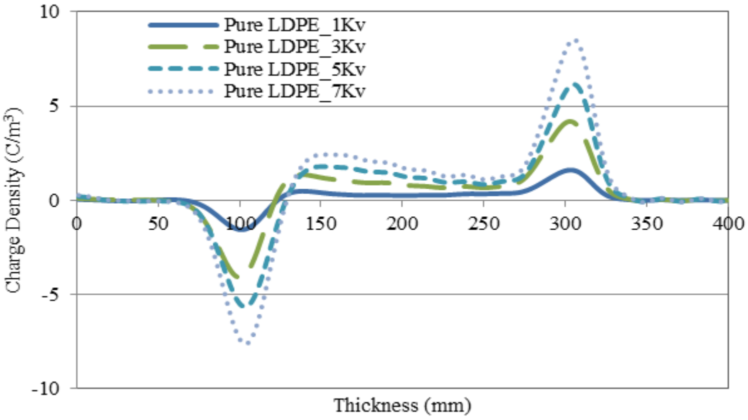


Figure 3. Space charge profiles for pure Low density polyethylene with varying DC electric field

3.2. Effect of zinc oxide nanoparticles on space charge distribution

Figure 4 shows space charge distribution relevant ZnO/LDPE nanocomposite films with 1%wt percentage of zinc oxide nanoparticles under varying dc electric fields; the charge density at both electrodes increases with increasing impulse applied voltages and there is high hetero-charge accumulated at cathode.

However, Figure 5 contrasts space charge distribution relevant ZnO/LDPE nanocomposite films with 5%wt percentage of zinc oxide nanoparticles under varying dc electric fields the charge density at both electrodes increases with increasing impulse applied voltages and there is high hetero-charge accumulated between electrodes. Figure 6 shows space charge distribution relevant ZnO/LDPE nanocomposite films with 10%wt. percentage of zinc oxide nanoparticles under varying dc electric fields; the charge density at both electrodes increases with increasing impulse applied voltages and there is high hetero-charge accumulated at cathode. Increasing zinc oxide nanoparticles from 5%wt up to 10%wt decreases charge density in the nanocomposite molecules.

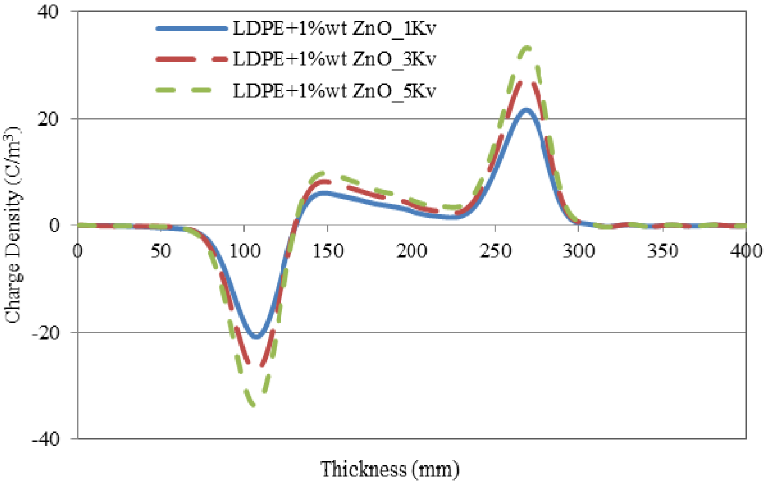


Figure 4. Space charge profiles for 1%wt. ZnO /LDPE nanocomposite films

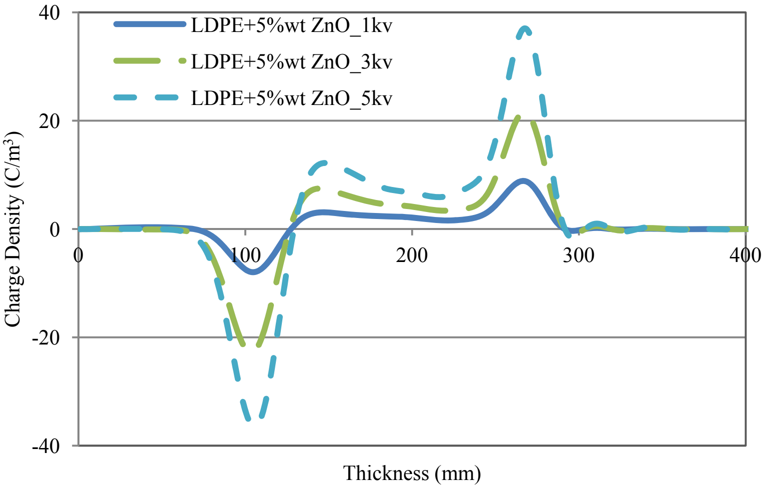


Figure 5. Space charge profiles for 5%wt. ZnO/LDPE nanocomposite

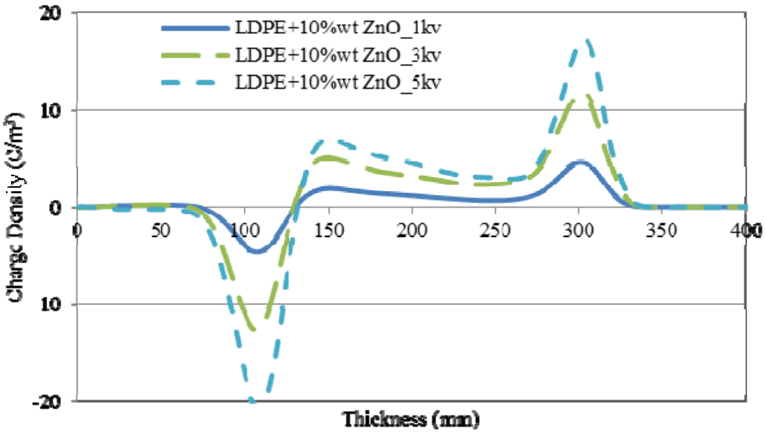


Figure 6. Space charge profiles for 10%wt. ZnO/LDPE nanocomposite films

3.3. Effect of clay nanoparticles on space charge distribution

Figure 7 shows space charge distributions relevant to Clay/LDPE nanocomposite films with 1%wt. percentage of clay nanoparticles under various dc electric fields.

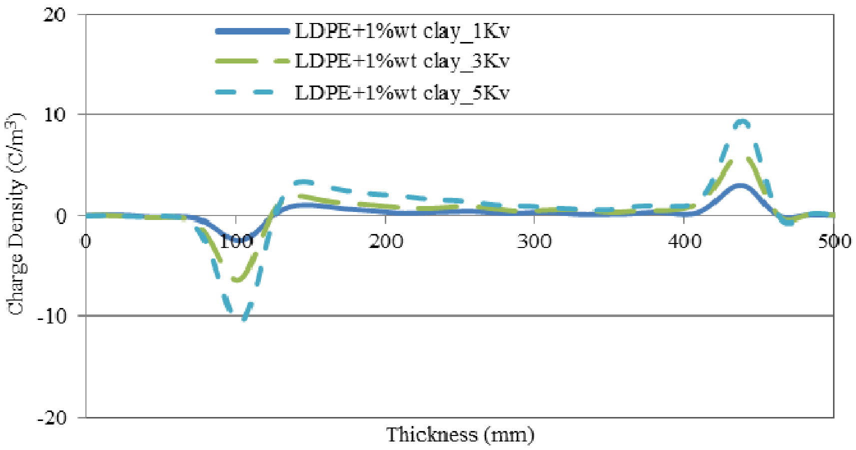


Figure 7. Space charge profiles for 1%wt. Clay / LDPE nanocomposite films

It is noticed that charging density at the anode is more than charging density at the cathode with increasing impulse applied voltages and there is a hetero-charge accumulated near the cathode.

However, Figure 8 shows space charge distributions relevant to Clay/LDPE nanocomposite films with 5%wt percentage of clay nanoparticles under various dc electric fields. The measurements imply the injection of impulse high voltages that depicts charge density at the anode and cathode is increased and there are hetero-charge accumulated charges between the both electrodes. It is cleared that increasing clay nanoparticles from 1%wt up to 5%wt increases charge density in the nanocomposite molecules.

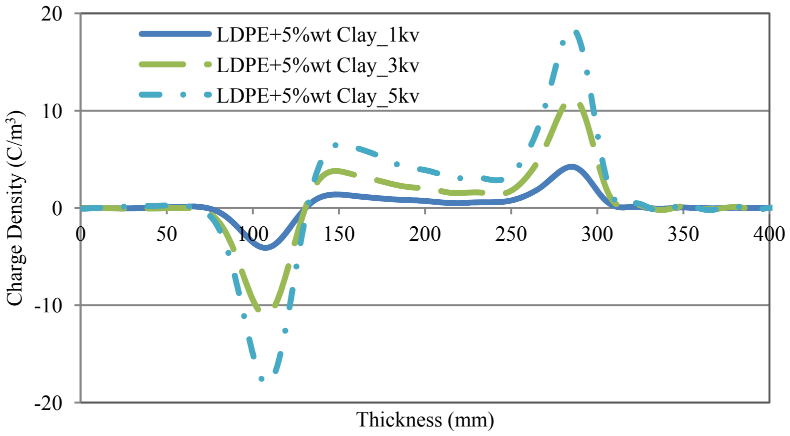


Figure 8. Space charge profiles for 5%wt. Clay/LDPE nanocomposite films

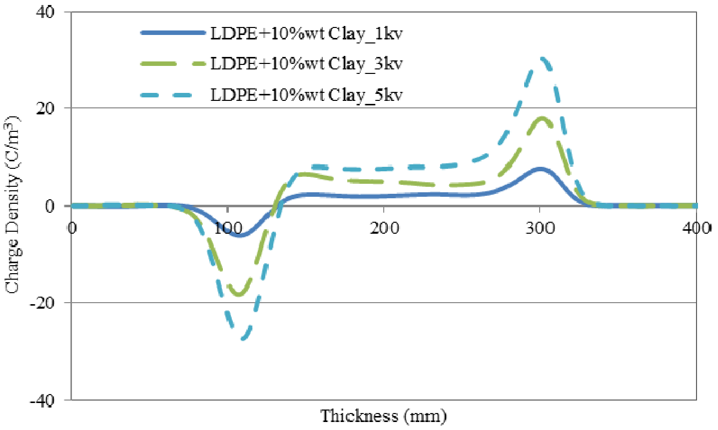


Figure 9. Space charge profiles for 10%wt. Clay LDPE nanocomposite films

Figure 9 shows space charge distributions relevant to Clay/LDPE nanocomposite films with 10%wt percentage of clay nanoparticles under various dc electric fields. The measurements imply the injection of impulse high voltages that depicts charge density at the anode and cathode is increased and there are homo-charge accumulated charges between the anode or cathode electrodes. It is noticed that increasing Clay nanoparticles from 5%wt up to 10%wt increases charge density in the nanocomposite molecules between electrodes.

3.4. Effect of aluminum oxide nanoparticles on space charge distribution

Figure 10 shows space charge distribution relevant Al_2O_3/LDPE nanocomposite films with 1%wt. percentage of aluminum oxide nanoparticles under varying dc electric fields; the charge density at both electrodes increases with increasing impulse applied voltages and there is high hetero-charge accumulated between anode and cathode. Also, Figure 11 contrasts space charge distributions relevant to Al_2O_3/LDPE nanocomposite films with 5%wt percentage of aluminum oxide nanoparticles under various dc electric fields. It is cleared that increasing clay nanoparticles from 1%wt up to 5%wt. increases charge density in the nanocomposite molecules, and so, the charge density at the anode and cathode is increased and there are hetero-charge accumulated charges between the both electrodes.

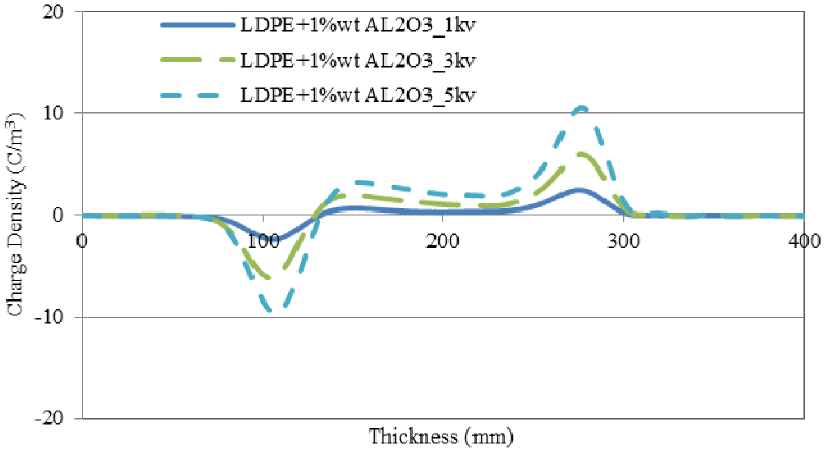


Figure 10. Space charge profiles for 1%wt. Al_2O_3 / LDPE nanocomposite films

Figure 12 shows space charge distribution relevant Al_2O_3/LDPE nanocomposite films with 10%wt percentage of aluminum oxide nanoparticles under varying dc electric fields; the charge density at both electrodes increases with increasing impulse applied voltages and there is high hetero-charge accumulated between anode and cathode. It is noticed that increasing Al_2O_3 nanoparticles from 5%wt. up to 10%wt. increases charge density in the nanocomposite molecules at both electrodes.

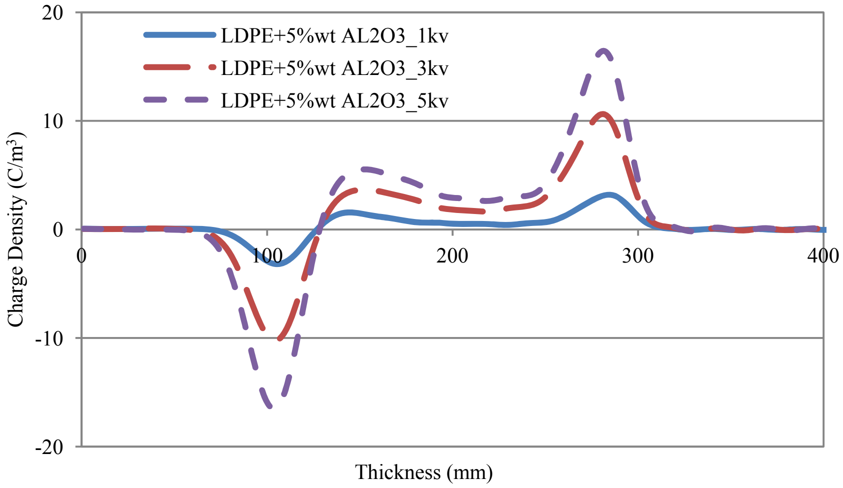


Figure 11. Space charge profiles for 5%wt. Al_2O_3 / LDPE nanocomposite films

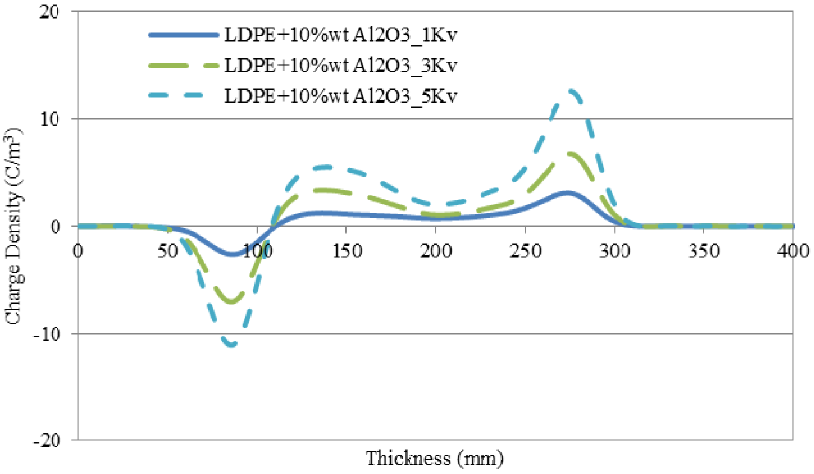


Figure 12. Space charge profiles for 10%wt Al_2O_3 / LDPE nanocomposite

4. Trends of nanoparticles for space charge distribution

First, it is an importance stage to be sure from homogeneity penetration of nanofillers in base matrix polymer (LDPE), as shown in Figure 2. **Second,** the effect of increasing concentration of ZnO, Clay and Al_2O_3 nanofillers in Low density polyethylene changes the dielectric constant properties as shown in Table 1. **Third,** it has been studied the effects of nanoparticles on the behavior of space charge accumulation in pure and nanocomposites with respect to impulse voltages and accumulated charges near the poles. Thus, Figure's (4-6) 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. Noting that, Figure's (7-9) depicts charging density at the cathode is more than charging density at the anode with increasing Clay nanoparticles percentage and impulse applied voltages; especially through 5%wt specimens and there is hetero-charge accumulated between both electrodes. Whatever, increasing Clay nanoparticles from 5%wt up to 10%wt increases charge density in the nanocomposite molecules at both electrodes. Although, The effect of raising concentration of Al_2O_3 nanofillers in Low density polyethylene increases its dielectric constant as shown in Table I; whatever as shown in Figure's (10-12), 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 5%wt specimens and there is hetero-charge accumulated at electrodes. Also, it is noticed that increasing Al_2O_3 nanoparticles from 5%wt up to 10%wt decreasing charge density in the nanocomposite molecules at electrodes.

5. Conclusions

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. Therefore, adding Clay and ZnO nanofillers in Polyvinyl Chloride decreases its dielectric constant, therefore; ZnO is more effective than Clay nanoparticles up to 5%wt in increasing the charge density at both electrodes with increasing DC impulse applied voltages and so increases hetero-charge accumulated electrodes. Whatever, adding Al_2O_3 nanofillers in Polyvinyl Chloride increases its dielectric constant and the charge density at both electrodes increases with increasing Al_2O_3 percentages up to 5%wt and impulse applied voltages, then; it increases hetero-charge accumulated between electrodes.

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