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Effect of ZnO Nanofiller in the XLPE Matrix on Electrical Tree Characteristics

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ABSTRACT The effect of nanofillers on the electrical tree behavior of polymeric materials, such as epoxy resin and polyethylene, has been investigated. Thus, research on electrical tree behaviour is extended but limited only to pure crosslinked polyethylene (XLPE) cable. Electrical tree properties are yet to be explored under various types of nanofillers in XLPE. In this paper, the electrical tree characteristics of XLPE containing zinc oxide (ZnO) nanofiller was reported. The nanofiller concentrations in XLPE were varied to 0.5, 1.0, and 1.5 wt%. Needle-plane electrodes were employed with 2 mm gap between the electrodes. A digital camera and microscope system were used to observe the structure and growth of electrical tree in XLPE. The tree inception voltage (TIV) of XLPE increased with the addition of ZnO nanofiller. The propagation length of tree growth improved compared with the unfilled XLPE. The TIV for the samples with 0.5, 1.0, and 1.5 wt% ZnO nanofiller improved by 7.69%, 14.65%, and 7.16%, respectively, relative to the unfilled XLPE.

INDEX TERMS Nanocomposite, tree inception voltage.

I. INTRODUCTION

Crosslinked polyethylene (XLPE) is a polymeric material that has been extensively applied as insulator in high-voltage cables owing to its excellent dielectric properties, high flexibility and mechanical strength, good resistance to chemicals, low cost and easy processing [1]–[3]. Nevertheless, the use of XLPE as an insulating material in underground and distribution cables is prone to unexpected weather and contaminations, leading to insulation breakdown.

The main failure mechanisms for high-voltage XLPE cable insulation is electrical treeing. When the tree instigates in a cable accessory, it grows through the insulation and then triggers cable failure [4]. Several methods have been attempted to restrain electrical tree development and improve the electrical tree resistance in high-voltage cable accessories. Such methods include improving material processing, adding treeing inhibitors, and altering the material composition [5].

With nanotechnology development, polymer composites containing nano-sized fillers exhibit great potential to enhance mechanical, electrical, and thermal properties without the need to modify polymer compositions and processing compared with neat polymers [6]–[9]. Improvement on electrical tree resistance in the polymer matrix (epoxy resin, polyethylene) containing nanofillers has been reported by few researchers [5], [6]. Furthermore, investigation on the electrical tree behavior is extended but limited to LDPE/alumina nanocomposite, XLPE/silica nanocomposite, and unfilled XLPE [7]–[12]. The effect of other nanofillers on XLPE composite is uncertain and should be investigated. In the present work, electrical tree behavior in XLPE containing ZnO nanofiller was investigated. The characteristics of XLPE electrical tree with different ZnO nanofiller concentrations of 0.5, 1, and 1.5 wt% were determined. Results of TIV, electrical tree growth, electrical tree length, and expansion

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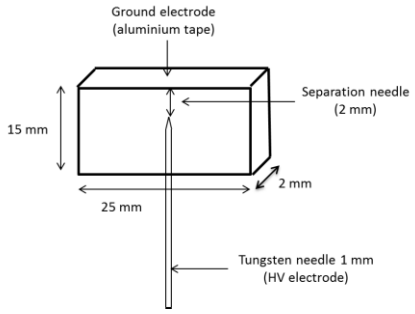


FIGURE 1. Diagram of rectangular slabs.

coefficient were discussed and compared with those of unfilled XLPE.

II. EXPERIMENTAL PROCEDURE

A. SAMPLE PREPARATION

XLPE containing 0.5, 1, and 1.5 wt% ZnO nanofillers were prepared as rectangular slabs with the dimension of 15 mm × 25mm × 2mm. The concentration was selected based on reports in previous research [13]–[15]. Figure 1 shows the diagram of rectangular slabs. The experiment used needle-plane electrodes. The tungsten needle electrode with 1 mm diameter, tip radius of 5 ± 1 μm, and tip angle of 30° was slowly inserted into each XLPE nanocomposite. Aluminum tape was used as a plane electrode, and the gap of needle-plane electrodes was 2 mm. Sample preparation was carried out in accordance to the method of Chen *et al.* [11] and was strictly controlled to prevent any mechanical stress and void formation in the needle tip region.

B. ELECTRICAL TREEING TEST

The schematic diagram of electrical treeing experimental setup is depicted in Figure 2. The set up consisted of a 50Hz 240 V/100 kV HV transformer, 10 MΩ limiting resistor, and 1000:1 capacitive divider. A step-by-step voltage of 1 kV with step duration of 60 s was utilized until electrical tree inception (TIV) was observed. The characteristics of electrical tree growth were investigated by applying voltage fixed at TIV for each specimen. Electrical tree growth was analyzed from the needle tip until the electrical tree reached 1 mm length. In addition, treeing images were recorded periodically using a real-time microscope digital imaging system comprising Leica microscope with a cooled light source to provide either transmitted or reflected illumination, a CCD camera, and a personal computer. The prepared sample was placed into the test cell, where the needle electrode was connected to the HV source, while the plane electrode was connected to the grounding. The test cell was filled with clean mineral oil to avoid external discharge and flashover during testing. Electrical treeing was observed by placing the test cell under microscope. Using this system, the treeing process was clearly observed and recorded in real-time without disturbance from the applied voltage stress.

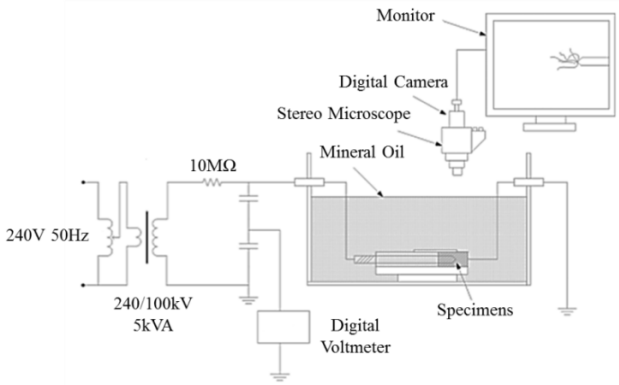


FIGURE 2. Electrical treeing experimental setup schematic diagram.

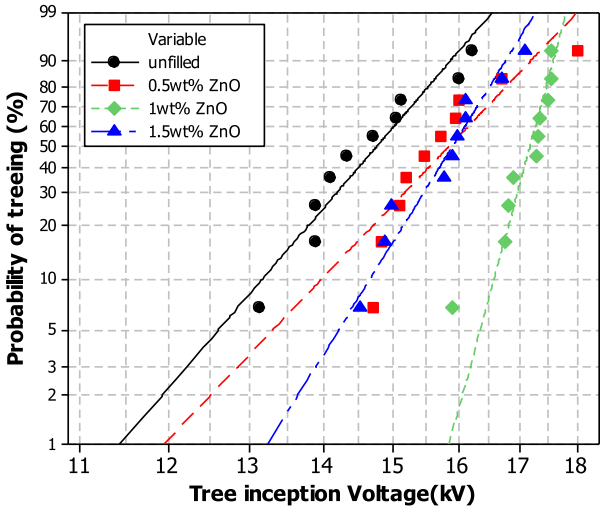


FIGURE 3. Plot of Weibull distribution for unfilled XLPE and XLPE with 0.5, 1.0, and 1.5 wt% ZnO nanofillers.

III. EXPERIMENTAL RESULTS

A. TREE INCEPTION VOLTAGE

Figure 3 shows the Weibull probability distribution of TIV for the unfilled XLPE and XLPE filled with various ZnO nanofiller concentrations. Based on the Weibull probability plot, the TIV probability was determined to be 63.2%. Table 1 shows the parameters for TIV for each sample. The scale parameter (h) and shape (b) denote the TIV characteristics of the samples at the failure probability of 0.6321 and scattering TIV values, respectively.

Figure 3 and Table 1 show that the addition of 0.5 wt% and 1.0 wt% ZnO nanofiller to XLPE slightly increased the TIV by 7.69% and 14.65%. However, further addition of ZnO decreased the TIV of the nanocomposite, but the value was still slightly higher than that of the unfilled XPLE.

B. ELECTRICAL TREE GROWTH

Figure 4 shows the typical image of electrical tree growth in unfilled XLPE and XLPE filled with 0.5, 1.0, and 1.5 wt% ZnO nanofillers. The images of electric tree growth at the TIV were captured until it reached the length of 1 mm from

TABLE 1. Weibull parameter of tree inception voltage for each sample.

Sample	Weibull parameter	
	Shape, *b*	Scale, *h* (kV)
Unfilled	16.61	15.09
0.5 wt% ZnO	14.94	16.25
1.0 wt% ZnO	52.76	17.30
1.5 wt% ZnO	23.08	16.17

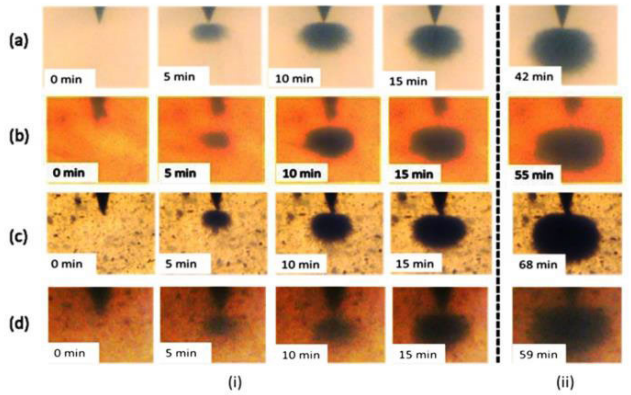


FIGURE 4. (i) Image of electrical tree growth for (a) unfilled XPLE and XPLE filled with (b) 0.5wt% ZnO, (c) 1.0wt% ZnO, and (d) 1.5 wt% ZnO; (ii) Image of electrical tree growth at 1 mm length after the TIV.

the needle tip. The result showed a bush-type tree appearing regularly in the unfilled XLPE and XLPE filled with ZnO at all concentrations. At 0 min, more than one main channel appeared at the needle tip. After 5 min, the electrical tree grew vertically and horizontally from one of the main channels and formed an elliptical dense tree structure. The dense bush type tree propagated slowly through the interval gap to the grounded electrode. After 10 and 15 min, the size of the bush-type tree increased and the electrical tree density intensified. Figure 4 (ii) illustrates the electrical tree image reaching 1 mm length after the TIV.

The electrical tree growth of the unfilled XPLE reached 1 mm length faster than that of XLPE filled with ZnO nanofiller. The addition of 0.5, 1.0, and 1.5 wt% ZnO nanofiller inhibited the rapid growth of the electrical tree, thereby increasing the time of electrical tree growth to 55, 68, and 59 min, respectively, compared with 42 min for the unfilled XLPE to reach 1 mm length from the needle tip.

Figure 5 shows the relationship between electrical tree propagation length and time for the unfilled XLPE and XLPE filled with ZnO nanofillers. The time was recorded from tree inception for 25 min duration. The results showed that XLPE containing 1.0 wt% ZnO had shorter electrical tree propagation within 25 min duration compared with the unfilled sample and those filled with other concentrations.

C. EXPANSION COEFFICIENT

The electrical tree growth behavior was described through a plot of expansion coefficient (D/L) against electrical tree time. D denotes the maximum width of electrical tree growth,

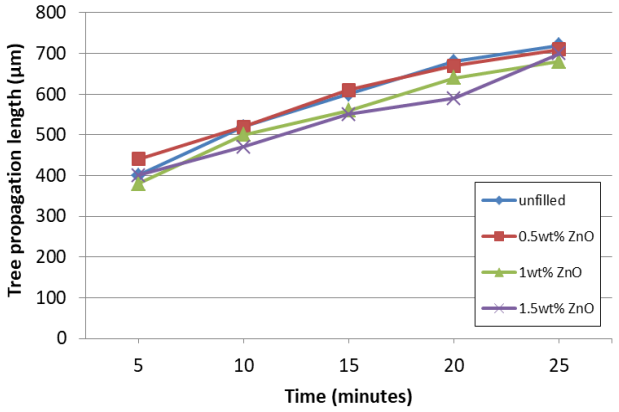


FIGURE 5. Relationship of tree length as a function of time for unfilled XLPE and XLPE filled with ZnO nanocomposites.

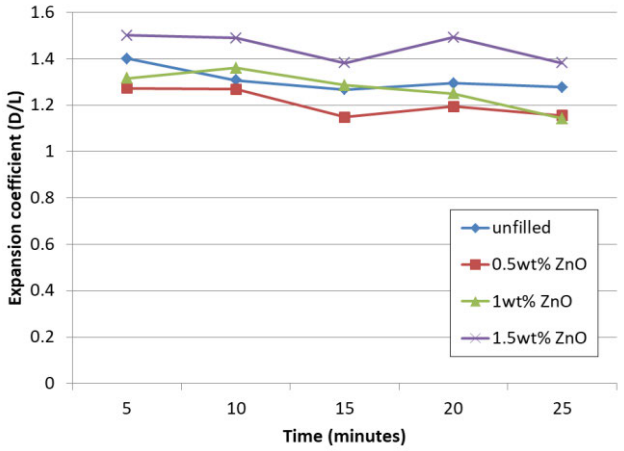


FIGURE 6. Relationship between expansion coefficient and treeing time for the unfilled XLPE and XLPE with ZnO.

and L denotes the maximum electrical tree length. Figure 6 shows the comparison of expansion coefficient characteristic within 25 min of electrical treeing for XLPE with ZnO nanofiller and unfilled XLPE. The analysis of the electrical tree type structure was based on three treeing phases in XLPE cable insulation [4].

Overall, XLPE with 1.0wt% ZnO nanofiller had the optimum result given that the expansion coefficient depicted similar value for vertical and horizontal axis creating the bush-type tree. The addition of nanofillers seemed to improve the nanocomposite electrical tree growth.

D. FRACTURE SURFACE MORPHOLOGY

The morphology of the fracture surface was analyzed using field emission-scanning electron microscope (FE-SEM). Figure 7 shows the SEM micrographs of microstructure of XLPE nanocomposites observed at 1000X magnification. At a high filler concentration of 1.5 wt% (Figure 7c), filler agglomeration was observed. The agglomerated filler exhibited poor interfacial adhesion, thereby reducing the efficiency of stress transfer. Moreover, the formation of large voids was expected at the interface or trapped in a cluster of fillers in

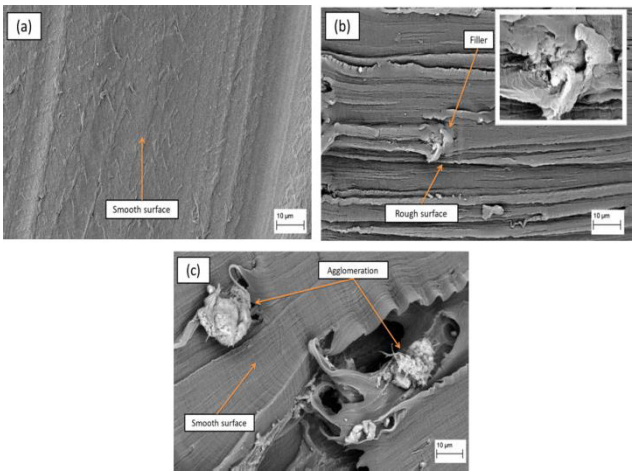


FIGURE 7. Fracture surface morphology of XLPE nanocomposites of (a) XLPE; (b) ZnO(1.0)/XLPE, and (c) ZnO(1.5)/XLPE.

high filler loading nanocomposites compared with that in low filler loading nanocomposites.

IV. DISCUSSION

The TIV result for the unfilled XLPE and XLPE filled with ZnO nanofillers can be explained according to the percolation model for electrical breakdown in insulating polymers reported by Wu *et al.* [16]. Nanofiller added in the polymer reacted as co-monomer, which can introduce new traps with high barriers. These barriers may lead to high critical field for percolation path formation, thereby increasing the TIV of the nanocomposite. The addition of ZnO nanofiller up to 1.0 wt% provided high barrier in the nanocomposite and thus required the high intensity of electric field to eliminate them as traps. Consequently, high voltage was required for the tree inception. The electrical behavior in the XLPE nanocomposites was further described by considering band theory in solid. Table 2 shows the value of band gap energy for ZnO nanofiller and XLPE composite measured by absorbance method using ultraviolet absorption spectra (UV-ViS). The band gap energy values for ZnO nanofiller and XLPE composite were 3.2 and 8.5 eV, respectively. With the band gap energy, the Fermi level for the base composite and nanofiller was determined using the following equation [17]:

$$E_c - E_f = kT \ln (N_c/n) \tag{1.1}$$

where E_c and E_f are energy at conduction band and Fermi level, respectively, k is the Boltzman constant, T is the room temperature, N_c is the effective density of states at conduction band, and n is the number of electrons. The position of Fermi level for ZnO nanofiller obtained from the calculation was 0.094 eV, which is below E_c.

The TIV was slightly reduced for 1.5wt% ZnO nanofiller compared with that of 1.0wt% ZnO nanofiller but still higher than the unfilled XLPE. This finding might be due to the filler dispersion, agglomeration, and overlapping, which increased the trap region density. Interaction occurred between loosely

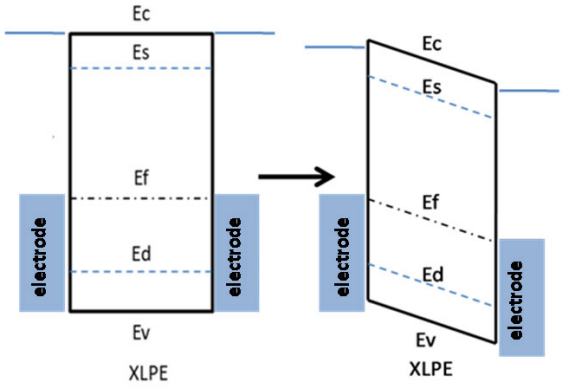
TABLE 2. Band gap energy of ZnO and XLPE.

Material	Band Gap energy (eV)
ZnO nanofiller	3.2
XLPE composite	8.5

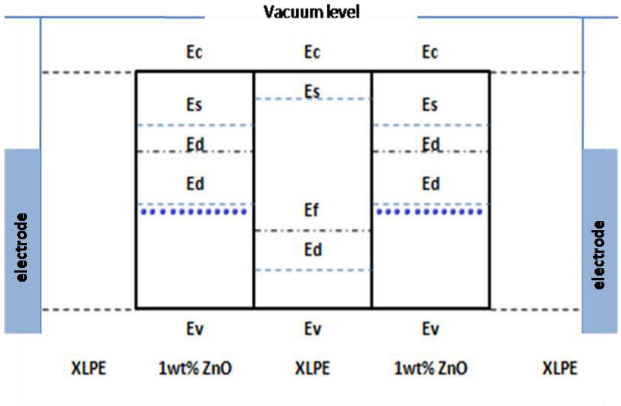
and tightly bound regions among the nanoparticles due to increment of trap density. Such interaction formed a conductive path region for the charge carriers, thereby reducing the critical field in the XLPE nanocomposite and the TIV result.

Given that ZnO nanofiller is a semiconductor, the position of Fermi level is near the conduction band; while for XLPE, the position is near the valence band. Figure 8 illustrates the band distribution of pure XLPE and XLPE nancomposites. The charge carriers from the needle electrode as well as free electrons in the polymer matrix under applied electric field required sufficient energy to accelerate across the band gap. The unfilled XLPE band was bent when the electric field was applied (Figure 8a). In pure XLPE composite, no blocking barrier existed, thereby enabling the charge carrier and free electrons to cross the band gap from the valence band to the conduction band, thereby initiating the growth of electrical tree in the polymer matrix. The first free electron travel across the band gap under the applied voltage reflected the tree inception voltage.

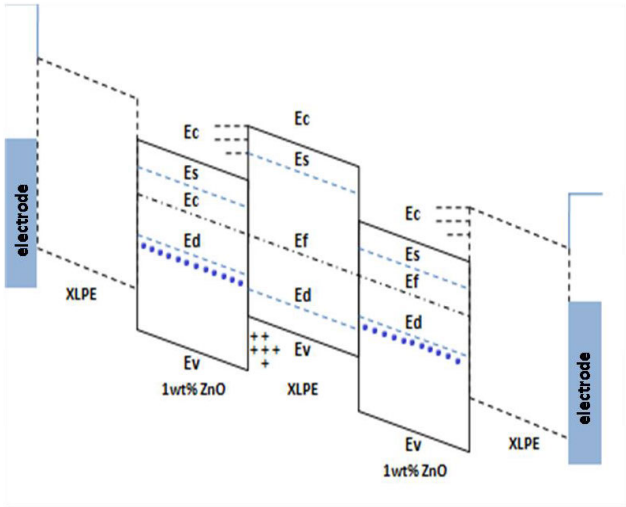
Figure 8(b) and 8(c) depicts the band distribution of XLPE/ZnO nanocomposite in cases without and with electric field application, respectively. The homogeneous dispersion of ZnO nanofiller in XLPE acted as an inhibitor to electrical tree inception voltage and electrical tree growth. The ZnO nanofiller in the XLPE composite introduced carrier traps. According to band theory, the band was bent when the electric field was applied [Figure 8(c)]. The respective Fermi levels of ZnO nanofiller and XLPE composite were aligned under the effect of EF to the same level of the Femi-level of the electrode. The conduction bands of the ZnO nanofiller at the nanocomposite interface and XLPE close to the grounding electrode become lower than that of XLPE, forming a blocking barrier for the injected electrons. At similar interface, the valence band also formed a blocking barrier for the holes. The band of holes trapped in the nanocomposite was lower than that in the XLPE composite. This phenomenon initiated the deep trap of free electrons in the XLPE nanocomposite. Furthermore, the conduction band of the nanocomposites became lower than that of XLPE at the other nanocomposite interface and XLPE close to the needle electrode; thus, no blocking barrier was formed. However, at the interface between the nanocomposite and needle electrode, another barrier for electrons and holes was formed. By considering this theory, the addition of ZnO nanofiller in XLPE introduced the deep trap of free electron in the band of holes trap. Consequently, high critical energy was required in the insulation gap to mobilize charge carriers from the needle electrode and free electrons in the polymer matrix through the insulation gap. Thus, the addition of ZnO nanofillers to



(a) Band distribution of unfilled XLPE before and after applied electric field



(b) Band distribution of nanocomposite before contact



(c) Band distribution of nanocomposite under a voltage

FIGURE 8. Band distribution of nancomposites (Ec: conduction band; Ev: valence band; Es: band of shallow trap; Ed: band of deep trap; Ef: Fermi level of insulation).

the XLPE composite indirectly elevated the energy level in the band gap for the electron to accelerate from the valance band to the conduction band, thereby increasing the TIV compared with that of the unfilled XLPE composite. In addition, the blocking barrier formed at the interface between the conduction band of the nanocomposite and XLPE near the grounding electrode and between the nanocomposite and needle electrode caused slow growth of electrical tree in XLPE containing ZnO nanofillers.

The TIV was slightly reduced for 1.5wt% ZnO nanofiller compared with that of 1.0 wt% ZnO nanofiller but still higher than the unfilled XLPE. This finding could be due to filler dispersion, agglomeration, and overlapping, which increased the trap region density. The interaction between the loosely and tightly bound regions among the nanoparticles occurred due to the increment of the trap density. The interaction between these bound regions formed a conductive path region for the charge carriers, thereby reducing the critical field in the XLPE nanocomposite and the TIV result.

The existence of the deep trap can be further explained by measuring depolarization current of dielectric. According to Simmons and Tam [18], in theory, when a dielectric with traps is introduced (experience polarization) and the source is removed at constant temperature, the discharge or depolarization current declines monotonically with time. From the depolarization current measurement as shown in Figure 9, it is apparent that the depolarization current of XLPE filled with ZnO nanofiller remains in the material after discharging process for some period of time. The depolarization current does not decrease with the same path of pure XLPE. The remaining current can be considered as a deep trap energized inside the nanocomposite. Under AC voltage, during the negative half-cycle, electrons are injected into the polymer as long as applied voltage is equal to or higher than the critical inception voltage. These electrons get trapped depending upon their energy. Some of these electrons recombine with holes, which were injected into the polymer during previous half-cycle, and remain trapped after polarity reversal. The site of recombination is referred to as the trapping centre. The process gives rise to light emission. When the polarity reverses, the injected species is holes and recombination occurs with electrons still residing in deep traps. This process is repeated during every half-cycle of the applied voltage [19]. The photoluminescence (PL) spectroscopy as a highly sensitive tool, it is a useful technique to represent electron trapped and hole trapped. Figure 10 shows the energy levels of nanocomposite for ZnO that represent the location of trap with the illustration based on analysis of interstitial, ZnO vacancy and oxygen vacancy defect on the nanocomposite.

However, further increase in filler concentration at 1.5 wt% ZnO nanocomposite in XLPE led to the nanoparticles interaction and agglomeration. The loosely bound region will overlap and yield a large conductive path region, and reduce the critical electric field. Under this condition, based on the band theory (see Figure 8c), the interaction at the barrier potential between the nanocomposite interface and XLPE may slightly reduce the barrier potential (reduction of critical electric field). This phenomenon allows the free electrons in the polymeric matrix to easily mobilize across the band gap to the conduction band, slightly lowering the TIV than that of 1.0wt% ZnO. The slight reduction of the barrier potential

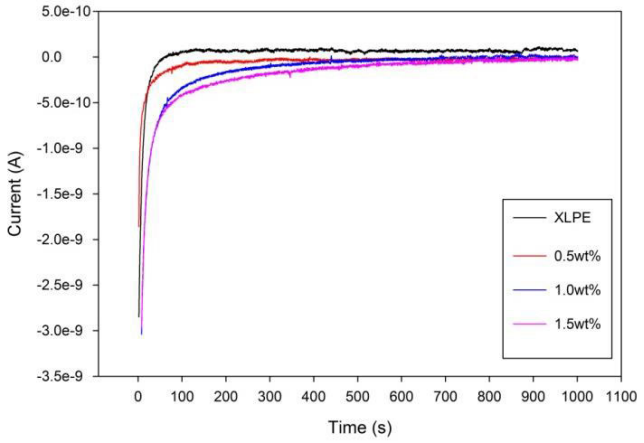


FIGURE 9. Depolarization current pattern for unfilled XLPE and XLPE with ZnO nanofiller.

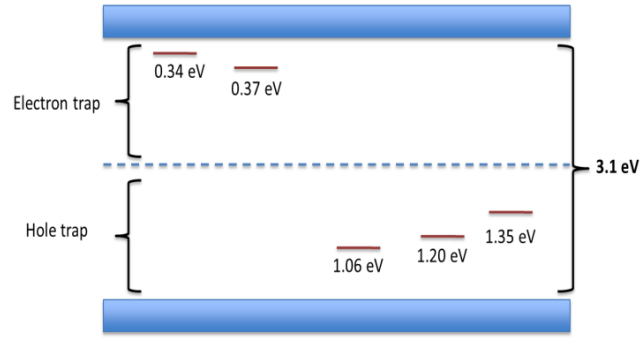


FIGURE 10. Energy levels of nanocomposite for ZnO that represent the location of trap.

may also affect the electrical tree growth to be slightly faster than that of 1.0wt% ZnO in XLPE.

From previous research, the trap density has been reported to increase as the nanofiller were added to the polymer matrix. It has been found that incorporation of a small amount of nanoparticles into polymers able to escalate the energy or deep traps density [20]–[22]. Polymer nanocomposite has been also found to exhibit better suppression for accumulation of space charge and higher breakdown strength compared to the neat polymer [23]–[26]. The relationship between traps and breakdown performance of polymer nanocomposites can be determined from the typical data of the energy and density of traps and breakdown strength that can be analyzed from many studies [23], [27]–[29]. The other reason for high electrical tree growth was related to space charges distribution of XLPE/ZnO nanocomposite as reported by Ramani *et al.* [30]. The average space charge for XLPE/ZnO was reported to become higher at 1.0wt% of ZnO nanofiller under the applied voltage. This is owing to the enhancement of charge injection when the voltage is increased. Hetero-charges are formed for XLPE and these charges can be diminished by adding the nanofillers. Since hetero charges are formed near the cathode, the electric field near the cathode is significantly enhanced, decreasing the potential barrier for electron injection. Hence, the injected holes are generally covered. As a result, XLPE/ZnO nanocomposite exhibits great ability in space charge suppression, which is significance for high voltage cable insulation development.

V. CONCLUSION

The effect of ZnO nanofiller in the polymeric matrix on electrical tree characteristic was investigated. The addition of ZnO nanofiller of 0.5 wt%, 1.0 wt%, and 1.5 wt% improved the tree characteristics by decelerating the electrical tree growth and increasing the TIV in the nanocomposite compared with that in the unfilled one. However, the addition of higher filler loading of 1.5 wt% ZnO nanofiller exhibits slight increase in the electrical tree growth propagation trend compared with the other filler concentrations possibly due to the filler agglomeration and large void formations at the interface or trapped regions.

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