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Effect of regular and core shell nano fillers on the partial discharge and tracking performance of low density polyethylene

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Abstract

Solid insulation materials are the backbone for all kinds of outdoor insulation applications. Environmental stresses degrade every outdoor insulation material with the passage of time. Internal partial discharges and external surface tracking are the two major factors governing the health of outdoor insulation. Best outdoor insulation is the one having lowest level of partial discharge and highest resistance to tracking. Improvement in tracking resistance of polymeric insulation materials has been a concern of many researchers. This study investigates the improvement in partial discharge and tracking resistance of LDPE under the effect of three different nano fillers including SiO$_2$, TiO$_2$, TiO$_2$@SiO$_2$. A very little has been reported on the effect of these nano fillers on the tracking performance of LDPE. TiO$_2$@SiO$_2$ is a novel core–shell nano filler. No study has reported the effect of these core–shell nano fillers on the partial discharge and tracking performance of LDPE. It has been observed in this study that core–shell nano fillers are much more efficient in improving partial discharge and tracking resistance of solid insulation materials than ordinary nano fillers.

1. Introduction

Partial discharge(PD) is a localized electrical discharge that only partially bridges the insulation between electrodes [1]. PD may be classified as internal and external discharges. External discharges include corona discharges and surface discharges. Internal partial discharges in solid and liquid insulation are caused by air voids (called cavity in case of solid and bubble in case of liquid) within the insulation.

External partial discharge on the surface of insulation material occurs where the tangential electric field is high enough to cause the progressive breakdown of insulation material [2]. Occurrence of PD affects the physical, electrical and mechanical properties of the insulation material. Progressive internal partial discharges form partially conducting channels known as trees. Most of the inorganic and organic dielectric materials are prone to partial discharges. Partial discharge monitoring is a good way to detect the insulation health levels. It also enables us to decide the schedules for preventive maintenance of solid and liquid insulation. Insulation designers always have a goal to reduce the partial discharge levels to minimum ever possible values. In Last 15 years there is a great interest of researchers to use different kinds of nano fillers (in some base dielectric materials) in order to achieve the lowest partial discharge magnitudes.

Nanocomposites (nano filler added polymers) have shown improvement in breakdown strength, mechanical strength, chemical resistance, reduced PD magnitude, high values of partial discharge inception voltage (PDIV) and partial discharge extinction voltage (PDEV) [3].

Zhe Li [4] studied the breakdown strength(BD) and PD resistance of micro and nano Alumina Al$_2$O$_3$ fillers added to epoxy resin and reported a decrease in breakdown strength and increase in partial discharge resistance of micro composites as compared to raw epoxy. The epoxy alumina nanocomposite demonstrated a PD magnitude of 280pC compared to raw epoxy which is 370 pC. Surface partial discharge characteristics of high density polyethylene (HDPE), low density polyethylene (LDPE) and polypropylene (PP) were investigated for...
three electrode arrangements [5]. HDPE showed least values of PD as compared to LDPE and PP. LDPE showed better PD resistance properties as compared to PP.

Dielectric strength, volume resistivity and permittivity of epoxy nanocomposites using TiO$_2$, ZnO and Al$_2$O$_3$ nano fillers were studied in [5]. TiO$_2$ filled in 0.2% and 0.5% in epoxy showed an increase in dielectric strength compared to raw epoxy. However, at 1% and 5% filler loading of TiO$_2$ the dielectric strength reduced compared to raw epoxy. The permittivity values for filler loading of up to 1% of ZnO were less than raw epoxy; whereas for filler loading of 5%, the permittivity was higher than raw epoxy. In general, TiO$_2$ and ZnO nanocomposites showed a decrease in volume resistivity value up to a certain filler loading and an increase beyond that point. The volume resistivity values for Al$_2$O$_3$ filler loading were high at filler loading up to 1% and then started to decrease.

Chemical resistance, flexibility, low DC conductivity, low cost and excellent insulating properties make Low Density Polyethylene (LDPE) ideal insulating material for underground distribution and transmission cables. The addition of small amounts of High Density Polyethylene (HDPE) to LDPE results in reduction of DC conductivity approximately 1 order of magnitude to about $10^{-15}$ S m$^{-1}$ at high electric fields of 30 to 40 kV mm$^{-1}$ and elevated temperature of 70 °C [7]. The addition of MgO nano fillers to LDPE matrix reduced the volume conductivity [8]. The lowest value of volume conductivity was observed for 3 wt% surface coated MgO nano fillers having value of $7 \times 10^{-16}$ S m$^{-1}$ as compared to $2 \times 10^{-13}$ S m$^{-1}$ of pristine LDPE polymer.

Montmorillonite (MMT)/SiO$_2$/LDPE multi element nanocomposites were examined for breakdown strength and electrical treeing resistance [9]. Results showed that (MMT)/SiO$_2$/LDPE multi element nanocomposites had improved breakdown strength of 116 kV mm$^{-1}$ compared to 108 kV mm$^{-1}$ for pure LDPE. The length of electrical tree growth in (MMT)/SiO$_2$/LDPE is 240 μm compared to 360 μm for pure LDPE at same voltage. LDPE/clay nanocomposites with different filler loadings were prepared to study the improvement in breakdown strength [10]. Breakdown strength increased from 1% − 5% filler loading and then decreased up to 15% filler loading. Highest value of breakdown strength for LDPE/clay nanocomposite was observed at 5% filler loading.

N.A Awang studied partial discharge performance of LDPE with plasma treated Boron Nitride (BN) nano fillers. LDPE/BN nanocomposites showed improved PD resistance compared to pure LDPE. PD magnitude of LDPE/BN nanocomposite was 3 pC compared to 23 pC for pure LDPE after 1 h of ageing [11].

PD resistance, PDIV and PDEV were investigated for LDPE/TiO$_2$ nanocomposites. LDPE/TiO$_2$ nanocomposite with 1% filler loading of TiO$_2$ showed least value of PD magnitude compared to pure LDPE [12]. PDIV and PDEV increased with increasing concentration of TiO$_2$ from 0.25% to 0.5% but decreased above 0.5%.

Surface, void and corona discharges of epoxy were investigated in [13]. The phase resolved partial discharge pattern (PRPD) of surface discharge revealed that the no of PDs, maximum charge magnitude and total charge per cycle increased with increase in voltage. PD magnitude of surface discharge was lower as compared to void discharges. PRPD of corona discharges depicted least PD magnitude as compared to surface and void discharges.

Yasuo Sekii evaluated degradation of LDPE by partial discharge in pure nitrogen (N$_2$) and mixture of nitrogen and oxygen (N$_2$ + O$_2$) gases. Atomic force microscopy (AFM) images showed formation of pits on the surface of LDPE specimens subjected to PD for 100 to 200 h. The depth of pits in the specimens increases with ageing time. Magnitude of pits for N$_2$ gas was approximately 420 nm as compared to 810 nm for N$_2$ + O$_2$ mixture. FTIR spectrum analysis detected presence of carbonyl groups in degraded LDPE specimens subjected to PD. With the increase in oxygen concentration in N$_2$ + O$_2$ mixture the magnitude of pits increased. Magnitude of pit for (N$_2$ + O$_2$) with 22% oxygen concentration was approximately 850 nm [14]. PD characteristics of LDPE samples were studied by addition of silica SiO$_2$ nano fillers. The no of PD pulses decreases with increasing nano filler loading. The no of PD pulses was 3900 for 4% and 750 for 8% SiO$_2$ filler loadings [15].

Breakdown and PD characteristics of LDPE in air/Nitrogen oxide (NO) environment with different humidity concentration were studied [16]. Breakdown time of 300 min in air and 130 min in air/NO environment for 75% humidity was reported. The PD magnitude for LDPE in air/NO atmosphere increased with increasing humidity.

The results of electrical tracking resistance and leakage current of Linear Low-Density Polyethylene with Natural Rubber (LLDPE/NR) nanocomposites with silicone oxide (SiO$_2$) and titanium dioxide (TiO$_2$) nano fillers were reported in [17]. It was clear that low leakage current means less development of carbon tracks.

Tracking resistance of raw XLPE and XLPE/ZnO nanocomposites was performed in [18] which revealed that virgin XLPE has better tracking resistance than XLPE/ZnO nanocomposite. The tracking time decreased with increasing filler concentration. The tracking time was 27 min at 4% ZnO filler loading compared to 45 min for raw XLPE.
The electrical tracking performance of different blends of linear low-density polyethylene (LLDPE) with natural rubber (LLDPE/NR) with and without alumina trihydrate (ATH) nano fillers was studied. Results show that a blend of 80% LLDPE, 20% NR without ATH has least values of leakage current, carbon track formation and degradation index as compared to LLDPE/NR blends with ATH. No study has reported the effect of core–shell nano particles on the tracking and partial discharge performance of LDPE. This study focuses on investigating the partial discharge and tracking performance of LDPE nanocomposites when used in concealed environments as an alternative to existing underground cable insulation materials under the effect of core–shell nano particles. The core–shell nano particles of titanium dioxide core with silicon dioxide outer shell (TiO$_2$@SiO$_2$) was used. In addition, ordinary silicon dioxide (SiO$_2$), titanium dioxide (TiO$_2$) nano fillers were also investigated in order to show their comparative performance w.r.t core–shell nano filler.

2. Materials and methodology

2.1. Materials
Low Density Polyethylene was used as a base polymer. LDPE used in this study has a melting point of 105 $^\circ$C and density of 0.922 g cm$^{-3}$. Nano fillers used include silicon dioxide (SiO$_2$), titanium dioxide (TiO$_2$) and titanium dioxide coated with silicon dioxide (TiO$_2$@SiO$_2$). The individual physical properties of these nano fillers are listed in table 1. All these nano fillers were dispersed into the base material in different percentages shown in table 2.

| Table 1. Physical properties of nano fillers. |
|---------------------------------------------|
| Properties | SiO$_2$ | TiO$_2$ | TiO$_2$@SiO$_2$ |
| Particle Size | 50–70 nm | 50–70 nm | 20–40 nm |
| True Density (g cm$^{-3}$) | 2.4 | 4.23 | 4.23 |
| Specific Surface Area (m$^2$ g$^{-1}$) | 180–600 | 35–60 | 40–50 |
| Purity (%) | 99.5 | 99 | 99.5 |
| Melting Point ($^\circ$C) | 1713 | 1843 | 2790 |
| Boiling Point ($^\circ$C) | 2950 | 2972 | 1840 |
| pH | 3.7–4.7 | 5.5–6 | 4.6–5.5 |

| Table 2. Nano fillers concentration. |
|--------------------------------------|
| NanoComposite | % of Nano Filler Loading |
|----------------|--------------------------|
| LDPE/SiO$_2$  | L$_{S1}$ L$_{S2}$ L$_{S3}$ L$_{S4}$ L$_{S5}$ |
| LDPE/TiO$_2$  | L$_{T1}$ L$_{T2}$ L$_{T3}$ L$_{T4}$ L$_{T5}$ |
| LDPE/TiO$_2$@SiO$_2$ | L$_{TS1}$ L$_{TS2}$ L$_{TS3}$ L$_{TS4}$ L$_{TS5}$ |

The electrical tracking performance of different blends of linear low-density polyethylene (LLDPE) with natural rubber (LLDPE/NR) with and without alumina trihydrate (ATH) nano fillers was studied. Results show that a blend of 80% LLDPE, 20% NR without ATH has least values of leakage current, carbon track formation and degradation index as compared to LLDPE/NR blends with ATH.

2.2. Materials preparation
The blending of LDPE and nano fillers was done using direct dispersion method (solution mixing method). Direct dispersion can be achieved using melt mixing [20, 21] or solution mixing [22, 23]. Initially, all the nano fillers used in this study were heated at 100 $^\circ$C for 5 min to remove any residual moisture. The nano fillers were then added in weighed quantity of ethyl acetate and mixed by the magnetic stirrer for 15 min and then ultrasonicated for 20 min. Weighed quantity of LDPE base matrix was then added in the ethyl acetate/nano filler solution, ultrasonicated for 20 min and then magnetically stirred for 15 min at 60 $^\circ$C and 700 rpm. Ethyl acetate gradually evaporated from the solution, leaving behind LDPE nanocomposites blend with nano fillers in LDPE base matrix. The LDPE nanocomposite blend was then placed in stainless steel molds and heated at 125 $^\circ$C in an oven for 20 min. The samples were then left for curing in the oven at 30 $^\circ$C for 20 min. Two different groups of samples were prepared with different sizes. The first group samples were circular with a dia of 60 mm and thickness of 2 mm. The second group samples were rectangular with dimensions (120 × 50 × 6 mm). Circular samples were used for partial discharge analysis according to IEC 60270 standard and rectangular samples were used for tracking analysis according to IEC 60587 standard. The details of the prepared nanocomposites for different filler loading are listed in table 2.
2.3. Methods of analysis
Partial discharge characterization and tracking performance analysis was performed. The details of the test setups are given below:

2.3.1. Partial Discharge measurement setup
To investigate PD characteristics (Partial Discharge Apparent Magnitude, Partial Discharge Inception and Partial Discharge Extinction voltages), high voltage AC 20 kV was applied to the test object using plane rod configuration. PD characteristics were recorded according to IEC 60270 standard \[1\]. The control desk takes input from the 230 volts, 50 Hz AC supply and regulated output is generated which is fed to the high voltage transformer. The output generated by high voltage transformer is measured by measuring capacitor and given back to control desk. High voltage output from HV transformer is given to the test object and coupling capacitor connected in parallel with it. Any PD occurring in the test object generates a high frequency signal which is passed by the coupling capacitor to the coupling quadripole. Output of the coupling quadripole is given to the PD meter which calculates its magnitude, frequency and phase resolved pattern. The PD setup is shown in figure 1.

2.3.2. Tracking analysis setup
The tracking analysis was carried out in accordance with IEC 60587 standard \[24\]. The scheme of the setup is shown in the figure 2 \[25\]. For tracking all the samples were fixed at an inclined plane of 45° having a distance of 50 mm between the electrodes. Contaminated solution was prepared by dispersing 0.1% of wetting agent and 0.002% NH₄Cl in de-ionized water. The conductivity of contaminant solution was 2500 $\mu$S cm$^{-1}$. The flow rate of 0.15 ml min$^{-1}$ of contaminant was adjusted using peristaltic pump. Filter paper was fixed at the upper electrode to give the uniform contaminant solution over the surface of nanocomposite sample. Filter paper and electrode proportions were selected according to the IEC-60587 standard. A transformer with bridge rectifier and filter capacitor of 1 $\mu$F was used to generate 2.5 kV high HVDC. Leakage current of LDPE nanocomposites was recorded. Leakage current measurement block was connected in series with test sample. This block consisted of series resistor with comparator unit and associated data logging in an attached computer. All prepared samples were tested on this setup by setting an upper limit of time up to 5 h or maximum value of leakage current up to 60 mA or sample ignition whichever occurs first.
2.3.3. Leakage current measurement setup
The leakage current measurement setup consists of HV transformer, rectifier circuit, filter capacitor, test specimen and high precision electrometer. The rectified, filtered DC voltage is applied to the test specimen in series with high precision electrometer which measures the leakage current flowing through the test specimen under high voltage stress.

3. Results and discussion

3.1. Partial Discharge Analysis
Partial discharge inception voltage (PDIV) is lowest voltage value at which partial discharge occurs in a test arrangement where the test voltage applied to the test object is increased gradually from a lower value. Partial discharge extinction voltage (PDEV) is the voltage lower than PDIV at which no partial discharges occur, when the voltage applied to the test object is decreased from a higher value [1]. The values of PDIV and PDEV for different nanocomposites were measured in accordance with IEC 60270 standard. The results are shown in figures 3(a)–(c).

Results show that in case of LDPE/SiO2 nanocomposite highest value of PDIV (2.76 kV) and PDEV (1.89 kV) occurs at 0.8% filler loading. In case of LDPE/TiO2 nanocomposite highest value of PDIV (2.58 kV) and PDEV (1.79 kV) occurs at 0.4% filler loading. In case of LDPE/TiO2@SiO2 nanocomposite highest value of PDIV (3.21 kV) and PDEV (2.21 kV) occurs at 0.6% filler loading. All the nanocomposites of LDPE/SiO2, LDPE/TiO2 and LDPE/TiO2@SiO2 have shown a higher value of PDIV and PDEV than pure LDPE. So, it can be concluded that inclusion of all these nano fillers in LDPE have improved the PD resistance properties of LDPE to varying extents. The percentage at which best resistance to partial discharge inception and partial discharge extinction is achieved is different for each type of nano filler.

The phase resolved partial discharge (PRPD) pattern of LDPE nanocomposites at different filler loadings are shown in figures 5–14. The addition of nano fillers improved the PD resistance characteristics of LDPE nanocomposites. PRPD patterns of LDPE/SiO2 nanocomposites showed better PD resistance characteristics as compared to raw LDPE. Lowest PD magnitudes and PD pulses per cycle were observed at 0.8% filler loading. PD magnitude and PD pulses per cycle increased at 1% filler loading.

The PD characteristics of LDPE/TiO2 nanocomposites are shown in figure 9–11. PRPD results show that LDPE/TiO2 nanocomposites showed better PD resistance as compared to raw LDPE base matrix. Least magnitude of PD discharges and PD pulses per cycle were observed at 0.4% filler loading. The conductive nature of TiO2 nano fillers resulted higher PD magnitude and PD pulses at higher filler loadings.

The PD characteristics of LDPE/TiO2@SiO2 nanocomposites are shown in figures 12–14. The PRPD characteristics among all LDPE nanocomposites investigated, LDPE/TiO2@SiO2 nanocomposites showed highest PD resistance as compared to other LDPE nanocomposites. Least magnitude of PD discharges and PD pulses per cycle was observed at 0.6%. The PD magnitude and PD pulses per cycle increased at higher filler loading 1%.

Cumulative apparent PD magnitude of LDPE nanocomposites is given in figure 15. Results show that SiO2, TiO2 and TiO2@SiO2 nano fillers gave least average PD magnitude at 0.8%, 0.4% and 0.6% wt. having values of 5.13 pC, 6.98 pC and 4.20 pC respectively. LDPE/TiO2@SiO2 nanocomposites exhibited enhanced PDIV, PDEV and PD resistance as compared to other LDPE nanocomposites. The shell of SiO2 around TiO2 in TiO2@SiO2 core–shell nano filler results in the formation of double layer between the polymer matrix and nano filler which plays a very vital role in the enhancement of dielectric properties of base matrix. The double layer of TiO2@SiO2 nano filler traps charge injections from electrode and excessive charge injections is restricted by repulsive forces which forms an electrical barrier [26]. The barrier effect of TiO2@SiO2 imparts enhanced dielectric properties to LDPE base matrix.
3.2. Tracking Performance

The outdoor insulators experience different types of environmental stresses which result in its progressive degradation. These environmental stresses include dust, moisture, UV light, humidity and lightning strokes etc. In the presence of dust and moisture conductive paths may develop on the surface of insulation under high voltage stress. Excessive flow of leakage current through the conductive path results in severe burning and erosion of insulation material. Therefore, such an outdoor insulation material is desired which exhibits high tracking resistance. In general tracking resistance refers to the voltage level at which a conductive track is developed on the surface of insulator in the presence of a contaminant liquid. The tracking performance of LDPE samples were investigated in accordance with IEC 60587.

3.2.1. Visual characterization of tracking

All the samples experienced arcing on their surface because of the contaminant and high voltage stress. The carbon tracks and pits were formed between the HV electrode and ground electrode due to continuous arcing on the surface of samples [27]. The samples having least amount of tracking and pitting among all are shown in figure 16 and the ones with highest amount of tracking and pitting are shown in figure 17.

![Graphs showing PDIV and PDEV of LDPE/SiO₂, LDPE/TiO₂ & LDPE/TiO₂@SiO₂ nanocomposites.](image)
From figure 16, it can be seen that the amount of carbon deposited on 0.4% TiO2 sample is higher as compared to raw LDPE and other LDPE nanocomposites. Carbon deposits on 0.8% SiO2 sample were less as compared to TiO2 nano filler samples. Least amount of carbon was deposited on 0.6% TiO2@SiO2 sample.

From figure 17, it can be seen that 1% TiO2 samples exhibit highest amount of carbon deposits as compared to raw LDPE and other LDPE nanocomposites. 0.2% SiO2 samples have shown lower carbon deposits as compared to TiO2 nanocomposite. 0.2% TiO2@SiO2 sample have shown lower amount of carbon deposits than raw LDPE and other LDPE nanocomposites.

Among TiO2 samples the carbon deposits on 1% are higher than 0.4% filler loading. The carbon deposits on 0.2% SiO2 sample are higher than at 0.8% filler loading. 0.2% TiO2@SiO2 sample have shown higher carbon deposits as compared to 0.6% filler loading.
In general, among all nanocomposite investigated highest surface erosion and pitting was shown by 1% LDPE/TiO$_2$. 0.2% LDPE/SiO$_2$ nanocomposite shows reasonable anti tracking properties and 0.6% TiO$_2$@SiO$_2$ nano filler have best anti tracking properties.

The scanning electron microscopy (SEM) image of LDPE/TiO$_2$@SiO$_2$ nanocomposites for 0.2% and 1% filler loading is given in figures 18 and 19. The EDX analysis of LDPE/TiO$_2$@SiO$_2$ nanocomposite for 1% filler loading is given in figure 20. At higher filler loading the agglomeration of nano fillers is visible in figure 19.

The composition of the elements of EDX analysis are given in table 3.
3.2.2. Leakage current after 2 h

The current which flows from the surface of the insulation even in healthy condition is called leakage current [28]. Leakage current (LC) directly affects the insulation properties if its value increases from a certain threshold point it causes a progressive degradation in insulation materials [29]. LC peaks after 2 h of tracking are shown in figures 21–23.

Increasing filler concentration causes a decrease in LC values [30]. The leakage current of LDPE/\( \text{SiO}_2 \) nanocomposites is given in figure 21. In case of LDPE/\( \text{SiO}_2 \) nanocomposites least LC value of 32 mA at 0.8% filler loading and the highest LC value of 38 mA at 0.2% filler loading was observed. The highest leakage current value shown by LDPE/\( \text{SiO}_2 \) nanocomposite is still lower than raw LDPE having 48 mA.

Figure 9. PRPD pattern of 0.2% LDPE/\( \text{TiO}_2 \) nanocomposite.

Figure 10. PRPD pattern of 0.4% LDPE/\( \text{TiO}_2 \) nanocomposite.
The LC peaks of LDPE/TiO2 nanocomposites are shown in figure 22. In case of LDPE/TiO2 nanocomposites least LC value of 34 mA at 0.4% filler loading and the highest LC value of 51 mA at 1% filler loading was observed. The highest value of leakage current shown by LDPE/TiO2 nanocomposite is higher than raw LDPE having peak value of 48 mA. The conductive nature of TiO2 nano filler results in higher leakage current value than raw LDPE and other LDPE nanocomposites.

The LC peaks of LDPE/TiO2@SiO2 nanocomposites are given in figure 23. In case of LDPE/TiO2@SiO2 nanocomposites least LC value of 26 mA at 0.6% filler loading and highest LC value of 36 mA at 0.2% filler loading was observed. The highest leakage current value shown by LDPE/TiO2@SiO2 nanocomposite is lower than raw LDPE (48 mA) and all other nanocomposites investigated.
The average leakage current was recorded for 5 min every 30 min. The average leakage current of raw LDPE and other LDPE nanocomposites is shown in figure 24. Results show that generally average leakage current increases with time. The increase in average leakage current is different for raw LDPE and LDPE nanocomposites. The average leakage current value of SiO$_2$ nanocomposites at 0.8% was less than 0.2% filler loading and raw LDPE. TiO$_2$ nano fillers exhibited lower values of leakage current at 0.4% than at 1% filler loading. The values of leakage current of TiO$_2$ fillers at 1% filler loading are highest among all nanocomposites investigated. TiO$_2$@SiO$_2$ nanocomposites have shown least values of leakage current at 0.6% among all nanocomposites.
3.2.3. Instantaneous leakage current versus voltage

A direct relationship exists between increasing voltage and respective increase in LC values. Leakage current of circular samples of LDPE/SiO₂, LDPE/TiO₂ and LDPE/TiO₂@SiO₂ nanocomposites was measured for a voltage threshold range of (5–30 kV).

The LC values versus voltage for all LDPE/SiO₂ nanocomposites are shown in figure 25. The highest value of leakage current was observed at 0.2% filler loading at 30 kV after which the value of leakage current gradually decreased. The least value of leakage current for LDPE/SiO₂ nanocomposite occurred at 0.8% filler loading at 30 kV. Therefore, 0.8% filler loading is the optimum point for LDPE/SiO₂ nanocomposite.

The LC values versus voltage for all LDPE/TiO₂ nanocomposites are shown in figure 26. The conductive nature of titanium dioxide (TiO₂) nano fillers results in more increase in LC with increasing voltage compared to LDPE/SiO₂ and other nanocomposites. The highest value of leakage current of LDPE/TiO₂ nanocomposite was
observed at 1% filler loading at 30 kV. Whereas least value of leakage current was observed at 0.4% filler loading at 30 kV.

The LC values versus voltage for all LDPE/TiO$_2$@SiO$_2$ nanocomposites are shown in figure 27. Decrease in leakage current was observed by increasing filler loading of TiO$_2$@SiO$_2$ nano fillers up to 0.6% at 30 kV. The least value of leakage current occurred at 0.6% filler loading having a value of 5.29 nA. The value of leakage current increases at 1% filler loading at 30 kV. A same trend was observed when encapsulated nano particles were used with other base materials as reported by some other studies e.g. the leakage current values of Zepoxy/TiO$_2$@SiO$_2$ nanocomposites were investigated in [31] showed that the value of leakage current decreased with increasing filler concentration. Enhanced insulation attributes of SiO$_2$ and high polarizability of TiO$_2$ resulted in lower values of leakage current at higher filler concentration. Least value of leakage current was observed at 0.8% filler

Figure 17. (a) (i) Raw LDPE sample before tracking, (ii) After tracking, (iii) After surface cleaning. (b) (i) 1% TiO$_2$ sample before tracking, (ii) After tracking, (iii) After surface cleaning. (c) (i) 0.2% TiO$_2$@SiO$_2$ sample before tracking, (ii) After tracking, (iii) After surface cleaning. (d) (i) 0.2% SiO$_2$ sample before tracking, (ii) After tracking, (iii) After surface cleaning.

Figure 18. SEM image of 0.2% LDPE/SiO$_2$@TiO$_2$ nanocomposite.
Figure 19. SEM image of 1% LDPE/SiO$_2$@TiO$_2$ nanocomposite.

Figure 20. EDX image of 1% LDPE/TiO$_2$@SiO$_2$ nanocomposite.

Figure 21. Leakage current of LDPE/SiO$_2$ nanocomposites.

Table 3. Composition of elements of 1% LDPE/TiO$_2$@SiO$_2$ nanocomposite.

| Element   | 0wt% | 0.2wt% | 0.8wt% |
|-----------|------|--------|--------|
| Carbon(C) | 78.69% | 78.69% | 78.69% |
| Oxygen(O) | 18.89% | 18.89% | 18.89% |
| Silicon(Si) | 0.97% | 0.97% | 0.97% |
| Titanium(Ti) | 0.83% | 0.83% | 0.83% |
| Calcium(Ca) | 0.62% | 0.62% | 0.62% |
loading having a value of 0.27 nA. Study of Leakage current characteristics of epoxy/TiO$_2$@SiO$_2$ nanocomposites exhibited lowest values at 0.6% filler loading [32]. Generally, the values of leakage current for circular LDPE nanocomposites have shown an increase in leakage current with increasing voltage threshold.

3.3. Volume resistivity
Volume resistivity is the opposition to the flow of current through the body of an insulating material [33]. The higher value of the volume resistivity of an insulating material, lower the value of leakage current flowing through the insulation. Volume resistivity was measured in accordance with ASTM D257 standard using high
precision electrometer. The comparison of resistivity values for LDPE base matrix with different nanocomposites of LDPE/SiO$_2$, LDPE/TiO$_2$ and LDPE/TiO$_2$@SiO$_2$ are given in figure 28.

Results show that raw LDPE matrix has a resistivity value of $2.84 \times 10^{14} \Omega$-cm. The value of resistivity of LDPE/SiO$_2$ nanocomposites increases with increasing filler concentration up to 0.8% filler loading having a value of $3.126 \times 10^{14} \Omega$-cm and then decreases to $2.964 \times 10^{14} \Omega$-cm at 1% filler loading. The value of resistivity of LDPE/TiO$_2$ nanocomposite decreases with increasing filler concentration having a maximum value of $2.81 \times 10^{14} \Omega$-cm at 0.4% and minimum of $2.5 \times 10^{14} \Omega$-cm at 1% filler loading. Resistivity values of LDPE/TiO$_2$@SiO$_2$ nanocomposite is maximum ($3.26 \times 10^{14} \Omega$-cm) at 0.6% and minimum ($3.05 \times 10^{14} \Omega$-cm) at 1% filler loading. LDPE/ SiO$_2$@TiO$_2$ nanocomposites have shown highest resistivity values as compared
to raw LDPE and all other LDPE nanocomposites. Next highest resistivity values have been shown by LDPE/SiO$_2$ nanocomposites. LDPE/TiO$_2$ nanocomposites have shown least resistivity values.

### 3.4. Dielectric strength

The maximum voltage stress required to produce a dielectric breakdown of the insulating material is referred as dielectric strength of an insulating material [34]. Dielectric Strength has a unit of volts per unit thickness. The higher the value of dielectric strength of a material the more efficient insulator it becomes. DC Breakdown strength was measured in accordance with ASTM D3755 standard. The dielectric strength of LDPE base matrix with different nanocomposites of LDPE/SiO$_2$, LDPE/TiO$_2$ and LDPE/TiO$_2$@SiO$_2$ is given in figure 29.

The raw LDPE has a dielectric strength of 26.92 kV mm$^{-1}$. The non-conductive nature of SiO$_2$ nano fillers results an increase in dielectric strength of LDPE/SiO$_2$ nanocomposites up to 0.8% filler loading having a value of 28.23 kV mm$^{-1}$ and then decreases to 28.17 kV mm$^{-1}$ at 1% filler loading. The dielectric strength of LDPE/TiO$_2$ nanocomposites increases with increasing filler loading up to 0.4% constituting a value of 27.53 kV mm$^{-1}$ and then decreases with increasing filler concentration having least value of 27.18 kV mm$^{-1}$ at 1% filler loading. The LDPE/TiO$_2$@SiO$_2$ nanocomposites exhibits an increase in dielectric strength with increasing filler loading up to 0.6% filler loading and then declines up to 1% filler loading having values of 28.46 kV mm$^{-1}$ and 28.27 kV mm$^{-1}$ respectively. The values of dielectric strength for all LDPE nanocomposites remain higher than raw LDPE base matrix.

### 4. Conclusion

The partial discharge and tracking characteristics of LDPE were investigated by addition of different nano fillers. The addition of nano fillers in the base matrix has shown to improve PD resistance and anti tracking properties of LDPE as long as percentages of fillers are low and well below 1%. Each nano filler has shown a different loading percentage at which it gives maximum improvement in the properties of the base material.

Among all three LDPE nanocomposites investigated LDPE/TiO$_2$@SiO$_2$ nanocomposites exhibit highest values of breakdown strength, volume resistivity and lowest values of leakage current at 0.6% filler loading.
LDPE/TiO2@SiO2 nanocomposite has shown highest PD resistance and anti tracking properties at 0.6% wt. The intra molecular band conduction and intra molecular hopping of mobile electrons are responsible for conduction of insulating polymers. The presence of semi conductive nature of TiO2 lowers down the hoping conduction barrier. Higher the filler concentration, lower the hoping conduction barrier and resistivity. Under high voltage stress the transition of insulator to conductor is easier. The high hydrophilic nature of TiO2@SiO2 nanoparticles and presence of impurities (like Al, Ca etc) increase at higher filler loading. EDX image of 0.8% LDPE/TiO2@SiO2 nanocomposite shown in figure 20 confirms presence of impurities.

The presence of impurities and semi conductive nature of TiO2 fillers dominate above 0.6% filler loading and tracking resistance start to deteriorate with associated increase in PD activity.

In general, it was observed that core–shell nano fillers are much more efficient in improving all the dielectric properties of solid insulation materials than ordinary nano fillers.

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