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PAPERS

Influence of nanofiller concentration on polypropylene nanocomposites for high voltage cables

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Polymeric insulation for HVDC cable is attracting more attention in the modern power transmission system. Especially, the thermoplastic material is desirable for power cable insulation because of its recyclability and ease of processing. Thermoplastic material development is a good alternative to cross-linked polyethylene in the future. Polypropylene has the advantage of avoiding by-products during cable production, which can minimize space accumulation and degassing costs. Therefore, this study investigates the influence of nanofillers on the structural properties of isotactic polypropylene. In addition, the proposed composite material's morphology, melting, dielectric permittivity, and breakdown strength are examined. Different weight percentages of inorganic nanofillers such as TiO_2 and ZnO are used to make nanocomposite thin films. With increasing filler concentration, the dielectric constant of the nanocomposite thin film increases. Apart from that, the dielectric loss of the TiO_2 nanocomposite thin film increases with weight percentage initially and it falls nearer to virgin material at a higher frequency. The breakdown strength of the nanocomposite materials shows a similar variation with filler concentration. TiO_2 is more resistant to deterioration than ZnO composite. Based on the results of the complete investigation, the TiO_2 nanocomposite is better suited for the insulation of HVDC cables.

Keywords: isotactic polypropylene, breakdown strength, titanium oxide, zinc oxide, thermoplastic, HF and superimposed AC

1 Introduction

The demand for power in the world is increasing day by day and also there is a drastic climate change due to the CO_2 emission in the environment during power generation by burning of fossil fuels [1–3]. On taking into account of environmental concerns and to meet the power demand most of the research focuses on alternate sources such as renewable energy. It is a big challenge to transmit the power without losses while transmitting the generated power from the generating station to the center of demand. High voltage direct current (HVDC) transmission is an efficient way to transmit large power over a long distance [1]. HVDC transmission system is economically less and losses are lower compared with HVAC transmission [2]. The first HVDC mass impregnated cable was put into the operation between Gotland Island and the Swedish mainland in 1954 [3]. HVDC transmission is an effective way to transmit bulk power from offshore to the converter end with reduced losses [4, 5].

The life of the cable is estimated with a key parameter, insulation. In the beginning, Oil Filled paper and mass impregnated paper are used as the insulation in the cables, later it is replaced by polymeric insulation [6]. Subsequently, Low-density polyethylene(LDPE) and High-density polyethylene (HDPE) were used as the insulation in the HVDC cable. However, due to poor mechanical properties and low withstand temperature it was replaced by cross-linked polyethylene (XLPE). XLPE insulation not only has properties like excellent mechanical strength but also has electrical properties hence it is used globally. In 1988, the first XLPE DC cable project came under operation between Gotland and Sweden [7]. XLPE is a thermoset material and hence cannot be recycled and reused. phenol, cumyl alcohol, acetophenone, and methyl styrene are the by-products of XLPE, which will introduce severe space charge accumulation and triggers the degradation of the lifetime of the cable [8]. Our motto is to investigate the recyclable material which should be friendly to the environment. Considering the demerits of XLPE and environmental concerns, the recyclable thermoplastic material is an alternate choice.

There are various stresses such as electrical stress, thermal stress, and mechanical stress that will influence the thermoplastic material during the transmission which will tend to degrade the life of the cable. Huang *et al* [9] dealt with a comprehensive analysis of the importance of thermoplastic materials such as isotactic polypropylene, propylene-ethylene block copolymer, and random copolymer for insulation by evaluating their potential. With recent evidence from the literature, we can conclude that Isotactic polypropylene is suited for insulation in HVDC cable applications. Isotactic polypropylene has the special feature of a high melting point and enhanced electrical properties such as breakdown strength and dc volume resistivity. Hence isotactic polypropylene is the finest choice to use as the insulating material instead of XLPE [10].

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Space charge accumulation is one of the most frequently stated problems associated in the HVDC cable insulating material. It will cause electrical field distortion and increase the local field, which will lead to partial discharge, faster degradation, and also premature breakdown [11]. Existing research recognizes that the role played by the addition of nanoparticles will effectively suppress the space charge accumulation in dielectric composites [12]. In modern times, nanotechnology plays a prominent role in insulating material. Recently investigators have wide focused on fine-tuning the dielectric properties of a material by incorporating nanoparticles. The addition of a considerable amount (3 wt%) of nanoparticles in the polypropylene will suppress the space charge accumulation [13–15].

From the broad examination of Montanari et al, the addition of nano-sized layered silicate exhibits improved electrical properties and space charge suppression [16, 17]. A good amount of work deals with the dielectric properties of LDPE and HDPE with nanoparticles but only a little research is focused on thermoplastic material [17–19]. It is reported that, incorporating the inorganic nanoparticles in the polymer matrix give a desirable effect on electrical properties and mechanical properties [20].

This paper gives a broad study on Isotactic Polypropylene (iPP) nanocomposite under various stresses. Andrea Cavallini et al reported that the harmonics, PWM-like waveforms on the power electronic circuits stress the insulation at the cable terminals [21–23]. The inorganic nanoparticles such as TiO_2 and ZnO are incorporated with the polymer matrix. The specimen is stressed under (i) HVDC (ii) high-frequency AC and (iii) superimposed stress (HVDC + high-frequency (HF) AC). From the examination, it is revealed that the nanofillers enhance the dielectric properties of the insulation material and hence iPP/nanocomposite is potential material for thermoplastic HVDC cable insulation.

2 Sample preparation

Isotactic Polypropylene (iPP) is the base material used for to study, which was supplied by Sigma Aldrich. iPP is in pellet form of melt index 12 g/10mins. The nanocomposite specimens are prepared using inorganic nanoparticles such as TiO_2 and ZnO (from Sigma Aldrich) with an average diameter of 50 nm with different weight percentages (viz.1 wt%, 2 wt%, 3 wt%, 4 wt%, and 5 wt%). The iPP pellet is completely dissolved using a non-polar solvent (Xylene) and the nanoparticles are mixed well in order to get a homogeneous form. The test specimen of thickness 100 m is prepared using spin coating. The sample is cured at the temperature of 70 °C

3 Measurements

The dispersion state of nanoparticles TiO_2 and ZnO in iPP polymer matrix was observed in the scanning electron microscope (SEM) model S3400N (Hitachi Make). The sample is sputtered with carbon black for making conductivity. The thermal and crystallization behavior of the polymer was investigated using differential scanning calorimetry (DSC), which was carried out in the instrument thermal analyzer instrument (Q20DSC). A small portion of the sample (5 mg) was cut down and placed in the aluminum pan. The aluminum pan was heated from room temperature to 200 °C with a heating rate of 20 °C/min to record the melting behavior. The measurements of melting behavior are investigated under the nitrogen atmosphere. Novel A high-frequency alpha acer high-frequency analyzer is used to measure the dielectric permittivity and loss of tangent of the sample. The sample of the thickness of 100 m was coated with gold on both sides of the surface to serve as electrodes. All the measurements were carried out in the range of 10^{-1} to 10^{-6} Hz at room temperature. The breakdown strength of the thin film sample at HVDC waveforms (14 kV DC), High-Frequency AC (10 kHz and 20 kHz), and superimposed of HVDC+ HF AC are carried out at room temperature. The electric field was applied at the rate of 0.5 kV/s until the breakdown of the sample. Five specimens were tested for each investigation and the average value is taken as the breakdown strength

4 Test setup

The sample is tested using two-electrode systems (sphere-plane). The bottom electrode is connected to the ground with the dimensions of 70 × 30 mm and a thickness of 15 mm. The high voltage source is connected to the top sphere electrode with a diameter of 25 mm. The specification of the sphere plane electrode is shown in Fig. 1 as per the IEC 60243-I standard.

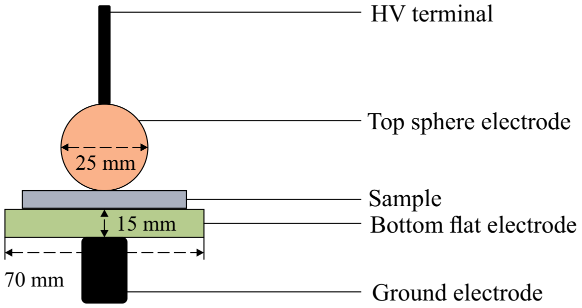


Fig. 1. Two-electrode test setup

To analyse the performance of the sample under AC stress, the sample of 100 μm thickness is stressed by the high voltage supply from the high voltage transformer (10 kVA, 230 V/11 kV, 50 Hz). In the same way, the sample is investigated under the output waveform of the (14 kV DC) source, the block diagram of which is shown in Fig. 2. Moreover, the sample is also investigated under the High-Frequency AC (10 kHz and 20 kHz) stress. To explore the real-time problem in the insulation during the

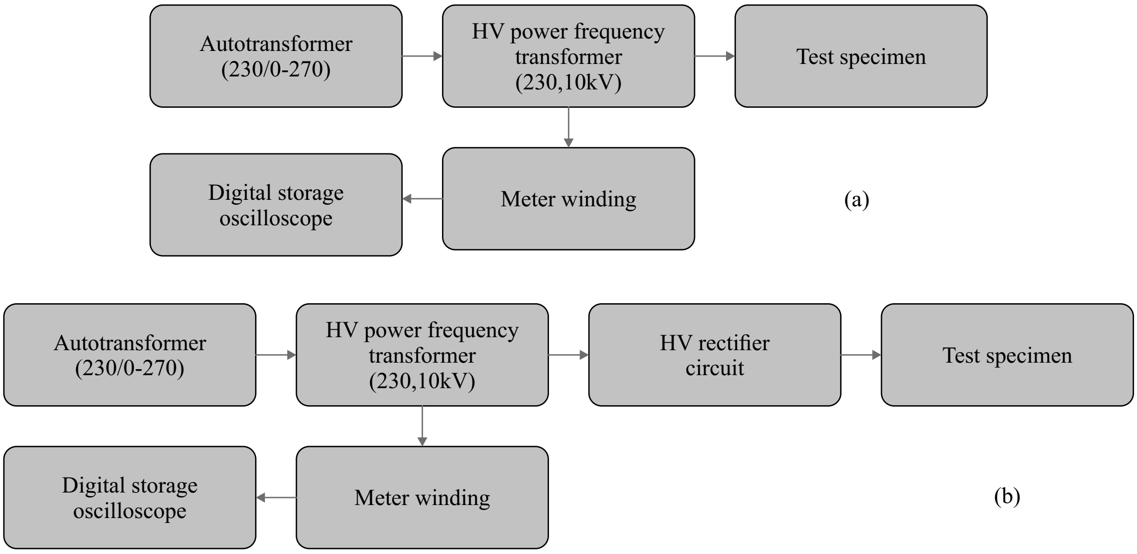


Fig. 2. Block Diagram of setups: (a) – high voltage power frequency, (b) – high voltage direct current (HVDC)

transmission of high voltage for long distances, the specimen is stressed simultaneously by both high-frequency AC and DC known as superimposed stress. Five samples are tested and the average is taken as the breakdown value.

5 Result and discussion

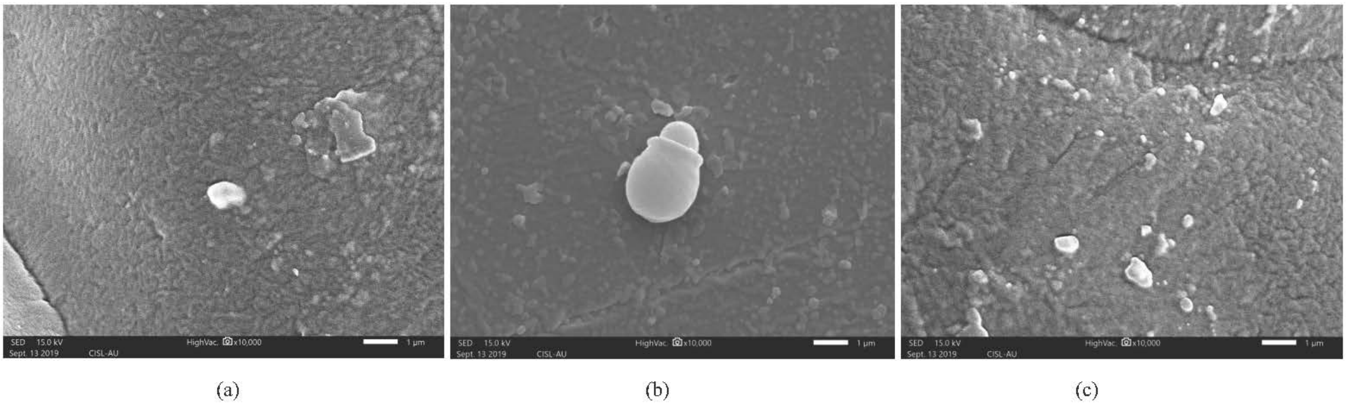
The cross-section of the nanocomposites was observed by SEM to characterize the dispersion of the different nanoparticles in PP matrix. The morphology and dispersion of the TiO_2 and ZnO particles in the polymer matrix are characterized by a SEM image. Figure 3 shows the SEM image of the PP nanocomposite at 3 wt%. The SEM image shows that the nanoparticles are well distributed in the polymer matrix and it has excellent bonding between the polymers. The nanoparticles are distributed in the polymer matrix with a dimension less than 240 nm without any large agglomeration. The nanoparticles in the polymer matrix are in the range of 170 nm, and 210 nm in TiO_2 and ZnO due to the nature of the particles.

5.1 *Thermal analysis*

Thermal performance is the key parameter that decides the withstand temperature of the thermoplastic insulation. Figure 4 shows the melting temperature of the TiO_2/iPP and ZnO/iPP composite. The melting temperature of the all iPP nanocomposite is extensively higher than the working temperature of XLPE (70°C). From the data, it can be concluded that the thermal property of iPP and ZnO nanocomposite is exactly suited for high voltage cable insulation applications.

5.2 *Dielectric properties*

The dielectric polarization and relaxation mechanism decide the breakdown strength of the nanocomposites. Dielectric permittivity and loss tangent of different fillers and weight percentage of iPP nanocomposite are shown in Fig. 5. The dielectric constant of the nanocomposite increases with the increase of the filler concentration for certain kinds of nanoparticles. From the literature, it is believed that the changes in the dielectric constant depend upon two reasons. First, the effect of nanoparticles



(a) (b) (c)

Fig. 3. The SEM image of (a) – iPP, (b) – TiO_2, and (c) – ZnO composites

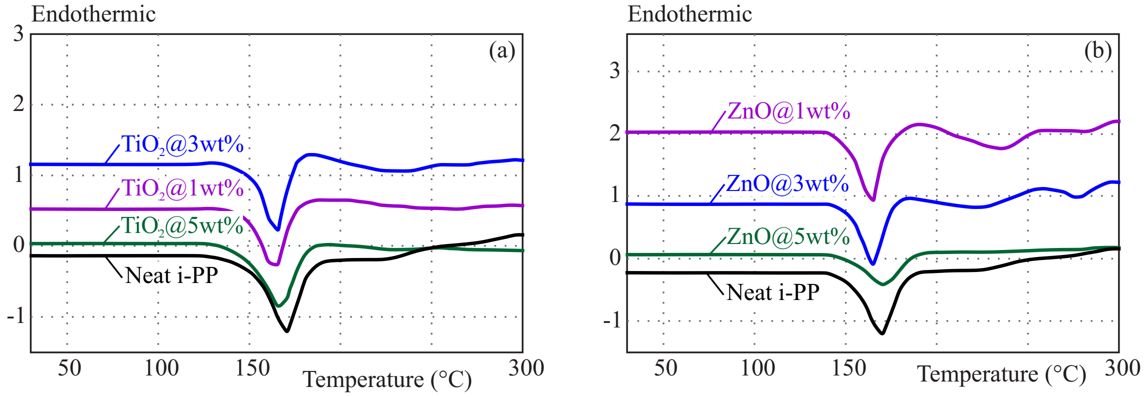


Fig. 4. Melting characteristics of (a) – TiO_2 and (b) – ZnO composites

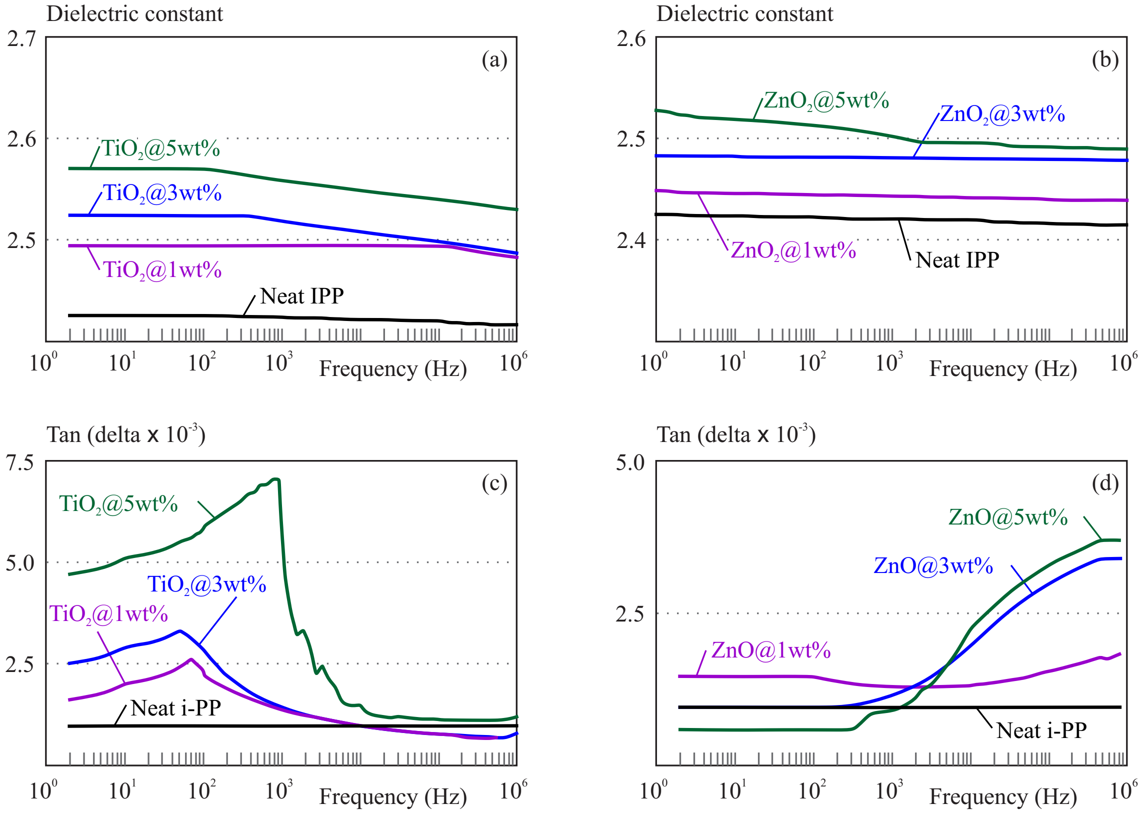


Fig. 5. Dielectric Spectra of iPP nanocomposites: (a),(b) – dielectric constant, (c),(d) – dielectric loss

in the polymer matrix will restrict the movement of the polymer chain which will tend to reduce the polarization of the polymer matrix [24, 25]. On the other hand, the dielectric constant increases due to the interfacial polarization occurring at the interface of the polymer matrix and nanoparticles [26]. The nanoparticles used in this paper have a higher dielectric constant than the base material hence this would increase the dielectric constant of the composites. There are also some reports that the inclusion of nanoparticles in the polymer can decrease the dielectric constant [24]. The dielectric characteristics vary with the weight percentage of different nanoparticles. The dielectric constant of TiO_2 and ZnO nanocomposites slightly increases from its base material with an increase in filler concentration and also at a lower frequency. From Fig. 5, the loss of tangent of the TiO_2 increases significantly at lower frequencies and then it came low at higher frequencies for higher concentration. This may be due to higher dielectric constant value and interfacial polarization plays an important role in TiO_2 nanocomposites. Whereas for the ZnO it is vice versa.

5.3 AC breakdown strength

The breakdown strength of the material decides the performance of the dielectric material. The test specimen is first stressed with a power frequency AC voltage source. The breakdown strength of the insulation is influenced by key parameters such as partial discharge and space charge distribution. Accumulation of space charge and its distribution in the bulk dielectrics degrade the life of

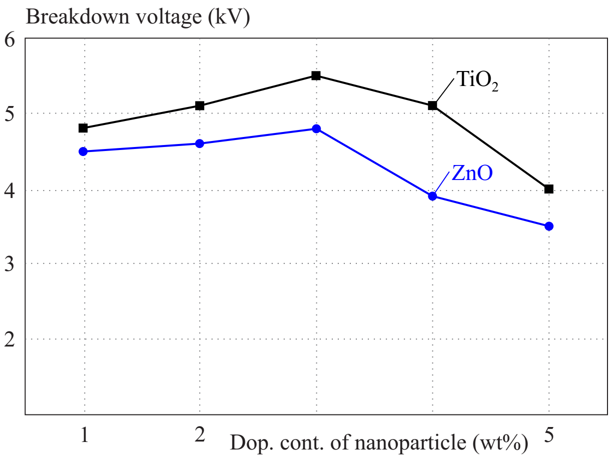


Fig. 6. Breakdown voltage of TiO_2 and ZnO nanocomposite under power frequency AC stress

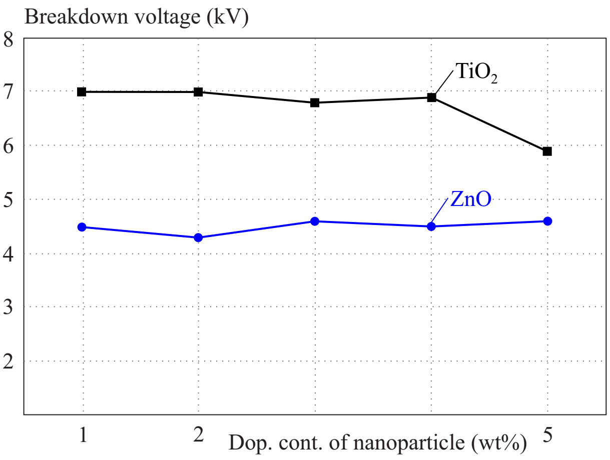


Fig. 7. Breakdown voltage of TiO_2 and ZnO nanocomposite under DC stress

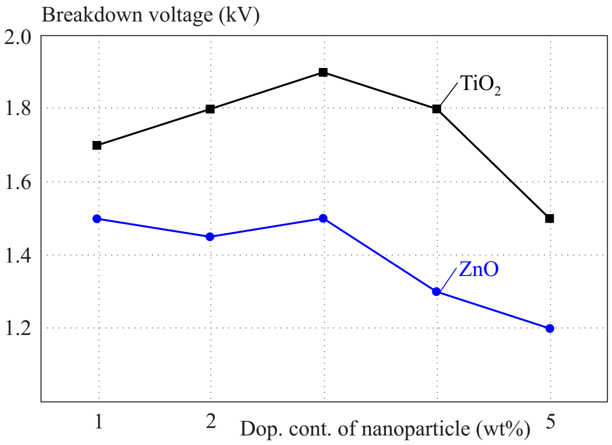


Fig. 8. Breakdown voltage of TiO_2 and ZnO nanocomposite under HF AC stress (10 kHz)

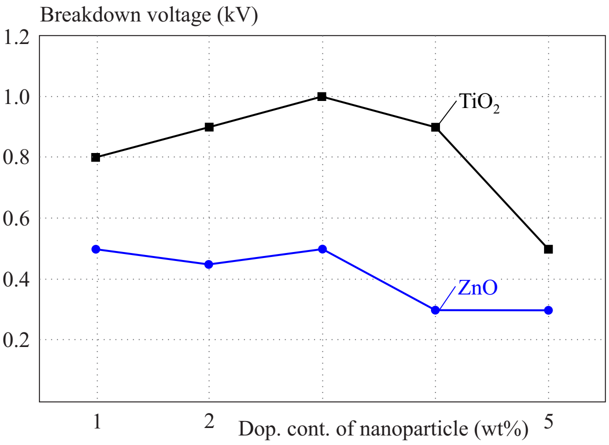


Fig. 9. Breakdown voltage of TiO_2 and ZnO nanocomposites stressed under HF AC (20 kHz)

the cable. It is reported that the space charge distribution in the polymer matrix is suppressed by incorporating the inorganic nanoparticles [11].

From the test result shown in Fig. 6, up to 5 wt% TiO_2/iPP nanocomposite has comparatively higher breakdown strength than the ZnO/iPP nanocomposites. Introducing nanoparticles in the polymer matrix will increase the high surface-to-volume ratio, indicating there is an excellent interfacial area between filler and polymer matrix. The performance of the nanocomposite is affected by the weak point which is due to the loose interface between the filler and polymer matrix [27]. From the examination, the breakdown strength of the TiO_2/iPP and ZnO/iPP nanocomposites increase with an increase in weight percentage of the filler concentration and start to decrease after 3 wt%. From Fig. 6, the doping at 3 wt% appears critical weight percentage. In comparison with neat iPP, the TiO_2/iPP nanocomposites have higher breakdown strength (5.5 kV) at 3 wt%, and ZnO/iPP is slightly higher (4.8 kV). The interaction between the nanoparticles created a rigid structure which probably delays the discharge flow up to the critical weight percentage (3%). After reaching the critical weight percentage the breakdown values decreases, which may be due to the agglomeration of nanoparticles.

5.4 *DC breakdown strength*

Under the DC stress, the breakdown strength of the polymer insulation increases with an increase in nanoparticle doping. The free charge carrier and energy of the charge carrier are the two parameters that strongly decide the DC breakdown. When applying the high voltage, the homocharges are injected by the cathode in the polymer. In the DC electric field, the energy of the charge carrier gets accelerated which would strike and break the polymer chain, this will tend to trigger initial partial discharge and finally leads to breakdown [28].

The effect of nanoparticles in the DC electric field is shown in Fig. 7. The breakdown voltage of TiO_2/iPP is 7.3 kV and it maintains almost constant up to 4 wt% and suddenly decreased at 5 wt%. The nanoparticles in the polymer matrix will act as a scattering source and it would reduce the charge carrier mobility and carrier energy by creating trapping in the charge carrier paths. Other than a small variation in the 2 wt% the breakdown

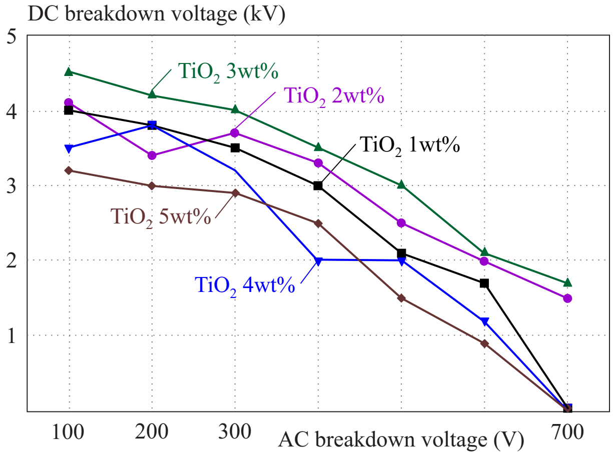


Fig. 10. Breakdown Voltage of TiO_2 nanocompositess stressed under HVDC + HF AC waveform of 20 kHz

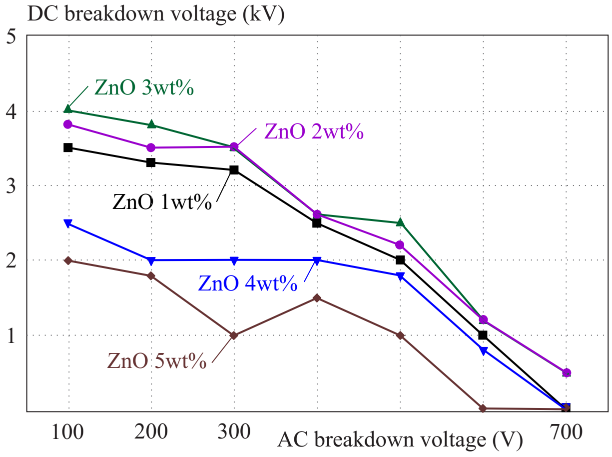


Fig. 11. Breakdown voltage of ZnO nanocomposites stressed under HVDC+ HF AC waveform of 20 kHz

Table 1. Breakdown voltage of the iPP polymer and its composites

Type	Stress		Filler	Breakdown at 3wt% (kV)
Neat	HVAC		NA	4.6
	HVDC			7
	HFAC	10kHz		1.02
		20kHz		0.60
	Super-imposed	DC+ 20 kHz		*
Composite	HVAC		TiO_2	5.5
			ZnO	4.8
	HVDC		TiO_2	7.3
			ZnO	4.6
	HFAC	10 kHz	TiO_2	1.90
			ZnO	1.5
		20kHz	TiO_2	1
			ZnO	0.5
	Super-imposed	DC+ 20 kHz	TiO_2 ZnO	*

* Decreases with increasing AC voltage

value of ZnO/iPP nanocomposites almost remains constant. However, ZnO/iPP composite showed lower breakdown strength in all weight percentages compared to neat iPP.

5.5 *High-frequency AC stress*

The test data for various filler concentrations at 10 kHz and 20 kHz is shown in Fig. 8 and Fig. 9. At 10 kHz and 20 kHz the breakdown value of TiO_2/iPP nanocomposite increase with an increase in weight percentage of filler concentration up to 3 wt% and then decreases with an increase in the filler concentration up to 5 wt% similar to the power frequency of AC stress. From the comprehensive analysis, the breakdown strength of TiO_2/iPP when compared with neat iPP is 90% and 67% more when stressed under at 10 kHz and 20 kHz respectively.

The reason behind the lower value of the breakdown voltage at the high-frequency stress is due to the internal heating produced during the reorientation of the dipole in the polymer matrix and the inception of PD at a lower voltage in the void initiating the earlier breakdown [29, 30]. The neat iPP has a higher breakdown voltage than the 5 wt% concentration at 20 kHz.

It is observed that there is no appreciable change in ZnO/iPP nanocomposites up to 3 wt% but beyond 3 wt% it decreases. The breakdown voltage profile revealed that TiO_2/iPP and ZnO/iPP appears similar variation at 10 kHz and 20 kHz. It is noted that in 10 kHz the ZnO/iPP nanocomposite is 50% higher than the breakdown value of neat iPP whereas in 20 kHz, the breakdown strength of the ZnO composite is lower than that of neat iPP at different filler concentrations.

The dissipation factor is the main reason for the lower breakdown strength of the ZnO. The dissipation factor of ZnO is increases with an increase in frequency whereas that of TiO_2 is quite opposite to it. From the reported results, 3 wt% of TiO_2/polyamide exhibits a resistant to partial discharge activity [28].

5.6 *Superimposed stress*

The power generated from the wind power station is converted into DC at the generating station and again DC is converted back to AC at the load side. During transmission, along with DC voltage, there may be some harmonics and other high-frequency components present probably, influenced by the power electronics circuits. In real-time, cable undergoes combined stress by the DC with harmonics and other high-frequency components.

Thus the superimposed stress on the cable will accelerate and degrade the life of the cable. From Fig. 10 and

Fig. 11 it is noted that the influence of AC voltage on the DC breakdown value. The breakdown value of TiO_2 /iPP DC stress along with high-frequency superimposed stress (20 kHz)is 50% to 65% lower than then pure DC. Whereas for the same condition ZnO/iPP nanocomposite is 45% to 90% low. The severity of the DC voltage, when superimposed with a high-frequency component, is revealed in this test results.

6 Conclusions

The cable may endure multiple stresses during power transmission from the generating station to the load center. In particular, superimposed stress can shorten the cable's lifespan. To address this issue, isotactic polypropylene base material with TiO_2 and ZnO nanocomposite insulation materials are investigated in this study for the HVDC cables. The breakdown studies of the sample are carried out using the sphere to flat electrode method according to the IEC 60243-I standard. The sample is examined under DC stress, power frequency AC stress, and superimposed stress in an experimental setup to determine the maximum stress exerted on the insulation. Moreover, the impact of high frequency 10 kHz and 20 kHz in the HVDC cable is analyzed. The outcomes show that the stresses in TiO_2 perform better than ZnO up to 3 wt% concentrations. As a result, TiO_2 will be the potential recyclable material for HVDC cable applications.

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