

Chapter 9

Potential Applications of XLPE Nanocomposites in the Field of Cable Insulation



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1 Introduction

Cables and insulated wires are used in a wide range of products. Typical applications include power wires, communication cables, equipment wiring, automotive wiring, signal and control cables, and construction cables [1]. In the basic form, the system can be isolated by a conducting material surrounded by a dielectric layer, but in the case of optical dielectric conductor cables, the optical signal must be confined to the fiber core with a jacket that reflects the optical signal. Papers used in telegraph wires were known to be one of the earliest insulating materials reported in 1795 [2]. Since the nineteenth century, telegraph cables have been embedded in grooved wooden rails covered with strips of wood. Later, the development of vulcanized natural rubber occurs for applications like arc lighting [3]. In 1950s, polyethylene was introduced into distribution cables and subsequent transmission cables [4]. Applications of extruded synthetic insulation materials continue to grow,

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and improvements in materials offer greater incentives for their use [5]. Examples of these extruded materials are

Polyvinyl chloride (PVC),

Polyethylene (PE),

Ethylene-propylene elastomers (EPM and EPDM).

Initially, PVC and PE were unable to access in high voltage applications. However, for low voltage applications, the use of PE and PVC without cross-linking provided a cost-effective insulating material. Polyethylene (PE) is one of the commonly used plastic, mainly for packaging. Polyethylene is Polyethylene s a thermoplastic polymer composed of long hydrocarbon chains with different chain lengths (C_2H_4) nH_2 . High-density polyethylene generally has a melting point in the range of $T = 120\text{--}130\text{ }^\circ\text{C}$, and low-density polyethylene has a melting point of about $105\text{--}115\text{ }^\circ\text{C}$. Also, it exhibits good chemical resistance. They are highly resistant to both strong acids and bases, and too mild oxidants and reducing agents. Polyethylene (in addition to cross-linked polyethylene) can be dissolved in aromatic hydrocarbons or chlorinated solvents at high temperatures. Polyethylene is a homopolymer or copolymer in which ethylene is the main constituent of the monomer. It is one of the versatile polymers which offers high performance compared to other polymers. PE is a semi-crystalline polymer which possesses low-to-moderate strength and immense toughness. On raising the crystallinity, the properties such as stiffness, elastic limit, mechanical and thermal properties also increase. At low temperatures, PE exhibits excellent strength. PE is relatively inexpensive and exhibits excellent strength at low temperature. PE is highly resistant to both solvents and acids [6].

Low-density polyethylene (LDPE) is manufactured under high pressure and it possesses a long chain branch. This particularly affects the rheological behavior of the alloy. Recovery of flexible alloys increases with the number of long-chain branches. Samples of polyethylene with the same average molecular weight under high split load deviate further from Newton's behavior and have better accessibility. Highly branched, high molecular weight polymers generally cause gel formation and reduce the optical properties of the film. LDPE can be produced by free-radical polymerization. Due to the high degree of branching in LDPE proves, chain do not pack well in the crystal structure, which leads to low-density properties ($0.9100, 940\text{ g/cm}^3$). The intermolecular forces are small due to low instantaneous dipole-dipole interactions. As a conclusion of the above properties, LDPE shows low strength and high ductility [7]. The high degree of branching with long chains provides molten LDPE with unique and desirable flow properties. Its applications include both rigid containers and plastic films such as plastic bags, pipes and films.

The cross-linking process of polyethylene, resulting in high-density cross-linked polyethylene (XHDPE) or cross-linked low-density polyethylene (XLPE), extends the use of PE, increases the operating limit at high temperature, and results in good mechanical properties. It could be due to the creation of 3D structures. This structure improves the tensile strength and thermal stability of the polymer and also

improves hardness and chemical resistance. PE cross-linking is activated in three different ways:

- Radiation
- Addition of peroxide or addition of silane.

The addition of silane, which was patented by Dow Corning in the late 1960s, did not change the crystallinity of the base polymer observed by Venkatraman and Kleiner, and also made the material more flexible by cross-linking (Si-O-Si) instead of making the process more difficult [8]. This is especially interesting because it guarantees each Si atom has three active sites, which allows it to attach to six network bridges up to six polymer chains. However, optimization of production conditions is useful for better performance when mechanical, chemical and thermal resistances are required. Currently, cross-linked polyethylene is used in various industries, such as the cable and wire industries, hot water pipes, steam-proof food packaging. Along with these, most studies on XLPE are based on, cross-linking low-density polyethylene (LDPE) with peroxide or silane.

Due to high operating temperature, performance and well-controlled extrusion technology cross-linked polyethylene (XLPE) plays a major role by replacing oil paper insulated systems and in high voltage AC cables. The polyethylene (PE) is the base matrix and can be cross-linked by peroxide. Cross-linking is considered necessary, but the low-density PE (LDPE) exhibits a more significant melting at the temperature of about 100 °C, leads a loss in its mechanical stability [(9)]. On the contrary, linear high-density PE (HDPE) shows a high melting point and achieve a higher operating temperature. XLPE is now widely used in the field of HV cable insulation. Today, one of the most commonly used cross-linking methods was a radical reaction with an organic peroxide. But the by-products remaining from the cross-linking process announce some degradation in electrical properties which creates space charge accumulation. To reduce these byproducts, XLPE insulation cables need degassing process.

2 Insulation Materials

2.1 *Natural Rubber (NR)*

NR will act as effective thermostable insulating material when they are compounded and cured properly. A variety of inorganic/organic materials has been used as fillers, anti-degradants, plasticizers, vulcanizing agents to improve the dielectric properties of the NR. Traditionally, up to 25 kV of natural rubber has been used for insulation. Later, it can be replaced by saturated hydrocarbons. These materials exhibit good resistance and constant electrical behavior [10].

2.2 *Styrene Butadiene Rubber (SBR)*

It is a copolymer of butadiene and styrene and styrene butadiene rubber is commonly used in applications like low voltages. The electrical properties mainly depend on the ratio of particulate filler, the morphology of the particles and the method of processing and forming and possible interactions between the conductive and the nonconductive phases [11].

2.3 *Butyl Rubber*

The processing of butyl rubber is similar to the NR with the complex procedure and the final properties are comparable to the NR. The temperature tolerance of butyl rubber is 85 °C and for the NR is 60 °C. Used for sulfur vulcanization, because it is a copolymer of isobutylene and isoprene. Due to its saturated Skelton, butyl rubber offers betterment over natural rubber. So, a modified version of natural rubber is used in both insulation and lining applications [12].

The polymeric insulating materials, listed as

- Thermoplastics,
- Cross-linked elastomers,
- Thermoplastic elastomers.

2.4 *Other Polymeric Materials*

For a long time, polymeric insulation materials have been used with great success in underground power transmission of extruded high voltage alternating current (HVAC) cables. Later, in the 1950s, prime 110 kV extruded polymeric HVAC cable came into the field, and then polymer insulation materials have been extensively recycled in HVAC cables. Japan was the first one who use the prime 500 kV extruded HVAC cable. The outgrowth of high voltage direct current (HVDC) cables is far behind the HVAC cable. The aftereffect of extruded polymeric HVDC cable is not that too long but it shows large advances in power transmission better the HVAC cables, which includes, extruded polymeric HVDC cable shows lower transmission loss compared to HVAC cables, avoids the oil leaks appeared to be one of the most environmental hazards, also it has low maintenance cost compared to OF and MIND cables. Along with this, the advancement of polymeric HVDC cables also contributes to modern power transmission [3].

3 Properties of Nanocomposites for Cable Insulation

An improvement in insulation performance is expected when nanocomposites are used in place of the traditional microcomposite due to the enhanced area of interface between filler and the polymer system. Electrical properties which play a significant role in cable insulation are discussed below.

3.1 Conductivity

Conductivity, the ability of materials to conduct electrical current is greatly influenced when nanofillers are incorporated into the base matrix. Yan et al. studied the effect of incorporation on carbon black in XLPE on electrical conductivity and showed that the dependence of conductivity on DC electric field was slow to increase up to 20 kV/mm above which nonlinear characteristics were observed. The effect of temperature was not much pronounced up to 90 °C. The inclusion of carbon black proved to inhibit space charge accumulation and at the same time reduces electric field distortion thereby improving the conductivity of the prepared composites [13]. The effect of modified and unmodified silica nanoparticles in XLDPE was studied by Zang et al. which showed an increase in values with temperature [14]. Similar studies were conducted for magnesium oxide incorporated cross-linked low-density polyethylene (XLDPE) [15] and TiO₂ nanoparticle filled XLDPE matrix [16].

3.2 Permittivity

The main property in dielectric parameters is the relative permittivity and the effect of the addition of nanoparticles to XLDPE on dielectric properties has been studied in detail. Various factors affect the dielectric properties such as temperature, moisture, frequency along with material properties. It has been shown that the addition of nanosilica particles in XLDPE improves the dielectric properties [17, 18]. However, the effect of humidity on such nanocomposites at different frequencies must be investigated to select the material for industrial purposes. In such a study, it was observed that as the incorporation of silica nanoparticles increases the moisture absorption of the composite, a reduced breakdown strength and increased loss was observed [19].

3.3 *Electric Treeing and Water Treeing*

Electrical treeing is a pre-breakdown in insulation systems and a primary cause for failure leading to damage of systems especially in high voltage insulations [20]. Stress points such as protrusions, contaminants initiate the development of these channels which thereafter grows through continuous partial discharge activity within the medium [21]. The addition of nanofillers is found to suppress the electrical treeing of polymer nanocomposites for high voltage applications [22]. Addition of zinc oxide and aluminum oxide nanofillers in XLPE has proved to inhibit the electrical treeing in the nanocomposites up to 1% concentration, beyond which the addition of fillers propagates the tree growth. Similarly, the addition of SiO₂ nanoparticles inhibited the electrical tree growth [23].

Water trees are plume-like structures consisting of water-filled cavities developing from point of stress which can initiate electrical treeing leading to electrical breakdown [24]. The study of XLPE nanocomposites containing ZnO and Al₂O₃ and SiO₂ nanoparticles proved to improve the water treeing characteristics of the composites [25]. In a similar study, the addition of Montmorillonite clay particles in the composite is found to suppress the water treeing behavior due to the layered structure formed by the addition of nanoparticle [26].

4 **Future Breakthrough of HVDC Cables**

There are three types of HVDC cables which includes

1. Mass impregnated (MI)
2. Self-contained fluid filled (SCFF)
3. Cross-linked polyethylene

Reliable, cost-effective and environmentally responsible power grid by extruded polymeric cables is preferred as a mean for electric energy transportation. From the use of HVAC and HVDC cable networks will be able to raise their operating voltage levels which can withstand the resulting increase in electric stress. XLPE matrix with fewer impurities is preferred. For the advancement of direct current (DC) applications, General Electric Global Research Center developed a new insulation material, which uses nanoclay particles to support ethylene propylene rubber (EPR) insulation results N-EPR [27]. When compared to XLPE insulation, N-EPR exhibits space charge propagation. Because of the structural pattern of N-EPR insulation, it is possible to limit the electrical enhancement. In addition to these, N-EPR provides higher power density and neglects the lengthy and costly degassing needed for XLPE insulation. Cross-linked polyethylene (XLPE) is widely applied for the cable insulation due to its very good dielectric properties, processing feasibility, desirable chemical properties and ecofriendly. These properties help in the transmission of electricity and the distribution of electric energy

[28]. Also, the high resistance to thermal shocks and tuneable thickness makes it a suitable candidate for the cable industry. These properties were utilized for the application of XLPE in underground electric cables. In addition to the advantages associated with XLPE, it is also capable of absorbing high loadings of fillers (e.g., carbon black, silica) compared to uncross-linked polyethylene which becomes brittle on the incorporation of fillers. The addition of NPs to insulation material results in a marginal improvement in its dielectric, electrical, thermal properties. The reason is that by the formation of cross-links, the particles are bonded and trapped within the polymer matrix [29]. As a result, levels of filler that are disadvantageous and make the polymer brittle would impart reinforcement in XLPE [30]. By cross-linking of polyethylene, there can be an improvement in properties (Table 1) which can be applied in cable installation, heat-resistant food packaging (up to 200 °C), foams for thermal insulation, and chemical-resistant seals.

5 Current Perspective

The space charge, trapping and detrapping of electrons followed by the release of energy can lead to partial discharge and aging of insulation materials. Dielectrics made up of nanocomposites have been extensively reported and shown to have many desirable properties under DC field. Properties such as space charge and DC breakdown strength can be improved. The most common nanofillers that have been reported are MgO, SiO₂, Al₂O₃ and TiO₂. The mechanism behind the improvement in dielectric properties is believed to be charge trap characteristics modified by nanofillers [34]. The interface developed in nanocomposites between polymer matrix and nanoparticles can be the main reason for the improvement of electrical

Table 1 Cross-linking of polyethylene and its change in properties [30–33]

S. no	Property of polyethylene	After cross-linking of polyethylene
1	Melt index	Decrease
2	Density	No changes/decrease
3	Molecular weight	Significantly increased
4	Tensile strength	No changes/slightly increase
5	Elongation-at-break	Decrease
6	Impact resistance	Significantly improved
7	Abrasion resistance	Greatly improved
8	Stress crack resistance	Greatly improved
9	Elastic properties	Greatly improved
10	Environmental stress crack resistance	Increase
11	Resistance to slow crack growth	Increase
12	Temperature resistance	Greatly improved
13	Chemical resistance	Significantly increased

properties. The dispersion of nanoparticles in semi-crystalline polymers is important and not explored much. If there is no good dispersion of nanoparticle in polymer matrix, there will be agglomeration. Distances between adjacent spherulites are so small that agglomeration of the introduced nanoparticles may act as passages between spherulites in the amorphous area.

Etching method can be used to treat the nanoparticle incorporated polyethylene. Proper surface modification by desirable coupling agent can improve the dispersion of nanoparticle, spherulite structure and can prevent the overlap between interaction zones. Commercial (AC grade) XLPE and its nanocomposite are also tested to use it for DC application. The incorporation of nanofiller (e.g., nanosilica) is reported to be improved its breakdown strength on the DC step test [35].

SiC nanoparticles are reported to suppress the space charge accumulation at a concentration of 1 wt%. Further increase in concentration inhibited the effect of NPs on the space charge accumulation [36]. In the low concentration of NPs, there will be a large distance between NPs. According to previous reports on MgO XLPE composites, they reported deep traps which were surrounding MgO nanoparticles in nanocomposites which attracted charges to MgO NPs. An NP which is surrounded by electrons or holes can be considered as electrically neutral [37].

Moisture combined with electrical stress is another factor which affects the aging of the insulation material. This process can affect the partial discharge on the material followed by disintegration. There can occur a significant reduction in flash voltage when the water is combined with impurities on the insulation surface. The major factors that affect the characteristics of insulation materials are conductivity of the droplets, insulation surface roughness and the moisture droplet arrangement. As the conductivity of the moisture content is increased, there is a decrease in flashover voltage as shown in [38].

The nanocomposites made by MgO/XLPE (0.5 wt%) reported having higher flashover results compared to PE. This explains that the NP addition improves the flash overvoltage. This is due to the hindering of movement of surface charges by NPs [38]. The ratio of NPs to the polymer matrix depends on the type of NPs. As discussed earlier above the optimum concentration the property of insulating material can deteriorate. Surface modification of insulation material can improve the adhesion between nanofillers and polymer matrix which in turn improves the dispersion of NPs. This, in turn, will have a great improvement in breakdown strength (AC/DC), electrical and water tree resistance, permittivity and dielectric loss.

Figure 1 shows the SEM images of XLPE/silica composite that represents that nanofillers are not dispersed inhomogeneous manner. Some of the samples in each wt% show agglomeration (Fig. 1a and b). Figure 1c and d shows that the nanoparticle is better dispersed in the polymer matrix. The octylsilane-modified XLPE can promote chemical adhesion between NPs and polymer matrix. It is essential to understand the role of nanofillers in the hindrance of partial discharge (PD). From Fig. 2a, we can see that unmodified nanofillers are not well dispersed. This can cause a decrease in PD frequency of encounter with NPs (Fig. 2 denoted by black color line). This, in turn, can affect the lifetime of insulation material.

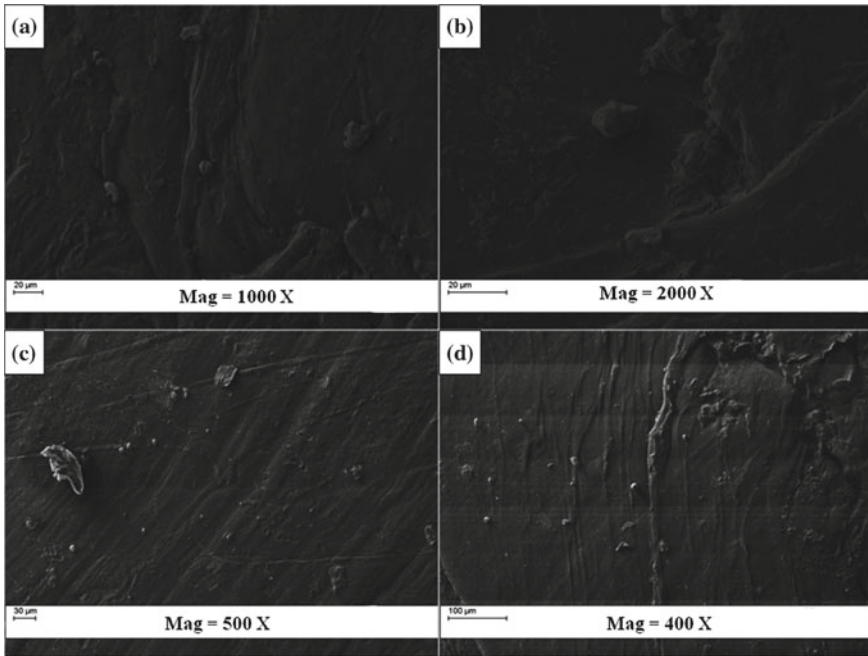


Fig. 1 SEM images of (a) XLPE/silica nano 2 wt% agglomerated nanocomposite, (b) XLPE/silica nano 3 wt% agglomerated nanocomposite, (c) XLPE/silica nano 2 wt% Octylsilane-modified nanocomposite, (d) XLPE/silica nano 3 wt% Octylsilane-modified nanocomposites (39)

Agglomeration of NPs leads to the segregation of nanofillers at several points. Hence, there will be easy propagation of PD to ground electrode in unmodified samples as shown in Fig. 2b. Formation of the electrical tree will lead to the degradation of XLPE matrix. Previous reports have shown an improvement in electrical treeing resistance of another polymer matrix (e.g., epoxy resin, polyethylene) with the loading of NPs [40–42]. Mansor et al. [43] studied the influence of ZnO and Al₂O₃ NPs on electrical treeing properties of XLPE matrix. When 1 wt % of ZnO and Al₂O₃ NPs was added, there was improvement insulation which might be due to the inhibition of the rapid growth of the electrical tree with a tree inception voltage (TIV) of 10.3 and 11.6%.

The modification of the polymer surface using a coupling agent can improve the dispersion and the distribution of NPs as shown in Fig. 2c. This can delay the propagation of PD and improves the lifetime of insulation material. The change in morphology of the final nanocomposite is also important to determine the final property of the material. NPs addition can lead to the decrease in surface energy or degradation of polymer which can cause secondary or incomplete crystallization.

Like PD, water tree phenomenon, first discovered in 1969, is another factor which reduces the lifetime of XLPE cables [44, 45]. The water tree problem was widely researched between 1980 and 1990. The reasons for the development of

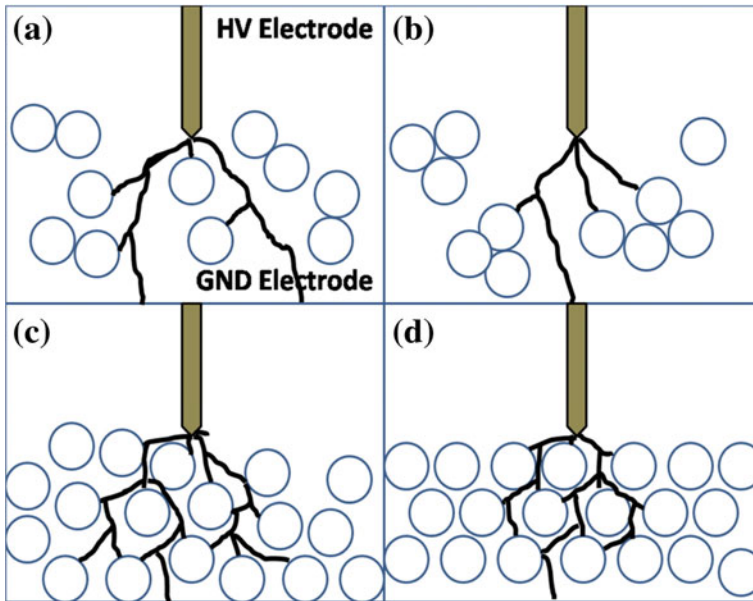


Fig. 2 schematic representation of propagation of partial discharge (PD) in (a) unmodified, (b) agglomerated (c) surface-modified, (d) ideally dispersed XLPE nanocomposite [39]

water tree are cable crosslink process by steam curing, extrusion of screens, semiconductors and insulations processes, and the cables lying in wetlands. The water tree is reported to be in the shape of microscopic scale voids which is connected to nanoscale and is like dendritic patterns discussed before (Fig. 3) [46]. Water trees are visualized as the presence of voids, contaminants which results in the formation of defects and lead to a powerful alternating electric field [47]. These deformations decrease the breakdown voltage and the impulse strength of the cable. The electrical tree is observed as the last stage of water tree growth before electrical breakdown. Because of the creation of an electric tree, the power cable is reported to fail in 15 days [48]. Phenylmethyldimethoxysilane (PMDMS) and titanium tetraisopropoxide catalyst (TTIP) were also studied in MV cables to resist this phenomenon [48].

Shirazi et al. [48] studied the use of phenylmethyldimethoxysilane (PMDMS), trimethylmethoxysilane (TMMS), ferrocene and sodiumdodecylsulfate with titanium tetraisopropoxide (TTIP) and α -alumina nanofiller for the reduction of water treeing behavior. They reported that the alumina nanofiller-improved the dielectric characteristics of degraded XLPE. During the addition of NPs, the process of lamellar thickness can also get accelerates which in turn can increase the heat of diffusion. The rearrangement of lamellae during the addition of NPs can be easily characterized using the DSC curves as shown in Fig. 4. By using a wire-plane test object for evaluating material resistance to electrical treeing, a relatively large

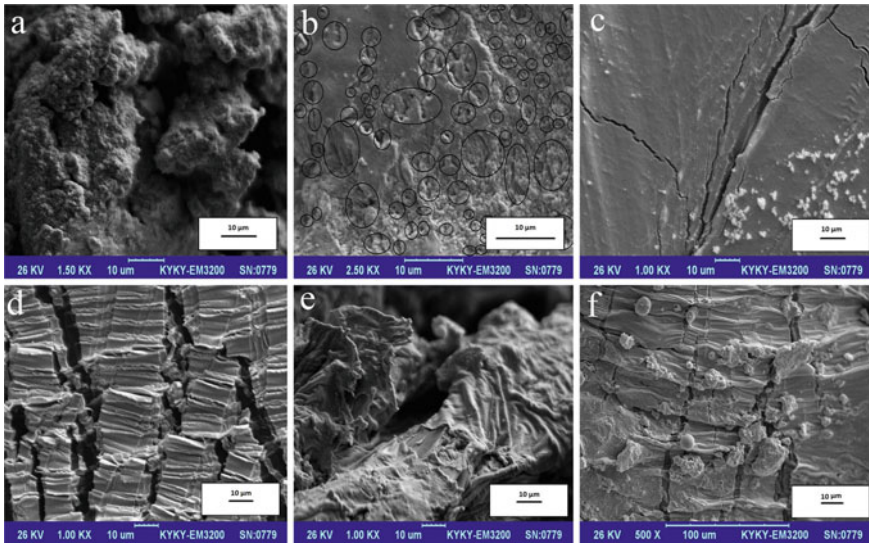


Fig. 3 Microstructure image of aged cable insulation and semiconductor material. (a) Water tree micro-voids. (b) Water tree aged area. (c) Cracks on aged insulation surface. (d) Electrical breakdown channels in the aged insulation. (e) Close-up view of aged inner semiconductor material. (f) Aged area of inner semiconductor material (48)

volume of the material is stressed, presumably also in its weakest points, and the growth of electric trees is not forced to one specific location like in the needle electrode objects. It thus provides an opportunity to better explore the influence of material structure and constituency on electrical tree inception. Several promising stabilizers of benzil-, thioxanthone-, fullerene- and melamine-types have also been tested and characterized. Majority of them exhibit a positive impact on the resistance to electrical treeing in cross-linked polyethylene, ranging from 20% up to well above 100% of the increased tree initiation field. It was found through correlating these test results with molecular modeling of electronic properties of the stabilizers that the electron affinity of stabilizing molecules generally correlates well with their stabilizing efficiency. This indicates that electron scavenging is here the dominant mechanism. Also, the addition of nanofillers to polyethylene-based materials yields a similar, though not equally strong effect nanocomposites at a filler content of 3 wt %. It appears, when considering our investigations and available literature data, that the effect is not related to the chemical nature of the nanofiller but rather to the formation of polymer-particle interfacial layer that provides deeper charge trapping sites and reduced long range molecular mobility.

Simulation studies were also carried out to analyze the electrical and mechanical properties of XLPE nanocomposites using different NPs [49, 50]. Nanoclay is widely applied for the production of nanocomposites. They have a multilayer structure with nanometer thickness with excellent electrical properties [51, 52]. The nanocomposites with clay exhibit good shielding of polymer matrices from heat and

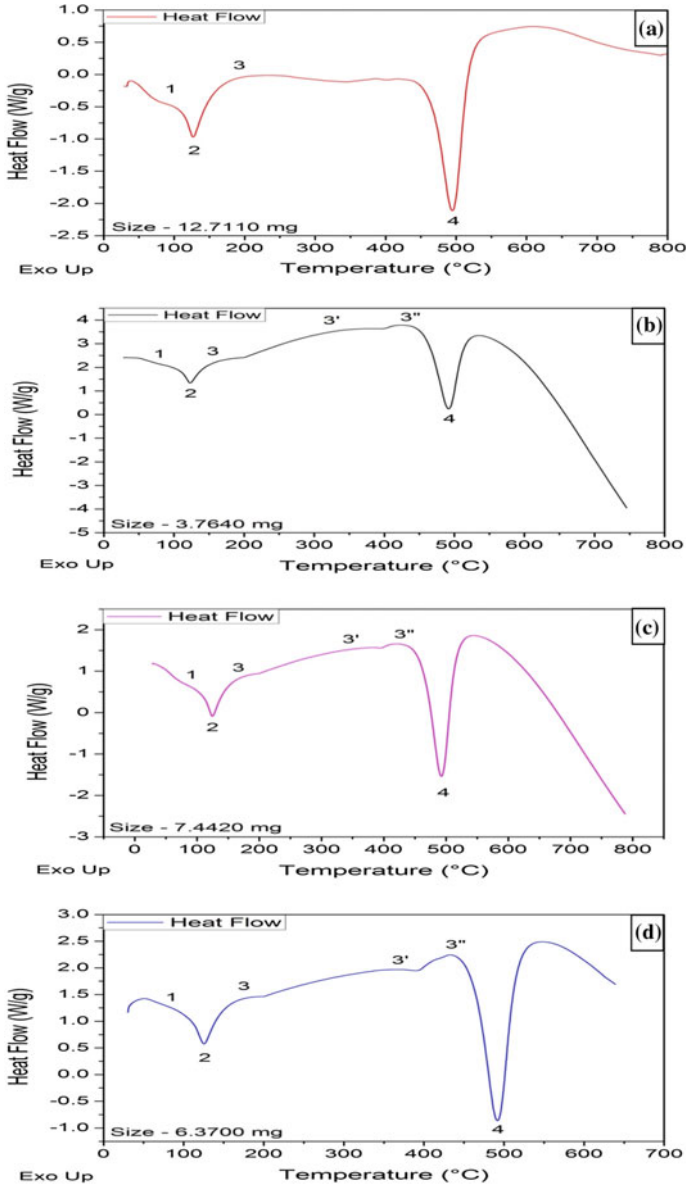


Fig. 4 Differential scanning calorimetry (DSC) of (a) Virgin XLPE, (b) nano 1 wt% unmodified, (c) nano 1 wt% agglomerated, (d) nano 1 wt% Octylsilane modified [39]

Table 2 Other studies on the XLPE-based nanocomposite

Material	Filler content	DC breakdown strength	Thermal activation energy	Space charge	Other significant result	References
XLPE/Al ₂ O ₃	10 wt%		0.99 eV			[57]
XLPE/MgO	3 wt%			75% suppression		[58]
XLPE/SiO ₂	2 wt%	increased			Temperature dependent conductivity	[59]
XLPE/BN	0.1 wt%			Suppressed space charge		[60]
XLPE/ZnO	2 wt%				1 wt% and 2 wt % improved the TIV	[61]
XLPE/SiO ₂	1.5 wt%				improve the tree growth resistance of the polymer	[62]

mass transfer due to its char structure with exfoliated silicate layers [53, 54] which also helps in flame retardancy mechanisms.

Said et al. [55] studied the clay XLPE composites with a different ratio. 1% clay composition exhibited a 14% increase in electrical properties, 20% higher of electrical capacitance and dielectric constant compared to pure XLPE. This structure material could, therefore, be used dominantly for nuclear power plant. Furthermore, the high voltage and low dielectric loss of XLPE cables insulation are one of the related important advantages. The higher of both resistance to thermal disfigurement and aging feature of the XLPE cable, permit it to carry large current under normal (90 °C), emergency (130 °C) or short circuit (250 °C) conditions [56].

Earlier reports have shown that the incorporation of nanoparticles (e.g., MgO, and SiO₂) which improved the electrical properties and reduced the space charge accumulation in the PE-based matrix [6, 7]. However, the high content of the nanoparticles had a negative impact on the PE matrix due to the introduction of impurities and defects [8] (Table 2).

6 Conclusion and Future Outlook

The addition of nanofillers to cross-linked polyethylene has gained attention in the high voltage research field toward the improvement of insulation materials since the beginning of the last century. The current need for research is the development of systems suitable for direct current–long distance transmissions. Such high-end application material needs to have good physical strength, degradation performance, and high insulation integrity while being available at an economic

economical cost. Many of the disadvantages of using PE-based cables have been covered by the introduction of XLPE-based nanocomposites. However, many questions are yet to be answered. The proper dispersion of the nanofillers needs to be ensured to obtain the enhanced properties, also the detailed mechanism of action of such composites needs to be studied. Hence, proper selection of the nanofiller, process and design are highly essential in order to extend the life of high voltage equipment and also to reduce the maintained cost.

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