Application of Cold Plasma in Nanofillers Surface Modification for Enhancement of Insulation Characteristics of Polymer Nanocomposites: A Review

NORHAFEZAIDI MAT SAMAN[ID], **MOHD HAFIZI AHMAD**[ID], **(Member, IEEE), AND ZOLKAFLE BUNTAT**

Faculty of Engineering, School of Electrical Engineering, Institute of High Voltage and High Current (IVAT), Universiti Teknologi Malaysia, Johor Bahru 81310, Malaysia

Corresponding author: Mohd Hafizi Ahmad (mohdhafizi@utm.my)

ABSTRACT Polymer nanocomposites have gained much attention among researchers due to their excellent characteristics, such as high thermal resistivity and the ability to withstand higher electrical stress. Introducing nanoparticles into the polymer matrix can further improve the insulation characteristics. However, the effectiveness of polymer nanocomposite to suppress an electrical tree, enhancing the partial discharge, and increase the breakdown strength is still a question mark due to the agglomeration issue of nanoparticles, which causing the nanoparticle to be dispersed non-uniformly within the polymer matrix. Plasma treatment is one of the methods that potentially replace conventional modification techniques such as chemical functionalization and heat treatment to improve the dispersion of nanoparticles, nanocomposite bonding, and the compatibility between polymer-nanoparticle interfaces. By altering the surface of nanoparticles using cold plasma with a glow discharge mechanism, the surface morphology of nanoparticles can be modified in terms of chemical structure, which indirectly solves the nanoparticle agglomeration. This study presents a review on the application of cold plasma in surface modification to enhance insulation characteristics of polymers nanocomposite. Various polymer hosts combined with different untreated and plasma-treated nanofillers are also reviewed. The trends on nanomaterial surface modification using the cold plasma technique based on its operating pressure to improve the dielectric properties of polymers nanocomposite are also discussed accordingly.

INDEX TERMS Cold plasma, nanofillers surface modification, polymer nanocomposites, plasma treatment.

I. INTRODUCTION

Recently, multiple studies and research of polymer nanocomposites have been conducted among the researchers to obtain the most usable and high-quality insulation medium for high and medium voltage application.

Polymer nanocomposites combines a polymer matrix and inclusions with at least one dimension such as length, width, or/and thickness in the nano-meter size range known as nanofillers [1]–[3]. Previously, micrometre size particles have been employed by several researchers to be used as

an additive material that combined with a pure polymer to improve the quality of insulator in several aspects such as to suppress an electrical tree prolongation, enhance partial discharge resistance, and increase the dielectric breakdown strength.

Nowadays, nanoparticles have become the size of choice for composites filler as they promise enhancement of the insulator's properties, and less amount is needed compared to micro-particles used in micro composites. Besides, nanocomposites also exhibit better mechanical, electrical, and thermal properties than micro composites [4], [5].

Based on previous research, the type of nanofiller used has been varied in terms of material, quantity, and mechanism

of surface modification to obtain different levels of electrical properties improvement. On the other hand, several types of polymer host are generally used as a subject in the study of nanocomposites such as low-density polyethylene, high-density polyethylene, cross-linked polyethylene, polypropylene, silicon rubber, and epoxy resin.

However, nanofiller surfaces and polymer matrix compatibility is the main issue due to the agglomeration of nanofiller [5]–[7]. Agglomeration of nanofiller formed due to the coarse accumulation of nanoparticles has affected the homogeneity of nanoparticles dispersed within the polymer matrix. The dispersion of nanofiller into polymer matrices became non-uniform due to the agglomeration of nanofillers [8]–[12]. Uniform and homogenous dispersion of nanofillers are crucial in enhancing polymers' nanocomposite insulation characteristics such as dielectric permittivity, space charge, direct current conductivity, dielectric losses, partial discharge resistance, dielectric breakdown strength, and electrical tree endurance.

In addition, the uniform dispersion of nanofillers into polymer matrices is also influenced by the surface characteristic of nanoparticles. Numerous comprehensive studies have revealed that uniform dispersion can be achieved by increasing the surface hydrophilicity of the nanoparticle [13]–[15]. Meanwhile, functionalizing a specific functional group such as carbonyl and carboxyl onto the nanoparticle's surface could increase the hydrophilicity of the nanoparticle, which leads to obtaining better uniformity of dispersion.

Furthermore, the insulation characteristics of polymers nanocomposite are also influenced by the characteristics of interfacial regions between organic and inorganic compounds. Other than uniformity of dispersion, interfacial bonds' strength is also the crucial factor in enhancing the insulation characteristics [16]–[18]. Preparation of polymer nanocomposite by using a direct mixing process without introducing surface modification technique might result in the formation of weak interfacial bonds and small deep trap sites, limiting the ability of organic-inorganic nanocomposites to suppress the space charge [19]–[22]. Thus, the insulation characteristics did not experience significant improvement compared to the pure host polymer.

The study on surface modification techniques has been intensively conducted since a few decades ago. The main purpose of those studies is to enhance the insulation characteristics by overcoming the agglomeration issue, increase the hydrophilicity of nanofiller surfaces, and strengthen the interfacial bonds. Surface modification can be categorized into three techniques: chemical treatment, heat treatment, and plasma treatment. Chemical treatment is the technique that uses chemical agents and/or solvents to functionalize certain chemical functional groups on the nanofiller surfaces since chemical treatment required chemical solvent, which was difficult to be decomposed and toxicity in some cases. Thus, the trend of surface modification study was shifted to heat and plasma treatment.

Heat treatment is also used to alter the surface morphology by drying the water molecule and moisture on the nanofillers. The justification of this technique is to reduce the hydrogen and hydroxide ions on the surface of the nanofiller to decrease the local electric field. This is because hydrogen and hydroxide ions have higher conductivity which may degrade the insulation characteristics of polymers nanocomposite [23]. However, this technique has a drawback that can damage the nanofiller's surface structure, especially when the amount of heat exposed onto the nanofiller exceeding the minimum amount of heat can be withstood by the nanofiller. Thus, cold plasma treatment seems to provide effective surface modification on the nanofiller by providing a self-repelling process to overcome the agglomeration of nanofiller and increase the hydrophilicity of nanofiller to enhance the dispersion uniformity.

Besides, cold plasma can also provide anchoring sites to enable covalent bonds between the organic and inorganic phases [24]–[27]. The insulation characteristics of polymers nanocomposite can be further improved by obtaining uniform and homogenous nanofiller dispersion in polymer matrices. Therefore, plasma treatment promises a reliable result in improving the insulation characteristics by reducing nanofiller agglomeration, functionalizing certain compatible functional groups, and strengthening the interfacial bonds.

This paper reports a review on the application of cold plasma treatment in nanofiller surface modification and the significant improvement of the insulation characteristics of polymer nanocomposite filled with plasma-treated nanoparticles. The effectiveness of cold plasma treatment has also been pointed out to give an overview sentiment for future study.

This scientific review began by describing the composition of polymer nanocomposites. The constituents, physical, chemical structures of the nanocomposites are also dismantled to deliver the basic concept of nano-composition. Also, the classification of nanocomposites material based on its natural characteristics of the compound has been described with an appropriate example of each listed class. The classification contents are structured in general so that a clear picture of the potential of nanocomposites with different classes, including their areas of application, can be distinguished and explained in detail.

This paper has been organized to describe the performances of dielectric insulations nanocomposites. In detail, the performance of nanocomposites was discussed according to the previous collective studies related to the investigation of electrical treeing prolongation, partial discharge endurance, and dielectric breakdown strength since these properties are required to be considered in designing a good insulation medium. The current limitation of the surface modification technique was also included in this review's structure, which explains the limitation of chemical functionalization and heat treatment technique in developing effectively treated nanocomposites insulating materials.

The limitation of cold plasma treatment was also critically reviewed at the culmination of this review based on justifications and claims made in the previous studies. The concept of cold plasma was also discussed as the main topic distinguished according to the operating condition of the plasma production. Besides, the effectiveness of cold plasma treatment as a surface modification technique to enhance insulation properties of the nanocomposites was also presented so that the recent performance of cold plasma treatment can be criticized scientifically.

The critical review of the nanocomposites has been summarized in the table to determine the trend of research regarding the insulation properties developed by using cold plasma treatment concerning the technique of surface modification. The climax of this review highlights the major challenges and future directions of cold plasma as an intensive technique in enhancing the insulation properties of polymer nanocomposites.

II. POLYMER NANOCOMPOSITES

A. CONSTITUENTS OF POLYMER NANOCOMPOSITES

Particularly, polymers nanocomposites have received much attention due to the addition of inorganic nanoparticles to develop insulation materials with tailored properties. A polymer nanocomposite is a composition that is formed by dispersing a certain amount of nanometer-sized fillers within a polymer matrix [1]–[3], [28], [29]. Fig. 1 illustrates the basic composition of polymers nanocomposite. The size of the nanofiller is normally from a few nanometers to 100 nm. The amount of the nanofiller distributed within the polymer matrix can be determined based on a measurement parameter known as weight percentages (*wt%*). Different amounts of nanofiller (weight percentage) used will produce different outcomes and change the properties of the nanocomposites. The main purpose of introducing nanofiller into polymer matrices is to enhance the insulation characteristics of polymers [19], [20], [28], [29]. The insulation characteristics promoted to be enhanced including dielectric breakdown strength, partial discharge resistance, dielectric losses, electrical tree prolongation, and thermal conductivity.
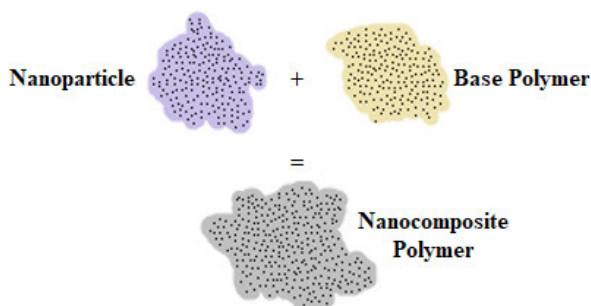

FIGURE 1. Composition of polymer nanocomposites.

Generally, polymers nanocomposites have three main constituents: polymer matrix, nanofiller, and interaction zone, as shown in Fig. 2. These three constituents are required to be
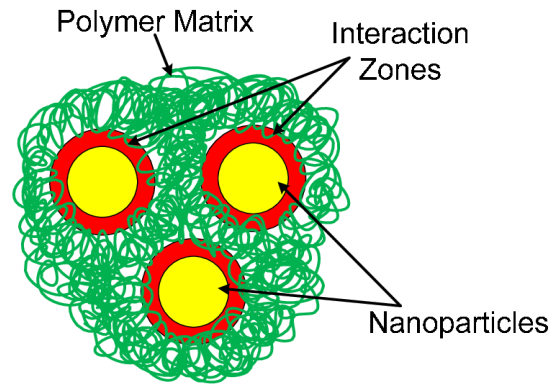

FIGURE 2. Constituents of polymer nanocomposites [33].

considered to understand the structure of polymer nanocomposites. Commonly, the nanofillers used to disperse into a polymer matrix consist of higher relative permittivity than the matrix substrate [30]. Besides, the interaction zone between the polymer matrix and nanofillers plays an important role in polymer property enhancement. Nanofillers also act as a barrier to the vibration of the polymer chain [31], [32].

Theoretically, the polymer chains will vibrate when the electrical stress is applied to the polymer insulation. The rapid vibration of polymer chains will initiate the sliding motion of chains, which causes the polymer chains to collapse and excite the electron avalanche to form across points where electrical stress is applied. Introducing nanofiller into polymer matrices is a comprehensive technique to reduce the vibration of polymer chains and obtain better stability of polymer chains.

Besides, nanofillers also work as a barrier to the injection of space charge to decrease the local electric field distortion. However, nanofillers' compatibility and polymer matrix are crucial for obtaining a well and homogeneous nanofiller dispersion into the polymer matrix. Thus, multiple surface modification techniques have been introduced to produce the most effective interaction zone, directly achieving a certain improvement level.

B. PHYSICAL STRUCTURE OF NANOFILLERS

Nanofillers are essential in determining the physical properties of polymers nanocomposites because they increase the molecular weight and limit the translational motions of the chains to each other [31]–[33].

Newly, the study on the type of nanofiller made from various materials has been intensively conducted among researchers worldwide to obtain the most effective nanofiller that can be used to form high-quality polymers nanocomposites. Generally, nanoparticles can be classified based on their dimension: zero-dimensional, one-dimensional, two-dimensional, or three-dimensional [34]. The illustration of nanofiller into those different dimensions is shown in Fig. 3.

In zero-dimensional (0D) nanomaterials, all dimensions are measured within the nanoscale (no dimensions are larger than 100 nm). Most commonly, 0D nanomaterials are
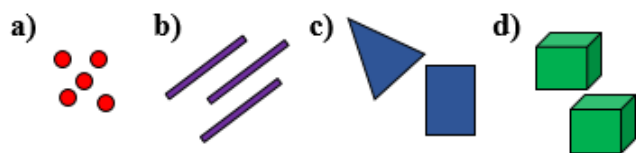
FIGURE 3. Classification of nanofiller (a) zero-dimensional, (b) one-dimensional, (c) two-dimensional, (d) three-dimensional (nano-meter sized of fillers which are less than 100 nm).

nanoparticles [34]. The characteristic of a one-dimensional (1D) nanofiller refers typically to the thin platelet or lamellar particles. Besides, two-dimensional (2D) nanofiller are represented as rod or/and whisker particles, while organic oxides in rectangular or spherical shapes can be classified into three-dimensional (3D) nanofiller.

As defined, the nanofillers added to the polymer matrices are very small in terms of quantity which is less than ten *wt*% compared to conventional microcomposites which, contain 50 *wt*% of micro fillers [4], [5], [35], [36]. Thus, polymer nanocomposites becomes selective due to the small quantity of nanofillers needed to obtain better properties enhancement than micro composites.

C. CHEMICAL STRUCTURE OF NANOFILLERS
Chemically, nanofillers can be categorized according to the chemical identities of the compound. In general, the category of nanofillers consists of carbon-based nanofillers, layered nanoclays, porous and hollow nanoparticles, nanocellulose, and nanoparticles of metallic alloys. Carbon nanotubes and graphene are among the types of carbon-based nanofillers that are often used to be dispersed to form comprehensive polymers nanocomposite. Carbon nanotubes consist of carbon fibres with nano-meter sized of diameter and micro-meter sized of length. The structure of carbon nanotubes consists of an enrolled graphitic sheet formed through the planar-hexagonal arrangement of carbon atoms distributed in a honeycomb-like lattice [37].

Carbon nanotubes can be classified depending on their preparation method. The classification of carbon nanotubes is either single-walled carbon nanotube or multi-walled carbon nanotube [38]. Single-walled carbon nanotube forms by rolling up a single graphene layer into a seamless cylinder [39]. In contrast, multi-walled carbon nanotube forms by arranging two or more concentric cylindrical shells of graphene sheets coaxially around a central hollow core. The properties of both classes of carbon nanotubes exhibit a strong covalent bond between carbon atoms and their arrangement in cylindrical nanostructures [40].

In contrast, graphene consists of a hexagonal arrangement of a single atomic thick sheet, in which the carbon atoms are covalently sp_2-bonded [41]. The primitive graphene cell comprises two non-equivalent atoms, and these two sub-lattices are translated from each other with 1.44 Å of carbons distance [40]. The preparation of graphene can be classified into several chemical intercalated graphite methods by using thermal expansion technique, the exfoliation of graphite

using a micromechanical process, deposition of chemical vapour, and chemical reduction of graphene oxide [42].

Nanoclays are also one of the types of nanofiller usually used to be filled into the polymer matrix. Generally, nanoclays are produced from layered silicates or mineral clays with traces of metal oxides and an organic compound. Hydrous aluminium phyllosilicates are a common type of clay mineral used as nanofillers with various iron, alkali metals, alkaline earth, magnesium, and other cations [43]. Montmorillonite, saponite, laponite, hectorite, sepiolite, and vermiculite are examples of nanoclays that are often used comprehensively [43]. Montmorillonite (MMT) consists of an alumina octahedral sheet flanked by two layers of tetrahedral silica sheets, in which hydrated exchangeable cations occupy the spaces between the lattices. Regular van der Waals gap, known as an interlayer or gallery, is formed due to the stacking of clay layers. The isomorphic substitution within the layers produced negative charges counterbalanced by the earth alkaline and alkali cations located inside the interlayers. The level of the negative charge of the nanoclay is characterized according to the capacity exchange of cation [44].

Porous and hollow nanoparticles also another type of nanofillers that have been chosen to be dispersed into the polymer matrix to enhance the properties of polymers nanocomposite. Halloysite nanotube is a hollow micro and nano-tubular structure of aluminosilicate [45]. Structurally, halloysite nanotube more likely to the kaolinite, which weakly held and led the intercalation of water molecules monolayer. It is composed of SiO_4 tetrahedral sheets with the shared edge of AlO_6 octahedral sheets. Thus, the chemical composition of the halloysite nanotube is almost like nanoclays, while the geometry of the nanotubular is almost similar to carbon nanotubes. Halloysite nanotube tubularly formed with the nano-meter scale of diameter and micro-meter scale of length. Zeolite is a faujasite-type framework of aluminosilicate with 1.3 nm of cavities diameter interconnected to the 0.74 nm of pores. The cubic cell of the aluminosilicates unit is composed of 192 (Si, Al)O_4 tetrahedrons [46]. Usually, only the large surface area of internal micro-porous of zeolites is considered, while the external surface area of micrometre-sized zeolites is neglected to design polymers nanocomposite [47].

Nanocellulose is a cellulose-derived material with at least one of its dimensions in the nano-meter scale. Recently, nano-cellulose has been listed as a comprehensive new bio-nanomaterial to fill into a polymer matrix, which promises a significant enhancement in insulating properties. The acid hydrolysis process is the key to form monocrystals cellulose by isolating the crystalline cellulosic regions [48]. Nanocellulose can be categorized into two chemical identities such as nanofibril and nanocrystal celluloses. Nanofibrils can be produced by applying the mechanical shearing force to a cellulosic fibre slurry. This process is called the de-structuration strategy. Generally, the mechanically induced de-structuration strategy is implemented using different shearing equipment such as homogenizer,

microfluidizer, or ultra-fine friction grinder. Nano-fibrillated cellulose is usually obtained as an aqueous suspension [49]. The width of nano-fibrillated cellulose is lying up to 100 nm with higher than one μm of the length depends on some factors such as the source of cellulose, defibrillation, and pre-treatment process [50].

In contrast, nano-crystallized cellulose is produced by applying a controlled strong acid hydrolysis treatment process to cellulosic fibres. This process will cause the dissolution of amorphous domains and longitudinally cutting the microfibrils. Thus, this process is known as the chemically induced de-structuration strategy. Nano-crystallized cellulose was also obtained as an aqueous suspension [50]. Commonly, the nano-crystallized cellulose tends to form rod-like nanocrystals or also known as whiskers. The geometrical dimension of nano-crystallized cellulose is affected by the source of cellulose and its conditions of hydrolysis. The production of nano-crystallized cellulose can be prepared from sulfuric acid. This process usually will form a sulphate functional group with negatively charged on the surface of nano-crystallized cellulose. The average length and width of nano-crystallized cellulose dimensionally up to a few nanometers [50]. In the selection procedure of nanoparticles, the ratio of the length to the width of nano-crystallized cellulose being the crucial parameter required to be considered to obtain a high surface area reaction [49].

Recently, the capability of metal hydrides alloys to enhance the properties of polymers nanocomposite has caused this type of nanofiller intensively used. The reversibly chemical reaction between metal hydrides alloys and hydrogen causes this type of nanofiller to be utilized in the solid-state storage of the gas. Metal hydrides alloys subjected to repeated hydride/dehydration cycles suffer from a pulverization phenomenon caused by a significant difference between hydride and the metallic compound [51]. Thus, repeated hydrogen loading/unloading cycles produce free metal powder particles on a nanometric scale.

D. CHEMICAL AND PHYSICAL COMPOSITION OF NANOFILLERS ON INSULATION CHARACTERISTICS

The incorporation of nanoparticles into polymer matrices has been explored as a strategy to produce nanocomposite materials with the superior enhancement of electrical, mechanical, and thermal properties. However, the prevalent issue that arises is the inappropriate nanofiller dispersion, which stifled the improvement of these properties. For instance, carbon nanotubes are one of the nanoparticles that difficult to be dispersed into polymer matrices. The agglomeration of carbon nanotubes during the low polymer/carbon nanotube preparation process causes the interaction between both materials to become weaker [52]. Thus, this clearly proved that the processing technique and conditions influencing the dispersion state of carbon nanotubes within polymer matrices. Therefore, the state of carbon nanotubes required to be modified to functionalize certain chemical functional groups. This indirectly able to improve interfacial interaction [52], [53].

The chemical reactivity between the carbon nanotubes and polymer matrix is increased by coating the chemical functional group on the surface of carbon nanotubes. Thus, the functionalized carbon nanotubes produced better interface regions, consequently forming a better self-mechanism of load transfer from the polymer matrix to the functionalized carbon nanotubes [40]. Chemical surface functionalization is a comprehensive technique implemented using nitric and sulfuric acids (HNO_3/H_2SO_4). This is a promising technique to functionalize carboxylic acid groups (-COOH) on the surface of carbon nanotubes [40]. As a result, functionalizing carboxylic acid groups on carbon nanotubes surfaces can alter the surface nature of carbon nanotubes, creating significant compatibility between carbon nanotube and the polymer matrix [54].

In improving the insulation properties of polymers nanocomposite, the employment of secondary particles such as clay is also another technique used intensively to achieve better dispersion of carbon nanotubes within polymer matrices [53]. Besides, instead of using a single type of polymer matrix, introducing blending polymer/polymer as a matrix also exhibits an excellent agreement to enhance the dispersion of carbon nanotubes. Polymer/polymer blends with carbon nanotubes have been observed to have better electrical and thermal properties compared to unfilled polymer/polymer blends [55]. The strength of electrical conductivity will be changed abruptly depending on the distribution and dispersion of carbon nanotubes within the polymer matrix due to the formation of a three-dimensional network of dispersed carbon nanotubes.

Literally, the electrical conductivity of polymers nanocomposite depends on the concentration of nanofillers. At the critical concentration of nanofiller, the electrical conductivity of the polymers nanocomposite is increased, surprisingly. This critical concentration of nanofiller is also inferred from the limit of the electrical percolation state. However, the further increases of carbon nanotubes dispersed within the polymer matrix exceed the critical concentration will cause a moderate increase in electrical conductivity [54], [55]. The amount of carbon nanotubes required to achieve an electrical percolation state for polymer/polymer blends usually less than the amount of carbon nanotubes needed to obtain the same state with a single polymer matrix. However, the region where the carbon nanotubes are dispersed must be selectively in the matrix phase or between the polymer/polymer blend [56]. The double phenomenon of electrical percolation can be achieved by forming the polymer/polymer blend nanocomposites with co-continuous morphology. Collectively, the concentration of carbon nanotubes and the final morphology of the polymer/polymer blends are crucial factors to the limit of the electrical percolation state. Both factors significantly act as a function of the nano-composition, determination of processing conditions, and compatibilizer steps [55].

One factor that drives graphene has been used intensively as a nanofiller to be dispersed within the polymer matrix

is the extremely high surface area with 2630 m^2g^{-1} [57]. Besides, graphene is also a promising carbon-based nanofiller due to the high thermal and electrical conductivity, such as 5000 Wm^{-1} and 6000 Scm^{-1}, respectively [57]. The nature of graphene with gas impermeability is also why this type of carbon-based nanofiller has been chosen to improve the insulation properties of the polymer. These properties are the primary consideration because graphene's overall factors greatly enhance the mechanical, electrical, thermal, and gas barrier properties of polymer nanocomposites [57].

Chemically, graphene's effectiveness as a nanofiller depends on the exfoliation process of bulk graphite to be translated into specific single sheets. The preparation process of individual exfoliated graphene sheets can be categorized into chemical and mechanical techniques such as chemical exfoliation, chemical vapour deposition, and mechanical exfoliation [58]. The effectiveness of each technique can be evaluated according to the results of chemical functional groups coated on the surface of graphene. However, graphene preparation usually involves the chemical oxidation process to produce graphite oxide first, followed by the reduction and mechanical exfoliation process [59]. Based on the chemical behaviour of graphite oxide, it is non-conductive and naturally will swell and disperse in water due to the hydrophilicity characteristic of its morphology. This carbon-based nanofiller seems to be modified by functionalizing the surface of graphene sheets with certain chemical functional groups to improve its dispersibility in polymers and organic solvents [57]. Thus, the better dispersion of graphene nanofillers can be achieved by overcoming the agglomeration issue, in which avoiding the formation of large trap sites of space charge on the dielectric materials to strengthen the dielectric field strength of polymers nanocomposite filled with nanometre-sized graphene.

Incorporating exfoliated nanoclays into the polymer matrix leads to an increase in the insulating properties, such as improving the stiffness, fire retardancy, and space charge barrier with only a small volume fraction of nanoparticles required [60]. Layered Nanoclays have proven effective in reinforcing fillers for a single or blend polymer because of their lamellar structure and high surface area with 750 m^2g^{-1} [61]. Generally, smectite clays or layered silicates have been chosen to be dispersed into the polymer matrix due to their superior properties such as swell, high cation exchange capacities, and cost-effectiveness. Besides, montmorillonite is another example of layered nanoclay that is intensively used to fill into the polymer matrix due to this nanofiller's beneficial properties, such as high surface and reactive area chemically well-known intercalation or exfoliation process, and high availability potential [62].

Naturally, the surface morphology of montmorillonite is hydrophilic, while characteristic most of the polymers tend to be hydrophobic. As a result, the dispersing process of montmorillonite into the polymer matrix is mismatched or incompatible, cause the nanofiller to agglomerate. Therefore, the surface modification technique must be done on the montmorillonite to form organophilic characteristics of nanoclay, which may improve the compatibility of nanocomposites mixture. Regularly, the surface modification technique executed to alter the surface morphology of layered nanoclays is the cations exchanging process in the interaction regions by using alkylammonium or alkyl-phosphonium salts as dioctadecyl dimethyl ammonium bromide. The exchange of inorganic cations with organic ions on the surface galleries of layered nanoclays effectively extends the nanoclays exterior corridor. As a result, the layered nanoclays will be intercalated easily into the polymer chains.

In addition, silanization is also a comprehensive modification technique used to alter the properties of layered nanoclays. This technique uses a reactive extrusion process to provide strong interactions between the nanoclay and the polymer matrix through the fostered covalent bonds during the modification process of nanoclay with organosilanes. For instance, 3-aminopropyltrimethoxysilane (APTES) and vinyltrimethoxysilane (VTMS) are widely used as silane agents to modify the surface galleries of layered nanoclays [63]. Consequently, the modified-layered nanoclays to be dispersed into the polymer matrix will have better insulation properties due to the formation of strong covalent bonds and well-dispersion nanoclays, which may improve insulation diagnosis properties as partial discharge endurance, dielectric field strength, and electrical treeing prolongation.

Porous and hollow nanoparticles are also categorized as good nanofillers since they promise innovative possibilities for preparing polymers nanocomposite [64]. In polymers nanocomposite, halloysite nanotube is one of the fillers found to have a great potential to increase mechanical, thermal, electrical, non-flammability significantly [64]. Besides, halloysite nanotube is also being used intensively in many applications by considering the shape, surface area, size, and length to diameter ratio. In addition, zeolite is another type of porous and hollow nanoparticle that promotes an improvement on the polymeric materials by enhancing the space of intercrystalline, forming a better external and internal surface area creating the volume and pores of the exposed muzzles. As a result, the insulation properties of filled polymeric materials experienced a significant improvement in terms of endurance, especially.

The nanofibril and nanocrystal celluloses are the nano-compounds with such renewable, recyclable, non-toxic, and sustainable properties because carbon footprints are low. Hence, both types of cellulose nanoparticles can be concluded as green nanofillers that significantly enhance polymeric materials. In nanoparticles of metallic alloys, the fragmentation of particles conductively may increase the surface area of the metal particles, and this consequently the properties of polymeric materials gaining a comprehensive improvement after dispersing this type of nanoparticle into the matrix.

E. CLASSIFICATION OF NANOCOMPOSITES MATERIAL

Nanocomposites can be categorized based on functional materials such as electrical, thermal, optical, electrochemical, and catalytic behaviours. However, in certain cases, nanocomposites can also be categorized based on the structure of the materials, which contribute more to their mechanical properties. Different component materials of nanocomposites would obtain different functional determination.

Another way to classify nanocomposites is through the identity of composition materials. Nanocomposites can be classified into several classes: polymer/ceramic nanocomposite, inorganic/organic polymer nanocomposite, inorganic/organic hybrid nanocomposite, polymer/layered silicate nanocomposite, polymer/polymer nanocomposite, and bio-nanocomposite.

Polymer/ceramic nanocomposite comprises ceramic materials such as silica and bioactive glasses used as nanofillers dispersed within the pure polymer. For instance, low-density polyethylene/silica nanocomposite and K-carrageenan/calcium phosphate nanocomposite is the polymer/ceramic nanocomposite that shows properties intermediate between organic and inorganic polymers ceramics. The primary goal of preparing polymer/ceramic nanocomposite is to achieve combinations of properties from polymer and ceramic components [65].

Besides, inorganic/organic polymer nanocomposite comprises a polymeric matrix and a plurality of metal nanoparticles. Epoxy/clay nanocomposite is an example of inorganic/organic polymer nanocomposite. Generally, the organic component is referred to as the polymer host, and the inorganic component is referred to as the nanoscale materials. This combination brings together the advantages of both components.

Inorganic material is a good component in terms of rigidity and thermal stability [66], [67]. In comparison, the organic polymer is an exciting material to be used as an insulation medium due to its flexibility, ductility, and processability and can be an effective dielectric medium [68], [69]. Thus, introducing inorganic nanoscale fillers into organic polymer hosts is intended to achieve both component properties' advantages.

Hectorite and montmorillonite are among the most commonly used smectite-type layered silicates to prepare polymer/layered silicate nanocomposites. The main reason why layered silicates are coated onto the nanofiller surfaces is that nanofiller's hydrophilic characteristic in nature makes them difficult to disperse uniformly into a polymer matrix [70]. Thus, silicate layers coated onto nanofiller surfaces would result in better dispersion, enhancing the properties of this nanocomposite due to biocompatibility between layered silicate nanofiller and polymer matrix [71].

Polymer/polymer nanocomposite is a composition that combines two substrates of polymers. Commonly, polymer/polymer nanocomposites are prepared by using in situ polymerization technique. The aims of producing this composition are to obtain a nanocomposite using both polymer properties [72].

The study trend regarding this composition also focuses on confining the polymerized phase to the nanoscale domain and characterizing the dispersion. Recent progress demonstrates polymer blends and interpenetrating networks via supercritical carbon dioxide substrates such as polystyrene-based. However, this composition is slightly more challenging to prepare due to the larger polymer chain size and higher mobility of the product due to stronger interactions with the matrix phase.

Recently, inorganic/organic hybrid nanocomposites have drawn special attention due to their extraordinary properties and widespread applications in diverse fields such as dielectric insulation, cell imaging, drug delivery, photo-thermal therapy, bio-sensing, catalysis, energy storage, and gas sensing. In some literature, inorganic/organic hybrid nanocomposites are also referred to as polymers nanocomposite that are filled by two or more types of nanofillers [73]–[76]. The main purpose of this composition is to obtain a better performance of dielectric insulation depending on the properties of multiple types of nanofillers used to be dispersed into the matrix. For instance, Chao *et al.* has used silicon nanoparticles-modified epoxy as a dispersing agent to obtain a uniform dispersion of silver nanowires into the epoxy matrix [73]. Inorganic/organic hybrid nanocomposites are promising polymers nanocomposite, consisting of great properties such as high thermal conductivity, high glass transition temperature, high electrical insulation, and low viscosity [73], [74]. However, this category of polymers nanocomposite is rarely used in designing dielectric insulations. In addition, inorganic/organic hybrid nanocomposites also appeared as active layers for electronic devices, such as memristor and transistor. The term 'hybrid' refers to the combination of inorganic and organic components that form a single material called nanocomposite with improved and unique properties than the individual components. The purpose of this combination is also to obtain a composition with both component properties. Graphene is the most recognized as one of the potential organic components, whereas silica is a popular material of inorganic components for inorganic/organic hybrid nanocomposites. Ziyu *et al.* [77] have introduced benign carbon dots/silk protein hybrid nanocomposites as a phototunable carrier trapping to fabricate an optical transistor synapse, which provides a comprehensive avenue for bio-inspired neuromorphic computing. The study revealed that after doping carbon dots into silk film, the potential value of the non-projected zone has decreased, and the potential change induced by electron injection has increased, implying that the carbon dots play a vital role in the carbon dots/silk hybrid nanocomposites which work as a mechanism of charge-trapping in the development of phototunable bio-electronic memory devices [77], [78]. In another study, Lv *et al.* also pointed

out that inorganic/organic hybrid nanocomposites such as all-inorganic caesium lead bromide (CsPbBr$_3$) perovskite quantum dots/poly-methyl methacrylate (PMMA) hybrid nanocomposites film effectively trap charge to dominate the properties of the photonic synapse [79].

Like other types of nanocomposites, the properties of an inorganic/organic hybrid nanocomposite are also dependent upon the particle morphology and interfacial region formed between both components [80]. Thus, the study trend regarding this nanocomposite class focuses on the synthesis and surface modification approach to obtain a compatible mixture between organic and inorganic compounds.

Bio-nanocomposite is also one of the nanocomposites developed for application in dielectric insulation, food packaging, biomedical, and pharmaceutical. In the last few decades, nano cellulose-based bio-nanocomposite has a great potential to incorporate with polymer matrix, which promises an effective improvement of insulation endurance. Aulia *et al.* conducted a study to identify the tensile properties of bio-nanocomposites polymer by dispersing two types of nanofillers which is nano-silica and nano-alumina powder, into bio-polymeric materials produced by blending low-density polyethylene and silicone rubber, in which the results obtained exhibits the addition of nanofillers into bio-polymeric insulator resulted into stiffer than unfilled composition [81]. For instance, bio-nanocomposite can be prepared by mixing the bio-polymer and solution of swollen nanoparticles. Through that, the polymer chains will intercalate and displacing the solvent with the interlayer of the silicate. Upon solvent removal, the intercalated structure remains, forming a bio-polymer/layered silicate bio-nanocomposite. Same as previous, bio-nanocomposite polymers are also rarely chosen as insulation material compared to other classes. In addition, the bio-nanocomposite is an ecological strategy for a greener electronic and dielectric insulation medium. Xing *et al.* pointed out that bio-nanocomposite material system has been applied in various areas, including resistive memory, flash memory, nano-biotechnology, biosensors, biofuel cells, artificial organs, and artificial synapses, and artificial neural network simulation [82]. In fact, introducing nanoparticles into bio-hybrid systems may generate new mutant materials and add new perspectives to the development of materials science and organic electronics [82].

III. DIELECTRIC INSULATION OF POLYMER NANOCOMPOSITES

Based on the previous research that has been conducted regarding the nanocomposites study. The most common nanoparticle used as a filler in nanocomposites for electrical insulation is carbon nanotubes, silicon dioxide, nanoclay (i.e., montmorillonite, organomodified nanoclay, sepiolite, etc.), and metallic oxides (i.e., zinc oxide, alumina titanium oxide, etc.).

Calebrese *et al.* [83] reported that silica nanofillers had been used widely together with numerous polymer matrices,

and the composition has shown positive improvement significantly. Besides silica, the other nanoparticle that can be categorized into metallic oxide is also used as a nanofiller to develop dielectric nanostructure materials. Metal oxide can be considered a good nanofiller to be merged within a polymer matrix since they have good electrical insulating behaviour and can be dispersed easily due to their hydrophilic characteristic [84], [85].

Metal oxide is a group of nanoparticles that consist of a combination of metal (positively charged ion, cation) and oxygen (negatively charged ion, anion) elements. The most common metal oxide used as a nanofiller is magnesium oxide, zinc oxide, and alumina. Those metal oxides usually can be produced into both quasi-spherical shapes and whisker particles. The metal element and oxygen element are chemically bonded as an ionic bond in terms of the chemical bond. Based on previous research, metal oxide has promising improvement in thermal conductivity, whereas the nanocomposite's ability to sustain the high operating temperature of medium and high voltage cable [86], [87].

A. ELECTRICAL TREEING

In the study of electrical trees, the historical development of electrical insulation has mainly been based on the introduction of polymeric materials, which often contain various additives. Polymer nanocomposite is another initiative to improve the performance of polymer insulation [19]–[22], [28], [29]. Recent research showed that the nanofiller with appropriate surface compatibility with polymer matrices could improve the performance of polymer insulation, especially in terms of electrical tree prolongation.

Zheng *et al.* [21] employed a density functional theory method in their study to investigate the significant role of nanometer-sized silica as a voltage stabilizer for power cable insulation. The study was found that the nano-silica additive can restrict the movement of the polyethylene chain through Van Der Waals physical interaction. In addition, the particular modified surface of silica, such as incompletely hydroxylated, boron-doped, and oxygen vacancy defect on the top layer of nanosilica, could induce the migration reaction and consequently affecting electrical tree growth in the cross-linked polyethylene chain [21].

The nanoscale additives can effectively suppress the space charge accumulation and inhibit the electrical tree growth of power cable insulation by constraining the movement of the polymer chain and trapping hot electrons [21], [22]. Hot electrons are usually formed due to the energy conversion from kinetic energy (movement of a polymer chain) to heat [21], [22]. In this study, nitrogen-doped silica with a completely hydroxylated surface, such as shown in Fig. 4, is the most promising additive due to its strongest adsorption abilities to cross-linked polyethylene and transferring charge as well as the weakest chemical activity.

Su *et al.* [88] also conducted an extensive study to identify tree characteristics in silica/silicone rubber nanocomposites under low temperatures. The study reported that the
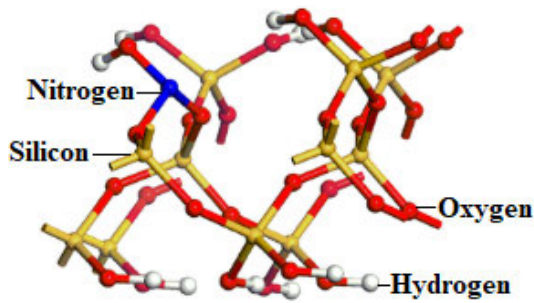
FIGURE 4. The nanosilica model with nitrogen-doped and complete surface hydroxylation [21].

addition of nanoparticles into silicone rubber could improve the insulating properties compared with undoped material. The samples were prepared by mixing nano-silica into room temperature vulcanized (RTV) silicone rubber, containing 0, 0.5, 1.0, 1.5, and 2.0 *wt%*. The experiment was conducted in the range of temperature from −30 °C to −90 °C. Alternating current voltage with a frequency of 50 Hz was applied between a pair of needle-plate electrodes to initiate the electrical tree at different experiment temperatures. The experiment results indicated that both nanoparticles and low temperature are important factors of the treeing process in silica/silicone rubber nanocomposites [88]–[90]. The distribution of tree structures depends on the content of nanoparticles and temperature [88]–[90]. Nano-silica can effectively repress tree growth, and the optimum content with the lowest tree growth speed is 1.5 *wt%* [88]. Crystallization caused by the changing temperature also influences the treeing process [88]–[90].

Alapati and Thomas [91] performed electrical treeing studies in polymer nanocomposites. In the study, epoxy resin was chosen as a host polymer, while nano-silica was used to fill within the epoxy resin matrix. Treeing experiments were performed at a constant alternating current voltage of 20kV, 50Hz on epoxy samples without any fillers and silica/epoxy nanocomposites with 1% by weight nano-silica. Times for tree inception and tree growth patterns were studied. The results show that adding a small amount (1% by weight) of nano-silica particles in the epoxy resin can improve the treeing resistance by delaying the tree inception time and the time required by the tree to reach the opposite electrode [91].

The initiation of electrical trees usually formed due to the existence of the void, crack, or/and inclusion between the surface of needle tip and nanocomposites polymer [91]–[93]. After the initiation of the tree, it starts to grow through the base polymer and continues to propagate towards the nanoparticle due to the local enhancement in the electric field [91]–[93]. The presence of hydroxyl groups on the nanoparticle surface forms hydrogen bonding with the polar groups present on the base polymer. It has been reported that hydrogen bonding forms a weak chemical bond with conductive nature because of polar groups [91]. The interfacial region between the nanoparticle and polymer
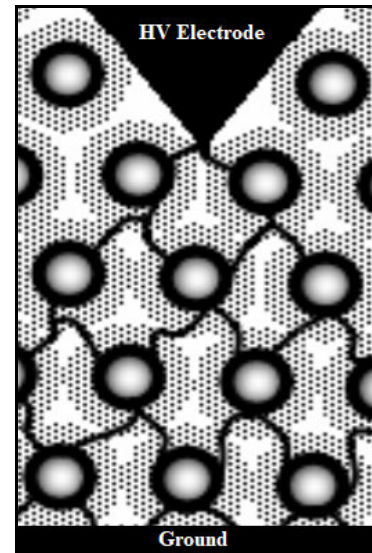
FIGURE 5. Physical model of electrical tree growth in polymer nanocomposites [91].

matrix has higher conductivity than the polymer base or the nanofiller because the density of hydrogen bond forms near the nanoparticle surface is higher than in the other region [91].

Once the tree channel reaches the nanoparticle, it propagates through the interfacial region due to the conductive nature of that region. After propagating along the interfacial region, the tree channel continuously propagates to another nanofiller through the polymer base. Thus, the pattern of tree propagation tends to be more *zigzag* and slow down the tree propagation with a greater number of branches, as shown in Fig. 5. The tree channel propagates around the nanoparticles and erodes the polymer region around the nanoparticles due to continuous discharges inside the tree channel [91]. When the tree channel diameter reaches the inter-particle distance, the nanoparticles enter the tree channel. The ingress of nanoparticles further delays the tree propagation by obstructing the discharge avalanche inside the tree channel [91]–[93].

B. PARTIAL DISCHARGE
Polymer nanocomposites have recently attracted attention among researchers in electrical insulating applications from energy storage to power delivery. However, the partial discharge has always been a predecessor to major faults and problems in this field. In addition, there is a lot more to explore, as neither the partial discharge characteristic in nanocomposites nor their electrical properties are not clearly understood. For instance, nanofillers in nanocomposites such as silica, alumina, and Titania play a significant role in providing a good approach to increase the partial discharge resistance of nanocomposites [94].

Tanaka [95] carried out a comprehensive experimental investigation of cross-linked polyethylene and its nanocomposite with fumed silica in the CIGRE Working Group

D1.24 cooperative tests conducted by members covering partial discharge resistance. The experiment was set up to measure the erosion speed due to partial discharge events. A voltage of 10 kVrms (250 Hz) was applied to three kinds of cross-linked polyethylene samples of 1 mm thickness for time up to 750 h (50 Hz equivalent time). Applied voltage corresponds to 5 times partial discharge inception threshold. A needle-plane electrode system with an air gap of 0.1 mm and a tungsten needle electrode of 1 mmφ diameter for partial discharge erosion evaluation. The result obtained in this study showed that filled cross-linked polyethylene exhibits much-improved endurance to partial discharge (i.e., 0.001 mm^2/h of average erosion speed) than unfilled cross-linked polyethylene (i.e., 0.0016 mm^2/h of average erosion speed), and the best performance was possessed by cross-linked polyethylene filled with surface-heat treated filler (i.e., 0.0006 mm^2/h of average erosion speed) [95].

In another study, Tanaka *et al.* [96] also performed research to enhance partial discharge resistance of epoxy/clay nanocomposite prepared by newly developed organic modification and solubilization methods. In the study, frequency accelerated partial discharge ageing of epoxy nanocomposite with 5 *wt%* additions of clay was investigated compared to epoxy without clay in partial discharge erosion depth. Layered silicate (LS) or clay is organically modified through hydrochloric acid processing and ion exchange processing by organic modification or solubilization methods. In the organic modification method, layered clay is organically modified with octa-decyl amine without a swelling process added and dispersed to neat epoxy.

On the contrary, organically modified clay is swollen in dimethylacetamide (DMAc) to be added into neat epoxy in the solubilization method. Scanning electron microscopy observation was made to evaluate the degree of dispersion of layered clay in nanocomposite specimens prepared by the solubilization method. In the scanning electron microscopy photo, nanofillers did not agglomerate in structure and were comparatively homogeneously dispersed [96]. The surface of the specimen is eroded due to partial discharge exposure, and erosion depth is selected as a measure of partial discharge resistance. The result confirmed that even the small content like 2 *wt%* would work well against partial discharge degradation [96]. It was shown that the highest partial discharge resistance was indicated by micro-nano mixed epoxy nanocomposite followed by solubilization method nanocomposite and organic modification method nanocomposite. While pure epoxy (reference erosion depth) showed the lowest partial discharge resistance.

Nanoparticles consist of three-layered cores with different characteristics and morphology compared to polymer matrices. Each core has different roles, but only two roles are distinctive and prominent for the characteristics of partial discharge resistance. The first role indicates the direct process where the base polymers are segmented into three-dimensional nano-meter scales by the nanofillers [95], [96].

The second role indicates an indirect process and an important process where the ability of nanofillers to hold physical tightness with pure polymer matrix and apparent enlarged volume due to the formation of interaction zones [95], [96]. Both aspects (refer to the first and second roles) must be mutually coordinated to increase partial discharge resistance. The formation of three cores would reduce the interstices between neighbouring fillers [95], [96]. Consequently, the strength of polymers nanocomposite in terms of partial discharge endurance can intensify.

The erosion mainly happens in the polymer region of nanocomposite substances. This is because inorganic nanofillers are far more partial discharge resistive than organic polymers [95], [96]. Thus, the partial discharge will attack pure organic polymer regions preferentially. Through that, the segmentation of organic substances (pure polymer) into many small areas and volume is effective against partial discharge attack [95], [96]. This configuration will help to increase partial discharge resistance.

C. DIELECTRIC BREAKDOWN STRENGTH

As mentioned, dielectric breakdown strength is one of the most important properties of insulation material. The dielectric breakdown strength of nanocomposites depends heavily on the nanofiller content, and even small quantities can cause improvement [97], [98]. This is linked to the maximum interface volume achieved already at low nanofiller concentrations. Homogeneous nano-dispersion is the key for dielectric breakdown strength increase and reliable results [97], [98]. Often the most profitable nanofiller quantities are below 5 *wt%* [97]. The same kind of behaviour can be seen with different voltage shapes, but the increase is more significant with direct current voltage. Dielectric breakdown strength has a maximum value concerning the nanofiller amount (below 5 *wt%*) and decreases after that. Filler quantities around 5 *wt%* and even lower are the most attractive considering the dielectric breakdown strength of nanocomposites [97].

Tanaka [95] experimented with investigating the dielectric breakdown strength of cross-linked polyethylene/silica nanocomposites. Alternating current dielectric breakdown strength tests were performed using a test cell with 2 spherical electrodes (diameter 30 mm) in silicone oil at room temperature. Data were obtained in the form of two parameters of the Weibull distribution (i.e., the scale parameter and the shape parameter). They represent the breakdown strength α [kVrms/mm] and the data scatter β, respectively. The applied voltage was increased at the rate of 500 Vrms/s (60 Hz). About 40 numbers of breakdown measurements were made on each of the 3 samples ($\sim$1 mm thick), and data outliers were removed from statistics. The results obtained showed that the highest alternating current breakdown strength was obtained with cross-linked polyethylene filled by functionalized nanofiller after heat treatment [95].

IV. RELATIONSHIP BETWEEN THE PHYSICAL AND CHEMICAL COMPOSITIONS OF POLYMER NANOCOMPOSITES AND INSULATION CHARACTERISTICS

A. PHYSICAL COMPOSITIONS OF POLYMER NANOCOMPOSITES

The main purpose of incorporating the nanofillers into the polymer matrix is to combine insulation properties in between the nanofillers and the neat polymer. Hence, the enhancement of insulation properties promised by adding the nanofillers into the matrix must be understood physically. A few physical factors must be considered to produce a better performance of insulation properties, especially in terms of electrical properties. Structurally, interfacial regions formed in between the nanofillers and polymer matrix is the crucial factor that contributes to the significant improvement of insulating properties [99]–[101]. This is related to the radius size of the fillers, in which reducing the size of the fillers forms a larger specific surface area, and this physically will increase the formation of interfacial area. Thus, the interfacial area between the nanofillers and polymer matrix will have a higher surface-to-volume ratio [102]. As a result, the insulation properties of the polymer nanocomposites will further extend to the comprehensive level.

The interfacial region formed through the incorporating process act as a barrier to the transported charge. Charge transportation is an important aspect required to enhance the dielectric field strength and the electrical erosion of the polymer nanocomposites [103], [104]. This has been proven by Montanari *et al.* [105] that pointed out that the new relaxation process has occurred related to the charge trapping at the interfacial region. Consequently, reducing the transportation charge due to the formation of the interfacial region also affects the magnitude of interfacial charge by exhibiting fewer charge forms in the polymer nanocomposites [100]. Besides, the decay of dynamic charges faster in the polymer nanocomposites than the pure polymer due to the formation of the interfacial region effectively serves as a barrier to the charge carried in the insulation materials [100]. Besides, the existence of nanofillers in the polymer matrix also makes the space charge accumulated on the polymer nanocomposites become lower and redistributed due to the presence of homopolar charge adjacent to the cathode [106]. This could be why nano-meter scale fillers are introduced to replace micro-meter scale fillers since space charge accumulation is higher and not redistributed due to the charge adjacent to the cathode being heteropolar.

However, the concentration of nanofillers is required to be considered to produce optimum and comprehensive enhancement of insulation properties. As referring to the physical conception, the interfacial region being the crucial constituent in producing the most effective composition of polymer nanocomposites. In fact, the concentration or amount of nanofillers incorporated into the polymer matrix affected the interfacial region formed. The higher the amount of nanofillers dispersed into the polymer matrix, the larger specific interfacial area will be formed. However, there is

a specific limit of the amount of nanofillers called critical loading, which indicates the maximum amount of nanofillers that need to be adhered to obtain an improvement in the insulating properties effectively.

Previously, in some literature, the critical amount of nanofillers defined as not to exceed 10 wt% because the further increase might cause an adverse effect to the properties [107]. Related to the interfacial regions, dispersing more than 10 wt% of nanofillers might cause the nanofillers distributed too close to one another. This might cause the self-agglomeration of filled nanoparticles and consequently reducing the specific interfacial region. Worse, this may destroy the natural properties of polymer nanocomposites as insulators because reducing interfacial region will cause the mechanism of trapping charge to occur ineffectively as compared to larger interfacial region formed with the loading of nanofiller less than 10 wt%.

In addition, the agglomeration of nanofiller also occurs due to the imperfection of the processing/dispersion technique [108]. The agglomeration of nanofillers in a polymer matrix will cause the formation of the interfacial region to reduce, usually indicated by the decrement of surface area to volume ratio significantly. This also might compromise the unique nanofiller properties in the polymer. Therefore, the processing technique needs to be executed in detail by considering mechanical mixing and ultrasonication-mechanical mixing technique in a high shear force mechanical mixer [108]. Follow by the second stage of dispersion by using ultrasonic agitation. The trapped air bubbles and moisture also need to be removed through the degassing process [108]. Furthermore, nanofillers' agglomeration can also be resolved using the surface functionalization technique discussed comprehensively in this review.

The interfacial region has been pointed out as a key in improving insulating properties due to its role in trapping the charges distributed in the polymer nanocomposites. The trapping charge mechanism will reduce the accumulation of space charge and minimize the distortion of the local electric field. This brings positive implications to the insulation properties, such as improving the relative permittivity, dielectric losses, dielectric breakdown strength, dissipation factor, partial discharge, surface tracking, erosion resistance, and electrical treeing prolongation.

B. CHEMICAL COMPOSITIONS OF POLYMER NANOCOMPOSITES

The chemical composition of polymer nanocomposites is closely related to its physical composition. As highlighted, the agglomeration of nanofillers in a polymer matrix being a major factor stifling the improvement of insulation properties of polymer nanocomposites. Commonly, the agglomeration of nanofillers is happened due to the incompatibility between the surface of nanofiller and polymer matrix chemically [5]–[12]. Naturally, most nanofillers' surface morphology is hydrophilic, while most of the polymers tend to be hydrophobic [51]. Hydrophilic is a characteristic

or entity of a molecule that chemically reactive to the water molecules and tends to be dissolved by water molecule [13]–[15], [70], [84], [85]. At the same time, hydrophobic is a property of a molecule that chemically not undergoing a reaction with water molecule [109], [110]. Thus, the dispersing process of nanofillers into the polymer matrix is mismatched or incompatible, cause the nanofiller to agglomerate. Organophilic represents the molecules on which the surface consists of covalently linked organic moieties [51]. Hence, the surface modification technique must be done on the nanofillers to form organophilic characteristics, which may improve the compatibility of polymer nanocomposites mixture [51].

Chemical modification is a comprehensive technique widely used since this technique significantly improves the insulation properties of polymer nanocomposites through the covalent attachment modifier that prevents desorption from the particle surface [111]. The modification of the interaction region by using chemical pre-treated nanofillers can be done by functionalizing modifiers containing reactive groups. This technique effectively constructs chemical bonding between the nanofillers and the polymer matrix, indirectly strengthening the merging mechanism of the nanocomposites mixture [111]. The coupling agent is the most intensive treatment used to enhance the compatibility between inorganic fillers and organic matrices. The ubiquitous coupling agents used for this purpose are silane, titanite, and zirconate. Collectively, the attachment of silanes on the nanofillers offers the reduction of hydrophilic properties of the nanofillers [63], [112]. In addition, grafting macromolecules onto inorganic particles is also a technique implemented to alter the interfacial characteristics between treated nanofillers and the polymer matrix. The grafted polymer properties can be strategized depending on the graft monomers' species and the grafting process's condition. Furthermore, the grafting technique also capable of strengthening the fragile agglomerated nanoparticles and forms a microstructure of nanocomposites that consists of nanofillers and homo-polymerized polymer.

The compatibility of nanofillers and polymer matrix also can be achieved by functionalizing a chemical functional group on the surface of nanofillers such as hydroxyl (OH-) and carboxyl (C(=O)OH) functional group [21], [113]. This technique can be done by using either chemical functionalization or plasma treatment. The functionalized chemical groups will further strengthen the interfacial region between the nanofillers and polymer matrix by forming covalent bonds; thus, a rigid interfacial region can be produced to maximize the trapping charge mechanism [108]. Hence, improve the insulation properties by minimizing the distortion of the local electric field on the polymer nanocomposites.

V. LIMITATION OF SURFACE MODIFICATION TECHNIQUES

Surface modification is one of the subjects required to be considered to obtain effective insulation performance of poly-

mer nanocomposite regardless of the types of polymer host and nanofiller used to form the nanocomposites. Several surface modification techniques are currently used to alter nanofillers' surface morphology, such as surface functionalization, coupling agent, intercalation, heat treatment, plasma polymerization, and plasma treatment. The purpose of all techniques is to enhance nanofiller surface structure by overcoming the agglomeration of nanofiller and incompatibility between nanofiller and base polymer surfaces.

Fig. 6 shows the model of agglomerated polymers nanocomposite. Specifically, the enhancement of insulation performance can be achieved by resolving agglomeration issues and creating compatible surface structures between nanofiller-base polymers [5]–[12]. Fig. 7 shows the model of a compatible mixture between surface-treated nanofillers and polymer matrices. For instance, the insulation characteristics promising improvement through surface modification techniques are partial discharge, dielectric breakdown strength, electrical tree, space charge, etc.
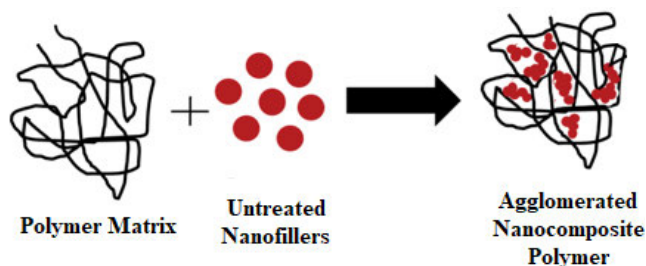

FIGURE 6. The model of agglomerated polymer nanocomposites filled with untreated nanomaterials.
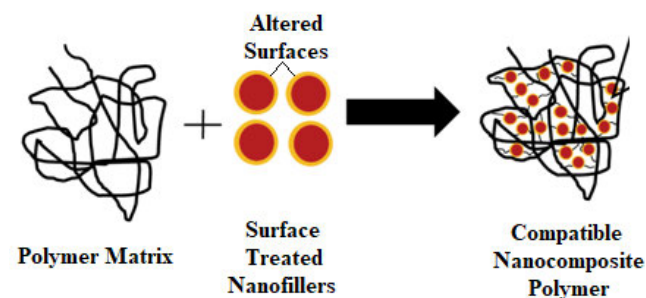

FIGURE 7. The model of compatible polymer nanocomposites filled with surface treated nanomaterials.

A. LIMITATION OF CHEMICAL FUNCTIONALIZATION

Surface functionalization, coupling agent, intercalation, and plasma polymerization are techniques that use chemical agents and/or solvents to functionalize certain chemical functional groups on the nanofiller surfaces [114]–[116]. Since this method required chemical solvent, which is difficult to decompose and toxic in some cases, surface functionalization methods are not suggested to be implemented, especially in the manufacturing industry of cable. Besides, the by-products formed during the chemical reaction of those techniques are

not guaranteed either harmful or not to the human and eco-environment. Thus, heat treatment and plasma treatment have been introduced to replace those techniques which are more eco-friendly.

B. LIMITATION OF HEAT TREATMENT

Heat treatment is also one of the techniques used to alter the surface morphology of the nanofiller. This is intended to obtain a uniform dispersion of nanofiller within the polymer matrix. However, this technique has a drawback that can damage the nanofiller's surface structure, especially when the amount of heat exposure on the nanofiller exceeds the minimum amount of heat that can be detained by the nanofiller [117]. Thus, an alternative to those techniques explained above is plasma treatment which deemed to provide an effective surface modification process on the nanofiller due to eco-friendly, providing a self-repelling process, high effectiveness, effective to form and strengthen interfacial covalent bonds but has not been explored thoroughly in this regard [112], [118], [119].

C. LIMITATION OF PLASMA TREATMENT

Dealing with the limitation of plasma treatment systems is a challenging task when choosing plasma treatment as a nanofiller surface modification technique. Cold plasma has been practiced widely in the study of nanofiller surface modification. However, the previous collective studies only highlighted the duration of treatment with filamentary plasma discharge. Only a limited number of studies are concerned about the discharge mechanism toward the effectiveness of insulation performances. Glow plasma discharge is a uniform discharge, while filamentary plasma discharge is a non-uniform discharge of gases [120]. Uniform discharge is expected to overcome incompatibility issues effectively compared to non-uniform discharge.

Nowadays, plasma technology has been chosen as an alternative method to improve the uniformity of nanoparticle dispersion within polymer matrices. Plasma treatment also helps to improve the molecular structure of nanofillers so that the distribution of nanoparticles within the polymer matrices is dispersed homogeneously [112], [118], [119]. As per our knowledge, the specific design of the cold plasma system for this purpose still has not existed yet. Therefore, the most effective cold plasma system with a glow discharge mechanism must be designed and developed by concerning any other factors such as optimum voltage and frequency of plasma generation and the structure of the plasma chamber.

Plasma surface treatment is also being employed in numerous researches to modify the structural surface of nanoparticles. However, a limited study was conducted which employed cold plasma with a glow discharge mechanism to treat nanofiller, which were next to be merged within the polymer matrix. Therefore, a specific cold plasma reactor needs to be used to treat the surface of a nanofiller before it is dispersed within the polymer matrix.

VI. COLD PLASMA

Plasma is a state of an ionized gaseous substance that becomes highly electrically conductive to the point that having long-range electric and magnetic fields dominates the substance's behaviour. Plasma is a technology that can be used to treat a certain condition of the chemical phenomenon to improve the properties of the system [121]. In other words, plasma treatments are useful for modification and treatment processes towards a certain mechanism, system, chemical structure, chemical process, etc.

Interestingly, plasma treatment can be applied to improve the dispersion of the nanoparticle, nanocomposite bonding, and the compatibility surface between polymer matrices and nanoparticles [112], [119]. However, several factors such as suitability, types of plasma, and harmful sub-product generated from the plasma treatment process need to be considered before carrying out the treatment process. As mentioned, plasma treatment is an alternative method that is useful to modify the surface of the nanoparticles. Thus, this section discusses the behaviour of cold plasma based on low and atmospheric pressure discharge conditions.

A. LOW-PRESSURE PLASMA

Low-pressure plasma is discharges made under low gas pressures compared to the gas at atmospheric pressure. Fig. 8 illustrates the setup diagram for plasma production under low-pressure conditions. This type of discharge benefits from less power requirement of sustenance of the discharge as the rate of volume-recombination is low [122]. At low pressure of gases, it is easier to achieve uniform discharges at once to produce a better quality of plasma [122], [123]. Thus, the materials processing proceeds at the same rate over large substrate areas. In addition, low-pressure plasma also produces high concentrations of reactive species that can etch and deposit into thin films [123].
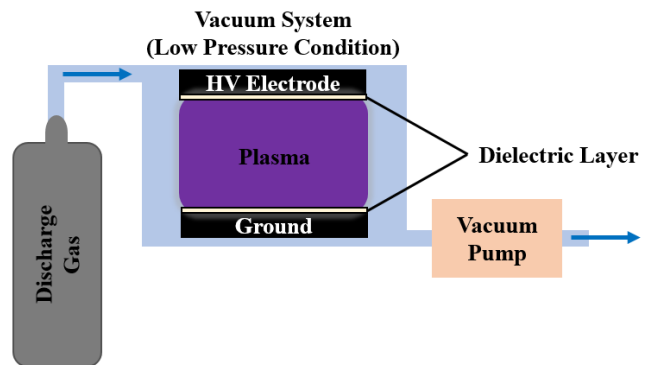
FIGURE 8. Setup diagram for plasma system under low-pressure conditions.

Usually, the prevalent background gas used is argon since it has a low potential of ionization in which it is easier to initiate and sustain the discharge process. The thermal-sensitive substrate also does minor damage since it is unreactive, monatomic, and extremely low boiling point [124]. The ions

produced in the plasma discharge will accelerate toward a substrate to make a directional etching of submicron features. In the other study, the various uses of low-pressure plasma reactors can change gas feed ratios, gas flow, time of treatment process, and power density. Due to those benefits, low-pressure plasma is the most common type of plasma discharge widely employed in industrial applications such as semiconductor manufacturing and material processing.

The frequency of low-pressure plasma reactors usually depends on the geometric properties of the reactor itself. However, the very common low-pressure plasma reactors are using microwaves at 2.53 GHz. This is because microwaves can operate at lower pressures and produce higher plasma densities [125]. Even though low-pressure plasma promises a good plasma treatment for this application, operating the plasma at reduced pressure has several drawbacks, such as requiring complicated and expensive vacuum systems. Besides that, the vacuum system also often requires a maintenance process which means consuming high expenditure. On top of that, the capacity and size of the nanofiller to undergo treatment are also limited by the size and capacity of the vacuum chamber. On the other hand, a low-pressure plasma reactor is not applicable to be practiced at the industrial levels as it requires a low controlled pressure system to achieve the plasma discharge state due to higher cost and great difficulty to operate as a continuous process.

B. ATMOSPHERIC PRESSURE PLASMA

Atmospheric pressure plasma is one of the plasma categories that discharge under atmospheric pressure (i.e., 1.01325×10^5 Pa). In order to overcome the drawbacks of low-pressure plasma reactors, atmospheric pressure plasma has been introduced since it promised the solution over low-pressure plasma drawbacks. The main advantages provided by this type of plasma discharge are the elimination of vacuum systems, reduced operation and maintenance cost as well as possibility to be employed in the continuous treatment system. The type of atmospheric pressure plasma discharge can be categorized into two main regimes: filamentary or micro-discharge plasma and atmospheric pressure glow (APG) discharge [126]. Micro-discharges are obtained either by using a corona system, dielectric barrier discharge, or plasma jet configuration [127].

One of the reactors to produce plasma discharge at atmospheric pressure is dielectric barrier discharge (DBD). Fig. 9 shows the structure of a dielectric barrier discharge reactor to produce cold plasma at atmospheric pressure conditions. Dielectric barrier discharge has been used widely in ozone production, surface treatment, and nanofiller treatment to improve insulating materials. Dielectric barrier discharge consists of two parallel plate electrodes, with at least one of these electrodes covered by a dielectric layer made up of insulating materials. The distance between the electrodes is maintained and limited to certain millimetres in width for a stable plasma production [128]–[130].
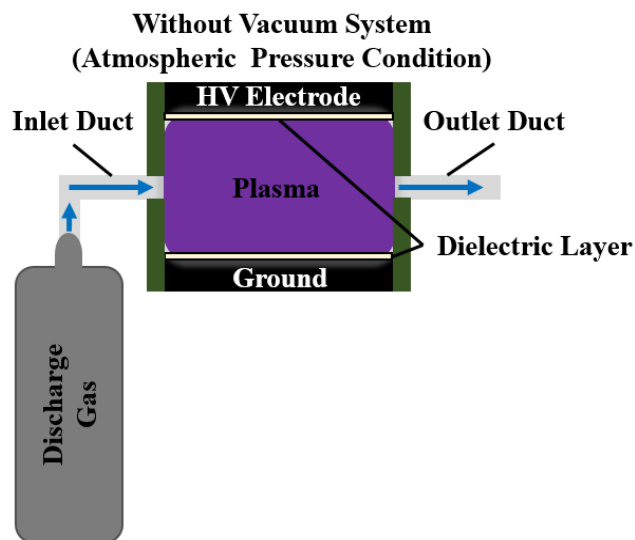

FIGURE 9. The basic structure of the dielectric barrier discharge reactor operated under atmospheric pressure conditions.

Dielectric barrier discharge can be ignited by using either a sinusoidal or pulsed power generator. The type of discharge in this configuration could be glowing or filamentary depends on the gas composition, voltage, and frequency excitation. As mentioned, filamentary discharge is formed by micro-discharges that are statistically developed on the surface of the dielectric layer while using an inert gas such as helium as plasma gas may produce glow discharge [131].

The dielectric layer plays an important part in limiting the discharge current and preventing the arc transition enabling the system to operate with a continuous or pulsed mode. This layer contribution ensures the streamers are distributed randomly and equally on the surface of the electrode so that the homogeneous treatment can be achieved. The streamers are produced from the accumulation of an electron on the dielectric layer [132], [133].

VII. SURFACE TREATMENT OF NANOFILLER BY COLD PLASMA

Plasma is a form of matter in which many electrons wander freely around the nuclei of an atom. The plasma properties are changed in electronic density or temperature depending on the energy supply and the amounts of energy transferred to the plasma [134]. Plasma can be categorized into certain categories based on these two parameters: electron density and electron temperature [134], [135]. Fig. 10 shows the characteristic of plasma discharge.

Plasma treatment is also chosen to be one of the processes that promise enhancement on the characteristic of the insulation. For surface modification, plasma treatment has promoted modification of wettability, printability, and adhesion of the treated materials [137]–[139]. Plasma treatment also can be used to treat the structure of the nanofiller surface, which indirectly improves the effectiveness of polymers nanocomposite as an insulation medium.
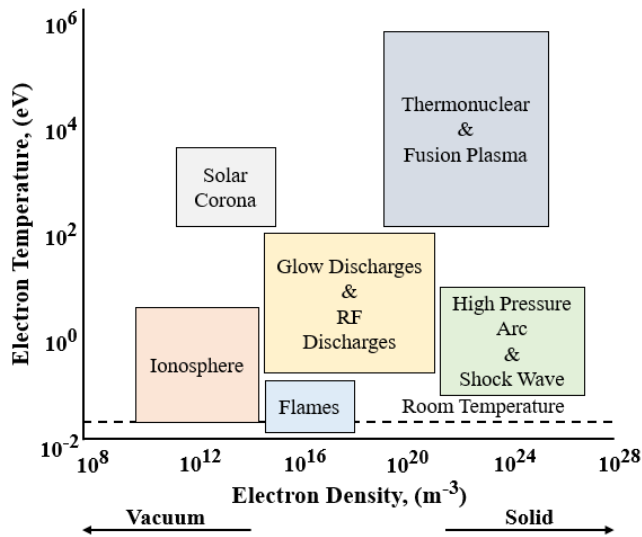
The type of plasma used for the treatment must be chosen based on the suitability and effectiveness offered by that particular plasma. The most effective form of plasma used for surface modification on nanofillers is glow discharge and radio frequency (RF) discharges [140]. However, plasma characteristics must also be studied to identify which type of plasma can work effectively for this treatment. In this section, the treatment of nanofiller surfaces using cold plasma is discussed based on the pressure of the discharge process.

Low-pressure plasma treatment can be done by exposing the nanofillers to the plasma plume produced under a low-pressure condition, usually operated inside a closed vacuum system. In contrast, atmospheric pressure plasma treatment is executed by exposing the nanofillers to the plasma plume forms under atmospheric pressure conditions. Usually, the duration of plasma treatment, type and flow rate of discharge gases, voltage, and frequency of supply are varied to obtain different plasma treatment mechanisms on the surface of nanofillers. The treated nanofillers under the different configurations of plasma production are then placed inside an airtight container, and then all the samples are characterized by using spectroscopy instruments such as X-ray Photoelectron Spectroscopy (XPS) and Fourier Transform Infrared Spectroscopy (FTIR). The waveform of voltage supply and discharge current was also recorded to characterize the mechanism of plasma discharge during the treatment process. Voltage-discharge current and Lissajous figure waveforms are usually used to characterize the plasma discharge. Besides, optical emission spectroscopy is another equipment used to characterize plasma discharge according to the spectrum of discharge species that occurred through the plasma discharge process. The treated nanofillers then dispersed in a polymer matrix using appropriate processing techniques such as direct mixing to form polymer nanocomposites.

A. LOW-PRESSURE PLASMA TREATMENT

In 2010, Hody *et al.* [141] conducted a comprehensive study to functionalize silicon carbide nanofiller with carboxylic groups by synthesis it is using low pressure (i.e., 40 Pa) plasma at radio frequency (i.e., 13.56 MHz) discharge. A magnetic stirrer was used to make sure the surfaces of the nanofiller are treated uniformly. The coating process was handled in an O_2/hexamethyldisilazane (HMDSN) mixture. The x-ray photoelectron spectroscopy (XPS) analysis showed that the O_2/ hexamethyldisilazane working gas resulted in a coating evolving from a polymer-like structure to a more inorganic SiOx-like structure as the oxygen ratio increased.

The result showed that coated silicon carbides are suitable to be further dispersed within the polymer host due to the surface morphology of silicon carbide after undergoing a low-pressure plasma coating process were being more inorganic SiOx-like structure. This indicates that the nanofiller and polymer host's surface compatibility can be enhanced by coating the carboxylic functional group on the surface of the nanofiller [141].

Compatible surfaces between nanofiller and polymer host will further be formed homogeneous and uniform dispersion of nanofiller into a polymer matrix, thus forms effective deep trap sites to reduce the local electric field on the polymer nanocomposite filled with coated nanomaterial [19]–[22], [141]. Therefore, the insulation properties such as dielectric breakdown strength, partial discharge, and electrical tree are compromising to be enhanced by introducing this alteration technique.

Low-pressure plasma is also effective in obtaining homogenous dispersion of nanoparticles with a polymer host. Haye *et al.* [142] employed low-pressure plasma with discharge at radio frequency to synthesize nickel/carbon nanocatalysts from solid precursors. Three combinations of mixing working gas were used containing argon: nitrogen (Ar: N_2, 5:2 sccm), argon: ammonia (Ar: NH_3, 5:4 sccm), and argon: oxygen (Ar: O_2, 5:2 sccm) with different flow rate ratio to determine the most effective synthesize of nickel/carbon nanocatalysts under different plasma chemistries. Low-pressure plasma is also used to functionalize carbon substrates such as carboxyl (-COOH), carbonyl (C=O), and cyanide (C≡N) functional group on nanofiller surfaces [142].

The radio frequency power was varied between 90 to 200W to obtain a complete process of plasma decomposition. The result was revealed that plasma chemistry and conditions strongly influence the organometallic compound decomposition and the size and the oxidation state of the homogeneously dispersed nickel nanoparticles, which obtained through a low-pressure plasma processing technique [142].

Introducing plasma treatment also helped increase the flux, whereas doping modified the surface properties of the nanoparticle [143]. Meanwhile, plasma treatment can increase the porosity and roughness of nanoparticles. Agarwal *et al.* [143] applied low-pressure plasma treatment

with a glow discharge mechanism to coat and modify the surface of nano-cobalt. The absorption bands of C-O and C=O are attributed to the creation of unsaturated -C=C- bonds after plasma treatment [143]. C-C and C-H bands' intensity also decreases, which represents the cross-linking phenomenon enhanced after plasma treatment. This indicates the successful process of surface functionalizing on the nanofiller by improving the hydrophilicity of nanofiller surfaces. Thus, the synthesis of polymer nanocomposites using nano-cobalt and polymethyl-methacrylate (PMMA) formed effective and homogeneous nanofiller dispersion into polymer matrices [143].

Jiansirisomboon *et al.* [144] experimented with investigating the effect of low-pressure plasma sprayed on alumina/silicon carbide nanocomposite. Plasma spray coatings are useful to provide surface resistance to corrosion. Low-pressure plasma spray is also considered a comprehensive technique to coat and transform nano-scale substrates into equilibrium metastable nanocomposite [144]. The study also revealed that the area percentage of nanofiller porosity and surface roughness could be further increased by using low-pressure plasma spray [143], [144]. Indirectly, this might improve the surface morphology of nanofiller and form compatible interfaces between hybrid nanofiller and polymer matrices.

Besides, low-pressure plasma is also efficient in providing significant oxidation that resulted in nanofiller surfaces with high hydrophilicity [145]. Norrman *et al.* [145] pointed out that exposing polyphenyl-sulfone surfaces to low-pressure plasma at 5 x 10^{-3} Torr will result in the maximal occurrence of the oxidation process on the surfaces of nanofiller. The oxidation reaction consists of two oxidation products which are C=O and (C=O)-O.

Both oxidation products are readily formed at 500W and increase slightly for harsher plasma conditions. The formation of both oxidation products on the surface of the nanofiller may increase the surface hydrophilicity and wettability of the nanofiller, which enhances the compatibility between nanofiller and polymer host interfaces [137]–[139], [145]. Therefore, the low-pressure plasma-treated nanofiller is expected to be dispersed uniformly better than the dispersion of untreated nanofiller into polymer matrices.

Xu *et al.* [109] reported that low-pressure plasma with particular functional monomers could obtain stable and effective modification techniques to functionalize chemical bonds. Low-pressure plasma is also useful to alter the surface roughness of nanofiller [109], [143], [144].

Spyrides *et al.* [146] used low-pressure plasma with argon and oxygen as a discharge gas to alter the surface roughness of ultra-high molecular weight polyethylene. In addition, low-pressure plasma is also used to improve the adhesion properties of polymers [146].

In fact, the wettability, chemical species, and surface morphology of polymer materials can also be modified using plasma treatment at low-pressure discharge conditions [147].

Mandolfino *et al.* [147] revealed that low-pressure plasma treatment is an effective technique for the surface preparation of polymers to create bonded joints.

The surface morphology of treating polymers was affected by the plasma power, exposure time, and type of working gas [148]–[150]. In terms of surface morphology, carbonyl, carboxyl, and hydroxyl are the possible polar groups introduced in the outermost layers of the surface by reactions with plasma species. All the polar groups functionalized on the surface of the polymer would affect the character of the treated surfaces, which increases the hydrophilicity of the surface.

Low-pressure plasma also significantly decreased the water contact angle of the polymer surfaces, especially pronounced for specific parameter sets. Consequently, low-pressure plasma can increase the surface free energy of polymers compared to untreated polymers [151], [152]. Increasing surface free energy occurred due to the functionalization of polar groups on the surface of low-pressure plasma-treated polymers.

For instance, oxygen-containing functional groups were formed on the treated surfaces due to the ionization of working gas, which is the principal cause of the enhanced wettability. Furthermore, the strength of chemical bonds on the nanofiller surface is also a crucial aspect that needs to be considered to obtain sufficient adhesive bond strength with relatively high surface energy. Iqbal *et al.* [153] revealed that low-pressure plasma had effectively functionalized dispersive components on the treated surfaces. However, identical polar groups forming on the low-pressure plasma-treated surfaces are not comprehensive as atmospheric pressure plasma treatment.

Since there are many advantages and impressive modification mechanisms provided by plasma treatment operated at low-pressure conditions, the low-pressure plasma becomes progressively common as the new surface modification technique. Indeed, plasma surface modification is also not required for water and chemical solutions because the chemical reaction produced from the ionization of discharge gases is developed from an electrical streamer discharge.

As mentioned, low-pressure plasma also has certain drawbacks, which are criticized and highlighted in some studies. Iqbal *et al.* [153] claimed that low-pressure plasma treatment is less effective than atmospheric pressure plasma treatment to improve surface energy and bonded joint strength. Besides, low-pressure plasma also required a complex vacuum system, difficult and complicated to control. Hence, low-pressure plasma is also not applicable to the industrial level since it is not cost-effective to produce mass products with a large-scale vacuum system. Thus, the plasma discharge at atmospheric pressure seems to need to be explored to counter back the drawbacks of low-pressure plasma and enhance nanofiller's surface morphology to form a compatible and well-dispersed nanofiller into a polymer matrix.

B. ATMOSPHERIC PRESSURE PLASMA TREATMENT

Atmospheric pressure plasma is preferred among researchers rather than low-pressure plasma because of the simple process of this system. In other words, the process of nanoparticle treatment becomes more accessible and more applicable by using atmospheric pressure plasma. The untreated nanofiller will be exposed to the plasma until the particle is homogeneously exposed and coated to become a treated nanofiller. The treated nanofiller is then mixed within the polymer matrix by using multiple stages of a conventional method.

Recently, Yan *et al.* [112], Yan [154], Yan *et al.* [155]–[159] reported the study of plasma application in high voltage insulations. Specifically, the study was conducted regarding the improvement in terms of an electrical tree. In the study, custom designs of atmospheric-pressure plasma reactors have been employed to treat nano-silica, which exists as a nanofiller. The specimen configuration was designed by placing 2 mm of tin-coated copper coil electrode above the top of the nanoparticle sample while the ground electrode was placed under the reactor to form a dielectric barrier discharge system.

Cold atmospheric-pressure plasma was generated for the plasma generation part using a 350 kHz radio frequency (RF) power supply with a maximum 5 kV peak-to-peak output voltage. However, the ideal specification of the plasma generator used in this study is 4 kV peak-to-peak radio frequency voltage. The working gas used for the discharge medium is helium since it can decompose to discharge uniformly. To obtain a reliable outcome, the nanofillers were treated by exposing the sample of the nanofiller for 30 minutes until it was homogeneously coated before mixed within an epoxy resin chain.

As expected, the studies also revealed that plasma treatment enhanced the compatibility and the properties of interfacial regions between epoxy resins filled with nano-silica. In terms of modification, the treatments were improved to increase the strength of chemical bonds and reduced the presence of weak chemical bonds while maintaining excellent uniformity of dispersion [112], [154]–[159]. Therefore, the properties of nanocomposite improved even with the low percentage weight of nanofiller.

In organic-inorganic nanocomposites, interfacial regions are primarily influenced by the dispersion uniformity of nanoparticles and the strength of interfacial bonds between the nanoparticles and the polymer matrix [159]. The insulating performance of organic-inorganic dielectric nanocomposites is highly influenced by the characteristics of interfacial regions [159].

In a study by Yan *et al.* [155], polyethylene oxide (PEO)-like functional layers were prepared on silica nanoparticles through plasma polymerization. Silica/epoxy resin nanocomposites are subsequently synthesized with these plasma-polymerized nanoparticles. Uniform atmospheric pressure plasma was generated with 5 kV peak-to-peak of applied voltage and 350 kHz of operating frequency. The untreated nanosilica is then exposed to the plasma plume to treat the nanoparticles. Based on the results, the concentration of each bond in the epoxy resin nanocomposites filled with untreated nanosilica remained similar to pure epoxy resin.

This mainly indicates the nanosilica was dispersed ineffectively within the epoxy resin matrix and failed to improve the interfacial structure of the nanocomposite due to the poor filler-matrix interaction [154], [155]. However, the concentration of each bond contained in the epoxy resin nanocomposite filled with plasma-treated nano-silica shows significant changes.

For instance, the concentration of C-O bonds of the nanocomposites with plasma polymer coated nano-silica increases dramatically (i.e., 36.15 %) than nanocomposites filled with untreated nanosilica (i.e., 16.14%). This enhancement happens due to the nucleophilic substitution reaction between the surface oxygen-containing functional groups on the nanoparticles and the epoxide rings in the pre-polymer resin [154], [155].

In the same study, it is found that plasma at low power (i.e., 10 W) can significantly increase the concentration of C-O bonds on the surface of silica nanoparticles. This plasma polymerized thin layer not only improves the dispersion uniformity by increasing the hydrophilicity of the nanoparticles but also provides anchoring sites to enable the formation of covalent bonds between the organic and inorganic phases [154]–[156], [158], [159]. Result obtained in this study was to reveal the improvement of electrical treeing resistance.

Musa *et al.* [160] investigated the electrical tree performance and the effect of filler concentration of silicone rubber nanocomposites filled with atmospheric pressure plasma-treated nanosilica. Atmospheric-pressure plasma was used to treat the silica nanofiller surfaces to enhance compatibility between the nanofiller and the silicone rubber matrix. The result showed that plasma-treated silica nanoparticles were uniformly well dispersed and formed strong covalent bonds with the molecules of the silicone rubber [160]. The plasma-treated nanocomposites were able to resist the electrical treeing better than the untreated nanocomposites.

Yan [154] experimented with investigating partial discharge (PD) characteristics of epoxy resin nanocomposites filled with atmospheric pressure plasma-treated nanosilica. Electrical tests showed improved partial discharge characteristics with plasma-modified nanofillers. In their study, partial discharge was measured using the direct coupling detection method, where the charge displacement caused by partial discharge activities is detected with measurement impedance or quadripole (Z_m).

The result obtained in this study has shown that plasma-treated nano-silica, where dispersed within an epoxy resin matrix, can suppress the partial discharge activities [154]. The partial discharge level curve was categorized into three stages in the electrical treeing process, namely 1-void formation, 2-tree propagation, and 3-ultimate breakdown. The partial discharge levels of the endurance test of untreated

TABLE 1. Summary studies conducted to enhance insulation properties of polymers nanocomposites using cold plasma technique.

Researchers	(Polymer Host/ Nanofiller)	Type of modification	Type of experiment conducted	Significant improvement
Friedrich *et al.* [163] (1993)	No Filler -Polyethylene. -Polypropylene.	Low-pressure plasma with a glow discharge	Material characterization	- Maximum adhesion. - Able to functionalize C=C, C-O-, C-OH, C=O, and COOH functional groups.
Shi *et al.* [164] (2002)	No Polymer Alumina	Plasma polymerization coated ultra-thin Pyrrole.	Surface morphology	- Energetically stable polymorph.
Sanchis *et al.* [113] (2006)	No Filler Low-density polyethylene	Low-pressure plasma	Material characterization, ageing test, crystallinity, & degradation temperature	- Longer lifetime. - No significant effect on crystallinity. - Improve degradation temperature. - Improve wettability. - Promote polar species (Carbonyl, Carboxyl, & Hydroxyl).
Hosseini *et al.* [165] (2010)	Polyvinyl chloride: styrene-butadiene (95:5)/Silver	Plasma treatment (Tetrahydrofuran)	Surface morphology & surface charge density	- Increase density of deposited nano-silver. - Highly uniform distribution of nano-silver. - Enhance membrane potential, surface charge density, transport number, perm selectivity.
Mukheriee *et al.* [166] (2010)	Epoxy resin/barium nitrate	Plasma treatment	Surface morphology	- Improve interfacial region between inorganic (nanofiller) – organic (polymer matrix). - Sufficiently strong to permit synthesis between inorganic-organic compounds.
Huang *et al.* [167] (2010)	Epoxy resin/ Mn3(Cu0.6Si0.15Ge0.25)N	Plasma treatment	Thermal conductivity	- Increase thermal conductivity. - Improve thermal stability.
Yan *et al.* [112] (2010)	Epoxy resin/silane-coated silica	Atmospheric pressure plasma treatment	Dielectric strength & Electrical ageing	- Increase dielectric strength. - Extended electrical ageing.
Yan *et al.* [154] (2013)	Epoxy resin/silica	Low-pressure plasma treatment	Partial discharge, dielectric strength, electrical ageing resistance, space charge, dielectric constant, & electrical treeing.	- Improve partial discharge endurance. - Increase dielectric strength. - Extended electrical ageing. - Reduce space charge accumulation. - Increase dielectric constant. - Reduce the rate of growth of electrical treeing.
Yan *et al.* [155] (2013)	Epoxy resin/silica	Plasma polymerization coated by poly(ethylene oxide) like film	Electrical ageing, electrical treeing, dielectric constant, & surface morphology.	- Longer endurance against electrical ageing. - Delay initiation electrical treeing. - Radial extent electrical treeing. - Reduce dielectric constant over a wide range frequency. - Better dispersion due to formation of C-O bond on filler-matrix.
Yan *et al.* [156] (2013)	Epoxy resin/silica	Atmospheric pressure plasma treatment	Dielectric constant, dielectric strength, & Electrical ageing.	- Lower dielectric constant at high frequency. - Increase dielectric strength. - Prolonged lifetime under high electrical stress.
Yan *et al.* [158] (2014)	Epoxy resin/silica	Plasma polymerization coated by Poly(ethylene oxide) like film	Surface morphology, electrical treeing, & dielectric constant	- Increase hydrophilicity. - Improve dispersion uniformity. - Enable anchoring sites to form covalent bonds between inorganic & organic phases. - Increase C-O bond on the nanofiller surface. - Improve electrical treeing. - Decrease dielectric constant.
Yan *et al.* [159] (2014)	Epoxy resin/silica	Plasma polymerization coated by Poly(ethylene oxide) like film	Electrical ageing, dielectric constant, & space charge.	- Better resistance against electrical ageing. - Decrease dielectric constant. - Mitigate space charge to built-up.
Musa *et al.* [160] (2016)	Silicone rubber/silica	Atmospheric pressure plasma (dielectric barrier discharge) filamentary	Electrical treeing & surface morphology.	- Improve electrical treeing resistance. - Enhance compatibility between nanofiller-polymer matrices. - Formed a strong covalent bond between nanofiller & polymer molecules.
Kumar *et al.* [110] (2016)	Ultra-high molecular weight polyethylene/multiwalled carbon nanotube	Plasma treatment (dielectric barrier discharge)	Surface morphology	- Transform hydrophobic to hydrophilic due to carboxyl groups bonded on nanofiller surfaces. - Hydrophobic results in less contact with biological fluids during physiological interaction. - Reduce surface roughness.
Liu *et al.* [17] (2016)	Poly(3,4-ethylenedioxythiophene)-poly(styrene sulfonate)/titania	Atmospheric pressure plasma. Synthesize in an aqueous solution.	Surface morphology.	- Reduce agglomeration. - Improve dispersibility and stability.
Awang *et al.* [161] (2017)	Low-density polyethylene/boron nitride	Plasma (dielectric barrier discharge) filamentary	Partial discharge (CIGRE Method II)	- Improve partial discharge (lower partial discharge magnitude).
Awang *et al.* [161] (2017)	Low-density polyethylene/boron nitride	Plasma (dielectric barrier discharge) filamentary	Alternating current dielectric breakdown strength	- Increase alternating current dielectric breakdown strength.
Mathioudaki *et al.* [168] (2018)	No Polymer Zinc oxide, alumina	Low-pressure plasma Polymerization (Cyclopropylamine)	Surface morphology	- Reduce agglomeration. - More polarizable. - High hydrogen bond. - Weaker electronegativity.
Awang et al. [162] (2019)	Low-density polyethylene/boron nitride	Plasma (dielectric barrier discharge) filamentary	Partial discharge & dielectric breakdown strength	- Reduce the magnitude of average discharge. - Reduce the number of partial discharges. - Increase dielectric breakdown strength.

and treated nano-silica were measured and confirmed the time of void formation, time of tree propagation. A lifetime

of the plasma-treated nano-silica/epoxy resin nanocomposite sample is prolonged (i.e., 19 hours of a lifetime) compared

to untreated nano-silica / epoxy resin nanocomposite sample (i.e., 14.7 hours of a lifetime) [154].

Awang *et al.* [161] reported a comprehensive study on the effect of atmospheric pressure plasma-treated boron nitride on partial discharge characteristics of low-density polyethylene. The purpose of this study was to improve the performance of the low-density polyethylene insulation against partial discharge by adding plasma-treated nano-boron nitride into the low-density polyethylene matrix. This study was performed by following CIGRE Method II at 7 kVrms applied voltage. The results revealed that the partial discharge resistance of low-density polyethylene nanocomposite was highly achieved by treating the nanofillers with cold plasma compared with the untreated boron nitride nanofillers. It is showed where the magnitude of partial discharge of sample low-density polyethylene/5 *wt%* of plasma-treated boron nitride nanocomposite is lowest (i.e. $\approx$ 2 pC at 600 s) followed by low-density polyethylene/5 *wt%* untreated boron nitride nanocomposite (i.e. $\approx$ 8 pC at 600 s) and pure low-density polyethylene (i.e. $\approx$ 27 pC at 600 s) [161].

Yan [154] also conducted a comprehensive study to investigate the dielectric breakdown strength of epoxy resin filled with atmospheric pressure plasma-treated nano-silica. The configuration of the electrodes and the samples are designed based on ASTM Standard D149.

As mentioned, homogeneous nano-dispersion is the key for dielectric breakdown strength increase and reliable results. Therefore, the primary purpose of plasma treatment is to improve the homogeneity dispersion of nano-silica within the epoxy resin matrix by altering the surface morphology of the nanofiller. Based on results, epoxy resin filled with plasma-treated nano-silica showed the highest breakdown strength at 63.2% of probability failure with 150 kV/mm breakdown strength, followed by epoxy resin filled with untreated nano-silica (i.e., 125 kV/mm). In contrast, the breakdown strength of unfilled epoxy resin is 55 kV/mm [154].

Awang *et al.* [162] also studied the effect of alternating current breakdown strength enhancement of low-density polyethylene nanocomposites using atmospheric pressure plasma. In this study, the surfaces of boron nitride nanoparticles were treated with atmospheric pressure plasma discharge to strengthen the interface between the low-density polyethylene matrices and boron nitride nanoparticles. The treatment of boron nitride nanofillers was done in the plasma chamber, which applied a dielectric barrier discharge configuration system. The gap spacing was kept at 3 mm, and it was generated by a 50 Hz of alternating current power supply with a maximum applied voltage between 7 to 8.5 kVrms. Helium gas was used as discharge gas with a flow rate of 1 litre/min, which was applied inside the plasma chamber. The plasma power was consumed at 3 to 15 W. The obtained results were analyzed with 2-parameter Weibull distribution. Results showed that the alternating current breakdown strength of the low-density polyethylene nanocomposites is improved when the nanofillers were modified using plasma treatment

(i.e., 173 kV/mm at 63.2% of probability failure) as compared to the untreated samples (i.e., 158 kV/mm at 63.2% of probability failure) [162].

C. SUMMARY ON PLASMA MODIFICATION TECHNIQUE TO ENHANCE INSULATION PROPERTIES

Based on the analysis of the various technical publications presented in the previous section, a few conclusions regarding the nanocomposite materials and plasma treatment are drawn as follows in Table 1. The majority of presented studies focus on improving the insulation properties of nanocomposite by manipulating the type and percentage weight of nanoparticles. Thus, a promising approach for the improvement purpose of polymer nanocomposites is still required in terms of electrical tree prolongation, partial discharge resistance, and breakdown strength enhancement.

Despite various configurations of composition that have been performed to investigate the insulation properties of polymer nanocomposite, there is still a limited number of publications, mainly using plasma treatment as an alternative method to modify the molecular surface of nanoparticles. It is only limited to intercalation and coupling agent methods. Less attention is paid to plasma treatment methods that can modify the surface morphology of nanoparticles.

Existing plasma systems used to treat nanoparticle surfaces are based on dielectric barrier discharge configurations. This configuration is inefficient and not suitable because it requires a complicated control system of discharge gas. As many nanoparticles also flow out of the treatment chamber with the flow of working gas, therefore the quantity of remaining treated nanoparticles in the treatment chamber would decrease. This drawback indicates that the existing configuration of dielectric barrier discharge is inefficient and needs to be improved.

Recently, research showed that the nanofiller with appropriate compatibility of polymer matrices could improve the performance of the nanocomposite insulation, especially in terms of electrical tree prolongation, partial discharge resistance, and dielectric strength enhancement. Thus, the surface of nanoparticles should have a hydrophilic characteristic to improve the compatibility, which means overcoming the agglomeration issue. The hydrophilic characteristic of nanoparticles can be obtained by treating the nanoparticle surfaces using plasma treatment. In fact, the effectiveness of this method still has not been emphasized by any researchers in detail.

VIII. CONCLUSION

Plasma treatment is one of the methods that can be used to improve the dispersion of nanoparticles, nanocomposite bonding, and the compatible surface between polymer matrices and nanoparticles. Incompatibility of molecular surface between nanoparticle and polymer matrix causing the structure of nanocomposites to become weak, thus reducing the effectiveness of the merging process. The studies conducted in which employed the cold plasma treatment on nanofiller

surface highlighted the significant improvement of insulation properties of polymers nanocomposite. Meanwhile, insulation properties of polymer nanocomposite filled with plasma-treated nanofillers compromising a better dielectric performance compared to polymer nanocomposite filled with untreated nanofiller.

Besides, the ability of cold plasma to replace conventional technique such as chemical functionalization and heat treatment seems to have a good potential due to the impressive result and outcomes from the cold plasma treatment that almost hit the same and even better level of effectiveness in altering the surface morphology of nanofiller to overcome the agglomeration issue. Specifically, the space charge, treeing lifetime (rate growth of electrical tree), partial discharge resistance, and breakdown strength are the parameters promoted to be enhanced through the surface modification of nanofiller using cold plasma. All the characteristics of insulation mentioned are proven improved by introducing cold plasma treatment with filamentary and glow discharge mechanism as a surface modification technique.

In terms of discharge conditions, atmospheric pressure plasma provides better effectiveness to functionalize chemical polar groups on the surface of nanofillers compared to low-pressure plasma. Previous studies have shown that atmospheric pressure plasma treatment successfully coats and functionalizes certain chemical functional groups such as the carbonyl and carboxyl groups. In addition, atmospheric pressure plasma is preferred to be implemented, especially on the industrial scale, compared to low-pressure plasma due to the unnecessary vacuum system.

The main challenges detailed out in this review are performance-related issues that arise in the cold plasma to improve the insulating properties of polymer nanocomposites as manuscripts were reviewed revealed the ability of cold plasma in improving compatibility between nanoparticles and the polymer matrix through the process of coating a certain chemical functional group on the surface of nanofiller before dispersing it into the polymer matrix. The effectiveness of cold plasma seems to require intensive consideration for structuring the standard and effective handling procedures in determining the steps and precise details of the cold plasma treatment process on nanomaterials, especially for insulating material applications.

The effective process of plasma treatment with a glow discharge mechanism will collectively overcome as many agglomeration issues as filamentary discharge, which directly improves the insulation properties of polymers nanocomposite. However, producing cold plasma with a glow discharge mechanism is one of the major challenges in implementing plasma treatment. This is because various factors need to be considered, especially producing a glow plasma discharge under atmospheric pressure conditions. Therefore, an appropriate cold plasma system specifically used to treat the surface of nanofiller is required to be developed by considering a few parameters such as optimum voltage, operation frequency, type and flow rate of discharge gas, and the geometrical

structure of atmospheric pressure plasma reactor together with the treatment chamber. Meanwhile, all configuration of parameters is important to determine the type of plasma discharge, effectiveness, and efficiency of plasma treatment on the surface of nanoparticles in terms of qualitative and quantitative. In addition, cost-effectiveness in developing a cold plasma system also needs to be considered since this system requires a disposable material that is a gas discharge to encourage the ionization process to occur.

Another challenge of the plasma release process is the lack of previous reports related to the by-product formed from ionization of discharge gas, which may decompose to form harmful molecules. Thus, this could be one of the trends for future research directions on cold plasma in producing an eco-friendly surface modification technique.

Besides, there is still no subsequent study to identify a comprehensive technique to control the coating process in terms of the type of chemical functional group that will be functionalized through a particular specification of cold plasma treatment and its significant role in enhancing insulation properties. Therefore, all the related factors, such as the type of discharge gas used, need to be acknowledged so that the type of chemical functional group coated on the surface of nanofillers can be inferred based on the composition of discharge gas manipulated.

Furthermore, the future direction of cold plasma treatment in improving polymers nanocomposite also tends to determine the most effective duration of plasma treatment, which is required to optimize the plasma time exposure. However, it depends upon the aims and objectives to be hit in its application. Introducing cold plasma treatment on nanocomposite technology also opens up the proliferation of research related to the type and optimum amount of plasma-treated nanofillers required to enhance the properties of nano-composition based on its utilization.

REFERENCES

[1] D. R. Paul and L. M. Robeson, "Polymer nanotechnology: Nanocomposites," *Polymer*, vol. 49, no. 15, pp. 3187–3204, Jul. 2008.

[2] A. D. de Oliveira and C. A. G. Beatrice, "Polymer nanocomposites with different types of nanofiller," in *Nanocomposites—Recent Evolutions*. London, U.K.: IntechOpen, 2019.

[3] W. S. Khan, N. N. Hamadneh, and W. A. Khan, "Polymer nanocomposites-synthesis technique, classification and properties," in *Science and Applications of Tailored Nanostructures*, P. D. Sia, Ed. Altrincham, U.K.: OCP, 2008, ch. 4, pp. 50–67.

[4] J. Moya, S. Lopezesteban, and C. Pecharroman, "The challenge of ceramic/metal microcomposites and nanocomposites," *Prog. Mater. Sci.*, vol. 52, no. 7, pp. 1017–1090, Sep. 2007.

[5] M. Pluta, A. Galeski, M. Alexandre, M.-A. Paul, and P. Dubois, "Polylactide/montmorillonite nanocomposites and microcomposites prepared by melt blending: Structure and some physical properties," *J. Appl. Polym. Sci.*, vol. 86, no. 6, pp. 1497–1506, Nov. 2002.

[6] N. El Miri, M. El Achaby, A. Fihri, M. Larzek, M. Zahouily, K. Abdelouahdi, A. Barakat, and A. Solhy, "Synergistic effect of cellulose nanocrystals/graphene oxide nanosheets as functional hybrid nanofiller for enhancing properties of PVA nanocomposites," *Carbohydrate Polym.*, vol. 137, pp. 239–248, Feb. 2016.

[7] B. Yuan, Y. Hu, X. Chen, Y. Shi, Y. Niu, Y. Zhang, S. He, and H. Dai, "Dual modification of graphene by polymeric flame retardant and Ni(OH)$_2$ nanosheets for improving flame retardancy of polypropylene," *Compos. A, Appl. Sci. Manuf.*, vol. 100, pp. 106–117, Sep. 2017.

[8] F. Vahedi, H. R. Shahverdi, M. M. Shokrieh, and M. Esmkhani, "Effects of carbon nanotube content on the mechanical and electrical properties of epoxy-based composites," *New Carbon Mater.*, vol. 29, no. 6, pp. 419–425, Dec. 2014.

[9] G. A. Gelves, M. H. Al-Saleh, and U. Sundararaj, "Highly electrically conductive and high performance EMI shielding nanowire/polymer nanocomposites by miscible mixing and precipitation," *J. Mater. Chem.*, vol. 21, no. 3, pp. 829–836, 2011.

[10] G. Iyer, R. Gorur, R. Richert, A. Krivda, and L. Schmidt, "Dielectric properties of epoxy based nanocomposites for high voltage insulation," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 18, no. 3, pp. 659–666, Jun. 2011.

[11] S. H. Lim, K. Y. Zeng, and C. B. He, "Morphology, tensile and fracture characteristics of epoxy-alumina nanocomposites," *Mater. Sci. Eng., A*, vol. 527, nos. 21–22, pp. 5670–5676, Aug. 2010.

[12] A. Duongthipthewa, Y. Su, and L. Zhou, "Electrical conductivity and mechanical property improvement by low-temperature carbon nanotube growth on carbon fiber fabric with nanofiller incorporation," *Compos. B, Eng.*, vol. 182, Feb. 2020, Art. no. 107581.

[13] M. I. Baig, P. G. Ingole, J.-D. Jeon, S. U. Hong, W. K. Choi, B. Jang, and H. K. Lee, "Water vapor selective thin film nanocomposite membranes prepared by functionalized silicon nanoparticles," *Desalination*, vol. 451, pp. 59–71, Feb. 2019.

[14] M. Zhang, L. Ding, J. Zheng, L. Liu, H. Alsulami, M. A. Kutbi, and J. Xu, "Surface modification of carbon fibers with hydrophilic Fe_3O_4 nanoparticles for nickel-based multifunctional composites," *Appl. Surf. Sci.*, vol. 509, Apr. 2020, Art. no. 145348.

[15] N. Nasrollahi, V. Vatanpour, S. Aber, and N. M. Mahmoodi, "Preparation and characterization of a novel polyethersulfone (PES) ultrafiltration membrane modified with a CuO/ZnO nanocomposite to improve permeability and antifouling properties," *Separat. Purification Technol.*, vol. 192, pp. 369–382, Feb. 2018.

[16] M. Guo, J. Jiang, Z. Shen, Y. Lin, C.-W. Nan, and Y. Shen, "High-energy-density ferroelectric polymer nanocomposites for capacitive energy storage: Enhanced breakdown strength and improved discharge efficiency," *Mater. Today*, vol. 29, pp. 49–67, Oct. 2019.

[17] X. Liu, J. Luo, J. Fan, S. Lin, L. Jia, X. Jia, Q. Cai, and X. Yang, "Comprehensive enhancement in overall properties of MWCNTs-COOH/epoxy composites by microwave: An efficient approach to strengthen interfacial bonding via localized superheating effect," *Compos. B, Eng.*, vol. 174, Oct. 2019, Art. no. 106909.

[18] J. Nasser, K. Steinke, L. Zhang, and H. Sodano, "Enhanced interfacial strength of hierarchical fiberglass composites through an aramid nanofiber interphase," *Compos. Sci. Technol.*, vol. 192, May 2020, Art. no. 108109.

[19] H. Luo, X. Zhou, C. Ellingford, Y. Zhang, S. Chen, K. Zhou, D. Zhang, C. R. Bowen, and C. Wan, "Interface design for high energy density polymer nanocomposites," *Chem. Soc. Rev.*, vol. 48, no. 16, pp. 4424–4465, 2019.

[20] N. Karak, "Fundamentals of nanomaterials and polymer nanocomposites," in *Nanomaterials and Polymer Nanocomposites*. Amsterdam, The Netherlands: Elsevier, 2019, pp. 1–45.

[21] X. Zheng, Y. Liu, and Y. Wang, "Electrical tree inhibition by SiO_2/XLPE nanocomposites: Insights from first-principles calculations," *J. Mol. Model.*, vol. 24, no. 8, p. 200, Aug. 2018.

[22] R. Liao, G. Bai, L. Yang, H. Cheng, Y. Yuan, and J. Guan, "Improved electric strength and space charge characterization in LDPE composites with montmorillonite fillers," *J. Nanomater.*, vol. 2013, pp. 1–7, Jan. 2013.

[23] T. Tanaka, G. C. Montanari, and R. Mulhaupt, "Polymer nanocomposites as dielectrics and electrical insulation-perspectives for processing technologies, material characterization and future applications," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 11, no. 5, pp. 763–784, Oct. 2004.

[24] M. D. L. de la Torre and M. M. Guijarro, "Covalent bonds on activated carbon," *Eur. J. Organic Chem.*, vol. 2010, no. 27, pp. 5147–5154, Sep. 2010.

[25] S. P. Pujari, L. Scheres, A. T. M. Marcelis, and H. Zuilhof, "ChemInform abstract: Covalent surface modification of oxide surfaces," *ChemInform*, vol. 45, no. 36, pp. 6322–6356, Sep. 2014.

[26] J. Cognard, "Some recent progress in adhesion technology and science," *Comp. Rendus Chimie*, vol. 9, no. 1, pp. 13–24, Jan. 2006.

[27] N. M. Sarkari, Ö. Doğan, E. Bat, M. Mohseni, and M. Ebrahimi, "Assessing effects of (3-aminopropyl)trimethoxysilane self-assembled layers on surface characteristics of organosilane-grafted moisture-crosslinked polyethylene substrate: A comparative study between chemical vapor deposition and plasma-facilitated *in situ* grafting methods," *Appl. Surf. Sci.*, vol. 497, Dec. 2019, Art. no. 143751.

[28] R. R. Prakash, S. Pandiarajan, P. Venkatesh, and N. Kamaraj, "Performance analysis of PMMA—TiO_2 nanocomposite dielectrics," in *Proc. Int. Conf. Emerg. Trends Electr. Comput. Technol.*, Mar. 2011, pp. 46–49.

[29] Y. Zheng, Y. Zheng, and R. Ning, "Effects of nanoparticles SiO_2 on the performance of nanocomposites," *Mater. Lett.*, vol. 57, no. 19, pp. 2940–2944, Jun. 2003.

[30] Y. Jin, N. Xia, and R. A. Gerhardt, "Enhanced dielectric properties of polymer matrix composites with $BaTiO_3$ and MWCNT hybrid fillers using simple phase separation," *Nano Energy*, vol. 30, pp. 407–416, Dec. 2016.

[31] P. Barber, S. Balasubramanian, Y. Anguchamy, S. Gong, A. Wibowo, H. Gao, H. Ploehn, and H.-C. Z. Loye, "Polymer composite and nanocomposite dielectric materials for pulse power energy storage," *Materials*, vol. 2, no. 4, pp. 1697–1733, Oct. 2009.

[32] M. H. Lean and W.-P.-L. Chu, "Modeling effect of ferroelectric nanofiller size on bipolar charge transport in nanocomposite film," *J. Polym. Sci. B, Polym. Phys.*, vol. 53, no. 19, pp. 1380–1390, Oct. 2015.

[33] M. H. Ahmad, N. Bashir, H. Ahmad, A. A. A. Jamil, and A. A. Suleiman, "An overview of electrical tree growth in solid insulating material with emphasis of influencing factors, mathematical models and tree suppression," *TELKOMNIKA Indonesian J. Electr. Eng.*, vol. 12, no. 8, Aug. 2014.

[34] Z. Lv, Y. Wang, J. Chen, J. Wang, Y. Zhou, and S.-T. Han, "Semiconductor quantum dots for memories and neuromorphic computing systems," *Chem. Rev.*, vol. 120, no. 9, pp. 3941–4006, May 2020.

[35] I. Pleşa, P. Noţingher, S. Schlögl, C. Sumereder, and M. Muhr, "Properties of polymer composites used in high-voltage applications," *Polymers*, vol. 8, no. 5, p. 173, Apr. 2016.

[36] M. Liang and K. L. Wong, "Improving the long-term performance of composite insulators use nanocomposite: A review," *Energy Procedia*, vol. 110, pp. 168–173, Mar. 2017.

[37] T. Torres, "Carbon nanotubes and related structures. Synthesis, characterization, functionalization, and applications. Edited by Dirk M. Guldi and Nazario Martí," *Angew. Chem. Int. Ed.*, vol. 50, no. 7, pp. 1473–1474, Feb. 2011.

[38] I. R. L. Machado, H. M. F. Mendes, G. E. S. Alves, and R. R. Faleiros, "Nanotubos de carbono: Potencial de uso em medicina veterinária," *Ciência Rural*, vol. 44, no. 10, pp. 1823–1829, Oct. 2014.

[39] P.-C. Ma, N. A. Siddiqui, G. Marom, and J.-K. Kim, "Dispersion and functionalization of carbon nanotubes for polymer-based nanocomposites: A review," *Compos. A, Appl. Sci. Manuf.*, vol. 41, no. 10, pp. 1345–1367, Oct. 2010.

[40] G. K. Maron, B. S. Noremberg, J. H. Alano, F. R. Pereira, V. G. Deon, R. C. R. Santos, V. N. Freire, A. Valentini, and N. L. V. Carreno, "Carbon fiber/epoxy composites: Effect of zinc sulphide coated carbon nanotube on thermal and mechanical properties," *Polym. Bull.*, vol. 75, no. 4, pp. 1619–1633, Apr. 2018.

[41] S. Lee, S. Lim, E. Lim, and K. K. Lee, "Synthesis of aqueous dispersion of graphenes via reduction of graphite oxide in the solution of conductive polymer," *J. Phys. Chem. Solids*, vol. 71, no. 4, pp. 483–486, Apr. 2010.

[42] V. Mittal and A. U. Chaudhry, "Polymer—Graphene nanocomposites: Effect of polymer matrix and filler amount on properties," *Macromolecular Mater. Eng.*, vol. 300, no. 5, pp. 510–521, 2015.

[43] M. Kotal and A. K. Bhowmick, "Polymer nanocomposites from modified clays: Recent advances and challenges," *Prog. Polym. Sci.*, vol. 51, pp. 127–187, Dec. 2015.

[44] S. S. Ray, "Recent trends and future outlooks in the field of clay-containing polymer nanocomposites," *Macromolecular Chem. Phys.*, vol. 215, no. 12, pp. 1162–1179, Jun. 2014.

[45] A. Kausar, "Review on polymer/halloysite nanotube nanocomposite," *Polym.-Plastics Technol. Eng.*, vol. 57, no. 6, pp. 548–564, Apr. 2018.

[46] W. Lutz, "Zeolite Y: Synthesis, modification, and properties—A case revisited," *Adv. Mater. Sci. Eng.*, vol. 2014, pp. 1–20, May 2014.

[47] S. Mintova, J.-P. Gilson, and V. Valtchev, "Advances in nanosized zeolites," *Nanoscale*, vol. 5, no. 15, pp. 6693–6703, 2013.

[48] R. F. Nickerson and J. A. Habrle, "Cellulose intercrystalline structure," *Ind. Eng. Chem.*, vol. 39, no. 11, pp. 1507–1512, Nov. 1947.

[49] A. Dufresne, "Cellulose nanomaterial reinforced polymer nanocomposites," *Current Opinion Colloid Interface Sci.*, vol. 29, pp. 1–8, May 2017.

[50] K. B. Azouz, E. C. Ramires, W. Van den Fonteyne, N. El Kissi, and A. Dufresne, "Simple method for the melt extrusion of a cellulose nanocrystal reinforced hydrophobic polymer," *ACS Macro Lett.*, vol. 1, no. 1, pp. 236–240, Jan. 2012.

[51] A. D. de Oliveira and C. A. G. Beatrice, "Polymer nanocomposites with different types of nanofiller," in *Nanocomposites—Recent Evolutions*. London, U.K.: IntechOpen, 2019.

[52] A. D. Oliveira, C. A. G. Beatrice, F. R. Passador, and L. A. Pessan, "Polyetherimide-based nanocomposites materials for hydrogen storage," *AIP Conf.*, vol. 1779, no. 1, pp. 1–6, 2016.

[53] L. Liu and J. C. Grunlan, "Clay assisted dispersion of carbon nanotubes in conductive epoxy nanocomposites," *Adv. Funct. Mater.*, vol. 17, no. 14, pp. 2343–2348, Sep. 2007.

[54] S. Goyanes, G. R. Rubiolo, A. Salazar, A. Jimeno, M. A. Corcuera, and I. Mondragon, "Carboxylation treatment of multiwalled carbon nanotubes monitored by infrared and ultraviolet spectroscopies and scanning probe microscopy," *Diamond Rel. Mater.*, vol. 16, no. 2, pp. 412–417, Feb. 2007.

[55] S. Monemian, S. H. Jafari, H. A. Khonakdar, V. Goodarzi, U. Reuter, and P. Pötschke, "MWNT-filled PC/ABS blends: Correlation of morphology with rheological and electrical response," *J. Appl. Polym. Sci.*, vol. 130, no. 2, pp. 739–748, Oct. 2013.

[56] M. Wegrzyn, B. Galindo, A. Benedito, and E. Giménez, "Effect of processing method on mechanical properties of PC/ABS-MWCNT nanocomposites," *Macromolecular Symposia*, vols. 321–322, no. 1, pp. 161–165, Dec. 2012.

[57] C. Wan and B. Chen, "Reinforcement and interphase of polymer/graphene oxide nanocomposites," *J. Mater. Chem.*, vol. 22, no. 8, p. 3637, 2012.

[58] R. Sengupta, M. Bhattacharya, S. Bandyopadhyay, and A. K. Bhowmick, "A review on the mechanical and electrical properties of graphite and modified graphite reinforced polymer composites," *Prog. Polym. Sci.*, vol. 36, no. 5, pp. 638–670, May 2011.

[59] C. C. N. de Melo, C. A. G. Beatrice, L. A. Pessan, A. D. de Oliveira, and F. M. Machado, "Analysis of nonisothermal crystallization kinetics of graphene oxide–reinforced polyamide 6 nanocomposites," *Thermochim. Acta*, vol. 667, pp. 111–121, Sep. 2018.

[60] F. Zaïri, J. M. Gloaguen, M. Naït-Abdelaziz, A. Mesbah, and J. M. Lefebvre, "Study of the effect of size and clay structural parameters on the yield and post-yield response of polymer/clay nanocomposites via a multiscale micromechanical modelling," *Acta Mater.*, vol. 59, no. 10, pp. 3851–3863, Jun. 2011.

[61] N. Bitinis, M. Hernandez, R. Verdejo, J. M. Kenny, and M. A. Lopez-Manchado, "Recent advances in clay/polymer nanocomposites," *Adv. Mater.*, vol. 23, no. 44, pp. 5229–5236, 2011.

[62] M. Kotal and A. K. Bhowmick, "Polymer nanocomposites from modified clays: Recent advances and challenges," *Prog. Polym. Sci.*, vol. 51, pp. 127–187, Dec. 2015.

[63] M. Raji, M. E. M. Mekhzoum, D. Rodrigue, A. E. K. Qaiss, and R. Bouhfid, "Effect of silane functionalization on properties of polypropylene/clay nanocomposites," *Compos. B, Eng.*, vol. 146, pp. 106–115, Aug. 2018.

[64] P. Pasbakhsh, G. J. Churchman, and J. L. Keeling, "Characterisation of properties of various halloysites relevant to their use as nanotubes and microfibre fillers," *Appl. Clay Sci.*, vol. 74, pp. 47–57, Apr. 2013.

[65] A. Coldea, M. V. Swain, and N. Thiel, "Mechanical properties of polymer-infiltrated-ceramic-network materials," *Dental Mater.*, vol. 29, no. 4, pp. 419–426, Apr. 2013.

[66] H. Wang, S. Meng, P. Xu, W. Zhong, and Q. Du, "Effect of traces of inorganic content on thermal stability of poly(methyl methacrylate) nanocomposites," *Polym. Eng. Sci.*, vol. 47, no. 3, pp. 302–307, 2007.

[67] O. Bera, B. Pilić, J. Pavličević, M. Jovičić, B. Holló, K. M. Szécsényi, and M. Špirkova, "Preparation and thermal properties of polystyrene/silica nanocomposites," *Thermochim. Acta*, vol. 515, nos. 1–2, pp. 1–5, Mar. 2011.

[68] B. P. Tripathi and V. K. Shahi, "Organic-inorganic nanocomposite polymer electrolyte membranes for fuel cell applications," *Prog. Polym. Sci.*, vol. 36, no. 7, pp. 945–979, 2011.

[69] C. Liao, M. Zhang, M. Y. Yao, T. Hua, L. Li, and F. Yan, "Flexible organic electronics in biology: Materials and devices," *Adv. Mater.*, vol. 27, no. 46, pp. 7493–7527, Dec. 2015.

[70] S. Sinharay and M. Bousmina, "Biodegradable polymers and their layered silicate nanocomposites: In greening the 21st century materials world," *Prog. Mater. Sci.*, vol. 50, no. 8, pp. 962–1079, Nov. 2005.

[71] Q. Zhou, K. P. Pramoda, J.-M. Lee, K. Wang, and L. S. Loo, "Role of interface in dispersion and surface energetics of polymer nanocomposites containing hydrophilic POSS and layered silicates," *J. Colloid Interface Sci.*, vol. 355, no. 1, pp. 222–230, Mar. 2011.

[72] R. K. Mishra, P. Mishra, K. Verma, and K. Joseph, "Manipulation of thermo-mechanical, morphological and electrical properties of PP/PET polymer blend using MWCNT as nano compatibilizer: A comprehensive study of hybrid nanocomposites," *Vacuum*, vol. 157, pp. 433–441, Nov. 2018.

[73] C. Chen, H. Wang, Y. Xue, Z. Xue, H. Liu, X. Xie, and Y.-W. Mai, "Structure, rheological, thermal conductive and electrical insulating properties of high-performance hybrid epoxy/nanosilica/AgNWs nanocomposites," *Compos. Sci. Technol.*, vol. 128, pp. 207–214, May 2016.

[74] Y.-X. Pang and S. N. B. Hodgson, "Ceramic/inorganic-organic nano-hybrid composites for thermally stable insulation of electrical wires. Part II: Properties of the composite insulated wires," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 27, no. 2, pp. 403–409, Apr. 2020.

[75] N. Raman, S. Sudharsan, and K. Pothiraj, "Synthesis and structural reactivity of inorganic–organic hybrid nanocomposites—A review," *J. Saudi Chem. Soc.*, vol. 16, no. 4, pp. 339–352, Oct. 2012.

[76] S. Paszkiewicz, K. Pypeć, I. Irska, and E. Piesowicz, "Functional polymer hybrid nanocomposites based on polyolefins: A review," *Processes*, vol. 8, no. 11, p. 1475, Nov. 2020.

[77] Z. Lv, M. Chen, F. Qian, V. A. L. Roy, W. Ye, D. She, Y. Wang, Z. Xu, Y. Zhou, and S. Han, "Mimicking neuroplasticity in a hybrid biopolymer transistor by dual modes modulation," *Adv. Funct. Mater.*, vol. 29, no. 31, Aug. 2019, Art. no. 1902374.

[78] Z. Lv, Y. Wang, Z. Chen, L. Sun, J. Wang, M. Chen, Z. Xu, Q. Liao, L. Zhou, X. Chen, J. Li, K. Zhou, Y. Zhou, Y.-J. Zeng, S.-T. Han, and V. A. L. Roy, "Biomemory: Phototunable biomemory based on light-mediated charge trap (Adv. Sci. 9/2018)," *Adv. Sci.*, vol. 5, no. 9, Sep. 2018, Art. no. 1870051.

[79] Y. Wang, Z. Lv, J. Chen, Z. Wang, Y. Zhou, L. Zhou, X. Chen, and S.-T. Han, "Photonic synapses based on inorganic perovskite quantum dots for neuromorphic computing," *Adv. Mater.*, vol. 30, no. 38, Sep. 2018, Art. no. 1802883.

[80] A. Haque, M. Shamsuzzoha, F. Hussain, and D. Dean, "S2-glass/epoxy polymer nanocomposites: Manufacturing, structures, thermal and mechanical properties," *J. Compos. Mater.*, vol. 37, no. 20, pp. 1821–1837, Oct. 2003.

[81] E. P. Waldi, M. H. Setiawan, A. Winarto, Y. Nugraha, M. A. Hafizi, and Z. Nawawi, "The tensile properties of alumina and silica bionanocomposite material for high voltage insulation," *IOP Conf. Ser., Mater. Sci. Eng.*, vol. 602, no. 1, 2019, Art. no. 012010.

[82] X. Xing, M. Chen, Y. Gong, Z. Lv, S.-T. Han, and Y. Zhou, "Building memory devices from biocomposite electronic materials," *Sci. Technol. Adv. Mater.*, vol. 21, no. 1, pp. 100–121, Jan. 2020.

[83] C. Calebrese, L. Hui, L. Schadler, and J. Nelson, "A review on the importance of nanocomposite processing to enhance electrical insulation," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 18, no. 4, pp. 938–945, Aug. 2011.

[84] S. W. Thomas, R. R. Khan, K. Puttananjegowda, and W. Serrano-Garcia, "Conductive polymers and metal oxide polymeric composites for nanostructures and nanodevices," in *Advances in Nanostructured Materials and Nanopatterning Technologies*. Amsterdam, The Netherlands: Elsevier, 2020, pp. 243–271.

[85] J. Njuguna, F. Ansari, S. Sachse, H. Zhu, and V. M. Rodriguez, "Nanomaterials, nanofillers, and nanocomposites: Types and properties," in *Health and Environmental Safety of Nanomaterials*. Amsterdam, The Netherlands: Elsevier, 2014, pp. 3–27.

[86] J. Yu, X. Huang, C. Wu, and P. Jiang, "Permittivity, thermal conductivity and thermal stability of poly(vinylidene fluoride)/graphene nanocomposites," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 18, no. 2, pp. 478–484, Apr. 2011.

[87] A. M. Pourrahimi, R. T. Olsson, and M. S. Hedenqvist, "Nanocomposites: The role of interfaces in polyethylene/metal-oxide nanocomposites for ultrahigh-voltage insulating materials (Adv. Mater. 4/2018)," *Adv. Mater.*, vol. 30, no. 4, Jan. 2018, Art. no. 1870025.

[88] J. Su, B. X. Du, T. Han, and H. Du, "Electrical performance of silicone rubber/SiO$_2$ nanocomposites under low temperature," in *Proc. Int. Symp. Electr. Insulating Mater.*, Jun. 2014, pp. 469–472.

[89] B. X. Du, T. Han, and J. G. Su, "Tree characteristics in silicone rubber/SiO₂ nanocomposites under low temperature," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 21, no. 2, pp. 503–510, Apr. 2014.

[90] B. X. Du, T. Han, and J. G. Su, "Effects of low temperature on treeing phenomena of silicone rubber/SiO₂ nanocomposites," in *Proc. Annu. Rep. Conf. Electr. Insul. Dielectr. Phenomena*, Oct. 2013, pp. 514–517.

[91] S. Alapati and M. J. Thomas, "Electrical treeing in polymer nanocomposites," *Int. J. Emerg. Electr. Power Syst.*, vol. 10, no. 2, pp. 1–5, Jun. 2009.

[92] S. Alapati and M. J. Thomas, "Electrical treeing and the associated PD characteristics in LDPE nanocomposites," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 19, no. 2, pp. 697–704, Apr. 2012.

[93] A. Sridhar and M. J. Thomas, "Electrical treeing in polyethylene: Effect of nano fillers on tree inception and growth," in *Proc. Int. Conf. High Voltage Eng. Appl.*, Oct. 2010, pp. 576–579.

[94] W. A. Izzati, M. Shafanizam, Y. Z. Arief, M. Z. H. Makmud, Z. Adzis, and N. A. Muhamad, "Comparative study on partial discharge characteristic in polymer-nanocomposite as electrical insulating material," *Appl. Mech. Mater.*, vols. 284–287, pp. 62–66, Jan. 2013.

[95] T. Tanaka, A. Bulinski, J. Castellon, M. Frechette, S. Gubanski, J. Kindersberger, G. Montanari, M. Nagao, P. Morshuis, Y. Tanaka, S. Pelissou, A. Vaughan, Y. Ohki, C. Reed, S. Sutton, and S. Han, "Dielectric properties of XLPE/SiO₂ nanocomposites based on CIGRE WG D1.24 cooperative test results," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 18, no. 5, pp. 1482–1517, Oct. 2011.

[96] T. Tanaka, Y. Ohki, M. Ochi, M. Harada, and T. Imai, "Enhanced partial discharge resistance of epoxy/clay nanocomposite prepared by newly developed organic modification and solubilization methods," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 15, no. 1, pp. 81–89, 2008.

[97] M. Takala, "Dielectric breakdown strength of polymer nanocomposites—The effect of nanofiller content," in *Proc. Nordic Insul. Symp.*, no. 23, Feb. 2018, pp. 1–4.

[98] T. Tanaka, "Dielectric nanocomposites with insulating properties," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 12, no. 5, pp. 914–928, Oct. 2005.

[99] M. Roy, J. K. Nelson, R. K. MacCrone, L. S. Schadler, C. W. Reed, R. Keefe, and W. Zenger, "Polymer nanocomposite dielectrics—The role of the interface," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 12, no. 4, pp. 629–643, Aug. 2005.

[100] J. C. Fothergill, "Ageing, space charge and nanodielectrics: Ten things we don't know about dielectrics," in *Proc. IEEE Int. Conf. Solid Dielectr.*, Jul. 2007, pp. 1–10.

[101] T. Tanaka, M. Kozako, N. Fuse, and Y. Ohki, "Proposal of a multi-core model for polymer nanocomposite dielectrics," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 12, no. 4, pp. 669–681, Aug. 2005.

[102] C. Green, A. Vaughan, G. Mitchell, and T. Liu, "Structure property relationships in polyethylene/montmorillonite nanodielectrics," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 15, no. 1, pp. 134–143, 2008.

[103] R. Smith, C. Liang, M. Landry, J. Nelson, and L. Schadler, "The mechanisms leading to the useful electrical properties of polymer nanodielectrics," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 15, no. 1, pp. 187–196, 2008.

[104] M. Roy, J. K. Nelson, R. K. MacCrone, and L. S. Schadler, "Candidate mechanisms controlling the electrical characteristics of silica/XLPE nanodielectrics," *J. Mater. Sci.*, vol. 42, no. 11, pp. 3789–3799, Jun. 2007.

[105] G. C. Montanari, F. Palmieri, L. Testa, A. Motori, A. Saccani, and F. Patuelli, "Polarization processes of nanocomposite silicate-EVA and PP materials," *IEEJ Trans. Fundam. Mater.*, vol. 126, no. 11, pp. 1090–1096, 2006.

[106] J. K. Nelson and Y. Hu, "Candidate mechanisms responsible for property changes in dielectric nanocomposites," in *Proc. IEEE 8th Int. Conf. Properties Appl. Dielectr. Mater.*, Jun. 2006, pp. 150–154.

[107] J.-H. Chang, "Comparative analysis of properties of PVA composites with various nanofillers: Pristine clay, organoclay, and functionalized graphene," *Nanomaterials*, vol. 9, no. 3, p. 323, Mar. 2019.

[108] K. Y. Lau, M. Piah, and M. Afendi, "Polymer nanocomposites in high voltage electrical insulation perspective: A review," *Malaysian Polym. J.*, vol. 6, no. 1, pp. 58–69, 2011.

[109] L. Xu, J. Deng, Y. Guo, W. Wang, R. Zhang, and J. Yu, "Fabrication of super-hydrophobic cotton fabric by low-pressure plasma-enhanced chemical vapor deposition," *Textile Res. J.*, vol. 89, no. 10, pp. 1853–1862, May 2019.

[110] N. N. Kumar, S. Yap, F. B. Samsudin, M. Khan, and R. P. Srinivasa, "Effect of argon plasma treatment on tribological properties of UHMWPE/MWCNT nanocomposites," *Polymers*, vol. 8, no. 8, p. 295, Aug. 2016.

[111] M. Z. Rong, M. Q. Zhang, and W. H. Ruan, "Surface modification of nanoscale fillers for improving properties of polymer nanocomposites: A review," *Mater. Sci. Technol.*, vol. 22, no. 7, pp. 787–796, Jul. 2006.

[112] W. Yan, Z. J. Han, B. T. Phung, and K. Ostrikov, "Silica nanoparticles treated by cold atmospheric-pressure plasmas improve the dielectric performance of organic-inorganic nanocomposites," *ACS Appl. Mater. Interfaces*, vol. 4, no. 5, pp. 2637–2642, May 2012.

[113] M. R. Sanchis, V. Blanes, M. Blanes, D. Garcia, and R. Balart, "Surface modification of low density polyethylene (LDPE) film by low pressure O₂ plasma treatment," *Eur. Polym. J.*, vol. 42, no. 7, pp. 1558–1568, Jul. 2006.

[114] M. Asgari, A. Abouelmagd, and U. Sundararaj, "Silane functionalization of sodium montmorillonite nanoclay and its effect on rheological and mechanical properties of HDPE/clay nanocomposites," *Appl. Clay Sci.*, vol. 146, pp. 439–448, Sep. 2017.

[115] M. Peyravi, M. Jahanshahi, A. Rahimpour, A. Javadi, and S. Hajavi, "Novel thin film nanocomposite membranes incorporated with functionalized TiO₂ nanoparticles for organic solvent nanofiltration," *Chem. Eng. J.*, vol. 241, pp. 155–166, Apr. 2014.

[116] S. W. Kim, T. Kim, Y. S. Kim, H. S. Choi, H. J. Lim, S. J. Yang, and C. R. Park, "Surface modifications for the effective dispersion of carbon nanotubes in solvents and polymers," *Carbon*, vol. 50, no. 1, pp. 3–33, Jan. 2012.

[117] A. R. Nesic and S. I. Seslija, "The influence of nanofillers on physical–chemical properties of polysaccharide-based film intended for food packaging," in *Food Packagings*. Amsterdam, The Netherlands: Elsevier, 2017, pp. 637–697.

[118] M. N. Belgacem and A. Gandini, "The surface modification of cellulose fibres for use as reinforcing elements in composite materials," *Compos. Interfaces*, vol. 12, nos. 1–2, pp. 41–75, Jan. 2005.

[119] B. N. Jung, H. W. Jung, D. Kang, G. H. Kim, M. Lee, S. W. Hwang, and J. K. Shim, "The fabrication of affinity improved nanocomposites with plasma treated polypropylene (PP) and alkaline cellulose nanofiber (CNF) suspension," *Polym. Test.*, vol. 85, May 2020, Art. no. 106352.

[120] L. Zhao, W. Liu, M. Xu, Y. Huang, Q. Zheng, S. Sun, and Y. Wang, "Study on atmospheric air glow discharge plasma generation based on multiple potentials and aramid fabric surface modification," *Plasma Processes Polym.*, vol. 16, no. 12, Dec. 2019, Art. no. 1900114.

[121] T. Shao, R. Wang, C. Zhang, and P. Yan, "Atmospheric-pressure pulsed discharges and plasmas: Mechanism, characteristics and applications," *High Voltage*, vol. 3, no. 1, pp. 14–20, Mar. 2018.

[122] C. Mandolfino, "Polypropylene surface modification by low pressure plasma to increase adhesive bonding: Effect of process parameters," *Surf. Coatings Technol.*, vol. 366, pp. 331–337, May 2019.

[123] Y. A. Lebedev, "Microwave discharges at low pressures and peculiarities of the processes in strongly non-uniform plasma," *Plasma Sources Sci. Technol.*, vol. 24, no. 5, Aug. 2015, Art. no. 053001.

[124] M. Laroussi, "Low-temperature plasmas for medicine?" *IEEE Trans. Plasma Sci.*, vol. 37, no. 6, pp. 714–725, Jun. 2009.

[125] B. M. Graves, "Synthesis of carbon nanotube materials from a microwave plasma," Ph.D. dissertation, Dept. Eng., Cambridge Univ., Cambridge, U.K., 2020.

[126] P. Lu, P. J. Cullen, and K. Ostrikov, "Atmospheric pressure nonthermal plasma sources," in *Cold Plasma in Food and Agriculture*. Amsterdam, The Netherlands: Elsevier, 2016, pp. 83–116.

[127] R. Brandenburg, "Corrigendum: Dielectric barrier discharges: Progress on plasma sources and on the understanding of regimes and single filaments (2017 plasma sources sci. Technol. 26 053001)," *Plasma Sources Sci. Technol.*, vol. 27, no. 7, Jul. 2018, Art. no. 079501.

[128] R. Foest, M. Schmidt, and K. Becker, "Microplasmas, an emerging field of low-temperature plasma science and technology," *Int. J. Mass Spectrometry*, vol. 248, no. 3, pp. 87–102, Feb. 2006.

[129] P. J. Bruggeman, F. Iza, and R. Brandenburg, "Foundations of atmospheric pressure non-equilibrium plasmas," *Plasma Sources Sci. Technol.*, vol. 26, no. 12, Nov. 2017, Art. no. 123002.

[130] Y. Sato, K. Ishikawa, T. Tsutsumi, A. Ui, M. Akita, S. Oka, and M. Hori, "Numerical simulations of stable, high-electron-density atmospheric pressure argon plasma under pin-to-plane electrode geometry: Effects of applied voltage polarity," *J. Phys. D, Appl. Phys.*, vol. 53, no. 26, Jun. 2020, Art. no. 265204.

[131] F. Denes, "Macromolecular plasma-chemistry: An emerging field of polymer science," *Prog. Polym. Sci.*, vol. 29, no. 8, pp. 815–885, Aug. 2004.

[132] A. Cristofolini, A. Popoli, and G. Neretti, "A multi-stage model for dielectric barrier discharge in atmospheric pressure air," *Int. J. Appl. Electromagn. Mech.*, vol. 63, pp. S21–S29, Jul. 2020.

[133] Y. Zhang, H.-Y. Wang, Y.-R. Zhang, and A. Bogaerts, "Formation of microdischarges inside a mesoporous catalyst in dielectric barrier discharge plasmas," *Plasma Sources Sci. Technol.*, vol. 26, no. 5, Apr. 2017, Art. no. 054002.

[134] J. Sornsakdanuphap, P. Suanpoot, Y. J. Hong, B. Ghimire, G. Cho, H. S. Uhm, D. Kim, Y. J. Kim, and E. H. Choi, "Electron temperature and density of non-thermal atmospheric pressure argon plasma jet by convective wave packet model," *J. Korean Phys. Soc.*, vol. 70, no. 11, pp. 979–989, Jun. 2017.

[135] P. Suanpoot, J. Sornsakdanuphap, H. S. Uhm, G. Cho, and E. H. Choi, "Using plasma propagation speed model for investigation of electron temperature of AR/N$_2$ in non-thermal atmospheric pressure indirect-plasma jet," in *Proc. IEEE Int. Conf. Plasma Sci. (ICOPS)*, Jun. 2016, p. 1.

[136] A. Kamal, "Design and experimental investigations of electrical breakdown in a plasma jet device and applications," Ph.D. dissertation, Dept. Elect. Eng., Benha Univ., Benha, Egypt, 2014.

[137] J. Žigon, M. Petrič, and S. Dahle, "Dielectric barrier discharge (DBD) plasma pretreatment of lignocellulosic materials in air at atmospheric pressure for their improved wettability: A literature review," *Holzforschung*, vol. 72, no. 11, pp. 979–991, Nov. 2018.

[138] P. Cools, L. Astoreca, P. S. E. Tabaei, M. Thukkaram, H. De Smet, R. Morent, and N. De Geyter, "Surface treatment of polymers by plasma," in *Surface Modification of Polymers*. Weinheim, Germany: Wiley, 2019, pp. 31–65.

[139] J. López-García, "Wettability analysis and water absorption studies of plasma activated polymeric materials," in *Non-Thermal Plasma Technology for Polymeric Materials*. Amsterdam, The Netherlands: Elsevier, 2019, pp. 261–285.

[140] H. Chim, J. L. Ong, J.-T. Schantz, D. W. Hutmacher, and C. M. Agrawal, "Efficacy of glow discharge gas plasma treatment as a surface modification process for three-dimensional poly (D,L-lactide) scaffolds," *J. Biomed. Mater. Res.*, vol. 65A, no. 3, pp. 327–335, Jun. 2003.

[141] H. Hody, J.-J. Pireaux, P. Choquet, and M. Moreno-Couranjou, "Plasma functionalization of silicon carbide crystalline nanoparticles in a novel low pressure powder reactor," *Surf. Coat. Technol.*, vol. 205, no. 1, pp. 22–29, Sep. 2010.

[142] E. Haye, Y. Busby, F. da Silva Pires, F. Bocchese, N. Job, L. Houssiau, and J.-J. Pireaux, "Low-pressure plasma synthesis of Ni/C nanocatalysts from solid precursors: Influence of the plasma chemistry on the morphology and chemical state," *ACS Appl. Nano Mater.*, vol. 1, no. 1, pp. 265–273, Jan. 2018.

[143] R. Agarwal, Y. Vijay, and K. Swami, "Reactive polymer surfaces for cell colonization," *J. Mater. Sci. Surf. Eng.*, vol. 1, no. 2, pp. 32–35, 2014.

[144] S. Jiansirisomboon, K. J. D. MacKenzie, S. G. Roberts, and P. S. Grant, "Low pressure plasma-sprayed Al$_2$O$_3$ and Al$_2$O$_3$/SiC nanocomposite coatings from different feedstock powders," *J. Eur. Ceram. Soc.*, vol. 23, no. 6, pp. 961–976, May 2003.

[145] K. Norrman, Y. Wang, E. Stamate, and W. Zhang, "Controlling surface properties of electrospun polyphenylsulfone using plasma treatment and X-ray photoelectron spectroscopy," *Heliyon*, vol. 5, no. 6, Jun. 2019, Art. no. e01943.

[146] S. M. M. Spyrides, F. S. Alencastro, E. F. Guimaraes, F. L. Bastian, and R. A. Simao, "Mechanism of oxygen and argon low pressure plasma etching on polyethylene (UHMWPE)," *Surf. Coatings Technol.*, vol. 378, Nov. 2019, Art. no. 124990.

[147] C. Mandolfino, E. Lertora, C. Gambaro, and M. Pizzorni, "Functionalization of neutral polypropylene by using low pressure plasma treatment: Effects on surface characteristics and adhesion properties," *Polymers*, vol. 11, no. 2, p. 202, Jan. 2019.

[148] A. Řezníčková, Z. Kolská, V. Hnatowicz, P. Stopka, and V. Švorčík, "Comparison of glow argon plasma-induced surface changes of thermoplastic polymers," *Nucl. Instrum. Methods Phys. Res. B, Beam Interact. Mater. At.*, vol. 269, no. 2, pp. 83–88, Jan. 2011.

[149] V. Švorčík, K. Kolářová, P. Slepička, A. Macková, M. Novotná, and V. Hnatowicz, "Modification of surface properties of high and low density polyethylene by ar plasma discharge," *Polym. Degradation Stability*, vol. 91, no. 6, pp. 1219–1225, Jun. 2006.

[150] P. Slepicka, N. S. Kasalkova, E. Stranska, L. Bacakova, and V. Svorcik, "Surface characterization of plasma treated polymers for applications as biocompatible carriers," *Exp. Polym. Lett.*, vol. 7, no. 6, pp. 535–545, 2013.

[151] N. Bhatnagar, "Effect of plasma on adhesion characteristics of high performance polymers," *Rev. Adhes. Adhesives*, vol. 1, no. 4, pp. 397–412, Nov. 2013.

[152] S. Tang and H. S. Choi, "Comparison of low- and atmospheric-pressure radio frequency plasma treatments on the surface modification of poly(methyl methacrylate) plates," *J. Phys. Chem. C*, vol. 112, no. 12, pp. 4712–4718, Mar. 2008.

[153] H. M. S. Iqbal, S. Bhowmik, and R. Benedictus, "Surface modification of high performance polymers by atmospheric pressure plasma and failure mechanism of adhesive bonded joints," *Int. J. Adhes. Adhesives*, vol. 30, no. 6, pp. 418–424, Sep. 2010.

[154] W. Yan, "Nanocomposite dielectric materials for power system equipment," Ph.D. dissertation, School Elect. Eng. Telecommun., Fac. Eng., Univ. New South Wales, Sydney NSW, Australia, 2013.

[155] W. Yan, B. T. Phung, Z. J. Han, and K. K. Ostrikov, "Dielectric performance of nanocomposites synthesized by poly(ethylene oxide)-like film coated silica nanoparticles by plasma polymerization," in *Proc. IEEE Electr. Insul. Conf. (EIC)*, Jun. 2013, pp. 424–428.

[156] W. Yan, B. T. Phung, Z. J. Han, and K. Ostrikov, "Plasma polymer-coated on nanoparticles to improve dielectric and electrical insulation properties of nanocomposites," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 21, no. 2, pp. 548–555, Apr. 2014.

[157] W. Yan, Z. J. Han, W. Z. Liu, X. P. Lu, B. T. Phung, and K. Ostrikov, "Designing atmospheric-pressure plasma sources for surface engineering of nanomaterials," *Plasma Chem. Plasma Process.*, vol. 33, no. 2, pp. 479–490, Apr. 2013.

[158] W. Yan, B. T. Phung, Z. J. Han, and K. Ostrikov, "Plasma functionalization of SiO$_2$ nanoparticles for the synthesis of polymer nano-dielectrics," in *Proc. IEEE 10th Int. Conf. Properties Appl. Dielectr. Mater.*, Jul. 2012, pp. 1–4.

[159] W. Yan, Z. Han, B. Phung, F. Faupel, and K. Ostrikov, "High-voltage insulation organic-inorganic nanocomposites by plasma polymerization," *Materials*, vol. 7, no. 1, pp. 563–575, Jan. 2014.

[160] F. Musa, N. Bashir, M. Ahmad, Z. Buntat, and M. Piah, "Investigating the influence of plasma-treated SiO$_2$ nanofillers on the electrical treeing performance of silicone-rubber," *Appl. Sci.*, vol. 6, no. 11, p. 348, Nov. 2016.

[161] N. A. Awang, M. H. Ahmad, Y. Z. Arief, I. H. Zakaria, and N. A. Ahmad, "The effect of plasma-treated boron nitride on partial discharge characteristics of LDPE," *Int. J. Electr. Comput. Eng.*, vol. 7, no. 2, p. 568, Apr. 2017.

[162] N. A. Awang, M. H. Ahmad, Z. A. Malek, M. A. B. Sidik, Z. Nawawi, M. I. Jambak, E. P. Waldi, and Aulia, "AC breakdown strength enhancement of LDPE nanocomposites using atmospheric pressure plasma," in *Proc. Int. Conf. Electr. Eng. Comput. Sci. (ICECOS)*, Aug. 2017, pp. 290–294.

[163] J. F. Hall, "History and bibliography of polymeric insulators for outdoor applications," *IEEE Trans. Power Del.*, vol. 8, no. 1, pp. 376–385, Jan. 1993.

[164] D. Shi, P. He, S. X. Wang, W. J. V. Ooij, L. M. Wang, J. Zhao, and Z. Yu, "Interfacial particle bonding via an ultrathin polymer film on Al$_2$O$_3$ nanoparticles by plasma polymerization," *J. Mater. Res.*, vol. 17, no. 5, pp. 981–990, May 2002.

[165] S. M. Hosseini, S. S. Madaeni, A. R. Khodabakhshi, and A. Zendehnam, "Preparation and surface modification of PVC/SBR heterogeneous cation exchange membrane with silver nanoparticles by plasma treatment," *J. Membrane Sci.*, vol. 365, nos. 1–2, pp. 438–446, Dec. 2010.

[166] N. Mukherjee, D. Wavhal, and R. B. Timmons, "Composites of plasma surface functionalized barium titanate nanoparticles covalently attached to epoxide matrices: Synthesis and evaluation," *ACS Appl. Mater. Interfaces*, vol. 2, no. 2, pp. 397–407, Feb. 2010.

[167] R. Huang, Z. Chen, X. Chu, Z. Wu, and L. Li, "Preparation and thermal properties of epoxy composites filled with negative thermal expansion nanoparticles modified by a plasma treatment," *J. Compos. Mater.*, vol. 45, no. 16, pp. 1675–1682, Aug. 2011.

[168] S. Mathioudaki, B. Barthélémy, S. Detriche, C. Vandenabeele, J. Delhalle, Z. Mekhalif, and S. Lucas, "Plasma treatment of metal oxide nanoparticles: Development of core-shell structures for a better and similar dispersibility," *ACS Appl. Nano Mater.*, vol. 1, no. 7, pp. 3464–3473, 2018.

NORHAFEZAIDI MAT SAMAN was born in Ipoh, Perak, in July 1995. He received the B.Eng. degree in electrical engineering from Universiti Teknologi Malaysia (UTM), Malaysia, in 2018, where he is currently pursuing the Ph.D. degree in electrical engineering. He is focusing on the plasma applications field and nanocomposite insulations. He also explores the insulation diagnostic study for medium and high voltage applications.

MOHD HAFIZI AHMAD (Member, IEEE) received the Ph.D. degree in high voltage engineering from Universiti Teknologi Malaysia. His Ph.D. works are related to partial discharge measurement in insulation. He was formerly a Visiting Researcher with the University of Leicester, U.K. In IVAT UTM, he serves as a Testing and Calibration Manager, and is involved in many calibrations and testing jobs related to high voltage equipment. He is currently working as a Senior Lecturer and a Researcher with the Institute of High Voltage and High Current (IVAT), Universiti Teknologi Malaysia (UTM). He is also a Senior Technical Advisor for Global Testing Services (M) Sdn. Bhd. Moreover, he has conducted several trainings and lectures on partial discharge fundamentals and measurement on primary electrical equipment for industries and Universities. He has conducted field and laboratory partial discharge measurements for transformer, switchgear, cable, and rotating machines using conventional and non-conventional methods. He is a member of CIGRE and IET U.K.

ZOLKAFLE BUNTAT was born in Pontian, Johor, in July 1964. He received the Diploma degree in electrical (communication) engineering and the B.Sc. degree in electronics and electrical engineering from the University of Strathclyde, U.K., in 1985 and 1988, respectively, the M.E.E. degree in high voltage insulator study from Universiti Teknologi Malaysia, in 1996, and the Ph.D. degree in high voltage and pulsed power engineering from Loughborough University, U.K., in February 2005. He is currently a Retired Professor with the School of Electrical Engineering, Universiti Teknologi Malaysia. His teaching and research responsibilities are in the areas of electrical power engineering, high voltage technology, electrical discharges, and ozone generation and technology. He had also given lectures at many high voltage short courses for industries. He had also published many research articles in indexed journals and many research papers in indexed conferences. His research interests include the generation of electrical discharges operating at atmospheric pressure, ozone generation, ozone application in medical, water, and wastewater treatment, rainwater harvesting, removal of NOx and SOx from diesel engine vehicles, and the process of agriculture produce and non-thermal plasma applications in medicine, environmental, and agriculture. He is a member of the International Ozone Association (IOA), the International Water Association (IWA), and the IEEE Nuclear and Plasma Sciences Society. He is also a member of the SIRIM Technical Committee for IEC, TC 115 high voltage direct current (HVDC) transmission for DC voltages above 100 kV.

• • •