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Electrical treeing in high voltage insulations: a review on nanocomposite insulating materials and their processing techniques

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Electrical treeing is one of the major causes of breakdown/failure of polymeric insulations. Nanocomposite materials play an important role in preventing insulation breakdown due to the presence of nanoparticles. Nanoparticles act as charge trapping sites and physical barriers to electrical treeing thereby hindering their growth. This study presents a review on electrical treeing studies on nanocomposite materials and processing techniques employed to mitigate insulation failure due to electrical treeing. Various polymers combined with different nanofillers as well as the most common processing techniques are also reviewed. Emerging trends on nanomaterial surface modification to improve electrical treeing resistance are also discussed.

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1. Introduction

Electrical tree is a pre-breakdown phenomenon that occurs in polymeric insulating materials used in high voltage insulation system [1]. Electrical treeing initiates due to imperfections in the insulations such as impurities, voids, defects, or conducting projections due to excessive electrical field stress within small regions of the insulation [2]. The propagation of a tree-like structure will eventually bridge the entire insulation thickness and thereby leading to insulation breakdown.

Consequently, addition of tree inhibitors, reinforcement and/or modifying the insulating material has been proposed by numerous researchers in order to increase the tree resistance of insulating materials [3-6]. Results from literature have shown that nanofillers are superior in inhibition of electrical treeing compared to the microfiller in polymeric insulating material [7-14]. However, there are problems related with these nanofillers as that they tend to agglomerate because of surface incompatibility between the polymer matrices and fillers [15]. As a result, the insulation performance of these nanocomposites is often significantly degraded. Several processing techniques have also been introduced to modify the surface of nanoparticles that are mixed with the polymers to improve the dispersion uniformity in the polymer matrices [16].

This paper presents a comprehensive review on nanocomposites materials used in high voltage insulations as well as the most commonly used methods to inhibit electrical treeing. The theory of nanocomposites material, roles of nanofillers and interfaces, previous research findings, sample configuration and recent research trends in electrical treeing of nanocomposite-based materials are discussed.

2. Nanocomposites material

Polymer nanocomposites are composites containing nano-sized fillers which are homogenously distributed within the polymer matrix by several weight percentages (%wt). As defined, the fillers added to the matrix are very small in quantity which are less than 10wt% compared to conventional microcomposites which contain 50wt% of filler [17,18]. Nanocomposites can be classified into several types such as conventional filled composite, mesocomposite, nano-micro composite and nano-meso composite [19].

Nanocomposites have the potential to improve the electrical, mechanical and thermal properties compared to the neat polymers [15,20-22]. Most nanocomposites can be classified as either functional materials (based on their electrical, magnetical and optical behavior) or structural materials (based on their mechanical properties). In terms of their mechanical properties, they can be further classified into polymer and non-polymer based. The non-polymer based includes metal/metal, metal/ceramic and ceramic/ceramic. While the polymer based include polymer/ceramic, inorganic/organic polymer, inorganic/organic hybrid, polymer/layered silicate, polymer/polymer and biocomposites.

However, in electrical treeing studies, the nanocomposites used comprise of inorganic/organic nanocomposites and polymer/layered silicate nanocomposites. Inorganic/Organic are polymer nanocomposites which comprise of a mixture of organic or inorganic polymers and metal oxides such as polymer/metal oxide nanocomposites. These types of nanocomposites exhibit a good performance in suppressing electrical treeing. Polymer/layered silicate nanocomposites received attention as one of the popular nanocomposites because they often perform good improvements in materials when compared with pure polymer and conventional microcomposites [23,24]. Sepiolite [25] and montmorillonite [26] are among the most commonly used layered silicates for the preparation of such nanocomposites.

Generally, polymer nanocomposites have three main constituents; polymer matrix, nanofiller and interaction zone as shown in Figure 1. The nanofiller are mixed homogenously into polymer matrix. The interaction between the matrix and filler which are the interfaces play important role in property enhancement of polymer. A further description and discussion of the polymer nanocomposite constituents are presented in the subsections below.

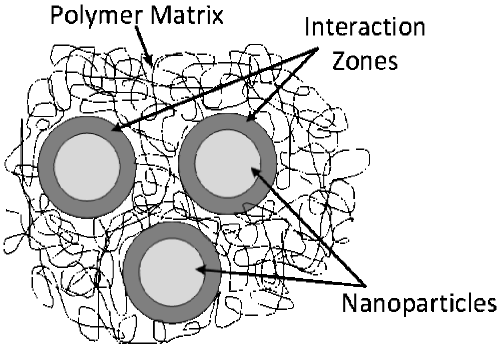

Fig. 1. Constituent of polymer nanocomposite [27,28]

a) Polymer matrix

Polymer is a substance that has a molecular structure consisting of structural units joined together by covalent bonds. Polymers can be classified as thermoplastics, thermoset and elastomers [29]. Elastomers are rubbers or rubberlike elastic materials such as silicone rubber or natural rubber. Thermoplastic polymers are hard at room temperature but become soft when heated and more or less fluid and thus can be molded. Polyethylene and its families are examples of thermoplastics. While, thermosetting polymers can be molded at room temperature or above, however when heated at higher temperature, they become hard and infusible. Epoxy resin commonly used in high voltage insulation is an example of a thermoset [30,31].

The structural characteristics are of paramount importance in determining the properties of polymers. Thermosetting polymers normally are made from relatively low molecular weight but when heated they become highly cross-linked thus becoming a hard and infusible polymer. A crosslink is a chemical bond between polymer chains. Crosslinks are important in determining physical properties because they increase the molecular weight and limit the translational motions of the chains with respect to one another [32].

With regards application in high voltage insulation, polymers on their own cannot perform greatly [33,34] and thus some additives must be added into the polymer matrix in order to enhance their performance. Such additives include filler, antioxidant and flame retardant depending on its function. Additives can alter the molecular structure or add a second phase in effect transforming the polymer into a composite material [30]. Performance of polymers have been shown to improve when additives such as fillers were added into them [35,36].

b) Nanofiller and its roles

Nanofillers (which are nano-sized particles) are the additives that are added to polymers to enhance the performance of the composites. Nanoparticles can be classified into one dimensional, two dimensional and three dimensional. One dimensional nanofiller is normally referred to as thin platelet or lamellar particles, two dimensional as whisker or rod particles and three dimensional as organic oxides in spherical or rectangular shapes. Figure 2 illustrates the nanofiller classifications and their sizes [29,37,38].

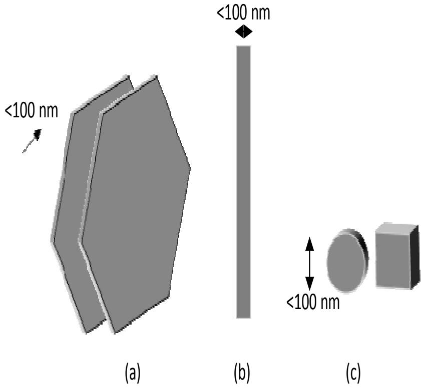

Fig. 2. Classification of nanofiller a) one dimensional b) two dimensional c) three dimensional [38] (nanofiller size less than 100nm)

The most common nanoparticles used in nanocomposites for electrical insulation are silicon dioxides, metallic oxides and hydroxides (alumina, titanium oxide, magnesium oxide, zinc oxide, etc.), nanoclays (montmorillonite, sepiolite, etc.) and carbon nanotubes (CNT).

Silicon dioxide (SiO_2) also known as silica has been known for its hardness. It is like a quasi-spherical shape with diameter ranging from a few nanometers to 100 nm. Nanosilica synthesis has improved significantly and widely used together with numerous polymer matrices [31].

Metal oxides are often used as nanofillers to build dielectric nanostructure materials. They have good electrical insulating behavior and can be dispersed quite easily due to their hydrophilic characteristic. Some examples of metal oxides include Titanium dioxide, TiO_2, also known as titanium oxide or titania which can be produced both in whiskers and quasi-spherical particles; Magnesium oxide, MgO, is formed by an ionic bond between one magnesium and one oxygen atom. Aluminum oxide, Al_2O_3 often referred to as alumina is another type of metal oxide used as a nanofiller to improve thermal conductivity of composites while zinc oxide, ZnO is an inorganic compound and rarely used as a nanofiller [31].

Nanoclays belong to the family of phyllosilicates also known as layered silicates. A silicate is a compound containing silicone (Si) and oxygen (O) forming a tetrahedral structure. Each tetrahedral has an excess of negative net electrical charge and the silicate has that charge balanced by some metal cations in order to achieve an electrically neutral compound. These metals (usually Fe, Mg, K, Na and Ca) have the fundamental purpose to bind together the different silicate tetrahedral structures. Phyllosilicates consist of two-dimensional layers where a central octahedral sheet of alumina is fused to two external silica tetrahedrons by the tip so that the oxygen ion of the octahedral sheet shares also the tetrahedral sheets [18,28,39]. Figure 3 shows the structure of phylosilicates.

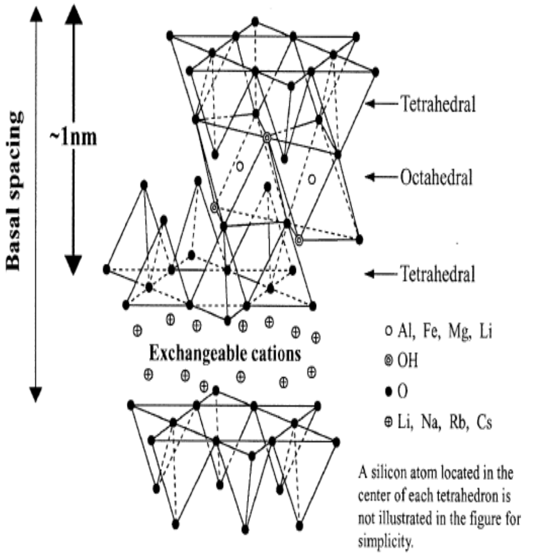

Fig. 3: Structure of 2:1 phyllosilicates [18].

Carbon nanotubes (CNT) are allotropes of carbon and comprise of hexagonal networks of carbon atoms. CNT present exceptional mechanical and thermal properties [40]. In addition, they can be electrical conductors, semiconductors or insulators depending on their chirality [41]. In polymers for electrical applications, they can be used to obtain semiconductive nanostructure materials having tuned electrical conductivity [38].

The presence of these nanoparticles in polymer matrix helps to prevent growth of electrical treeing as they act as an obstacle against treeing. As it is known, electrical treeing is initiated by charge carrier injection and extraction from embedded electrode into the polymer [42,43]. In nanocomposites, when high voltage is applied to them, the electrons (carriers) are injected from high voltage electrode. The free electrons present in the polymer get trapped in or around nanoparticles. These trapped carriers require high energy to move or get extracted from the polymer. This result in slower degradation rate and require more time from tree to initiate [35].

Nano-filled polymers can be differentiated from conventional micro-filled polymer by the filler content in %wt to be added with polymer matrix, filler size as well as distance between neighboring filler and surface area of the filler [18]. Nanoparticles have a small filler size as well as small distance between neighboring filler. Tree propagation in insulations filled with nanofillers takes longer time due to the fact that the nanofillers have higher discharge resistance and thus the tree propagates around the particle through the polymer matrix [35].

c) Interaction zone and its roles

Nanocomposites provide a promising future due to their advantages. The main advantage is the use of nanofiller. This is because the nano-sized filler provide larger surface area as the size of the particles become smaller. The surface area is the interfacial region of filler and polymer [44,45]. The multi-core model of polymer nanocomposites is as shown in Figure 1. There are three layers of the nanocomposites. The first layer is the transition layer tightly bonded to both inorganic and organic substances by coupling agents. The second layer is an interfacial region which consists of polymer chains strongly bound and interacted to the first layer and the surface of the inorganic particles. The third layer is a region loosely coupling and interacting to the second layer [46].

The interfaces play an important role in insulating process. The reduction of sizes of the nanoparticles results in larger interaction zone. The larger interaction zone modify the polymer behavior, change morphology and reduce internal field [47]. The interfaces act as an important part in electrical treeing suppression. When the tree initiates, it grows through polymer matrix and moves towards the nanoparticles. Once the tree channel reaches the nanoparticles, it follows a conductive interface region and starts to propagate along the interfaces and continues to propagate to other nanoparticles through the polymer matrix [35]. Due to larger surface area, more time is needed for the tree to propagate. Figure 4 shows a physical model of the electrical tree in nanocomposites [15].

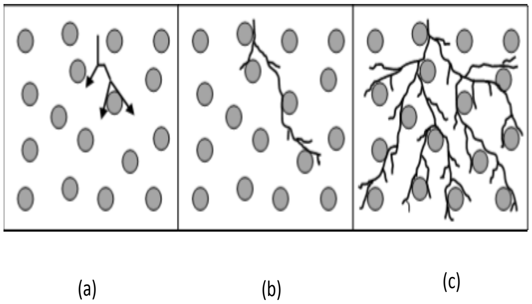

Fig. 4: Physical model of electrical tree formation in nanocomposites (a) tree propagation path (b) tree contact with nanoparticles (c) tree growth at interface between nanoparticles and polymer [15]

1. Previous studies on electrical treeing in nanocomposites material

Numerous researches have been conducted to investigate the electrical treeing performance of the nanocomposites. Results from the literature have shown that nanocomposites in general improved electrical treeing resistance in polymeric insulations. There are a lot of available polymers in existence, however in relation to electrical treeing studies, epoxy resins, polyethylene, silicone rubber and ethylene-vynil acetate will be discussed.

Epoxy resin is a commonly used polymer for high voltage insulation applications as it offers many advantages compared to other polymer such as strong adhesion, mechanical and electrical properties [48,49]. There are a lot of researches that have investigated the electrical treeing performance of epoxy resin used as the base polymer in high voltage insulations. Nanofillers such as silica, alumina, titania, zinc oxide have been used in epoxy resin-based polymers to improve the performance of the nanocomposites. Investigation by Raetzke et al [50] shows tree initiation time for epoxy nanocomposites was prolonged by one order of magnitude at high field (>10 kVrms) and two order of magnitude at low field (2-3 kVrms) compared to neat epoxy resins. In another study by Chen et al [51] investigating the tree initiation time of base epoxy resin (BER), microcomposite (MC), nanocomposite (NC) and nano- and micro-filler mixed composite (NMMC), results showed that at high field, tree initiation time for NC was high while at low field, NMMC showed higher tree initiation time compared to the other composites.

The concentration of the filler plays an important role in enhancing the treeing performance in epoxy resin nanocomposites. Epoxy resin mixed with SiO_2 filler were able to suppress the electrical tree growth and prolong the tree initiation time [12,52]. Alapati et al [35] showed that the tree initiation time and tree growth resistance increased with the increase in epoxy/Al_2O_3 nanocomposites with different filler loading. Even small quantity of nanofillers (0.1% wt) was sufficient to suppress the tree growth in epoxy nanocomposites. Microscopic treeing images captured in their study revealed that the unfilled epoxy nanocomposites exhibited a branch like shape and almost bridged the insulation while 3%wt filled nanocomposites was a bush like structure with a much shorter tree length implying the tree growth was inhibited. Ding et al [9] investigated that treeing in epoxy/ZnO nanocomposites and found that addition of ZnO nanoparticles into epoxy resin improved the resistance to electrical tree growth and increase the time to breakdown. Besides, the shape and size of the filler affected the treeing performance. Nagao et al [13] reported that tree growth was also influenced by filler shape. Round shape filler prevented tree propagation more significantly than rectangular shape, however, the tree initiation voltage were almost the same for both shapes. Iizuka et al [11] investigated the treeing breakdown time using different filler size. Nanofiller size of 12, 40, 90 and 300 nm with the same concentration of filler tested. Results showed that the optimum size of nanofiller used is the smallest size which is 12nm in order to obtain the maximum tree breakdown time.

Another type of polymer used in high voltage insulation is polyethylene (PE), which is widely used in underground distribution and transmission cables because of its excellent electrical insulating properties and good thermo-mechanical behaviour. PE can be classified into several types but the most commonly used in high voltage application are High Density Polyethene (HDPE), Low Density Polyethene (LDPE) and Cross Linked Polyethene (XLPE). Sridhar et al [53] investigated the PE/ SiO_2 nanocomposites for different filler loading. The tree inception voltage increase when the concentration of nanofiller increases. For base PE, the tree growth pattern forms a branch-like tree while for PE nanocomposites tree growth pattern form a bush-like tree.

Low Density Polyethylene (LDPE) is the low density version of PE that has excellent resistance to water, moisture and most organic solvents and chemicals. LDPE is the major type of insulating material of power cables. Guastavino et al [54] investigated treeing phenomena of LDPE using sepiolite (SEP) and montmorillonite (MMT). LDPE/MMT nanocomposites effectively slowed down the propagation of electrical treeing thus increasing the time to breakdown. The LDPE/SEP + MMT nanocomposite shows a much longer duration of time to breakdown compared to pure LDPE. Tiemblo et al [14] reported electrical treeing performance of LDPE mixed with SiO_2, fibrous and laminar silicates. In their study, results showed that LDPE/ SiO_2 reduced the electrical stability by decreasing the inception of the tree growth and increasing the tree propagation rate. LDPE/laminar silicate and LDPE/fibrous silicate had barrier effects on the propagation of the tree growth at voltages below 15 kV as it increased the duration of the electrical tree propagation and hence the dielectric stability. Likewise, Kurnianto et al [55] reported the improvement in treeing breakdown by improving resistance against partial discharge in LDPE filled with MgO nanoparticles. The time lag to breakdown increased with the increase in filler concentration thus suggesting that nanocomposites are resistive to partial discharge. Results from [14,54,55] showed that pure LDPE have the lesser resistance to treeing compared to LDPE nanocomposites.

For outdoor insulation, silicone rubber (SiR) is widely used in XLPE cable accessories which covers the voltage levels ranged from 10 to 500 kV due to its excellent electrical and mechanical properties. Pure SiR has poor electrical conductivity as the tree grows when the applied voltage is increased [33,34]. Therefore, to improve the SiR, nanofillers have been added to the silicone rubber (as base polymer). Du et al [10] investigated Room Temperature Vulcanized, RTV SiR/SiO_2 nanocomposites with the filler content from 0 to 5 wt%. The tree inception time and tree inception voltage of the nanocomposites increased with increasing filler loading. The highest filler concentration exhibited the highest tree inception time and inception voltage. However, the tree growth speed for SiR/SiO_2 nanocomposites with 3%wt had the slowest tree growth speed followed by 5%wt, 15%wt and pure SiR. Similarly, Xiang et al [56] reported studies on high temperature vulcanization (HTV) SiR/SiO_2 nanocomposites. Results from their study revealed that tree initiation voltage rose at initial stage and then later fell, the threshold was around 35% by weight with the increase of filler loading. Besides, Musa et al [57] investigate the SiR/TiO_2 nanocomposites. TiO_2 acted as a barrier to prevent tree propagation. However, further increase in the nanofiller concentration resulted in the magnitude of the partial discharge increasing as well as the number of tree branches due to the increase in number of discharges. The uses of organic fillers on silicone rubber polymer matrices were recently reported. One of the organic filler that has been studied for application in high voltage insulations is Montmorillonite (MMT). MMT exhibits excellent mechanical and electrical properties and has been reported to be a good electrical treeing inhibitor. A modified version of the MMT called Organo-Montmorillonite (OMMT) has also been reported to be good organic filler for high voltage insulation. Studies by Ahmad et al and Jamil et al [58-61] have shown that OMMT created a barrier to treeing propagation. SiR/OMMT nanocomposites had the highest tree initiation voltage compared to other filled nanocomposites [58]. Trapping sites that existed at interface between silicone rubber and OMMT nanofiller helped to increase the voltage for charge to pass through the barrier from the OMMT nanofiller. Figure 5 depicts the microscopic images of the electrical tree for pure/neat silicone rubber, silicone rubber nanocomposites (1%wt) and silicone rubber nanocomposites (3%wt) respectively.

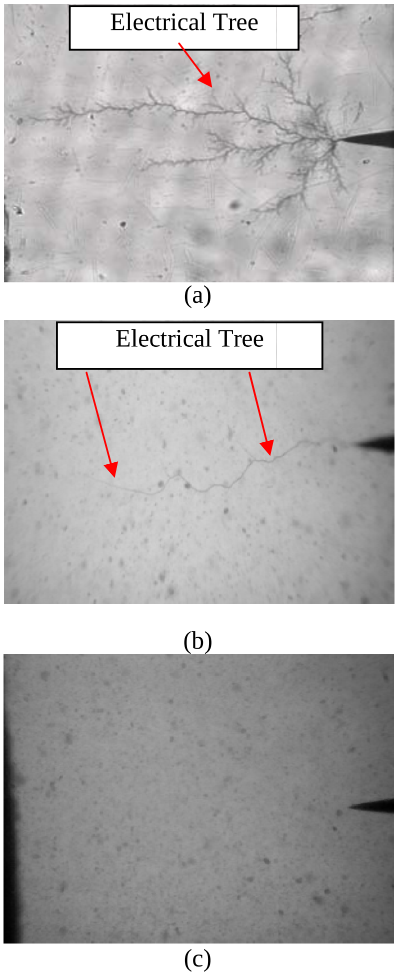

Fig. 5. Miscrocopic images of electrical trees of Silicone Rubber samples filled with OMMT nanofillers (a) Neat Silicone Rubber (b) Silicone rubber with 1% wt OMMT nanofiller (c) Silicone rubber with 3% wt OMMT nanofiller [58]

Another type of polymer used in cable accessories as heat shrinkable insulation, semi-conductive insulation jackets, and flame retardant insulation is Ethylene-Vynil Acetate (EVA). This polymer usually acts as a copolymer mixed with other polymers to improve its performance. Guastavino et al [62] investigated EVA mixed with unmodified and organically modified synthetic fluorohectorite, as well as organically modified natural MMT. EVA/modified MMT showed very high tree inception voltage. EVA nanocomposites containing Bohemite and MMT nanoparticles have also been investigated. Bohemite has a nanometric cube-like symmetry and MMT particles are a typical layered structure. The results show that the growth of the electrical treeing inside the original polymer can be altered significantly by the dispersion of inorganic nanoparticles.

The breakdown voltage decreased for EVA/MMT nanocomposite and it showed very good performance since the decrease of the breakdown strength is slight while the increase of the duration during aging tests was significant [63]. Electrical treeing performance of EVA/layered silicate was reported by Guatavino et al [64] whereby comparison between pure EVA with EVA nanocomposites in growth of electrical treeing was reported. The nanocomposite EVA/layered silicate showed superior performance in the presence of electrical tree compared to pure EVA.

2. Processing technique of nanocomposites

Even though the addition of additives or fillers to polymers showed a good performance in relation to treeing, however, the use nanofillers in polymers cause problems such as lack of adhesion between the nanofiller and the polymer matrix [15,65,66]. In order to improve particle dispersion, several techniques such as direct mixing method, intercalative method, coupling agents and surface modification of the nanoparticles or nanofiller by physical and chemical methods using surfactants are used [17]. The subsequent subsections present discussions on the common processing techniques employed in electrical treeing studies.

a) Direct mixing

Direct mixing method is a basic technique to improve the dispersion and distribution of the nanofiller in polymer. This method is very simple and chemical-free. The mixing process is employed during the addition of the nanofiller into the polymer. This method is used in the formation of nanocomposites for electrical treeing and it improves the electrical performance of the nanocomposites [67]. Direct mixing process is easy, low cost and suitable for mass production.

The preparation of direct mixing of the composites is as shown in Figure 6. The polymer is mixed with filler and then with the hardener. The mixing process of nanocomposites and microcomposites are almost the same. However, the preparation of nanocomposites involves applying shear force. Shear force is applied to the mixing process to disaggregate the nanofiller and enhance the dispersion [36,68].

Several studies on electrical treeing of polymer composites using direct mixing have been reported. Du et al [34] and Hosier et al [35] processed SiR (as base polymer) using the direct mixing method. The SiR samples were cast using the needle plane arrangement to test the treeing performance. Imai et al [36] used direct mixing method to study the performance of base epoxy resin, microcomposites, nanocomposites and nano-micro composites with layered silicate, SiO_2 and TiO_2.

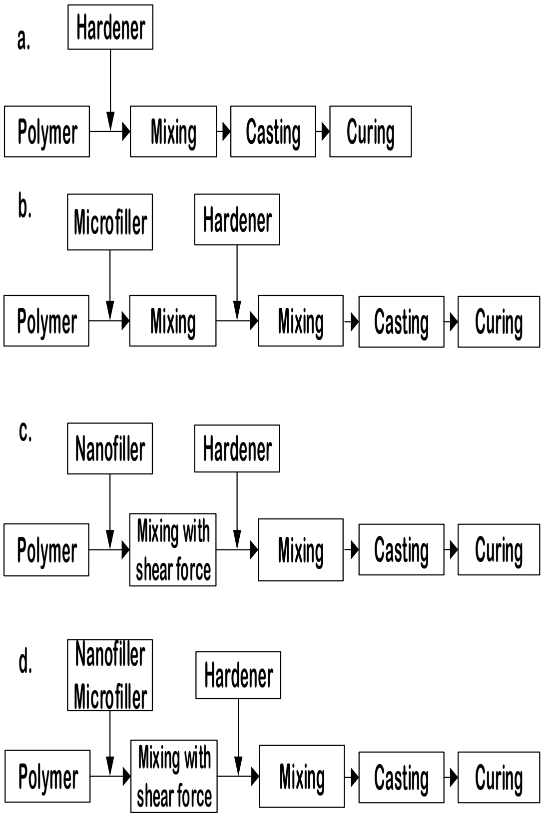

Fig. 6: Preparation schemes for a) base polymer b) microcomposites c) nanocomposites d) nano and micro mixed composites [36]

The nanofillers or microfiller or both were dispersed in the epoxy resin by mixing with shear force. The specimens were produced by a curing reaction with a hardener. Musa et al [57] investigated SiR/ TiO_2 nanocomposites using direct mixing methods. Similar processes have been applied to other polymer/metal oxide nanocomposites as well [7,9,10,12,36,57,68]. In these studies, the nanocomposites were mixed using mechanical disperser to obtain a homogeneous dispersion of nanofiller. Hardener was then added and mixed as well using a mechanical disperser. However, a drawback of the direct mixing techniques is that during the blending/dispersion process of nanocomposites, air bubbles inside the polymers sample are usually formed. To reduce the air bubbles, the samples are degassed. The void/bubble in the nanocomposites might be the starting point of the partial discharge initiation thereby leading to treeing phenomena. Partial discharge is known as a discharge of a gas-filled void in an insulation and it commonly partially bridges the insulation [2,15,69].

b) Intercalative method

Intercalative method is another processing technique employed in clay nanocomposites. This method intercalates the polymer and inorganic layered substances to enhance the dispersion of the nanocomposites. Figure 7 shows the intercalation and exfoliation process of clay nanocomposites. The intercalation method is sub-divided into three namely intercalation of polymer or pre-polymer from solution, in situ intercalative polymerization and melt intercalation [67].

Intercalation of polymer or pre-polymer from solution involves solvent such as water, chloroform, ethanol, acetone, methanol or toluene. The layered silicate is first swollen in a solvent. When the polymer and layered silicate solutions are mixed, the polymer chains intercalate and displace the solvent within the interlayer of the silicate. After the solvent removal, the intercalated structure remains, resulting in polymer/layered silicate nanocomposites.

In situ intercalative polymerization method use liquid monomer. The step is almost the same as intercalation of polymer. The layered silicate is swollen within the liquid monomer or a monomer solution so the polymer formation can occur between the intercalated sheets. Polymerization can be initiated either by heat or radiation, by the diffusion of a suitable initiator, by an organic initiator or catalyst fixed through cation exchange inside the interlayer before the swelling step.

Melt intercalation method involves annealing, statically or under shear, a mixture of the polymer and organic modified layered silicate above the softening point of the polymer. The layered silicate is mixed with the polymer matrix in the molten state. Under these conditions, if the layer surfaces are sufficiently compatible with the chosen polymer, the polymer can crawl into the interlayer space and form either an intercalated or an exfoliated nanocomposite. This technique does not require the use of solvent [39,70].

Numbers of researches have reported the good performance of intercalative method in electrical treeing studies. Guastavino et al [54] reported that the mixture of two nanoparticles, sepiolite and MMT having different aspect ratio using melt intercalation method seems to be a promising approach. It showed a much longer duration than pure LDPE and fewer problems in the adhesion between the needle and the nanocomposite than MMT only. In addition, Guastavino et al [71] used melt intercalation in EVA/layered silicate nanocomposites. The results show that the breakdown voltage and tree inception voltage improved with the intercalated nanocomposites compare to pure EVA. Epoxy/layered silicate shows a treeing process in nanocomposites that treated with intercalation method suppressed treeing compared to the pure epoxy resins [72-74]. Even though the use of this technique showed good result in improving the insulation, however it is not suitable for industrial use for mass production. The use of chemical solvent makes the process not environmental friendly, toxic, flammable and complicated to handle. Besides, intercalation technique is only suitable for use in polymer/clay nanocomposites only. Therefore, this technique cannot be the appropriate processing method for the nanocomposites.

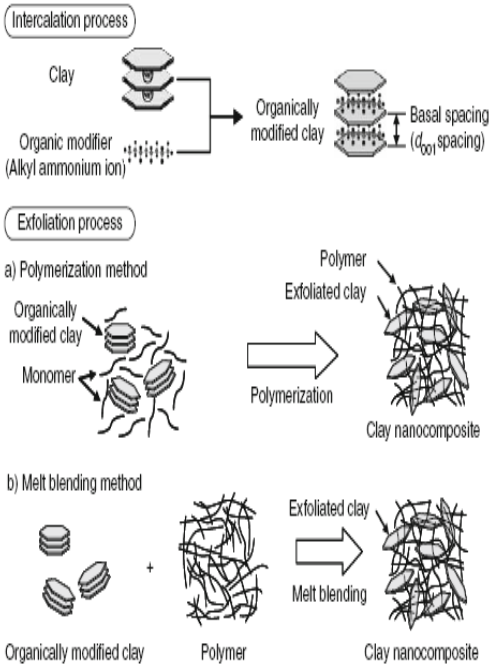

Fig. 7: Intercalation and exfoliation process of clay nanocomposites [39]

c) Coupling agent

Coupling agents are used to provide a stable bond between incompatible surfaces. The improved bond between inorganic component and the organic matrix polymer results in greater composite strength and longer endurance. The most commonly used agents include silane, zirconate and titanate to treat the filler [75]. Silane coupling agents have been in common use for decades providing enhanced adhesion between variety of inorganic substrates and organic resins. Silane coupling agents have the ability to form a durable bond between organic and inorganic materials and thus are used to enhance the adhesion of polymer-filler interfaces [76]. Another name for this method is silane coupling method due to the use of silane coupling agents such as ethoxysilane and methoxysilane. Fig. 8 shows the chemical structure of the silane coupling agents. Both the substances are hydrogen bonded by a monolayer. This bonding limits the motion of polymer outside the monolayer thereby extending the regions of immobile polymer in the interface. This means that the far-field influence is due to polymer entanglement trapping. This extended region can be 2 to 9 nm in thickness.

Silane coupling method has been used either in chemical or electrical application by researchers for treatment of polymer composites. Zhou et al [77] reported that use of coupling agents improved the interfacial bond strength between Aluminum (Al) particles and epoxy resin, and decreased the voids and defects at the phase interfaces. Todd et al [78] reported that the addition of silane coupling agents had significant effects on the dielectric constant of the interphase region but no effect on the dielectric constant values of the polymer phase. Iizuka et al [79] showed that silane coupling prolonged the treeing lifetime. However coupling agent method is similar to intercalative method as also uses chemical solvent thus limiting its application to research use and not suitable for industrial. In addition the processing of the coupling agents is difficult and need to be very meticulous when preparing the nanocomposites. Furthermore, some researchers have reported that this technique cannot guarantee good performance of polymers against electrical treeing. Nagao et al [13] and Kurnianto el al [80] reported that silane coupling did not change the tree inception voltage that result in propagation of electrical treeing.

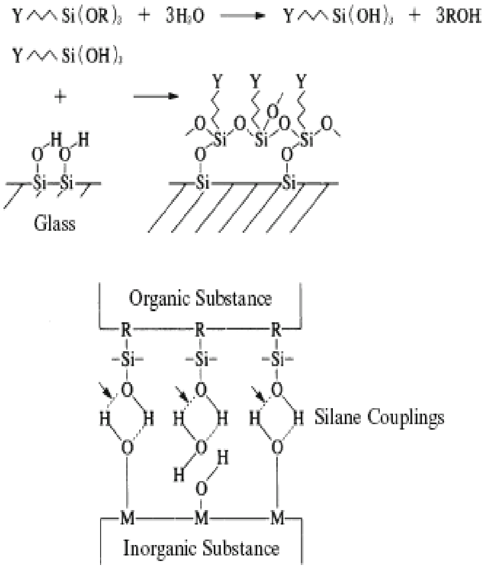

Fig. 8: Chemical structure of silane coupling [67]

2. Specimen configuration

Nanocomposites are usually cast into specimen to investigate their performance as insulating materials. There are several types of specimen configuration such as needle plane/needle plate, leaf like specimen and wire plane. All of the aforementioned configurations use needle in order to investigate the treeing process except wire plane. The three configurations are discussed below.

a) Needle plane/ needle plate

Needle plane or needle plate is a very common configuration employed in the study of electrical treeing in polymeric insulations. It uses a lot of nanocomposites during sample preparation. Needle plane configuration is based on a needle with small tip inserted into block sample of nanocomposites material placed on an electrode. The upper point of the needle is attached to high voltage supply and the lower electrode is connected to the ground. Fig. 9 (a) shows the needle plane configuration. The high divergent field present near the tip of the needle results in the initiation of electrical treeing which propagates towards the ground electrode. This configuration was introduced by Noto et al [81]. In their configuration, a block of size 14 x 23 x 6 mm of LDPE was used as a sample to test the tree initiation using DC and impulse voltage. A sewing needle was used as the needle electrode. The needle tip was ground to have an angle of 30^o and a curvature radius of 3 µm. The distance between the needle tip and the plane electrode at the bottom of the sample was 4 mm. The LDPE was kept in contact with the base plane electrode using silver paint. Similar configuration was used by Fukazawa et al [82] but their sample size is 25 x 16 x 6 mm with radius of curvature of 10 µm.

However, the most commonly used configuration is with the modification of ground plate for grounding purpose. Niederhuber et al [83] investigated the performance of epoxy resin/ SiO_2 using this configuration. The slab size was 25 x 6 x 10 mm with a needle with tip-radius of 5µm. The distance between the tip of the needle and the bottom of casting mold of ground electrode respectively was set approximately to 5 mm. Likewise, Imai et al [84] using steel rod with tip angle of 30^o and radius of curvature of 5 µm with the gap between needle tip and base being 3mm.

Guastavino et al [54,62] in their study of electrical treeing presented a modification of the needle plane configuration. The modification made with the needle electrode was connected to the ground while copper plate applied with high voltage source. The configuration is an upside down version of the normal needle plane configuration. The step is almost the same but the needle was 5 µm tip radius and the interelectrode distance is 3mm. This configuration is the commonly used due to ease in fabrication.

Needle plate method is based on the use of the needle in the configuration but rarely used in determining the treeing performance. Zhou et al [56] used this configuration as shown in Figure 9 (b). This configuration uses fewer amounts of nanocomposites compared to needle plane but the arrangement is too complicated. The stainless steel needles with diameter 250 µm, needle tip curvature radius 2µm were used as the high voltage electrode. The distance between the needle and the plate was 3 mm ± 0.1 mm.

Both needle plane and needle plate use the needle for treeing initiation. The needle is the crucial part as the size of needle tip radius plays an important role on the initiation of electrical treeing due to electric field enhancement. The electric field is needed to initiate the electrical treeing [69]. A drawback of these configurations is that voids or micro-voids are created when inserting the needle into the nanocomposite which may cause partial discharge.

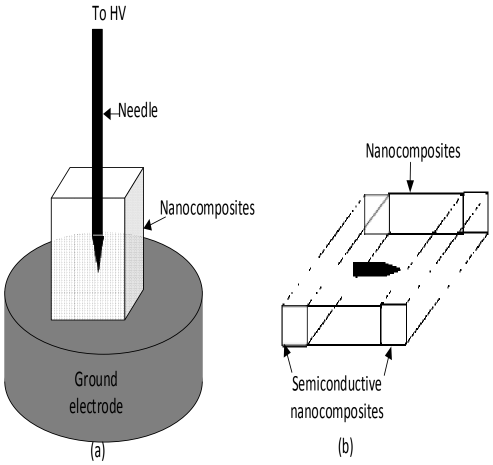

Fig. 9: Configuration of the specimen (a) needle plane (b) needle plate [83]

b) Leaf like specimen

Leaf like specimen were first introduced by Hozumi et al [85] but then modified by Nagao et al [13] and Kurnianto et al [86]. The leaf like specimen configuration is now the most widely used specimen configuration for treeing studies due to the fact that less amount of insulating material is needed compared to other methods [87]. However, this technique is a little bit complicated because there are many elements in the specimen; plus, the required element is small and thin in size. So, careful preparation and handling is needed. Similar to needle plane configuration, this specimen configuration also uses needle. Thus suitable needle tip size is of great concern. In this configuration, getting the required tip radius is a complicated process as tungsten wire (which is used as the needle) needs to be monitored when its tip is etched using chemical etching solution to get the required tip size.

Leaf like specimen introduced by Hozumi et al [85] showed a promising result in monitoring the tree initiation which is assumed to start approximately at 10 µm length. The semiconducting counter-electrode was made of 30-100 µm thick polyethylene sheets which contained carbon black. Several layers of films of 50-100 µm thick were piled and the needle inserted between the layers. Then it was hot pressed to a 100-300 µm. The thickness of the specimen was about 200-300 µm in order to obtain sufficient resolution and magnification for microscopic observation of tree initiation. The electrodes gap distance was fixed approximately at 1.0 mm and a cooper tape was used as ground electrode.

The configuration by Hozumi was further modified by Nagao et al [13] to investigate the effect of filler size in epoxy resins microcomposites. The needle electrode was a tungsten wire with diameter of 30 µm. Electrochemical etching method was performed to form a needle tip of 2 µm. The cover slip was placed on the resin at a proper distance by a film spacer. The specimen thickness was 200 µm and the electrode gap distance was adjusted to 1 mm.

Kurnianto et al [86] further modified the specimen and introduced a better version of leaf like specimen. The needle used was 0.25 mm in diameter tungsten wire. Sodium Hydroxide solution (NaOH) solution was used to aid the formation of the needle tip by using electrolytic polishing. Tip radius and tip angle of needle were 5µm and 30 degrees respectively. The gap distance between the needle tip and the plane electrode was adjusted to 2 mm. Thickness of spacer was fixed about 1.5mm. Fig 10 shows the leaf like specimen configuration introduced by Kurnianto et al [12].

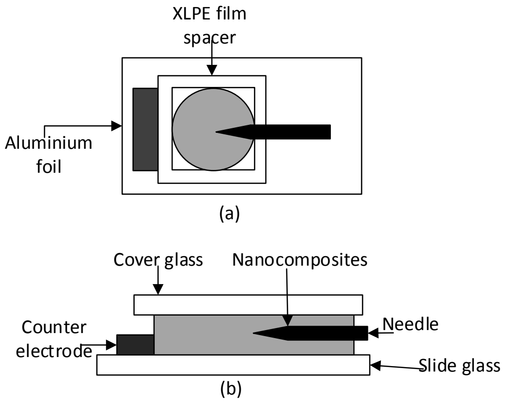

Fig. 10. Configuration of leaf like specimen (a) top view (b) side view [12,80]

c) Wire Plane

Wire plane configuration is a new specimen configuration used for investigating the treeing performance of polymers. Very few researchers have used this technique. This method avoids the use of needle in the specimen. This is because of the concern that needle may cause a micro void close to the tip or the tip can be fractured. These micro-voids create a potential problem since partial discharges developing in them would start the treeing process at a lower stress. The wire plane configuration is complicated and tedious in preparing because the tip of the wire is not sharp enough to be inserted into the nanocomposites. In addition, there are concerns that the wire is not thin enough to cause sufficient stress enhancement to initiate electrical trees.

Huuva et al reported the use of this type of configuration. Their specimen consisted of a semi conducting cross-linked polyethylene tab moulded between two flat sheets of the material to be studied. The thickness of the test object is maintained at less than 2 mm in order to provide sufficient transparency for optical observations for the development of electrical trees. An ultra-thin tungsten wire attached to a semi conducting tab forms the high voltage electrode. The tab itself operates as a connector between the external high voltage supply and the wire electrode. Tungsten wire has good electron emission ability, high elastic modulus and hardness and low thermal expansion. Wires with 10 and 20 µm diameters were used in their study. The counter-electrode is the bottom surface of the specimen. The distance from the wire to the bottom painted with silver paint is trimmed to 3 mm [88]. Figure 11 shows the configuration of the wire plane proposed by Huuva et al [88].

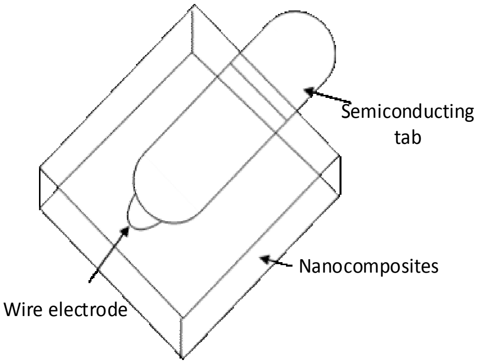

Fig. 11. Configuration of wire plane [88]

3. Plasma technology

Methods to improve particle dispersion, nanocomposite bonding and surface compatibility have been discussed earlier. However these methods have some drawbacks such as toxicity due to the use of chemical solvents, creation of voids during the processes or complicated procedure. In the quest for search of an alternative method to overcome these drawbacks, recently preliminary studies on surface modification of the nanoparticles using plasma has been reported. Plasma is clean (non-toxic), harmless and its application has shown promising results.

a) Fundamentals of plasma

Depending on the type of energy supply and the amounts of energy transferred to the plasma, the properties of the plasma change, in terms of electronic density or temperature. These two parameters classify plasmas into different categories as presented in see Fig. 12. Plasma can be classified depending on its electron density and electron temperature. Furthermore, plasma for surface modification to study electrical treeing resistance performance must be the plasma in the form of glow discharge and radio frequency (RF) discharges [89].

Low-pressure plasmas (LPP) have wide application in industrial application such as manufacturing and material processing. LPP generate high concentrations of reactive species that can etch and deposit thin films. The temperature of the gas is usually below 150 ºC, so that thermally sensitive substrates are not damaged. The ions produced in the plasma can be accelerated toward a substrate to cause directional etching of submicron features. In addition, a uniform glow discharge can be generated, so that materials processing proceeds at the same rate over large substrate areas [90].

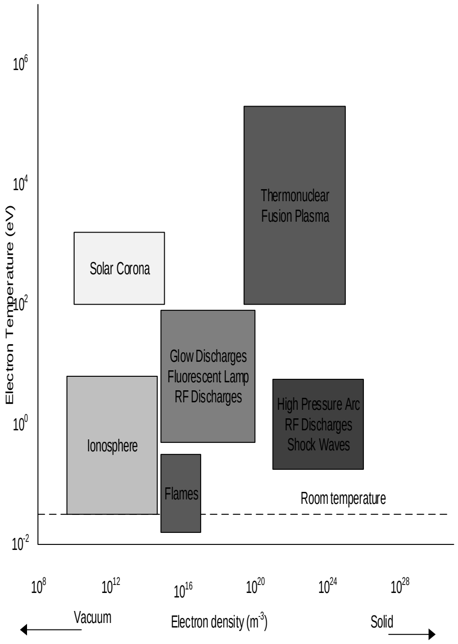

Fig. 12. Classification of plasmas [91]

LPP reactor usually is supplied with 27 MHz or commonly used are microwaves at 2.53 GHz. Microwaves allow working at higher pressures and giving higher plasma densities. On the other hand, audio frequency plasmas (kHz) cause a higher bombardment and radiation damage which is often not desired for the application [91,92]. Even though LPP perform a good plasma treatment, operating the plasma at reduced pressure has several drawbacks such as use of vacuum system. Vacuum systems are expensive and require maintenance. Also, the size of the object that can be treated is limited by the size of the vacuum chamber [93,94]. LPP presents many important advantages in order to treat surfaces of the materials. The various use of this type of reactors being able to change gas feed ratios, flow, time treatment, power density among others allow to tailor surface properties in a wide range. On the other hand, this technique presents an important drawback at industrial levels as it requires low pressure to achieve the plasma state which usually means a higher cost and increases the difficulty to obtain continuous processes [93-95].

In view of the above, Atmospheric Pressure Plasma (APP) has been proposed as it overcomes the drawbacks of LPP. The main advantages are the elimination of vacuum systems, reduction of costs, the possibility to use continuous systems and treatment of materials [96]. APP has been successfully employed in some fields such as etching, surface activation, ozone production and decontamination among others. APP could be divided in two main regimes, the microdischarge or filamentary plasma and the atmospheric pressure glow (APG) [97]. Microdischarges and filaments are obtained always when corona systems are used. In a corona system one electrode has a thin and sharp configuration which produces the discharge [98]. Another well known configuration for APP treatment is the dielectric barrier discharge (DBD). DBD systems have been used in a very wide range of applications such as excimer lamps, ozone production, surface treatment, and recently in nanofiller treatment to improve insulation [99,100].

The DBD device consists of two plane-parallel metal electrodes with at least one of these electrodes covered by a dielectric layer. The DBD reactor configuration is as shown in Figure 13. To ensure stable plasma operation, the gap which separates the electrodes is limited to a few millimetres in width. Plasma gas such as helium, argon or nitrogen flows in the gap [101,102]. The discharge is ignited by either a sinusoidal or pulsed power source. Depending on the working gas composition, the voltage and frequency excitation, the discharge can be either filamentary or glow [103]. A filamentary discharge is formed by micro-discharges or streamers that develop statistically on the dielectric layer surface. The use of helium as plasma gas may generate glow discharge [104]. The dielectric layer plays an important part by limiting the discharge current and avoiding the arc transition that enables to work with a continuous or pulsed mode and distributing randomly streamers on the electrode surface in order to ensure a homogeneous treatment. The streamer creation is due to the electrons accumulation on the dielectric layer [89].

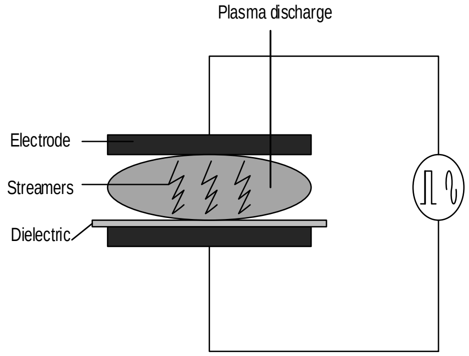

Fig. 13: DBD reactor configuration [99]

The nanoparticles treatment using atmospheric pressure plasma is a simple process. The nanofiller is treated using plasma until it is homogenously dispersed or coated. Then the nanofiller is mixed with the polymer matrix using direct mixing method. Finally the nanocomposites are prepared using needle plane arrangement, needle plate electrode or leaf like specimen.

b) Application of plasma in high voltage

Development of plasma has shown excellent results in engineering and biomedical field. In biomedical field, plasma can be found in the most critical application which is in tissue removal and coagulation. RF plasma applications for tissue removal and coagulation during surgery has an advantage as a safe energy source in human or animal bodies because nerve and muscle stimulation by current through the body ceases at 100 000 cycles/second (100 kHz). The traditional electrosurgeries used this RF to cut and coagulate tissue with minimal heat damage [105]. Recently, RF plasmas at atmospheric pressure developed for less heat damage, shorter treatment time, elimination of direct current flowing through the body to prevent site burn, and shorter post-operative healing time [106]. Plasma medicine is an emerging application and attracts attention from all researches. There are much more application of plasma medicine in [107-109].

In engineering application, plasma can be found in high voltage circuit breakers, ozone generators, high power CO_2 lasers, excimer lamps, and plasma display panels. High voltage circuit breaker is an automatically operated electrical switch designed to protect an electrical circuit from damage caused by overload or short circuit at substation. Historical breakers used a reservoir of compressed air at a pressure of about 2MPa and required expensive and bulky compressors. Then, the development of circuit breaker use SF_6 as the switching medium instead of compressed air. The main benefits is that the higher the arc current, the higher the pressure that is built up to drive the flow. SF_6 is suitable to increase pressure because one molecule decomposes into seven atoms. The pressure increase due to dissociation is supported by removal of nozzle material and metal evaporation from the contacts [110].

Formation of ozone in air starts when O_3 molecules decay rapidly at elevated temperatures. At the same time a relatively high pressure is required because O_3 formation an oxygen atom, an O_2 molecule and a third collision partner, O_2 or N_2. Oxygen atoms are generated by the dissociation of O_2 caused by electron collisions and by reactions of O_2 with excited nitrogen species [97]. Ozone offer many advantages especially in ozone drinking water and waste water treatment [111]. Ozone drinking water systems kill bacteria with ozone instead of the more common chlorine. Besides, ozone offer benefits in food industry. Ozone is suitable for washing and sanitizing solid food with intact and smooth surfaces and ozone-sanitized fresh produced [112]. Besides, ozone also helps in pollution control as ozone is a strong oxidant. It is easy to oxidize and decompose those toxic matters. So it has been used to purify the industrial wastewater.

c) Nanofiller surface modification using plasma

Plasma treatment for surface modification promotes wettability, printability and adhesion of the material. The application of plasma to nanoparticles in polymers for modification of their surface chemistry and morphology has been carried out by several researchers. Shi et al [113,114] investigated the performance of fluidized bed plasma reactor that can deposit ultrathin pyrrole film of Al_2O_3 nanoparticles. The results improve the adhesion of nanoparticles interfaces. The same method has also been used by [113] for treatment of polystyrene thin films on carbon nanofibres. The plasma coating greatly enhanced the dispersion of the nanofibers in the polymer matrix and it improved the strength of the nanocomposites.

Cho et al [115] employed plasma enhanced chemical vapor deposition (PECVD) with 360° rotating reactor to overcome nanoparticle aggregation and provide continuous agitation and mixing. Then the PECVD used to coat functionalize barium titanate ($BaTiO_3$) nanoparticles with reactive amine group. The epoxy/$BaTiO_3$ nanocomposites showed an improved mechanical strength compared to the untreated nanocomposites [116].

The application of plasma in high voltage insulations, specifically in electrical treeing application was recently reported by Yan et al. [117-122]. Yan et al [121] reported that the treatment of SiO_2 nanofiller using APP custom designed atmospheric-pressure plasma reactor. Cold atmospheric-pressure plasma was generated by a 350 kHz radio frequency (RF) power supply with a maximum 5 kV peak-to-peak output. A tin-coated copper coil electrode was placed 2 mm above the top of the nanoparticle layer. The ground electrode was placed under the reactor to form a DBD structure. Helium was used as the working gas for the discharge. The plasma was generated by a 4 kV peak-to-peak RF voltage. The nanofiller were treated for 30 minutes until it was homogenously dispersed or coated before mixed with polymer.

In addition, their studies revealed that the plasma treatment method enhanced the compatibility and the characteristics of the interfacial region between epoxy resins filled with nanosilica. The treatment produced stronger chemical bond and reduced the presence of the weak bond while maintaining excellent dispersion uniformity. Thus, the dielectric insulation properties improved even at low nanoparticles loading rate. However, a comprehensive study on the effect of the plasma treated nanoparticles mixed with the base polymer on the electrical tree characteristics such as tree growth, tree inception voltage, tree length et cetera was not carried out.

3. Conclusions

This paper has presented a review on nanocomposites material with focus on processing technique. The outcome of this review reveals the superiority of nanofillers as compared to microfillers in improving electrical tree performance of polymers. The importance of well dispersed nanoparticles on the improvement of mechanical and electrical properties has also been emphasized. The traditional methods of improving particle dispersion have also been presented. Although these methods have shown to be effective, however their major drawbacks include toxicity and complications of the method. The use of plasma has recently be advocated due to promising preliminary studies on improvement of treeing performance in polymers and overcoming the drawbacks of the traditional treatment method, however, a comprehensive study on this new method is lacking. In view of the foregoing, theauthors are proposing an in-depth electrical and chemical study on the electrical treeing performance of polymer nanocomposites treated with plasma.

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