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Applications of Polymer Nanocomposites in High Voltage Engineering: A Concise Review on Possible Applications and Questions Regarding Some Limitations

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Abstract

Polymer nanocomposites have been a topic of intensive research regarding High Voltage Engineering since the nineties of the last century. They present an alternative to conventional polymers since the latter were diagnosed with certain drawbacks. In this paper, a short review is given as to the basic characteristics of the polymer nanocomposites and some comments are offered as to the possible applications of the said materials in High Voltage Engineering.

Keywords: polymer nanocomposites, nanoparticles, partial discharges, insulation lifetime, functionalization, interfaces, interaction zone

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1. Introduction

Older insulating materials, such as paper, served well the high-voltage industry for most of the decades of the 20th century. The older insulating materials presented, among others, higher dielectric losses beyond certain high voltages and thus polymers were proposed as viable alternatives to the traditional insulating materials. For example, the relatively high dielectric loss of paper ($\tan\delta = 1 \times 10^{-3}$) is a disadvantage that increases in importance with increasing rated voltage for the cable and the loss becomes unacceptable at 400 kV. Polymers had higher breakdown strength, smaller dielectric losses and being flexible presented better mechanical and thermal properties. Conventional polymers served well for many years as insulating materials for high voltage applications [1-4]. However, such polymers had their own limitations, among others, due to their inherent cavities as well as to their sensitivity to electrical treeing, water treeing and partial discharges [5, 6].

Given that the needs of modern societies are even higher demands of consumption of electrical energy, the search for better insulating materials was intensified [7, 8]. As an alternative to the conventional polymers, a new group of materials emerged, namely, the polymer nanocomposites. Inorganic nanoparticles were added with some specific procedures to conventional polymers in order to form a new category of insulating materials. The seminal publication was that by T. J. Lewis back in 1994 [9] followed by numerous other publications [10, 11].

Polymer nanocomposites are conventional polymers in which nanoparticles in small wt% (or vol%) percentages are added with the aid of some processes. The appropriate selection of the conventional polymer, of the nature of the nanoparticles as well as their percentage wt% are critical parameters which define to a great extent the behavior of the nanocomposite w.r.t. its electrical properties [12]. Nanoparticles are particles with dimensions of less than 100 nm. If well mixed with the polymer matrix, they result in polymer nanocomposites with improved electrical, mechanical and thermal properties in comparison to conventional polymers [13]. Nanoparticles – because of their minute size - present a large area/volume ratio. This has significant consequences for the properties of polymer nanocomposites mainly because of the significant interaction zone between the nanoparticles and the polymer matrix [14]. It goes without say that the nanoparticles must be evenly distributed in the polymer since agglomerations of nanoparticles must be avoided [10, 15].

In the context of the present paper, there will be a short description of aspects of the polymer nanocomposites and some remaining problematic areas will be discussed. Possible applications of the aforementioned materials for the high voltage engineering will also be discussed.

2. The Historical Background

It was Nobel-prize winner R. Feynman, who in 1959 speculated that direct manipulation of individual atoms would result in a more robust form of synthetic chemistry. Feynman's view was a "vision of exciting new discoveries if one could fabricate materials and devices at the atomic/molecular scale" [16, 17]. Norio Taniguchi, a Japanese professor, was the first to coin the term "nanotechnology" in 1974 in order to describe novel semiconductor processes i.e., thin film deposition and ion beam milling. These processes exhibit characteristic control with nanometer precision [18]. The researcher M. Roco classified nanotechnology development in four successive generations, namely, those of products in nanoscale dimensions, nanomaterials and their effect on health, systems of nano-systems and molecular nano-systems [19]. Relatively recently, the importance of interaction zone in the case of nanoparticles in conventional polymers was emphasized in contradistinction to the interaction zone of microparticles [10]. The first industrial applications of polymer nanocomposites were in the car industry [20, 21].

Nanotechnology permeated relatively lately the field of insulating materials with applications in high-voltage engineering in comparison with the fields of semiconducting

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and biological applications. This had as a result the rather fast development of a whole range of new materials known as nanodielectrics and/or nanocomposites (terms that used are interchangeably when they refer to mixtures of polymers and nanoparticles) [11].

3. Conventional Polymers

Polymers are substances characterized from a large molecular weight and are multiples of simpler chemical units which are called monomers. Natural and synthetic polymers are created by polymerizing monomers. Polymers tend to form amorphous and semi-crystalline structures instead of complete crystals, and a number of them have excellent physical properties, such as high dielectric strength, toughness and high elasticity [22].

Polymers can be distinguished in thermoplastic polymers, such as polyethylene, polypropylene, polystyrene, and PVC, i.e. materials that can soften as the temperature arises, in thermosetting polymers, such as epoxy resin, i.e. materials that are hard and stable at higher temperatures, and in elastomers, which can elongate and can recover their initial form [22].

Of the aforementioned polymers, polyethylene (in fact more in its cross-linked form - XLPE) is mostly used in underground cables because of its high dielectric strength, thermal stability and chemical resistance [22]. Polypropylene has also very good mechanical, thermal and chemical properties and it has a satisfactory dielectric strength (which is, however, lower than that of XLPE). Epoxy resin is also a very good insulating material and it is widely used in numerous high voltage applications, such as switchgear and electrical machines [22, 23].

4. Critical Properties of Polymers

Mechanical properties consist a significant factor for the suitability of a polymer for some high voltage applications. Forces due to traction, deformation and bending, are among the parameters that may cause a polymeric insulation to be of very limited use [24]. Among the factors that determine the mechanical strength of a polymer are its molecular weight and crystallinity [7, 24]. Thermal properties are equally important since very often polymeric insulation is subjected to higher temperatures. Thermal conductivity is influenced, among other factors, by the structure of the chemical chain of the polymer and its crystallinity [25, 26]. Regarding the electrical properties of polymers, dielectric strength is a most significant property, which is affected by a number of factors. An important one is the applied voltage, its waveform, its rate of rise, its frequency, the shape of the electrodes and – generally – the shape of the technical arrangement. Additional ones include the polymer's molecular weight, the presence of humidity, the existence of protrusions, and the temperature [1, 2, 27, 28]. Important factors degrading the performance of polymers, such as partial discharges, have been studied extensively [2, 3, 29-32]. All the aforementioned properties are crucial for the high voltage applications since most electrical failures are preceded by mechanical or thermal malfunctions. Bending of insulation, sharp local temperature increases and/or decreases, among other factors, may precede an electrical failure. Examples of electrical failures that have as precursor mechanical or thermal stresses can be found, just to cite a few examples, in electrical machine insulation and also in high voltage cable insulation [2, 3, 23].

5. Nanoparticles

Nanoparticles when mixed with a polymer material result in a new material, the polymer nanocomposite. This is because, as the nanoparticles have dimensions in the range of the nanometers, when mixed with a polymer matrix, the interaction zone between the nanoparticles and the polymer becomes significant and the properties of the resulting composite resemble more those of the interaction zone and not those of the original constituents. Consequently, it is fair to say that a new material is formed, namely the polymer nanocomposite [10, 11].

Classification of nanoparticles is done in categories according to size, morphology, and properties, physical and chemical. Carbon-based-nanoparticles can be distinguished in carbon-nanotubes (CNTs) and fullerenes. CNTs are further categorized into single-walled carbon nanotubes (SWCNTs) and multi-walled carbon nanotubes (MWCNTs). CNTs contribute to the structural reinforcement of the conventional materials. Fullerenes are carbon allotropes with sixty or more carbon atoms forming a hollow cage structure [33, 34]. A disadvantage of the CNTs is that they form agglomerations of particles since they do not easily interact with the polymer matrix [35]. Ceramic nanoparticles are inorganic solids composed of oxides, carbides, carbonates and phosphates. Metal nanoparticles descend from metal precursors, and their synthesis is done by chemical, electrochemical, or photochemical methods. Semiconductor nanoparticles can exhibit both metal and non-metal properties. Polymeric nanoparticles are organic-based [33]. Layered nanoclays, such as montmorillonite, consist of layers whose thickness is not larger than 1 nm and can be classified as intercalated and exfoliated clays. In the former the polymer matrix penetrates the layers whereas in the latter the layers disperse in the polymer matrix [36, 37]. Other types of nanoparticles improving mechanical and thermal properties of nanocomposites are, among others, the nanotubes of halloysite [38] and the nanoparticles of zeolite offering improved chemical reactions [39].

It is very difficult to have a generalized approach as to the advantages and disadvantages of nanoparticles since there is not only a variety of nanoparticles and a variety of polymers but also a number of other factors affecting the resulting polymer nanocomposites, which have to do with their preparation (in-situ polymerization, solvent method, melt blending, sol-gel process) as well as with the treatment of the nanoparticles themselves (surface nanoparticle treatment) [11].

6. Polymer Nanocomposites

Polymer nanocomposite is considered a material with a polymer matrix in which one or more types of nanoparticles are dispersed [10, 11]. The main advantages of polymer nanocomposites are the high dielectric strength, the improved mechanical and thermal properties, and the improved permeability regarding humidity and gases [40]. Nanoparticles must be well and uniformly dispersed in the polymer matrix so that agglomerations of nanoparticles be avoided. Regarding high voltage applications, agglomerations may well be the source of partial discharges which in turn may contribute to the deterioration and ageing of the nanocomposite [7, 10]. Exfoliated structures render larger the interaction zone between the nanoparticles and the polymer matrix [41]. It goes without say that in order for the nanoparticles to be well dispersed in a polymer matrix, the two materials must be chemically compatible [42].

Production of polymer nanocomposites uses the methods listed. In situ polymerization is done by properly dispersing the nanoparticles in the monomer solution, ensuring the polymer will be formed between the nanoparticles, before the process takes place. Solution blending is a technique using ultrasonic irradiation, magnetic stirring or shear mixing in order to disperse the nanoparticles within the polymer. Melt blending is done by dispersing the nanofilters in molten polymer directly [15]. The sol-gel process is done at room temperature and the polymer may be introduced at the initial stage rendering thus the nanoparticles of sol more homogeneously dispersed [11]. Techniques of characterization of polymer nanocomposites render understandable their physical and chemical properties. Such techniques are the wide-angle X-ray diffraction (WAXD), small-angle X-ray scattering (SAXS), scanning electron microscopy (SEM), transmission electron microscopy (TEM), Fourier-transform infrared (FTIR), rheometry, differential scanning calorimeter (DSC), thermogravimetric (TGA), thermomechanical (TMA) and dynamic modulus analysis (DMA) [43, 44].

The explanation of the behavior of polymer nanocomposites has been the subject of numerous publications. Their behavior was basically explained on models assuming the existence of layers around the nanoparticles and the role of interfaces between the nanoparticles [9, 45, 46]. In fact, as was noted elsewhere [9], the interfacial interaction zone surrounding the nanoparticles results in a behavior which provides improved properties to polymer nanocomposites. Its characteristics include mobility, stemming from the physico-chemical bonds created on the common surface, and local conductivity, altered by the formation at the interface of a double layer. A considerable increase of the interfacial surface area is noted due to the dual mechanism [47]. In fact, the surface and interfacial area formed by nanoparticles is three orders of magnitude larger compared to one formed by microparticles [10]. The various proposed models regarding the behavior of polymer nanocomposites have been the subject, among others, in [12, 14, 48, 49]. Based on models such as those of [9, 46], the improved dielectric strength of polymer nanocomposites and their resistance to electrical treeing can be explained [50]. In the case of polymer nanocomposites, the nanoparticles act as minute barriers hindering thus the propagation of electrical trees. The nanoparticles obstruct the propagation of electrical trees and given the reduced interparticle distance, the dielectric strength will increases [50]. On the other hand, possible nanoparticle agglomerations (which are due to poor preparation process) may lead to additional partial discharge activity, further electrical treeing and a lowering of the dielectric strength [7, 10, 11]. It is true that polymer nanocomposites present improved properties compared with the conventional polymers fire resistance [15, 51], erosion and performance at higher temperatures [10, 52, 53].

7. Polymer Nanocomposites as Insulating Materials

Inorganic nanoparticles when mixed with polymers may render polymer nanocomposites that perform satisfactorily. Examples of such polymers with nanoparticles of Al_2O_3, SiO_2, TiO_2, glass fibers, and carbon nanotubes, presented improved electrical and mechanical properties [22, 54-57]. Epoxy resin and polyethylene are polymers that if combined with the appropriate nanoparticles may be used for a variety of high voltage applications, such as, underground cables, generators and transformers [22]. Experimental work previously performed indicated that with the reduction of the nanoparticle radius, an increase in the ratio of surface/volume is noted, which in turn means that there is a larger interaction zone. The chemical treatment of the nanoparticles as well as their uniform dispersion in the polymer matrix also play a most important role for the electrical behavior of polymer nanocomposites [58-60].

A substantial increase in partial discharge endurance was noted by using nanoparticles of amorphous silica in anhydride cured epoxy and XLPE with loadings of 5-6 vol% [61], whereas in [62] it was reported that partial discharge degradation was smaller when polyamide was mixed with 2 wt% nanoparticles than when polyamide was without nanoparticles. Agreeing qualitatively with [62], a later work confirmed that epoxy nanocomposite with 5 wt% additions of clay presented a far smaller erosion depth than the epoxy without nanoparticles [63]. Other researchers reported that even minute volume fractions of nanoparticles of either alumina (Al_2O_3) or titania (TiO_2) to epoxy resin increase the resistance of the material to degradation due to surface discharges [64]. Longer lifetimes were observed with epoxy resin samples with organophillic-modified ultra-clean montmorillonite nanoparticles compared with epoxy resin without nanoparticles [65]. Montmorillonite nanoparticles modified by Hexadecyl trimethyl ammonium bromide with silane and titanate coupling agents blended with high density polyethylene (HDPE) gave higher tensile strength and improved dielectric strength, volume resistivity and surface resistivity compared to HDPE without nanoparticles [66]. Lower contents of space charge and higher dielectric strength were observed with polypropylene (PP) and ethylene-vinylacetate (EVA) filled by nanosilicates in comparison with PP and EVA without nanoparticles. Such phenomena – according to the authors – may be due to the trapping of charges at the interfaces between the nanoparticles and the polymer matrix [67]. In yet another publication, it was indicated that space charges tended to be less in polymer nanocomposites [68].

Silicone rubber – a well-known insulating material for outdoor insulation – filled with nanoparticles of fumed silica had an increased erosion resistance, with the latter increasing even further with an increasing percentage of the nanoparticles [69]. Epoxy resin with nanoclay content of up to 5 wt% presented improved dielectric strength, tracking time and storage modulus compared with pure epoxy resin [70]. Experiments regarding the polymer nanocomposite insulation of dry-type transformers – mainly epoxy resin and polyester - were reported in [7]. Reports on cable insulation consisting of polyvinyl chloride (PVC) with the insertion of chemically modified silica (silicon dioxide, SiO_2) nanoparticles revealed an improved dielectric strength for both AC and DC conditions and an improved tensile strength and the Young's modulus compared with pure PVC [71]. Further work showed that on epoxy resin with nanoparticles, like TiO_2, ZnO and AI_2O_3 at low nanoparticle concentrations by weight an improvement in dielectric strength was observed [72]. Later research indicated that a combination of micro- and nanosilica filled epoxy samples may accumulate the smallest possible amount of space charge [73], echoing thus results of an earlier piece of work [74].

Polymer nanocomposites in enameled wires have been the subject of intensive research although there was an ongoing debate as to the stability of these materials for that particular application [75, 76]. As was pointed out in [76], enameled nanocomposite response depends on the level of applied voltage and waveform type, presence of humidity, and temperature levels. Short duration experiments indicated that a filled enamel sample with a ratio of 3wt% of Al_2O_3 exhibits higher levels of voltage inception and extinction, whereas a filling of the same sample with 1wt% of ZrO_2 provides higher breakdown strength [77]. Another work noted that alumina nanoparticles (with small loadings of 5-7.5 wt%) filled in polyamideimide showed a superior AC dielectric strength as well as longer voltage endurance at elevated temperatures [78]. The latter work was qualitatively in accordance with earlier research [79], which investigates polyesterimide and polyester and provides a comparison with their nanocomposite counterparts with a 1 wt% of nanosilica, 1 wt% of zincoxide and 3 wt% montmorillonite (MMT). Conclusions are that nanocomposites show better resistance to partial discharges, higher water absorption capability, and increased thermal endurance. A more recent work on silica dispersed in polyesterimide (PEI) enameled wire suggests a significantly longer lifetime w.r.t. neat PEI enameled wire in similar partial discharge conditions. It was remarked that in this case the percentage of silica nanoparticles with a content of 20 wt% gave optimum results, a content radically different and much higher than those reported before [80]. This can only stress the point that the content of nanoparticles in a polymer nanocomposite depends on the process of preparation, on the polymer matrix, on the chemistry of the nanoparticles themselves as well as on their functionalization.

As far as DC experiments are concerned, there were studies that pointed out that polymer nanocomposites present better electrical properties than pure polymers. In [81], the authors focused on polypropylene/poly (ethylene-co-octene) (PP/POE) nanocomposites with silica nanoparticles. Depending on the treatment of the nanoparticles, the said material showed a suppression of space charge accumulation, which in turn is a crucial factor for DC applications.

An improvement in electric field distribution was obtained in bushing insulation by adding nanoparticles of graphene oxide in epoxy resin with different percentages of 0, 0.05, 0.1 and 0.5 wt%. The authors cautioned, however, the use of introducing excessive nanoparticle content as the latter would diminish the DC dielectric strength [82]. Silicone rubber, an insulating material suitable for cable accessories, flexible and with excellent electrical and mechanical properties, mixed with alumina nanoparticles of concentrations 1 vol% and 2 vol% showed enhanced tree inception voltage and slower electrical tree growth rate. Mixed with halloysite nanoclay of approximately 3 vol% showed also an amelioration in tree inception voltage and slower electrical tree growth rate [83].

8. Practical Applications of Polymer Nanocomposites

Although the present review is a short one, there are strong indications that – as emphasized above – the behavior of a polymer nanocomposite depends, among other factors, on the chemistry of both the polymer matrix and the nanoparticles, on the processing of dispersing the latter, on their surface area/volume ratio as well as on their functionalization. In the previous three or so decades emphasis was given on laboratory experiments of polymer nanocomposites and on their possibility of whether these materials could present an alternative to conventional polymers [84, 85]. Consequently, there were some justified questions as to the long term behavior of these materials [7, 12]. Small percentages of either vol % or wt % of nanoparticles can be mixed with the polymer matrix since larger percentages may cause agglomerations [86]. As was remarked in [87], "an overarching concern … remains the ability to control nanoparticle dispersion by designing NP/polymer interfaces, by adapting typical processing protocols, and by preventing morphology evolution during use". The problems of dispersion of the nanoparticles were also pointed out in [88], where boron nitride (BN) nanoparticles were proposed for epoxy resin for application in machine insulation instead of alumina since the permittivity of BN is around 4 and thus closer to that of epoxy resin.

The dispersion of silica nanoparticles, in particular, in XLPE improved the electrical, mechanical and thermal properties of the polymer regarding possible application in power cables. Other nanoparticles such as alumina, TiO_2, and MgO can also be potential candidates in high voltage cables. Surface functionalization of the nanoparticles was again discussed as an important parameter to increase the compatibility with the polymer matrix [89-91].

One of the problems which arose was that of the difficulty of transferring experimental data from small scale experiments with polymer nanocomposites to large industrial equipment. This problem is not a new one since, e.g., questions regarding the effect of specimen size on the dielectric strength of insulating materials, were posed in numerous publications long time ago [92, 93].

Polymer nanocomposites showed promising results in a variety of possible high voltage engineering applications. Until now most of the experiments were performed in laboratories with small samples of the relevant insulating materials. This, however, changes since the electrical industry is already using polymer nanocomposites in enameled wires [94], in a 250 kV DC XLPE power cable [95], in yet another 525 kV cross-linked polyethylene power cable [96], as well as prototypes for rotating machine winding insulation and for GIS spacer insulation [97]. Enameled wires were made of polyamide-imide and nanoparticles presenting an insulation very resistant to inverter surges, practically unaffected by partial discharges and having higher mechanical strength than the conventional polyamide-imide [94]. The main advantage of the 250 kV DC XLPE cable having as insulation XLPE with nano-sized particles is that there is almost no space charge accumulation when stressed at 50 kV/mm for several days and that its lifetime under DC voltages is expected to be much longer [95]. A possible explanation of the reduction of space charge may be that although charges may occur around nanoparticles and thus increase the space charge at low fields, such an increase at low fields is not detrimental for the operation of the cable [11, 96]. Yet the other DC application of 525 kV with insulation of XLPE with nanoparticles operating at

90 ^{0}C gave also satisfactory results transmitting large capacities in excess of 2 GW [97]. Prototypes for rotating machine insulation and for GIS spacer insulation in the context of NEDO 2022 project have also been developed [98]. Another promising field of applications is the high voltage capacitor industry, where polymer nanocomposites such as polypropylene can be used notwithstanding the problems of nanoparticle agglomeration and of minute cavities [99].

Regarding the high-voltage high-energy capacitors, nanoparticles of high permittivity (~4000 at 25^0 C, dropping to ~2000 at 120^0 C), such as barium titanate ($BaTiO_3$) can be used [11, 100]. Poly(vinylidene fluoride) (PVDF) being a high permittivity polymer can be used in capacitor applications in combination with nanoparticles of CuPc [101]. Surface modified barium titanate nanoparticles in poly(vinylidenefluride-co-hexafluoropropylene) were also shown to have both high permittivities and high dielectric strength [102]. Nickel and barium titanate nanoparticles in polyamide systems were also studied, with both systems showing high permittivities for embedded capacitor applications [103]. Furthermore, a recent study reported that a maximum energy density greater than 30 J/cm^3 has been obtained by optimizing preparation procedures for the interfaces between polymer matrices and nanoparticles as well as capitalizing on the various polarization mechanisms connected with nanoparticles [104].

From the above, it is well established that the polymer nanocomposites find their way to high voltage applications [97, 98]. Polymer nanocomposites are materials that enhance cable's electrical properties, such as the dielectric strength, a property crucial for high voltage transmission. Polymer nanocomposites show promising in electrical applications with their use expected to increase in the future due to their improved electrical, thermal and mechanical properties. This is not to say that there are not still areas which have to be further researched: more precise control has to be exercised regarding the spatial distribution of the nanoparticles in the polymer matrix as well as the resulting interfaces, more effective methods of processing the polymer nanocomposites have to be explored and also a more thorough research on the long-term behavior of such insulating materials.

9. Conclusions

This short review indicates that although there is a wealth of experimental data on polymer nanocomposites with respect to the research in high voltage engineering, only relatively recently the use of the aforementioned materials started being used at an industrial scale. Polymer nanocomposites present a new class of promising insulating materials because of their highly interesting properties on the nanoscale range. A great deal of experimental work has shown that these materials may perform better than their conventional counterparts as far as the electrical, thermal and mechanical behaviors are concerned. One of the main questions w.r.t. the polymer nanocomposites was the lack of long term testing and their ageing behavior. The prospects, however, are high since high voltage cables are already in use as well as enameled wires, and efforts on prototypes of insulations for electrical machines and GIS are being made. Moreover, various nanoparticles in conjunction with polymeric materials have been studied for applications in the high-voltage capacitor industry. Since polymer nanocomposites have many advantages in comparison with the conventional polymers, it is to be expected that they will be used extensively in the high voltage industry in the years to come.

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