

Interfaces in High Voltage Engineering: A Most Important Question for Conventional Solid Insulating Materials as well as for Nanocomposite Polymers

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Abstract

Interfaces consist a most important part of conventional insulating systems at high voltages. They are considered to be problem areas which have to be dealt with. Numerous publications have contributed in rendering the mechanisms of interfaces understandable. On the other hand, interfaces in nanocomposite polymers seem to function in an entirely different manner from that in conventional insulating systems. The present paper reviews past work on both the conventional insulations and in nanocomposites. Differences regarding the interfaces are mentioned and discussed. Whereas interfaces in conventional insulating systems are to be avoided, interfaces in nanocomposite polymers seem to be desirable - at least - up to a certain percentage of nanoparticles in the base polymer. Although things are better understood in conventional insulating materials, more work has to be performed in order to clarify several aspects, such as space charges and electrical trees emanating from enclosed cavities. Needless to say that much more work has to be done in nanocomposites w.r.t. their modeling and possible explanations of the surprising performance of interfaces, a performance that deviates strongly from the performance in classical insulating materials.

Keywords

Breakdown, breakdown strength, conventional insulating materials/systems, nanocomposite polymers, partial discharges, electrical treeing/trees

Introduction

With some sort of exaggeration, a well known professor said once that "the problems of high voltage insulations are problems of interfaces" [1]. Interfaces result when there are two different insulating materials next to each other or when an insulating material meets a conductor. An interface may become, e.g., the source of partial discharges or even the cause for a complete breakdown of an insulating system. When two insulating materials of different dielectric constants have a common interface, then the material with the lowest dielectric constant will undergo the more intense electric stressing [2]. Depending also on other parameters, such stressing may result in a gradual deterioration of the insulating system and consequently in a complete breakdown [3].

In this paper, the questions raised by the existence of interfaces in high-voltage insulating systems are discussed. Interfaces play an important role in determining the robustness of an insulating system, when conventional insulating materials are used. Interfaces play also a vital role in insulating systems with nanocomposite polymers. Their functioning, however, is of another nature.

This short review is by no means exhaustive since the topic of interfaces is a vast one. It is the aim of the authors to give the gist of the problems and questions the researchers may face and to offer some comments. In the context of this paper, the terms "insulating material" and "dielectric material" or simply "dielectric" are sometimes interchanged meaning the same thing.

Interfaces in Conventional Insulating Materials

Dielectric breakdown in insulating materials depends on electrode configuration, insulating material thickness, electrode materials, presence of microcavities, temperature, pressure, nature and morphology of material under test, type of applied voltage, damage path (surface or volume) [4]. Various dielectric breakdown theories have been put forward [5]-[13]. No matter whether the proposed theory was based on cumulative impact ionization by electrons - creating thus positive space charges which distort the field distribution and weaken the dielectric [5] -, on the notion of "intrinsic breakdown" - according to which a large number of electrons trapped in energy levels due to lattice imperfections can transfer

energy to the lattice vibrations [6]-[8], on the "40 generations avalanche theory" [9], on the importance of space charges which modify the local electric field value [10], on the right assumption that the breakdown is a property of the dielectric material plus its electrode system [11] or on the theory based on the ionizing electrons and the hole traps [12], [13], the fact remains that all the above mentioned phenomena result from electric field intensifications, i.e. from either electrode imperfections or mismatch of dielectric materials. This brings us to the point mentioned in the Introduction of the present paper: that interfaces may create the conditions which may cause electric field intensifications.

The subject of partial discharges (PD) which may ensue because of electric field intensifications and/or because of gas (or foreign particle) inclusions in a solid dielectric material, has been studied by Mason in his fundamental publications [14]-[18]. Having in mind all the above, it is fitting to say that interfaces - created either by a mismatch of dielectric materials or because of intrusion of foreign particles and/or air cavities in the insulating material under

question -, are the problem areas of an insulating system.

As was pointed out quite early [19], interfaces play a most significant role in the discharge and breakdown processes: even if an insulation does not contain any cavities, at a sufficient stress "some event" releases gas with the subsequent formation of a cavity. The cavity is occupied by a gas discharge which increases the rate of gas formation with the subsequent growth of both the cavity size and the discharge intensity. The importance of the differing nature of interfaces was also stressed in another publication, where it was pointed out that damage in internal cavities in polyethylene is little compared to the electrode adjacent cavities of the same dimensions and tested under the same experimental conditions [20]. It is evident that in [20], interfaces between polyethylene and gas were compared with interfaces between polyethylene and electrodes, and the latter were found to be more dangerous and deleterious to the insulating material. On the same lines, Kreuger showed that with PVC-insulated cables, the number of discharges increased with increasing electric stress in the dielectric [21]. In yet

another paper, it was indicated that the nature of internal discharges was greatly affected by the assembly of the electrode system and the adhesion of the insulating tapes [22]. Discharges always start in the electrically weaker insulating material, as was commented in previous works [23]-[25].

Needless to say that phenomena related to PD, such as electrical treeing, are also closely connected to the mismatch of the dielectric constants of insulating materials and/or to the existence of gas cavities in their volume. Earlier papers indicated that the treeing phenomenon in polyethylene cables started from both inner and outer surfaces and also from solid particles and fibres [26]. Pioneering work performed with 15 kV and 22 kV polyethylene insulated cables reported that trees originated from contaminants and cavities, the tendency for tree initiation from a contaminant being probably more affected by the contaminant material than by the size, location or shape of the contaminant [27]. The importance of enclosed cavities in the initiation of trees was also reported more recently [28]. According to Ieda [29], tree propagation can be induced by internal gas discharge in the

tree. It is to be borne in mind that in numerous publications dealing with experimental work, the electrode arrangement that was used was a needle-plane electrode arrangement, indicating again that an electrode arrangement was chosen, with pronounced interfaces, in order to study the treeing phenomenon [30]-[33].

Interfaces which may play a role in determining the breakdown strength of an insulating material need not be only interfaces between insulating material and metal or between insulating material and gas cavity or contaminant. Differing phases may play also a role, as was noted in [34], where the interfacial domain of crystalline and amorphous phases may determine the various properties of semi-crystalline polymers, such as biaxially orientated polypropylene (BOPP). Stressing the importance of interfaces and experimenting with cross-linked polyethylene (XLPE), McKean showed that a considerable improvement in cable breakdown can be achieved by impregnation with silicone oil or diethyleneglycol. Such liquids can impregnate gas microspaces in the main insulation and thus result in an increase of the breakdown strength [35]. Similar observations were also

reported with polypropylene and polyethylene impregnated with suitable dipolar liquids [36].

Interfacial breakdown was studied with various electrode geometries and insulating systems consisting of paper typical for transformers and transformer oil [37]. It was reported that interfacial breakdown will occur if the paper is not carefully dried or if many gaseous micro-porosities are left in or on the paper. In [37], however, it was also noticed that using a carefully prepared paper-oil interface structure, the breakdown does not necessarily take place at the interface. Similar observations were made more recently by using silicone rubber interfaces, where both perpendicular and parallel to the applied electric field were investigated [38].

Conventional paper-oil cable insulation was studied quite early and the problems of interfaces were noted [39]. Alternative insulating systems, based mainly on polymeric materials, were proposed with considerable commercial success [40] - [43]. Modern cables with solid polymeric insulation did not avoid the problems of interfaces, namely those of extrusions of semi-conducting sheaths with the main insula-

tion or the inclusion of microcavities and/or impurities [44], [45]. Extruded cable insulation exposed to wet conditions suffered from electrochemical treeing and impurities greatly deteriorated its electrical performance [46]. Moreover, operating electrical stresses may also cause premature insulation failure in 15 kV polyethylene cables, if combined with unfavorable interface profiles and moisture [47]. Interfaces between polyethylene and small contaminants or microcavities may cause bow-tie trees in polyethylene cables [48].

Interfaces either perpendicular to the applied electric field or parallel to it or at an angle with it were dealt with in [49], where it was noted that such a variety of interfaces may be encountered in applications, such as capacitors, cables and in transformer windings. Composite insulating systems must preserve low dielectric losses. Higher dielectric losses may imply high ionic concentration in a solid/liquid insulating system, i.e. high ionic concentration in the liquid component of such a system [50].

Before concluding this section, it is fitting to mention the composite insulating systems of electrical machi-

nes, which consist mainly of epoxy resin and mica sheets. Previous work done in this context indicated that electrical treeing propagates through the epoxy resin and generally stops at the mica sheets, as mica is harder and electrically stronger than epoxy resin. The importance of such interfaces was reported before using a needle-plane electrode arrangement [51], where experiments were carried out without and with a mica sheet inserted in epoxy resin (Figs. 1 and 2). Evidently electrical trees were propagating more easily in the case of absence of the mica sheet and with much more difficulty with mica sheet.

Simulation work done recently showed that mica sheets prevent electrical trees from reaching the opposite electrode [52]. The purpose of mentioning experimental results regarding the composite system of epoxy resin/mica sheets is to show that the electrical trees propagate through the electrically weaker insulating medium. The simulation results indicate that even the slightest variations of dielectric constant may cause the electrical tree growth. In other words, the simulation data indicate that

local fluctuations of dielectric constant imply local - even microscopically minute - formations of interfaces, which in turn may mean local field intensifications, encouraging thus the growth of electrical trees (Figs. 3 and 4). Such observations w.r.t. the local variations of dielectric constant have also been reported for polyethylene [53], [54].

It is evident from all the above that interfaces in classical insulating systems seem to cause problems (possible dielectric constant mismatch, PD, treeing phenomena and ultimately risk of ultimate insulation failure). Due attention should be paid in choosing the insulating materials for specific applications and to the construction of the composite insulating system. Too many things depend on the quality of the construction of the interfaces [55]-[58], too many things that cannot be ignored. Interfaces in traditional insulating systems are considered as the weak aspects of such systems. Keeping this in mind and without exaggerating, it is not far from the truth if we state that an insulating system is as good as its weakest interfaces.

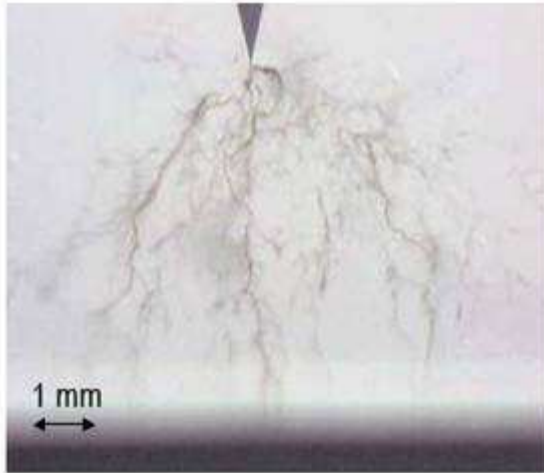


Fig. 1: Electrical tree propagation without the presence of mica sheet (applied voltage 28 kVrms, 50 Hz) (after [51])

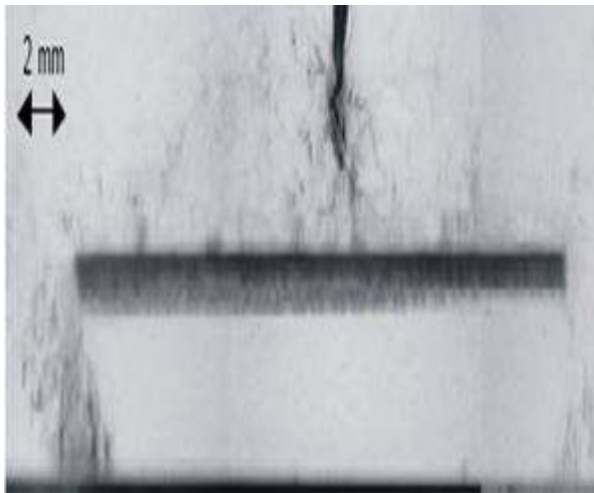


Fig. 2: Electrical tree propagation with the presence of mica sheet. The mica sheet increases the propagation time of the tree (applied voltage 28 kVrms, 50 Hz) (after [51])

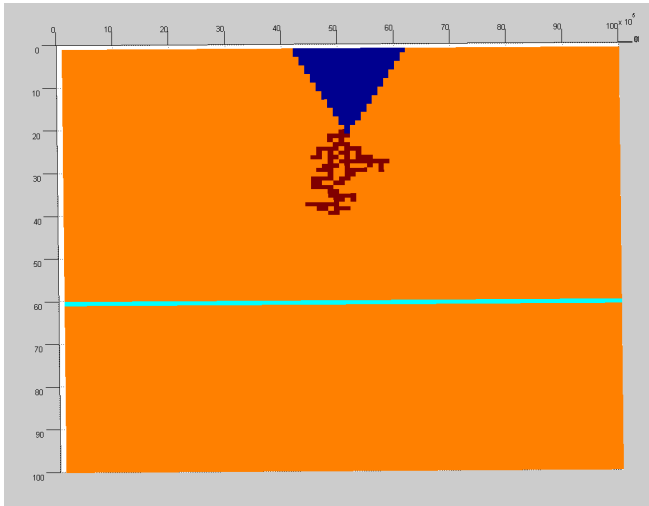


Fig. 3: Simulated electrical tree propagation with one mica sheet. Needle-plane electrode arrangement used

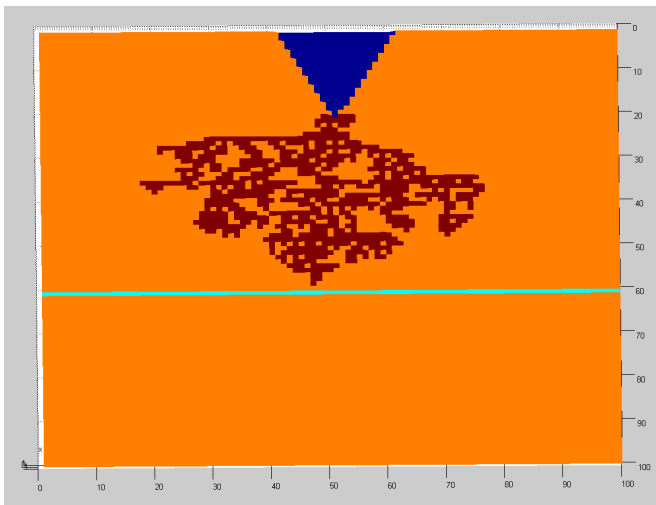


Fig. 4: Further expansion of electrical trees. Electrical trees stop at mica sheet. Needle-plane electrode arrangement used

It goes without say that this short review regarding classical interfaces does not by any means cover the whole subject and variety of solid insulating materials (for example, no mention in this paper was offered about the interfaces in outdoor polymeric insulators [59], [60] or in indoor polymeric insulators [61]). Both in the libraries and in the Internet, the interested reader may find practically tens of thousands of publications referring to the questions and problem areas of the solid insulating materials and insulating systems. What this short review tried to do is to show that interfaces, electric field intensifications, pre - breakdown phenomena (such as PD and electrical trees) and breakdown mechanisms are all interwoven and interrelated. Having said that, the next question related to interfaces, is whether they play the same detrimental role in the new generation of insulating materials, the nanocomposite polymers. This is to be examined in the following section.

Interfaces in Nanocomposite Polymers

More than twenty years ago, nanocomposite polymers came to our lives [62]. The first application of nanocom-

posites appeared in 1990, when Toyota Motor Corporation introduced nanocomposite nylon in their car industry [63]. Since that year, many car industries introduced the use of nanocomposite polymers. Use of nanocomposite polymers was noted in other industries, such as in the optics and the electronics industries as well as in the food industry. A seminal paper by Lewis gave the impetus for research also in the insulation branch [64].

For the electrical insulation, nanocomposite polymers are defined as conventional polymers in which particles smaller than 100 nm are added and dispersed in such a way that at the end one gets a homogeneous mixture [63]. The addition of such nanoparticles (the term "nanofillers" is also widely used) is being done in very small quantities, usually less than 10 wt%. Nanocomposite polymers consist of three components:

- a) the base polymer (or polymer matrix),
- b) the nanoparticles (or nanofillers) and
- c) the interaction zone (or interface zone) between the matrix and the nanoparticles [63].

Regarding the polymers used, these may be either thermoplastics, thermosettings or

elastomers. Nanoparticles may be classified w.r.t. their dimensions, and they can be distinguished as

- 1) mono-dimensional (i.e. extremely thin),
- 2) two-dimensional (nanotubes) and
- 3) three-dimensional (inorganic oxides).

The most usual nanoparticles for the purposes of electrical insulation are

- 1) silica nanoparticles SiO_2 ,
- 2) montmorillonite nanoparticles (layered silica),
- 3) metallic oxides such as Al_2O_3 , TiO_2 , MgO and ZnO and
- 4) carbon nanotubes.

Nanocomposite polymers can be obtained in two types of structures, namely,

- (i) intercalated nanocomposites (formed when there is limited inclusion of polymer chain between the clay layers with a corresponding small increase in the interlayer spacing of a few nanometers and
- (ii) exfoliated nanocomposites (formed when the clay layers are well separated from one another and individually dispersed in the continuous polymer matrix [65], [66].

As mentioned above, nanoparticles are added and dispersed in relatively small

quantities in the base polymer (usually no more than 10 wt%). Since nanoparticles are smaller than microparticles (smaller by three orders of magnitude), their interaction with the surrounding polymer matrix is much greater [62]. The so-called interaction zone is the main factor contributing in the improvement of the properties of the base polymer [67]. In the case of addition of nanoparticles into a polymer, the interfaces are far more numerous and far larger than in the case of microparticles. As the size of the added particles is reduced, the interface becomes larger and larger. The distance between the nanoparticles is also extremely small. It seems that interfaces determine to a great extent the properties of nanocomposite polymers.

The size of nanoparticles and the distance between them is of the order of magnitude on nanometers. Such particles may interact with the polymer matrix both physically and chemically in the nanometer scale. This has as consequence the appearance of properties that are somehow different from those we already know in a more macroscopic scale [68]. In contradistinction to the interfaces in classical insulating materials, and also to what we know

from classical high voltage textbooks, the improved insulating properties of the nanocomposite polymers are due to

- a) the large surface area of nanoparticles, which creates a large interaction zone,
- b) the changes in polymer morphology because of the large interaction zone,
- c) the changes in the space charge distribution and
- d) a dispersion mechanism [69].

Both the size of the nanoparticles and the chemical properties of their surface play an important role in determining the electrical, thermal and mechanical properties of nanocomposites. Needless to say that the chemical compatibility between the introduced nanoparticles and the polymer matrix is of paramount importance for the general properties of the nanocomposite [70].

One of the most significant characteristics of nanocomposite polymers is the increase of their breakdown strength as the size of the added nanoparticles tends to extremely small values. This increase is not in agreement with the conventional wisdom, which suggests that as the number of interfaces increases, the breakdown strength decreases dramatically

[2], [3]. Nanocomposite polymers seem not to agree with what we already know for classical insulating materials or systems [71]. Differences in breakdown strength between conventional epoxy resin and epoxy resin with nanoparticles was reported in [72]. Such observations were also noted later, when six different materials based on epoxy resin with various with and/or micro- and nanoparticles of alumina/silica were tested. It was shown that epoxy resin with nanosilica particles was the most suitable to obtain high values of breakdown strength [73].

Addition of the percentage of nanoparticles to epoxy resin up to a certain level favors the increase of breakdown strength both with a.c. and d.c. voltage, as was noticed in [74]. Why nanoparticles act in such a favorable way, despite the numerous interfaces they create? The increase of the breakdown strength may be due to

- (i) the increase of the surface area of the interfaces, which somehow alters the behavior of the polymer,
- (ii) the changes of space charge distribution inside the insulation structure,
- (iii) the dispersion mechanism, and
- (iv) the changing properties of the insulating material,

more specifically its volume resistivity, its $\tan\delta$ and its dielectric constant.

It is possible that the electrons moving in such a nanocomposite polymer, lose their kinetic energy because of the nanoparticles. Since the distances between the nanoparticles are extremely small, the electrons cannot acquire enough speed so that they can contribute to the breakdown process. Consequently, epoxy resin with nanoparticles presents a higher breakdown strength than conventional epoxy resin [74].

Normally the introduction of particles in polymeric materials has as result the introduction of defects and subsequently the worsening of its electrical properties. Nanocomposite polymers seem not to obey the above rule, as the mechanisms of conductivity during the breakdown process are influenced from the applied electric field, the dielectric constant of the nanoparticles and their number. The combined effect of these factors is difficult to fully understand at this stage and we need more work [71]. Similar results were obtained with epoxy resin with nanoparticles of TiO_2 as it presented a much higher breakdown strength than conventional epoxy resin [75].

On the other hand, electrical treeing propagation was found to be easier in conventional polymers than in nanocomposite polymers [76], [77]. Even a small wt% addition of nanoparticles affects in a positive way the electrical treeing resistance of the nanocomposite polymer. It seems that electrical tree propagation paths go through the base polymer and around the nanoparticles (experimental evidence for this was presented in SEM photographs published in [76]). Consequently, the more the nanoparticles in a polymer, the more difficult the formation of treeing paths. It seems that nanoparticles act as extremely small barriers, thus preventing the easy growth of electrical trees. Electrical trees propagate through the base polymer (in other words through the polymer matrix) and not through the nanoparticles. In some cases the electrical trees stop at the nanoparticles and they do not progress any further. Such observations were made in simulation studies recently [78]-[80].

Further research showed that a small percentage introduction of nanoparticles into a conventional polymer may increase its resistance to electrical treeing [81]. It is interesting to note

that nanoparticles may function as barriers preventing the tree growth even in minute quantities [82]. Earlier work indicated that as soon as the electrical tree touches the nanoparticle, the physico-chemical properties of it are such that very high energies are required in order to cause its deterioration [83]. Although the latter paper is an old one, it may give a clue as to why nanoparticles act as elementary barriers and why they prevent (or they delay) electrical treeing. More recently, a similar argument was given by some Japanese researchers [84].

Loading (i.e. the percentage of included nanoparticles expressed in wt%) plays also a vital role in determining the resistance to electrical treeing. More loading (i.e. more nanoparticles, that is more interfaces) implies a better resistance to treeing. This is probably because trees interact with many more nanoparticles and this delays their growth [85]. Another possible explanation was offered some years ago, where the authors proposed that in front of a tree a damage process zone is formed in a conventional polymer. Such a zone cannot progress easily when it meets nanoparticles [86]. Simulation data indi-

cated that loading affects the tree growth. The nanoparticle size plays also a role in delaying tree propagation, smaller nanoparticles offer a better tree resistance than the larger ones [79]. In Figs. 5-8 simulation results regarding the loading of nanoparticles as well as the size of nanoparticles are shown. It is evident that more loading makes tree growth more difficult. It goes also without say that smaller nanoparticles offer a better resistance to tree propagation.

There is no need to emphasize that there is also in the field of nanocomposite polymers a vast body of technical literature, too vast to be mentioned here in the context of this paper. From this short review it is obvious that interfaces in nanocomposites play an entirely different role from the one they play in classical insulating materials. Why is this so? This may be because the physics and/or chemistry somehow change in the nanoscopic world of such materials. Interfaces become highly desirable - at least up to a certain percentage of added nanoparticles. The surface area of the nanoparticles is huge if compared with that of microparticles for other high voltage applications.

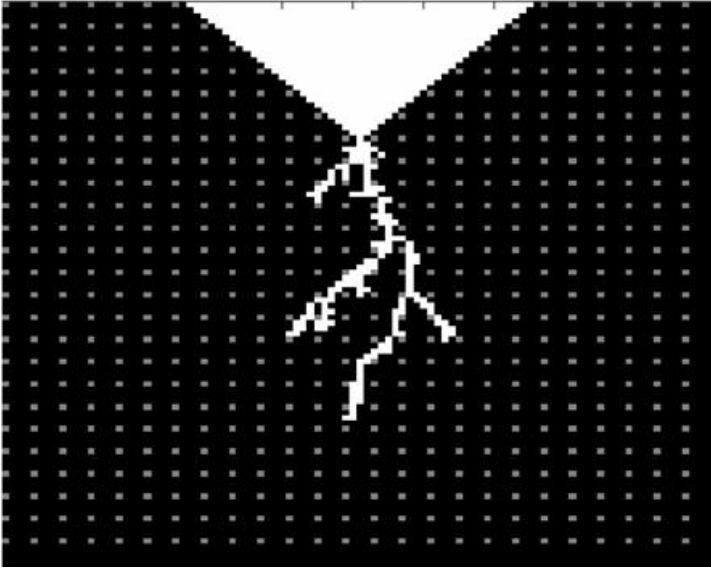


Fig. 5: Simulation with epoxy resin filled with TiO_2 nanoparticles (loading of 2 wt%, nanoparticle diameter 100 nm)

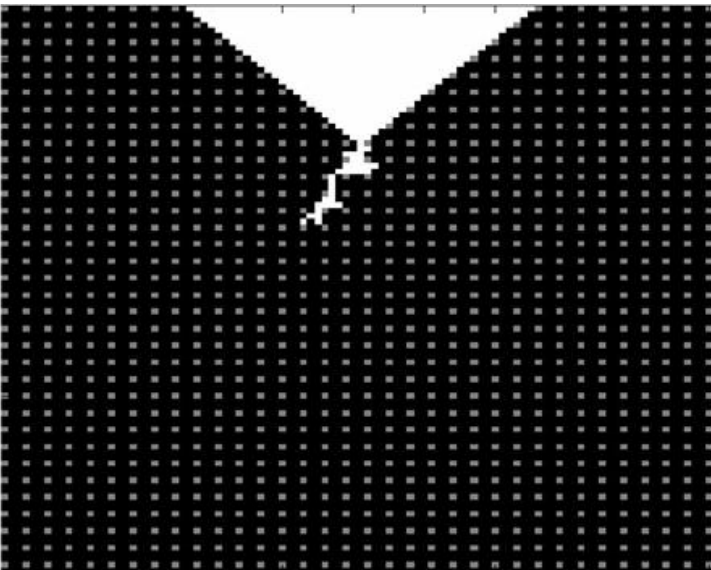


Fig. 6: Simulation with epoxy resin filled with TiO_2 nanoparticles (loading of 6 wt%, nanoparticle diameter 100 nm)

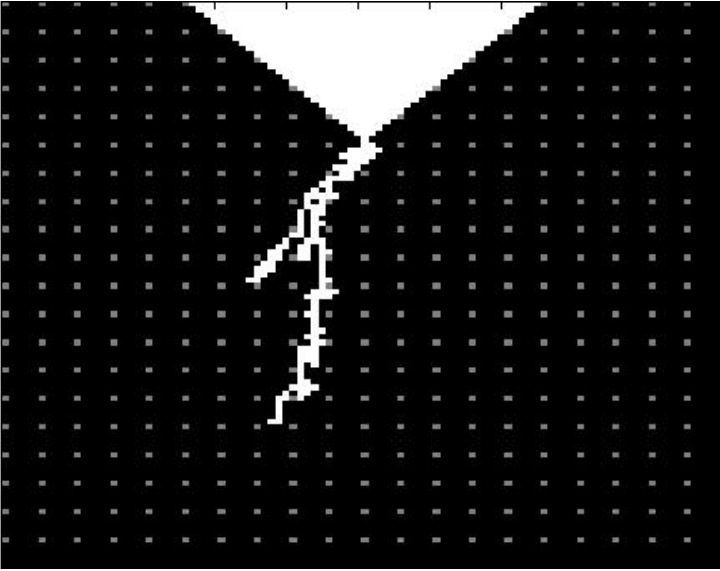


Fig. 7: Simulation with epoxy resin filled with TiO_2 nanoparticles (loading of 6 wt%, nanoparticle diameter 200 nm)

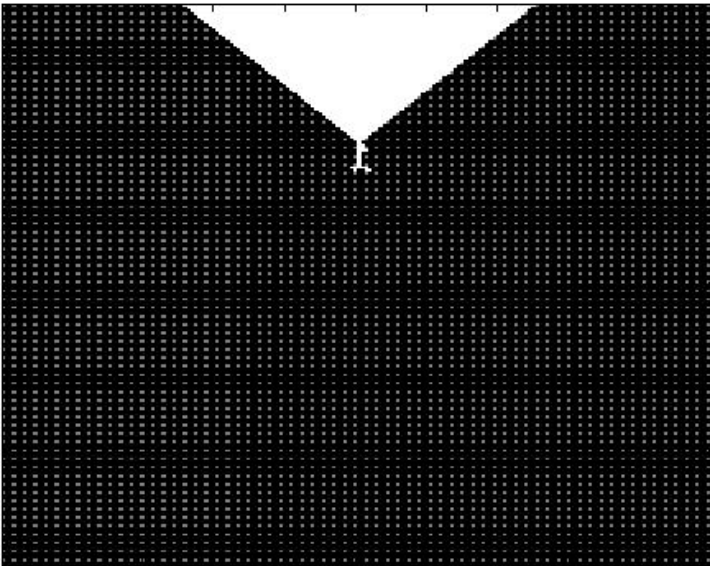


Fig. 8: Simulation with epoxy resin filled with TiO_2 nanoparticles (loading of 6 wt%, nanoparticle diameter 100 nm)

Models regarding the explanation of the functioning of nanocomposites were proposed as well as how the nanoparticles behave inside the base polymeric material [62], [87], [88]. Such models try to explain the higher breakdown strength values of nanocomposites and also their higher resistance to electrical treeing. The explanations seem to be plausible but more hard evidence is needed. Such evidence will be offered by many more photographs (SEM and TEM photos) showing in detail how the electrical trees circumvent the nanoparticles and how they propagate through the polymer matrix. Such photographs are very difficult to obtain.

Where all this leaves us? How can we understand in a unified way interfaces in both classical and nanocomposite insulating systems? Can the experimental results and simulations on electrical treeing with typical machine insulation ([52]) provide a hint also for a possible explanation in the nanoscopic world? A recent paper on interfaces posed some pertinent questions regarding classical and nanocomposite insulating materials [89]. From this paper it is obvious that although more questions are in need of an answer for the nanocomposites, the subject of

interfaces in classical insulating systems is by no means finished. For example, charging of larger interfaces, such as found in cable joints and terminations, needs to be further explored regarding the mechanisms of space charges. Another aspect in need of further discussion is whether electrical trees may emanate from enclosed cavities in conventional polymers. The latter question has been partly answered in [28], [90] - where some experimental evidence was offered as to the possibility of electrical trees stemming from enclosed cavities - but further experimental data is needed.

One thing that should not be forgotten - and it is common to conventional as well as nanocomposite polymers - is that an insulating system to a great extent is as good as its interfaces. This means that, no matter whether we deal with conventional insulating materials or nanocomposites, preparation and construction in both cases has to be not only careful but meticulous.

A last remark on the literature presented here: the interested reader may find that the authors dwell perhaps too much in the older scientific literature. This is not done because they tend to ignore the more recent re-

search: they simply would like to show that even in the old days, the problems were more or less the same. Moreover, they would also like to show that fundamental ideas - which are with us even today - on the various mechanisms in insulating materials came about quite early.

Conclusion

This review - by no means exhaustive - tackled the subject of the importance of interfaces both in conventional insulating materials and in nanocomposite polymers. Whereas interfaces are to be avoided in conventional materials, they seem to be a blessing in nanocomposites.

Whereas in conventional materials they cause problems of compatibility and sometimes high field intensifications with all the bad consequences such intensifications entail (i.e. PD, trees), in nanocomposites they seem - up to a certain loading - to be desirable and they prevent (or the delay) tree growth. Whereas in conventional insulating systems the introduction of more interfaces seems to cause sometimes insurmountable problems, the introduction of interfaces (because of the nanoparticles' introduction to the polymer matrix) seems to alleviate electrical trees and to distribute more evenly electric fields and space charges.

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