Chapter 8

Precursory Phenomena and Dielectric Breakdown of Solids

8.1. Introduction

The physical stresses to which dielectric materials are subjected do not, *a priori,* contribute to the early ageing of these materials or to their failure, and it is the fundamental reason for the undertaking of a test which guides the choice of test made. Experiments show that the joint action of physical or physico-chemical parameters leads to an unexpected evolution and breakdown of dielectrics used as electrical insulating materials (as we shall subsequently call them).

The evolution of the materials is related to the growth of stresses, such as temperature (which can have a positive as well as negative effect), high voltage, electrical field, environment (whether on earth or in space, where humidity and pollution in a broad sense can play a part), and radiation across the whole range of frequencies. New terms such as very high voltage and reliability appear and, because of these requirements, the use of natural materials such as wood, natural rubber, paper, etc, is significantly reduced.

Please note, however, that the use of paper-oil (PO) remains, particularly in the insulation of direct current high voltage cables. Due to its physical and chemical properties and thermal stability up to 700°C, muscovite mica is always used for high voltage systems and rotating machines.

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Obviously, chemistry has always played a large part in this evolution and as an example, cables (very well described by Arrighi [ARR 86] in the introduction to the Jicable conference in 1984). Indeed, this chapter notes the exponential growth over the years of the extent of high voltage cables insulated with chemically cross-linked polyethylene (XLPE); this polymer has the advantage of keeping up sufficient mechanical resistance at operating temperatures of 90°C to 100°C in exceptional circumstances, and 250°C in short-circuit conditions.

However, several causes of failure can appear: the manufacturing of the basic material as well as interfaces, accidental working conditions such as overvoltage, reverse voltage and the presence of inorganic impurities or water molecules [DEN 94]. Tests and standards can help avoid problems, and research can aid understanding and help in overcoming them. It should be stressed, however, that damage prevention in a closed system represents a major difficulty for the user. Readers could usefully consider Densley's [DEN 01] work on cables, an approach which we believe is applicable to other systems.

In the following sections, particular attention will be paid to organic solid materials, considering the important role they play in the wide world of electrical insulation: for the production of nuclear energy, energy transport, rotating machines, electronics and in space (where we include aeronautics and satellites).

8.2. Electrical breakdown

The fundamental aspects of electrical breakdown have been dealt with elsewhere so here we shall here only consider the final aspect: the destruction of the insulating material. However, first we will mention important syntheses published on the subject: very early on by Von Hippel [HIP 37], whose originality was to compare the breakdown of the three states of matter, then those by Whitehead [WHI 53], Straton [STR 61] and O'Dwyer [O'DW 79], whilst a particular interest was shown in alkaline halogens by Cooper [COO 63], [COO 66]; organic materials and their industrial applications have been studied by Mason [MAS 59], Fava [FAV 77] and Ieda [IED 80]; the different aspects of the dielectric breakdown of solids are detailed in more recent books, such as those of Coelho [COE 79], Nelson [NEL 83], and Dissado and Fothergill [DIS 92].

The dielectric breakdown of a system is an established fact which reveals either the simple puncture of the material or a perforation in the carbonization of all (or part) of the material, and sometimes damage to the conductive part which it was meant to be insulating. Carbonization is related to the current supplied by the source, as long as the incident did not send it off-circuit; obviously the flammable character of the decomposition products of some organic materials can contribute to the final degradation. At the microscopic scale, a perforation without carbonization can affect a weak part of the material if, locally, the melt temperature is reached. The phenomenon is particularly seen in both low density polyethylene (LDPE) and high density (HDPE) polyethylene films, chemically reticulated polyethylene (XLPE) or polypropylene (PP) (which is itself meant for the manufacture of capacitors). A simulation can be realized in a laboratory for bulky materials as well as thin films. The degradation of the material, however much hidden, locally removes or at least diminishes the dielectric quality in volume, and consequently could diminish the lifetime of the insulation.

As far as experiments are concerned, the notion of *intrinsic breakdown or strength* remains academic, as Blythe very rightly underlines [BLY 79] in his introduction on polymer breakdown, despite certain conceptual approaches which cannot ignore the insulating material, strictly speaking, and its particular utilization [CUD 87].

The maximum electrical voltage an insulating material can withstand before getting perforated defines its *dielectric strength*, which is expressed (under the International System of Units (SI)) in MV/m, kV/mm, or V/µm, depending on the type of insulating material or dielectric concerned, i.e. depending on whether we are dealing with bulky materials (cables) or thin layer (films, semi-conductors etc.). Indeed, the breakdown voltage V_b, of a dielectric depends on its thickness e, and the resulting breakdown field F_b is the ratio V_b / e. Consequently, the growth of V_b should in principle follow that of e, (remembering that, for a cylindrical structure such as a cable, the relationship becomes $F_b = V_b / \ln(r_0 / r_1)$, ($r_0$ and r_1 being respectively the external and internal radii). But what contradicts this simple relationship? Dielectric breakdown tests have a long history, as pointed out by Bartnikas in Chapter 3 of his book [BAR 87], such that they take the form of a rite if we want to obtain a correct value of the rigidity of a dielectric. Depending on the application, the tests can be undertaken under direct or alternating voltage for different frequencies, since the growth rates of the test voltage can be variable [MAS 73]. The specimen thickness is to be specified when it is related to the electric field value. An example is given in Table 8.1 for vinyl acetate (EVA), an elastomer used in the insulation of photovoltaic cells [CUD 87].

e (mm)	F_b (kV / mm)
0.12	98.3
0.15	85.5
0.40	44.2

Table 8.1. *Values of the electrical field as a function of the thickness*

Cables (kV)	Field (kV / mm)
60 < V < 110	4 < E < 6
120 < V < 245	8 < E < 10
V > 300	12 < E < 15

Table 8.2. *Stress values for cables*

In agreement with one of our introductory remarks, tests have shown that this characteristic was: $200 > F_b > 260$ kV/mm for muscovite mica and $140 > F_b > 200$ kV/mm for phlogopite, the former being recognized as the better of the two materials [HEP 00]. For comparison only, Table 8.2 gives the stress values for high voltage (HV) and very high voltage (VHT) cables.

8.3. Precursory phenomena

Because the breakdown of a dielectric is produced in a very short time (in less than a second), the events which precede it particularly deserve our interest.

8.3.1. *Definition*

Following the Latin adjective præcursor (forerunner), the term precursory phenomena must be applied to all phenomena which we can detect before an irreparable breakdown. When dealing with a real system of solid insulation, two situations are presented: either the phenomenon appears at the surface and is therefore optically or electrically observable, or it is created inside the volume and only detectable by electronic detection. These are, for example, the respective cases of an insulating material, a cable or a motor.

Another approach has been proposed for cables used in the instrumentation and control of nuclear power stations. The insulating materials concerned are vinyl polychlorure (PVC), XLPE and ethylene copolymer-vinyl acetate (EVA), for example. Samples, in the form of a test cable, were introduced inside a reactor in sufficient quantities to allow periodic removal of samples over about 40 years. The samples are tested chemically, mechanically and electrically [BAM 99a].

The problems which arose have led to pragmatic studies, which have influenced manufacturers and equipment, and have had more fundamental influence on materials sometimes far from practical or commercial interests, such as ion crystals. This has obviously led very quickly to extending the range of materials studied to those used in construction, such as polyepoxy or polyethylenes for thick materials, polyethylene terephthalate (PTFE), polypropylene (PP) or polyethylene naphthalene (PEN) for films. Consequently, the term precursor, previously defined, will fortunately be applied to observable phenomena in the laboratory and allow advances in the understanding of the degradation and breakdown processes.

We thus move from investigating the black box constituting a transformer, or a motor or cable in service, a cable insulator under pressure, or a capacitor, towards consideration of the component itself: the insulating material or dielectric. The observations are made at the microscopic level and, owing to modern analysis techniques as well as mathematical models and techniques, we can imagine the processes of ageing and breakdown of the material. The term *precursor* then takes a wider meaning, because it reveals the origin of the problem which could be a gaseous cavity, or an impurity, which can be defined, or simply a semi-crystalline structure whose distribution is random, especially given that interfaces are created between materials of a different nature (metal/insulating material being an example).

8.3.2. *Potential precursors*

8.3.2.1. *The material*

The physical, mechanical or electrical properties which we expect from an organic insulating material depend particularly on its microstructure: thus, the empty volume hypothesis for polyethylene which was proposed by Matsuoka [MAT 61]. Depending on the degree of branching (CH3/100C), their concentration was estimated between 18 x 10^{-3} cm^3/g and 23 x 10^{-3} cm^3/g. At a scale of several tens of nanometers, a lamellar structure appears due to the withdrawal of polymer chains. The lamellae are organized in microstructures and constitute spherulites. The water tree problems in buried and XLPE insulated cables thus give dimensions to voids with diameters ranging between 5 and 10 µm for the biggest [MUC 76]. Observations situate the voids at the junction of two or three spherulites. The latter, of average diameter equal to 0.1 µm, can appear bigger in LDPE than in XLPE, but depend on the presence of antioxidants which play the role of nucleant agents (see Figure 8.1).

We shall note, on the one hand, that the analysis reveals voids of weaker sizes and, on the other hand, that in the case of polymers obtained by the chemical process of crosslinking, some residue such as dicumyl or acetophenone peroxide, for example, could be found in these spaces, despite a thermal process intended for their extraction at the end of manufacture. The concentration of these volatile products is estimated at between 500 and 2,000 ppm [DEN 94].

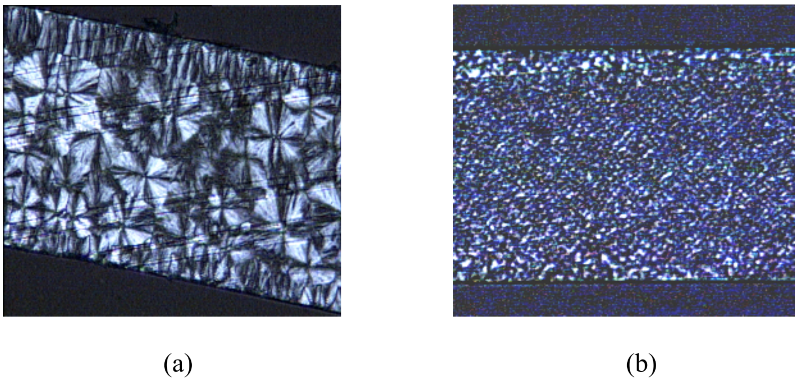
(a) (b)

Figure 8.1. *Spherulites in an LDPE (a) and an XLPE containing an antioxidant (b) [BER 98]*

Spherulites have an orderly structure and a high density compared with the space between them. If the space is dense, the field of electrical or mechanical stress is higher. The reduction of the spherulites' size therefore leads to the growth of the insulating material's breakdown field, as shown by Kolesov for PP and HDPE [KOL 80]. The presence of both amorphous and crystalline phases favors the diffusion of gas towards the amorphous phase and *a fortiori* towards empty spaces. With the exception of helium and hydrogen, gases contained in the atmosphere have neighboring diffusion constants [LAU 83]; this is another characteristic which can reveal the harmfulness.

Material is sensitive to temperatures and their variation, either during manufacture or whilst in use. Just to take XLPE as an example, this influence has been shown in laboratory models in miniature cables placed between plaque samples and the sample under investigation. Thus investigations in this area have shown annealing effects due to temperature rise, which leads to a densification of the polymer, and the formation of micro-cavities whose dimension are of the order of 100 nm. This non-homogeneous evolution obviously depends on the thermal gradient. In fact, considering the evolution of local stresses, competition takes place between quenching and annealing of the material, with an influence on the number and size of voids [KAR 89].

During manufacture, an important effort has been made to reduce the number of cavities and their size [YOD 73]. Thus from 10^6 cavities/mm^3 with diameters ranging between 1 and 5 µm in 1975 [KAG 75] we reached, about ten years later, a concentration of 10^3–10^7 cavities/mm^3 of the same size in cables insulated by XLPE [JAC 78], [BAL 84]. In his interpretation of dielectric breakdown, Artbauer drew a clear distinction between what are conventionally called cavities and holes or voids, these latter (in the amorphous phase of the polymer) having dimensions of a molecular order, therefore very much less than those of cavities which favor a specific degradation mode, as we shall analyze later [ART 96]. It is this theory of *free volume* that the author relies on to explain the electrical breakdown of a polymer.

8.3.2.2. *Impurities*

Despite the precautions taken to achieve good insulation, a post-mortem after an incident can reveal the presence of many sorts of impurities [MIN 84]. Tests in the laboratory have shown that metallic inclusions were less dangerous than mineral impurities [MOR 88]. However, the implementation of cables with more and more significant voltages (400–500 kV) has required a reduction of the use of organic materials (XLPE for this type of application), because of contamination with metallic impurities whose size can range between 10 and 100 µm, for example [BOS 03]. The site of manufacture also needs to be under close supervision; one example shows that the failure of a 275 kV cable was as a result of a joint contaminated by fibers. Laboratory tests comparing cotton, polyester, nylon and silk, have revealed the double danger of a hollow natural fiber such as cotton [KAM 92]. Indeed, this foreign body can have a hydrophilic character and most of all behave like a cavity since it is hollow. Tests carried out on polyethylene films within which silver microspheres had been moulded, have shown an important decrease of the breakdown field of the order of 37% when this type of contaminant reached a diameter of 20 µm, and 58% for 80 µm [STA 84].

When making connections and at the ends of high voltage cables, modern technology has led to the making of interfaces which can be in the presence of XLPE, a silicon rubber or polyepoxy. Generally speaking, an interface can be a weak point when next to an applied field, on the one hand because of the discontinuity of the physical properties of the insulation created (for example, a heterogenous response to thermal stress) or, on the other hand, because of the possibility of gas trapping. It is at this point in the insulation that the tangential component of the field is the most noxious, giving rise to the creation and propagation of an electrical tree. The example of a silicone–XLPE interface failure on a 150 kV cable in operation is given by Ross [ROS 99]. The diffusion of ionic impurities originated from semi-conductor screens and the ground in which certain high voltage cables are buried could be observed. Analyzes on samples extracted from non-aged cables or placed under voltage for several years, have revealed the presence of different types of ions (Al, Si, Fe, P, Mn, S, K, Fe, etc.) due to the diffusion of water coming from the ground in the case of cables not protected by a sheath [BOG 94], [CRI 87], [MAS 87], [MAS 92].

In the same way, conductive particles have been detected, no longer in the volume but on the surface of the charged resin playing the role of an insulating spacer. In general, bi-phenol A or cycloalyphatic resins are used, in which mineral charges representing about 70% of their weight are mixed. These charges are chosen from quartz, melted silicium, tri-hydrated alumina and sometimes combined mixture. These mechanical supports play a part in transport lines and circuit-breakers whose insulation is maintained by SF_6 under pressure. This type of contamination reduces the by-passing voltage by 50%, underlined by Cookson in a large, heavily referenced, review of gaseous insulation cables [COO 85]. The influence of free particles in transmission lines, sub-stations and circuit-breakers operating at 60 Hz, and insulated by sulfur hexafluoride at pressures of 300 and 500 kPa, shows the decreased conditions of the breakdown voltage related to the displacement of the impurity [CHA 89]. More recently, another well-documented analysis underlined the difficulty of removing the contamination of metallic particles in transmission lines and circuit-breakers insulated by SF_6 which can operate up to 750 kV. A continuous control is required for such systems; ecological considerations mean that, in certain countries, the use of nitrogen instead of sulfur hexafluoride should be envisaged [MOR 00]. Laboratory studies, with direct voltage, have been undertaken, during which metallic particles and insulators with different profiles were introduced in identical systems filled with N_2 or an N_2/O_2 mix. The generation of discharges and the breakdowns observed were determined by the displacement of particles [HAR 87]. A first review was proposed on the dielectric standard of gases related to the contamination by particles in 1981 [LAG 81]; a second, which considered the by-passing mechanisms along dielectrics in gases under pressure, appeared five years later [SUD 86].

As far as insulators maintaining the support of external high voltage conductors are concerned, their environment favors the same process. This process is observable from the outside, but if the erosion (by short-circuit) is at an advanced stage, we can only see the destruction. The discharge mode is dependent on the type of voltage (whether direct or alternating). In dry periods, dust constitutes sediment which, under the effect of rain, is transformed into a conductive film. The conductivity of this film is clearly modified by the formation of saline solutions in coastal regions; it is also variable as a function of the contents brought by surrounding pollution from industrial or agricultural spaces. Recent studies have shown the efficiency of cycloaliphatic resins (already mentioned), but comprising two dispersed phases. The improvement of mechanical properties is obtained by introduction of trihydrated alumina or silicium; we have noted the efficiency of a preliminary treatment of these charges with silane towards by-passing the mechanism on one hand and the preservation of the dielectric constant (~ 4) in a humid medium on the other. Damping measurements show there is no spreading of the conductive film previously seen [BEI 01]. The influence of non-soluble contaminants on external porcelain insulators gave rise to a modeling applicable to direct and alternating voltages, and in good correlation with the experiment it permits to limit the tests and maintain *in situ* control [SUN 96]. Finally, we should mention the problem encountered by certain external insulators, made of porcelain or a composite (EPDM) which are subjected to low temperatures. A more or less excessive crystallization covers the elements with a density varying from 0.3 to 0.9 g/cm^3, and the leak current doubles and by-passing probability increases. The parameters playing on this type of contamination are the wind and a saline atmosphere [FAR 95]. Despite its publication date, we could usefully refer to the synthesis of a workshop on transmission lines held in 1981 [EPR 81]. The recent use of optical fibers for the communication industry, and installed along the towers supporting high voltage lines, could be added to the previous research; indeed, on the surface of the fibers, by-passing and erosion phenomena appear (the example provided has LDPE coverings, charged with black carbon, insulating the fibers which are mounted along line supports of 161 kV subjected to sub-tropical climatic conditions [KAI 00]).

8.3.3. *Induced precursors*

So far we have not taken into account the influence of stresses undertaken in the system by the insulating material itself, which is important. Although these stresses could be considered when looking at the elements themselves, we are going to consider the evolution of the part they play during the operation of the system.

8.3.3.1. *Outgassing of the insulating material*

A number of experiments have been carried out on inorganic and organic materials, showing that the presence of a strong field could lead to their outgassing. We compared the behavior of these materials under vacuum (~ 10^{-4} Pa) and submitted them to a direct voltage of up to 150 kV. It should be remembered that the behavior of different organic materials varies; only methyl polymethacrylate (PMMA) is close to ceramic and reveals a gas emission from 120 kV. At the same time we noted a current increase of 10^{-7}–10^{-8} A preceding breakdown [AVD 67]; this behavior is in accordance with PMMA’s listing, where it appears within the highest values for the breakdown field and for cohesion energy density (CED). The parameter δ^2 (vaporization energy/molecular volume) characterizes the energy required to separate a compact structure in a large number of separated molecules [SAB 76].

A gas emission was also detected by mass spectrometry when films of polyethylene fluoride, polyethylene terephtalate and polyvylidene fluoride (PVDF) were submitted to fields ranging from 40 to 100 V/µm. This gas emission (H_2, CH_3, HF, etc.) is produced rapidly, so cannot be imputed to an increase in temperature [BIH 87]. A polarization of PVDF under fields of the order of 100 V/µm has revealed emissions (HF) during the suppression of the field at both polarities, a phenomenon which was not observed during the PET study [EBE 93]. In experiments whose goal was to study the conductivity of Nylon 66, it is interesting to note the synchronism of gas emission which stopped with the polarization application [SEA 68]. Despite the particular choice that these materials represent, experiments emphasize the influence of the electrical field and permit, for example, at least partly, the conductivity evolution of certain insulating surfaces to be justified, whatever the type of pressure surrounding them, as we have already seen. The behavior of the materials is particularly noxious to the vacuum-packed systems strength under high voltage. The avalanche of secondary electrons at the surface of the material under stress and preceding the breakdown has often been evoked to justify the adsorption of gas molecules preceding breakdown. These observations have led to the proposition of models in close agreement to the experiment [AND 80]. When the gas molecules remain in the material, we have also underlined the existence of free volumes likely to receive them; indeed, the local field, the defect size and the nature of the gas can contribute to the apparition and the preservation of the ionization process, which we shall see in the following section.

8.3.3.2. *Mechanical deformations*

The action of a unidirectional mechanical stress, an elongation for example, can generate ellipsoidal-shaped defects comparable to gaseous cavities, considering the little matter they contain. Wendorff chose polyoxymethylene to demonstrate this phenomenon; while the average dimension of defects, from 8 to 120 nm according to the direction of the stress, is independent of that stress, their concentration increasing exponentially with it [WEN 79]. In the domain which concerns us, it is the stress generated by the electrical field which causes the initiation of defects and possible fractures [BLO 69]. The joint action of charges and operating voltages leads to the deformation of the material and the bond break; and charges generate local fields getting combined to the macroscopic field in an additional or opposite manner. We consider the easiest of the deformations presented by the amorphous phase of the semi-crystalline material when dealing with a polymer. Amongst the three relaxation processes (generally named α, β, γ, in decreasing order of time), α plays a dominating role on the free volume and the creation of microcavities. A component perpendicular to the direction of the field contributes to the local dilation of the structure and therefore to an increase of the free volume [LEW 02], [JON 05]. When taking into account only the electromechanical force Maxwell applied at the interface of the conductor and the insulating material, the stress undergone by this latter is proportional to the square of the applied field ($P = ½\ \varepsilon_0\ \varepsilon\ E^2$). The presence of induced stresses has been shown by optical measurements on obviously transparent materials, i.e. free from mineral charges. We can mention for example polyepoxy [SAB 76], LDPE [LEW 93], XLPE [STA 55] [DAV 92] [DAV 94], and PET [MAM 04b]. We note, however that the purely mechanical effect subordinated to the action of the field and its frequency [ARB 86], depends on the nature of the insulating material, its chemical and physical structure, its crystallinity and its working temperature.

8.3.3.3. *The frequency*

The influence of the working voltage frequency of an electrical system on the ageing of insulating materials has been revealed in several commercial polymers. In dry materials, we have observed the breakdown voltage drop as well as the decrease of the lifetime, making the frequency vary between 60 Hz and 1 kHz. On the other hand, experiments show that the development of water trees present in cable insulating materials such as LDPE, XLPE and EPR increases in a significant way with the frequency and can lead to an early breakdown [CRI 98]. The limit frequency for tests on insulated cables by XLPE was estimated at 1,700 Hz [BAH 82].

8.3.3.4. *Irradiations*

In this section we consider two important areas: space and nuclear applications. In the latter, irradiation conditions are not very severe but long lasting or, conversely, contain important doses for a short time, pressurized water reactors being characteristic of both situations. The insulating materials used in cables appear on top of the polymer listing being subjected to irradiation tests [SCH 79], since the ethylene-propylene-hexadiene terpolymer 1-4 (EPR) insulating conductors, and chlorosulfonated polyethylene (HYPALON) play the role of internal and external sheath. These materials are usable until doses of 1 MGy (in the SI system, 1gray = 1 J/kg = 100 rads). Certain studies have shown that for outputs of doses less than 500 Gy.h^{-1}, degradation is controlled by oxygen diffusion and accelerated by temperature. We observe, for the EPR, an oxidated superficial layer and a cross-linking process in volume; this latter is unique in the case of HYPALON. We believe that the structure modification generated by irradiation and amplified by temperature leads to the loss of electrical properties for this type of material [GUE 92].

In space, in a vacuum environment, the parameters participating in the degradation of electrical insulating materials are even more numerous (electrons, protons, ions, photons, etc.) [FRE 82], [ROS 87], [LAG 90]; but the electrons and protons are of great toxicity considering their deepness of penetration, the most important one being electrons, and therefore of the creation of a space charge as a function of the stopping ability of the used materials [BAL 83], [BEE 81]. The breakdown processes concern the volume as much as the surface of the material and, in this latter case, we find again the conditions of surface discharges or by-passing which we have previously evoked [RID 82].

The term *precursor* for dielectric solids, used in embedded systems, can be applied when experiments are carried out on earth, in order to simulate at least one part of life conditions; however, it can only be used to attempt to explain the failure phenomena which have appeared in orbit, like in the case of experimental satellites such as SCATHA (Spacecraft Charging at High Altitudes) [REA 87]. More recent experiments called SPEAR (Space Power Experiments Aboard Rockets) have been carried out in order to verify the efficiency of devices capable of inhibiting the breakdown processes generated, and kept by outgassing of the materials and the effluent of the vehicle [RUS 92].

8.3.4. *Observed precursors*

Chapter 6 states the injection and the storage of charges in insulating materials under stresses, as well as the consequences. We shall examine here some behaviors of materials subjected to the electrical field stress and subordinated to induced precursors previously mentioned.

8.3.4.1. *Electroluminescence*

Although the light emission phenomenon of polymer films subjected to an alternating electrical field was observed over fifty years ago, a greater interest has been shown in this phenomenon much more recently to explain the degradation process of organic electrical insulating materials.

To avoid any discharge phenomenon on the surface of films placed under uniform stress, the tests were first carried out, under vacuum, on samples of LDPE, Nylon, PET, etc., one of the electrodes being composed of a conductor glass. This technique allowed the simultaneous recording of the applied voltage and the signal delivered by a photomultiplier detecting the light through the transparent electrode. The presence of light was imputed to the presence of carbonyle groups [HAR 67]. It was difficult to distinguish between the groups created because of the oxygen diffusion in the material and the groups inherent to the structure. In the following years, the same approach was undertaken with polyepoxy to study the dielectric breakdown phenomenon [COÏ 78]. More recently, in a synthesis article, Laurent [LAU 99] presented the radiative processes (fluorescence or phosphorescence) generated in the materials, particularly underlining the importance for ageing, of the presence of oxygen molecules to which an excitation can be transferred, giving rise to other reactions. This electroluminescence phenomenon is well associated with degradation reactions; it is an *indicator* of pre-breakdown and, even if it cannot be discovered on a real system, its study contributes to the understanding of breakdown mechanisms of solid materials. Detection thresholds differ according to whether the applied stress is direct or alternate: for the XLPE 80 kV/mm in direct voltage to 10 kV/mm in alternating voltage was achieved, and from 400 kV/mm to 70 kV/mm under the same conditions for the PET. Bamji estimates the highest detection sensibility for electroluminescence if we compare it to that of partial discharges, which is shown by the conversion of water arborescence into electrical arborescence [BAM 99b].

8.3.4.2. *Pre-breakdown currents*

The polarization of a certain number of organic insulating materials (PE, PET, etc.) in the form of films identical to those destined for the manufacture of capacitors, has often revealed current oscillations as the electrical field increases; the threshold field of the phenomenon decreasing when the testing temperature increases. This deviation from Ohm's Law could manifest itself for fields as weak as 25 V/μm applied to PET or LDPE films for example [PRE 71], [TOU 74], [GOF 82]. The warning signals of an impending breakdown can be interpreted without resorting to local defect hypotheses [JON 80]. In certain experiments, it is a rapidly increasing pre-breakdown current, of the order of 10^{-3} s before perforation, which has been recorded on several polymer films; LDPE and EVA make an exception to this behavior [MIZ 87], [HIK 90]. More recently, current measurements on LDPE plates of 1 mm thickness, submitted to direct voltages ranging from 10 to 60 kV, have revealed instabilities of transient current when the voltage was greater than 50 kV. These instabilities can be attributed to an injection of electrons at the cathode of this experimental capacitor. Simultaneous measurements of the space charge subordinated to the increase of the applied field show the coincidence of these instabilities with the period preceding the breakdown [VEL 96].

8.3.4.3. *Arborescence*

Two types of these defects can appear in insulating materials (and amongst energy transport cables in particular), with electrical arborescence perfecting the degradation achieved by water arborescence when this latter exists; statistical studies on cables found faulty after several years have demonstrated it [CHA 90]. We shall evoke here the observation which was first made on weak voltage cables (8 kV), insulated from XLPE, having aged 8 years. Physico-chemical analyzes have revealed oxidation marked in the regions where water arborescences were present; this same oxidation could be activated on samples placed in contact with high-temperature water. The outbreak of microcavities was observed, which become the seat of discharges, and lead to the breakdown by electrical arborescence [GAR 87]. This latter observation gets even more important when dealing with higher voltages and therefore operating temperatures of the order of 90°C. A second observation relative to this transition phenomenon calls into question impulsion due to lightning, as happened when 80 kV struck a 15 kV cable insulated with XLPE. The modeling of such a circumstance suggests a consequent warming-up of the water contained in water trees, the creation of an electromechanical stress due to the effect of the local field, and the creation of cavities becoming the seat of electrical discharges [BOG 98].

8.3.4.4. *Presence of electrical discharges*

In this section we will present a few representative examples of *observed precursors* taken from the numerous articles published on the subject. We also recommend reading reference books on measurements and their interpretation, in which it is noted (particularly by Timpe) that the first experiments relative to the toxic nature of gas bubbles in solids took place at the end of the 19th Century.

Timpe also records cable failures from as early as 1902 [TIM 79]. In 1936, Robinson, whose interest was focused on oil-impregnated high-voltage cables, already called into question the presence of cavities and ion bombardment of their walls [ROB 36]. In fact, ions, electrons and UV radiation appear when under the influence of alocal field, and the gas trapped in these cavities gets ionized. More recently, Bartnikas [BAR 02] records the history of what were first called *crown discharges*, and then *partial discharges*, and chiefly stresses the efforts realized over the years to detect this *precursor* of degradation and breakdown in all types of insulation. Bartnikas recalls that progress in electronics has enabled the use of detectors in variable ranges of frequency, from 30 to 400 kHz for routine measurements, and from 800 kHz to 1 GHz according to whether we are dealing with cables, rotating machines or gas insulated cables (SF_6) under pressure. The very fruitful association of the computer with analysis on current impulsions related to discharges must also be mentioned. These tracks are then correlated in the form of cavities, their number and their localization [GUL 92], [VOL 92]. Let us note finally that techniques have been developed to get a numerical analysis of the transient electromagnetic field [POM 02].

Laboratory studies have strongly influenced this progress by the approach of physical phenomena in ionized gases, as well as in degradation processes of the insulating material under the effect of discharges.

We would remark as an example that the evolution underlined at the beginning, i.e. the replacement of paper by a plastic film to constitute the dielectrics of a power capacitor, leads to an increase of the operating field until 60 V/µm, but limits the acceptable level of partial discharges to 20%. This same level is at 200% when the dielectric is constituted by a paper-oil association [HAN 92], [HAN 93]. The diagnostic test of a 15 kV cable insulated by EPR shows that the cavity of about 1 mm will generate a signal of the order of 10%, the minimum detection threshold. When the cable is the seat of arborescence, the form of impulsions, their width and their growth time are different from spherical cavities. However, the time separating the detection from the breakdown can only be defined from the amplitude of discharges, a large amplitude not being synonymous with harmfulness, nor that their outbreak is subordinated to the presence of a floating metallic particle. The experiment shows that the detection is specific to the equipment concerned and the insulating material [BOG 00]. We note that practical detections are much greater than those realizable in a laboratory, a level of 0.01% having been reached and the detection of the first arborescence branch (< 10 µm) made from 0.05% [LAU 93]. Let us note that amongst the disadvantages constituted by the discharges, certain types are difficult to detect; these are the *luminescent* or *pseudo luminescent* discharges which can appear in cavities trapped in the insulation of turbogenerators. These cavities containing hydrogen subjected to a neighboring field of the ionization threshold field could be the seat of this type of non-detected discharges [BAR 92]. Recent laboratory research has focused on characteristics of discharges appearing in electrical arborescence created in XLPE and at the EPOXY-EPR interface. The characteristics found are identical in both types of insulation; however, the threshold and the voltage of the discharge outbreaks appears to depend on the homogeneity of the bonding of both materials, and therefore of the presence or not of trapped gas at the moment of manufacture. This conclusion from laboratory tests is applicable to the manufacture of a cable. These are also the characteristics of discharge (growth time, width, etc.) which distinguish the origin of the defect (interface or core of the material) [DEN 01]. An investigation of about 25 years concerning the detection of discharges and its interest in the domain of rotating machines and particularly stator coils has recently been published by Stone which shows the possibilities of following the behavior of a system during its lifetime and of intervening before a breakdown [STO 04].

8.3.4.5. *The case of transformers*

The analysis of gases dissolved in power transformers in service, amongst them being paper and oil, can lead to their origin. A list of detectable defects has recently been presented by Duval [DUV 02]. It includes in particular the defects observed in service from the Defects of thermal origin are classified according to temperature, with 300°C < T < 700°C or > 700°C. A comparison is made with laboratory models in order to confirm the nature of analyzed hydrocarbons and their correlation with the amplitude of detected discharges. The decomposition products of cellulose must obviously appear as late as possible and, in order to avoid the oxidation of oil, the extraction of oxygen appears essential in the same way as particles suspended in the liquid phase [FER 02]; the prevention of this oxidation is generally secured by the addition of an antioxidant in the transformer oil.

8.4. Conclusion

We have seen that by taking the term precursor in the broadest sense, we have been led to define the causes of the breakdown of electrical insulating materials.

They have thus shown the difficulties of preventive detection, system failures and their possible life end. The revelation of the role of precursors before the material undergoes the stresses of operation has underlined the precautions required in the development of materials and equipment. During the use of this latter, parameters such as environment, temperature, direct or alternative voltages with their frequencies of use, are separately or in association with the actors of the lifetime. Of the three types of insulation (solid, liquid and gaseous), only the first presents the disadvantage of having an irreversible breakdown process. As we have seen, the control methods are not numerous but most of them are efficient owing to their analysis techniques. The contribution of laboratories is fundamental in this domain even though caution is required; different schools of thought are sometimes opposed to or otherwise distinguish themselves from ageing or breakdown in their relative outlines. This point was well made by Cygan and Laghari about ageing models in 1990 [CYG 90].

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