AGING OF POLYETHYLENE FOR HIGH VOLTAGE
CABLE INSULATION

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INTRODUCTION

The development of polyethylene in the Second World War as a low loss dielectric for communication cables raised hopes of reducing the cost of underground power transmission with solid dielectric cables. In this case and from then onwards, the United States has taken the lead in the development of new power cables. The first 15-kV cable with extruded polyethylene insulation went into service in 1955, and by 1963 a 138-kV solid dielectric cable was produced. The first short length of this cable was supplied in 1965 to Puerto Rico where it has been operating ever since. In the low voltage distribution field, polyethylene insulated cables have been a resounding success, but progress in the high-voltage area has been slow. Power companies have hesitated to adopt high-voltage solid dielectric cables because of a series of splice failures caused by thermal expansion and more splice and cable features on the Waltz Mill test site on load cycling to the exceptionally high conductor temperature of 130°C and more. A significant amount of 225-275 kV solid dielectric cable is in service in France, Sweden, and Japan. Fujikura is the first developer of a 500 kV XLPE cable system.

Substituting polyethylene or other polymeric tapes for paper promises significant reductions in cable charging current and dielectric loss. There may also exist some potential for lowering the thermal resistance of the high-voltage insulation. Serious studies of this topic began in 1960. The first tests of 275 kV cable samples with polyethylene tape insulation filled with SF_6 gas at 150 psi reported in 1967. As might be expected, these pioneering experiments were not entirely successful, but they provided sufficient encouragement to continue the research. A serious differential thermal expansion problem had been revealed, which on thermal cycling, caused premature breakdown of the insulation. Besides, the thermal resistivity of the gas-polyethylene combination turned out to be greater than anticipated. These difficulties were overcome with further research, compared with oil-paper cables, the new insulation system would reduce cable capacitance by 37 percent, dielectric losses by 95 percent and the thermal resistivity by 33 percent. A commercial 400 kV self-contained cable with SF_6 gas pressure assisted polyethylene tape insulation is being developed in Great Britain.

SOLID DIELECTRIC CABLES

At the 1976 IEEE Underground Transmission and Distribution Conference, Deschamps claimed that, ". . . at the present time in France, at the 225 kV level, the polyethylene insulated cable is superseding all its competitors." In the same year, CIGRE reported that approximately 200 km of 123 to 145-kV cross-linked polyethylene cable had been manufactured in Sweden and installed in Scandinavia and other countries. Both the French and Swedish experience with solid dielectric high-voltage cables appears to be very satisfactory. Although the first successful installations of this new type of cable were made in the United States in the mid 1960s, here solid dielectric transmission cables suffered a severe setback through a series of splice failures and adverse test results at Waltz Mill.

History has taught us that the success of a new type of high-voltage cable is not secure until at least 20 years of service experience are at hand. However, the growing understanding of breakdown in solid dielectrics, gives us reasons to be optimistic.

In 1969, the first high voltage cable with extruded polyethylene insulation became a part of the French 225-kV transmission network. By 1976, some 14 circuit-kilometers were in service, which are being produced by the SILEC Company. Its excellent performance record appears to be due to the conservative design.

To obtain a three-phase circuit capability of 300 MVA, the 225 kV cable is furnished with either an 805 mm^2 (1589 kcmil) copper conductor or a 1200 mm^2 (2368 kcmil) aluminum conductor. Both are of the standard four-segment construction familiar from pipe-type cables. High-molecular-weight plain polyethylene is coextruded with semiconducting (carbon filled) polyethylene layers on either side. All manufacturers of solid dielectric cables agree that firm adhesion of the shielding layers to the insulation is imperative. SILEC claims that this requires simultaneous bonding at a temperature of 200°C.

It may seem surprising that the high voltage solid dielectric cable relies on non-cross-linked and unstabilized thermoplastic polyethylene. This material was used in the United States in distribution cables and has a poor performance record. It is particularly vulnerable to water, and this fact may have decided the French manufacturers to enclose their cables in a conventional 3-mm-thick lead sheath. The insulation wall of 2.4 cm (0.944 in.) thickness is more substantial than in a paper cable, but quite highly stressed as far as extruded polyethylene goes.

The most conservative aspect of the French cable is the conductor temperature rating. Originally, 65°C was recommended by the manufacturers. Electricite de France appears to have relaxed this to 70°C. As cables are never subjected to constant full load over extended periods of time, the French 225-kV cables are unlikely to operate above 60°C. This will keep the differential thermal expansion between conductor and insulation wall within 1 percent, which prevents serious plastic deformation of the dielectric during load cycling. The lead sheath has to be protected against corrosion. SILEC chose for this purpose a 3.5-mm PVC jacket extruded directly on the lead.

Three advantages are claimed by the French utility for solid dielectric over self-contained oil-paper insulated cables. They are (1) lower manufacturing costs, (2) less maintenance of the dry cable system, and (3) easy adaptability to external water cooling.

The U.S. cable manufacturers advanced cross-linked polyethylene insulation to the stage where it could be employed in high-voltage cables. The General Electric Company, which was primarily responsible for the introduction of cross-linked polyethylene insulation, in addition filled the polyethylene with mineral or organic powder which acts as a voltage stabilizer. For some years, cross-linked polyethylene was promoted on the strength of its higher softening temperature and the higher conductor temperatures and current densities which this permitted. It was claimed that cables with this insulation could safely operate at 90°C and under emergency conditions at 130°C.

Vulcanized rubber was the first solid dielectric material that was applied to power cables. Rubber-insulated cables were employed by the New York Subway Commission as long ago as 1888. Wherever flexibility is of overriding importance, rubber-insulated cables are still in great demand. An example is trailing cables for mining machinery. Synthetic butyl rubber is being preferred to natural rubber. Not until the copolymerization of ethylene and propylene in an ordered structure became a practical reality did rubber insulation have a chance to compete with polyethylene in dielectric strength.

Ethylene propylene rubber (EPR) insulated cables were quickly developed in the 1960s up to 138 kV. EPR cables have since then been confined to distribution voltages below 100 kV.

A more significant property of EPR is the high softening temperature of 135°C, which is equal to that of cross-linked polyethylene. EPR is toughened with a filling of mineral powder, but it retains sufficient elasticity so as not to have to undergo plastic deformation during load cycling. This appears to be an advantage, yet at the same time, the small modulus of elasticity is potentially hazardous if the conditions for electromechanical breakdown are fulfilled.

TREEING IN POLYMERS

The prebreakdown process in polyethylene is called "treeing" because of the branched structure and the single stem of communicating fine channels that were left behind as evidence of a partial discharge of the dielectric. Discharge trees have been observed in virtually all solid insulating materials, including glass

and porcelain, but they are difficult to find unless the insulation is transparent. They have the appearance of three-dimensional Lichtenberg figures, and it is, therefore, tempting to think of them as having been created by electron avalanches and streamers.

The tree channels are approximately circular and typically 5 to 20 μm in diameter. The tree channel cross sections were photographed with a scanning electron microscope and were found to have rough walls to which molecular debris adheres. Several investigators have observed the growth of trees under an optical microscope. They found it to proceed in distinct steps, one or more branches being added at a time. The growth process could be speeded up or slowed down by raising or lowering the applied voltage. At sufficiently high voltage, a tree can be grown in a microsecond, whereas it may take years to develop under low dc stress. That every breakdown in polymers is preceded by tree formation is now widely accepted. No absolute proof of this fact appears to have been provided but much convincing evidence is available in the form of tree branches emanating from the much larger breakdown channels.

A tree developing at constant voltage in uniform or divergent fields does not inevitably lead to breakdown during the lifespan of a cable. This is true even if the tree grows from the low field region at the insulation shield into the stronger field near the conductor. Breakdown is likely to occur when a tree completely bridges the interelectrode gap, but it need not take place the instant the tunnel has been finished. At times, the leading branches of two trees growing from opposite electrodes have been observed to pass each other quite closely before breakdown took place.

Dielectric material excavated during the tunnelling operating is somehow converted to gas, which diffuses to the root of the tree or from the branches directly into the insulation. The gasification and the breaking of molecular bonds that this implies could possibly be explained by gas channel breakdown mechanism. Although free carbon has been detected on the walls of tree channels, they are usually not blackened like the much wider breakdown channels. Most trees are grey in appearance; few have color with blue, green, brown and yellow tints prevailing. Colorless trees also contained foreign elements, among them sodium, potassium, calcium and chlorine. From these experimental findings, it may be deduced that conducting and semiconducting matter is drawn into the tree from the root. If water is present at the root, this will also be pumped into the tree by electrostatic forces, and it is likely to bring with it dissolved salts. Trees of this kind have become known as "water trees." Wet and dry trees containing elements that do not belong to the dielectric material are also described as "chemical trees." Sulfur trees have received particular attention. They were found in cables containing copper conductors and were installed in locations where hydrogen sulfide gas was present and diffused through the polymer to the copper conductors. Corrosion products may then initiate treeing over an extended area and copper sulfide migrates into the tree channels.

Conflicting reports exist on the effect that water immersion has on tree growth in polyethylene–insulated cables. Most experience suggests that the wet environment reduces cable life.

It is a remarkable fact that all trees have their origin or root at a free surface of the insulation or at an interface between the insulation and electrodes or foreign particles. This proves that tree initiation is not only determined by electrical stress but has something to do with the property of surfaces. The convenient practice of studying tree growth from the point of a needle molded into the polymer specimen has highlighted the importance of local field enhancement at conductor surface projections. It is undoubtedly a factor contributing to the site selection for the tree root. Less voltage is required to grow a tree from a needle point when the tip is positive rather than negative.

Just as the dielectric strength of a material is greatest when exposed to dc and lowest when placed in an ac field, so the tree starting voltage is much higher for dc than for ac. Impulse voltage tree initiation lies between dc and ac values.

Applied electric stress cannot be the only factor governing tree growth because the tree channels do not follow the field lines of flux. The way branches grow simultaneously at different distances from the opposite electrode and in directions that can be almost perpendicular to the applied field is perhaps the most puzzling aspect of treeing in polymers. Figure 1 is an attempt to classify the appearance of trees. The most

photographed trees are those grown deliberately from the tip of needles. They resemble the shape of natural trees. Other trees have the appearance of a bush with many branches going directly back to the root. Bush trees are often observed when water is present at the root. Bow-tie trees grow in the middle of the dielectric wall from cavities and foreign particles. The streamer and broccoli trees were found in cables removed from service. The significant feature of the last two groups is the large number of roots per unit electrode area, which suggests that they are caused by area contamination rather than stress raising asperities. The similarity of the geometry of gas discharges (Lichtenberg figures) and polymer trees is striking. A noteworthy aspect of all these discharges is that, in spite of the irregular paths of discharge channels, the branches never cross each other and are all connected to a common root.

The early development of polyethylene-insulated cables revealed that the tree inception voltage could be increased by mixing staining rather than nonstaining antioxidant into the insulation compound. The results plotted in Figure 2 show this effect for varying amounts of DPPD (NN-diphenyl-P-phenolyne-diamine) antioxidant. The half-life refers to the time after which half the number of samples tested under identical conditions showed tree growth. This finding set in motion extensive research on polyethylene additives. The additives have become known as voltage stabilizers or tree inhibitors and the intention is to disperse them uniformly through the insulation. Although voltage stabilizers are not able to eliminate tree initiation, they retard tree growth sufficiently to have a significant influence on insulation life in the presence of some inevitable defects. Polysulfone has the highest tree initiation resistance; chemically cross-linked polyethylene is almost as good as commercially available voltage stabilized thermoplastic polyethylene, and polypropylene rubber has very low treeing resistance. The decomposition products of dicumyl peroxide, which is being employed for chemical cross-linking, have been found to be excellent voltage stabilizers, but they slowly diffuse out of the insulation and thereby lower the treeing resistance. Fillers also increase the treeing resistance of polyethylene and so does dispersed water vapor.

Since trees in polymers are precursors of complete insulation failure, we must expect all breakdown mechanisms to play a role in their formation. By the same token, it would be unrealistic to expect us to understand treeing any better than the breakdown of solid dielectrics. When they were first discovered, it was thought that trees were the result of erosion by discharges in cavities and the tree channels themselves. Sputtering of dielectric material under the impact of impinging positive ions should make the channels straight and parallel to the applied electric field. As this is not so, the sputtering explanation has to be abandoned. Actually, experiments showed that the first luminous event of the discharge takes place in the solid material rather than in the gas channel.

AGING OF POLYETHYLENE INSULATION

Aging processes in polyethylene (PE) are initiated and accelerated by such factors as electric field, temperature, moisture, ionizing radiation and light. Several of these factors can, either independently or simultaneously, play an essential role in cable aging, according to cable operating conditions and environment.

The aging processes lead to structural changes in the polymer. Irrespective of the aging factors being applied, oxidation and subsequent disintegration of macro-molecules is the most important, internal, degradation process [1]. Absorption of oxygen by PE is characterized by an initial period in which the rate of oxygen absorption is very low. This period, therefore, determines the speed of the oxidation process. After this period, the content of carbonyl groups increases rapidly. The number of oxygen compounds increases as well. The increases are, in general, maintained during the entire aging period. It has been postulated that electrical breakdown arose due to this PE oxidation or oxidation together with mechanical stresses.

The presence of oxygen bonds can be experimentally determined by means of spectrophotometric analysis in the infrared. The coefficient of absorption for a given band of radiation is a measure of qualitative and quantitative structural changes. This coefficient is defined in terms of a ratio of absorbed radiation to incident radiation and is given by,

$$A_\omega = \frac{\ln\left(T_0/T\right)}{T} \tag{1}$$

where T_0 is the value of background transmission, T is the minimum transmittance value, and $\omega = \lambda^{-1}$ is the wave number of the absorption band being used. These quantities are shown in Figure 3.

The changes in the absorption spectra are indicative of physical and chemical changes in the PE due to aging. Different spectral bands are sensitive to different physical and chemical changes. Therefore, infrared spectrographic analyses can be used to compare the composition of materials which have the same basic component but varying additives. It can also be used to compare similar materials constructed using different technologies and methods. Spectrographic investigations make it possible to perform the evaluation of internal structural changes occurring in polymers subjected to various aging processes. The technique, therefore, supplies information on the number and composition of carbonyl groups. In the work reported here, the absorption of infrared spectra at 990, 1720 and 3380 cm^{-1} was used to monitor PE structural changes.

Some publications mention the correlation between structural changes and electrical properties of PE, such as changes in the dielectric loss factor [3] or changes in electric conductance [4]. The work reported in this paper, as well as other papers of the authors [5,6], have as an objective to relate structural changes, measured spectrographically, to changes in electrical characteristics which are important to reliable cable operation. The electrical properties and absorption spectra were determined as a function of aging time and multifactor stresses.

Test Method

Experimental investigations were performed on specimens in the form of PE sheets and foil. Sheets of dimensions 15x15x0.5 cm^3 were molded for these investigations using polyethylenes used in the cable industry. These polyethylenes contained a voltage grading additives. The foil of 0.06 mm in thickness was made of PE initially used for the production of cable. The PE, as such, contained no voltage-grading material.

The specimens were subjected to various aging processes. Sheet specimens underwent thermal in air, thermal-water, thermal-chemical, voltage and voltage-water aging. The foil specimens were additionally aged by partial discharges and ultraviolet radiation. Thermal aging was carried out at a temperature of 90°C. A 5% (by weight) water solution of NaCl was used as an agent to simulate chemical aging. Foil thermal aging, under normal conditions, was performed at a temperature of 20°C, in a well-illuminated room, but in the absence of sunlight. Voltage aging was accomplished by using dc and ac voltages at a field value of 6 kV/mm.

The aging effect of partial-discharges was determined using a system with simulated discharge voids. This system consists of three foils of which the top and bottom layers were the foils being tested. The middle foil had a 2 mm diameter hole which formed the simulated void. Only the lower foil was evaluated to determine the effects of partial discharge aging. The aging voltage was three times higher than the partial discharge inception voltage. Aging was also performed in the presence of UV radiation, produced by a quartz lamp, located 50 cm from the sample.

In order to define the influence of aging agents on the electrical characteristics of PE, measurements of electric strength and of mean time to breakdown were made for unaged and aged specimens. The electric strength measurements were performed using 50 Hz alternating voltage.

For the specimens in the form of sheets, needle-plane electrodes were used to measure the breakdown voltage. These steel needle electrodes had a tip radius of 10 μm and were inserted in the insulation. The thickness of tested insulation was 2 mm. The dielectric strength determined is the maximum stress, at the needle electrode, at breakdown.

For the foil, these measurements were made directly by applying and measuring the breakdown voltage in a parallel electrode geometry. The breakdown voltage was determined by gradually increasing the applied voltage at a rate of 2 kV/min.

The analyses of structural changes were made using an infrared spectrophotometer. The spectrum was analyzed in the range of 4600 to 650 cm^{-1} (2.2 to 15 μm). The resulting spectrograms were used to determine the degree of crystallinity of the foil samples. The degree of crystallinity was defined by determining the mutual ratio of infrared transmittance in the 731 cm^{-1} band and the transmittance in the 1368 cm^{-1} range. The transmittance in the 731 cm^{-1} band corresponds to the contents of the crystalline phase in PE and the 1368 cm^{-1} band characterizes the amorphous phase in the PE.

Using spectrograms of selected PE samples, the percentage of crystallinity, H, was defined as

$$H = \frac{100\,D}{\dfrac{A^C_{731}}{A^A_{1368}} + D} \tag{2}$$

where $A^C_{731} = 1.53$ is the characteristic coefficient for the crystalline phase in the 731 cm^{-1} band,

$A^A_{1368} = 0.41$ the characteristic coefficient for the amorphous phase in the 1368 cm^{-1} band, and

$D = A731/A_{1368}$ the ratio of absorption values in the 731 and 1368 cm^{-1} bands.

Changes in absorption, in selected infrared spectral bands, are indicative of specific structural changes. Spectrographic data were obtained in several spectral ranges. These include: (a) the 3600 to 3350 cm^{-1} band, which is indicative of the presence of OH groups; (b) the 990 cm^{-1} band, which is indicative of the presence of C–H unsaturated bonds; (c) the 1720 cm^{-1} band, which is indicative of the presence of the C=O group in PE. The performance of absorption in this band is an important indicator of material oxidation.

RELATIONSHIPS BETWEEN PE ELECTRIC STRENGTH AND STRUCTURE

The relationships between changes in the structure of PE and changes in its dielectric properties, caused by material aging processes, are shown in the data to be presented. Data points shown were determined on the basis of 10 measurements. Values, representative of given sample lots, were established by making use of the Weibull distribution.

Specimens in the Form of Sheets

Electrical measurements show that under the influence of different aging agents, a change in electrical properties occurs. Figure 4 shows probability of failure as function of time to breakdown, after aging under different conditions. Thermal and voltage–water aging have been selected as factors which simulate the cable operating environment. The influence of only voltage aging (curves 2 and 3) on the probability of failure is small. The thermal aging in air (curve 4) has a smaller effect than in the case of thermal–chemical aging (curve 5). The most extreme aging (curve 6) was caused by voltage–water aging. This can be explained by postulating that without voltage, the water penetrated through the insulation only a relatively small distance from the water insulation boundary. However, the application of voltage caused penetration of the water to a larger insulation depth and, hence, a relatively large change in breakdown performance. Additional water–voltage aging data were obtained using three types of PE sheets subjected to voltage–water aging. These results are shown in Figure 5. In these data, the aging rates of different materials have been compared. All three types of PE had reduced electric strength and time to breakdown after voltage–water aging.

Effects of aging factors upon the infrared spectrum were also observed. Special data, obtained simultaneously with the electrical data of Figure 3, are shown in Figure 6. In this figure, the changes in absorption

at 1720 cm^{-1} (carbonyl band), as a function of time during voltage-water aging, are presented. It was found that the absorption increased as aging time increased, for all materials. The C-type PE shows the least change in absorption which implies the highest resistance to voltage-water aging.

Figures 7 and 8 present the relationship between the infrared absorption and changes in electric quantities. The electrical quantities were determined by the needle method. Figure 7 shows the absorption coefficient, in the three spectral bands, as a function of the maximum value of electric stress, at the needle electrode, at breakdown. It is important to observe that smaller breakdown fields correspond to larger absorption coefficients. This implies a definite relationship between A$_w$ and degradation of the PE electrical performance.

Figure 8 shows the absorption coefficient, for the three spectral bands, as a function of time to breakdown. It should be observed that small absorption values imply a long time to breakdown. Again, there is a clear correlation between the A$_w$ values and a degradation in electrical performance. The time to breakdown appears to be a stronger indicator of the relationships between dielectric properties and the polyethylene structure.

The data of Figures 7 and 8 show that the voltage-water aging caused an increase in infrared absorption in the 3600 to 3350 cm^{-1} band. The absorption in this band is due to the hydrogen and oxygen in the PE. An increase in this absorption can be postulated to be due to penetration of water into the insulation. The increase of absorption in the 1720 to 1140 cm^{-1} band was also observed. This has been determined to be due to progressive PE oxidation occurring during the aging process. The spectrum also showed an increased absorption in the 990 cm^{-1} band. This absorption has been determined to be due to tearing of macrochains in the PE and creation of unsaturated radicals.

Foil Samples

The foil samples have been aged in a manner similar to that used for sheet samples. However, a considerably widened range of environmental hazards was used for foil samples, including partial discharge and UV radiation factors. It was found that, for the applied range of environmental hazards, electrical and various types of structural degradation occurred in the PE. It is likely that the observed processes will occur under service conditions.

Figure 9 shows Weibull distribution functions for electric breakdown stresses in PE foil, aged by UV radiation. It was found that, as aging time increases, the value of foil breakdown stress decreases. A considerable increase in the scatter of the data also results during aging. This performance implies that there is an increasing number of locations with weakened strength within the tested material.

The permittivity, dielectric loss factor, and electrical strength of the foil were also determined as functions of aging time and the multifactor stresses. The relative permittivity was observed to change in a range from 2.26 to 2.32 for different types of aging. A more distinct relationship (compared to the change in permittivity) was obtained for the dielectric loss factor and electrical strength as a function of aging. Figure 10 shows the dielectric loss factor, as a function of aging time, for different types of aging. These data show that the largest changes occurred for aging by partial discharges and by UV radiation. The slowest process of degradation occurred for aging under normal conditions and thermal-water aging. Figure 11 shows the electric strength of the investigated PE foil, as a function of aging time, for several types of aging. These data indicate that the electric strength is also degraded most by partial discharges and UV radiation.

Changes of absorption, at selected infrared spectral bands, have been observed for the foil samples for each aging factor. Figure 12 shows absorption in the 1720 cm^{-1} band as a function of dielectric loss factor after aging. These data show the absorption increases as the dielectric loss factor increases, corresponding to aging. This is true for all aging methods shown. Figure 13 shows absorption in the carbonyl band, 1720 cm^{-1}, as a function of breakdown electric field, for various aging methods. It should be noted that the absorption is high in all cases if the breakdown strength has degraded to a low value as aging progresses. The strongest changes in absorption were found to be associated with partial discharge and UV radiation aging.

The degree of crystallinity is another quantity which can be obtained by means of the infrared analysis. Figure 13 shows the relationships between electric strength of PE foil and the degree of crystallinity for several types of aging. In the case of thermal-water aging and aging under normal conditions, it was found that the degree of crystallinity increases as the value of electric strength decreases due to aging. However, for aging by UV radiation, partial discharges and voltage-water and thermal-chemical, the degree of crystallinity decreases as the breakdown strength decreases due to aging. These results show that an estimate of the degree of PE aging cannot be based on the analysis of changes in the degree of crystallinity since, according to the data of Figure 14, a decrease (or increase) in crystallinity cannot be unambiguously interpreted in terms of aging and related degradation of electrical performance.

AGING OF PE CABLE INSULATION CAUSED BY THE OVERLOAD CURRENT

In earlier research, imperfections and weaknesses in polyethylene cable insulation have been generally assessed by partial discharge studies. The partial discharges have led to tree formation and propagation in the insulation. Consequently, the relationship between electrical and other properties and the morphology structure of polyethylene have been widely investigated.

Some of the earliest articles on morphology structure and its relation to the breakdown mechanism included those by Muccigrosso and Phillips [17] and Wagner [20]. Nitta et al [18] described the changes of breakdown voltage of preheated XLPE cables resulting from changing density of the polyethylene. One paper [4] described the morphology structure in different parts of high voltage cable. The morphology structure of the sample varied along the radius of the cable with the material becoming less crystalline away from the conductor. Bulinski et al [7] studied the effect of temperature aging and temperature cycling on times to breakdown of miniature XLPE cables. Yet, many factors that influence the mechanisms of treeing in polyethylene insulation required further testing. One of the most important factors that influences formation and the rate of growth of the trees is the morphology structure, i.e., the crystalline phase in the material.

The work done by Kellow and St-Onge [14] reports the investigation of XLPE insulated cables under emergency conditions. The cable temperature rose in response to an overload and as a result, some cables failed well below the permissible temperature under emergency conditions.

Ieda [12] discussed the breakdown mechanism of polymers and the electrical properties of polymers as a function of chemical structure, morphology structural irregularities and electric degradations caused by partial discharge.

In our previous paper [9], some results of the thermal influence on the morphology structure and on the electrical breakdown and dielectric losses were presented. In this paper, changes in morphology structure of polyethylene cable insulation and some selected electrical properties, caused by temperatures higher than long term in service permissible temperature, are reported.

Thermal overloads in cable insulation may appear during the system's faults while the cable is operating under overload current. The temperature of cable insulation then exceeds the long term permissible temperature. In such a case, the permissible time of the overloads has to be known so that a significant degradation of electrical and mechanical properties of the polyethylene will not take place.

A large amount of amorphous phase in the polyethylene cable insulation could also be created under service condition of the cable system. If, for some time, the cable is working under overload current, the temperature of the cable insulation increases and when it exceeds a given value, the decrystallization process will start. Above this temperature the amorphous phase volume increases.

After a successive cooling process of the polyethylene cable insulation, the crystallization will again leave a larger amount of polyethylene in the amorphous phase. It is suggested that if the temperature of the polyethylene insulation was rising to higher value and for a longer period, the amorphous phase volume will be larger. This process is known as the thermal aging mechanism, which reduces the crystallinity of the polyethylene cable insulation, leading up to the deterioration of electrical properties of the cable insulation.

To estimate the structure changes of the tested polyethylene, samples were cut from 15 kV PE power cables and were subjected to thermal overload at 90°C and at normal working voltage. The testing cycle consisted of 8 hours of sample loading followed by 16-hour intervals. The microscopic examinations of the samples were carried out every 50 to 100 hours of thermal overload.

The samples of polyethylene insulation selected for morphology investigation were prepared in such a way that the spherulite structure of the tested material would be displayed. The cut-out pieces of polyethylene from the thermally-aged cable samples were grinded and polished to a mirror-like finish. Later, the samples were etched with 1% KMO_4 in concentrated H_2SO_4.

The samples were coated with gold before placing them in a scanning microscope. The morphology structure of the polyethylene was studied using a scanning microscope, JEOL-JSM-50A type. The treeing process in polyethylene was observed using an optical microscope, DOCUVAL type, equipped with a TV camera, monitor, etc.

In order to define the time to breakdown and to study the treeing mechanism, needle test electrodes were used. The power cable sample of 70 mm^2 aluminum conductor 1.4 mm semiconductor shield on the conductor with thermoplastic polyethylene insulation 4.5 mm thick was subjected to 15 kV test voltage. The steel needle electrodes of 10 μm point radius were driven to such a depth that they would not damage the 2 mm thick insulation (checked by optical microscope). A 50 Hz ac voltage of 15 kV was applied to the system.

Morphology Structure After Thermal Aging

A typical picture of the morphology structure of the polyethylene cable insulation aged at an elevated temperature is shown in Figure 15. The amorphous phase regions are much larger than the diameter of the neighboring spherulites. This is a result of long time thermal aging effect of the polyethylene insulation.

As a result of thermal overload of the polyethylene cables, the following processes can be observed:

- increase of width of the amorphous phase between the spherulites
- formation of the amorphous phase regions (in the investigated areas of the spherulites concentration at the inner shield) with dimensions which exceed the largest diameter of the neighboring spherulites.

Dielectric Loss Factor and Crystallinity Degree

The change of dielectric loss factor or tanδ as a function of thermal overload time is presented for two different types of thermoplastic PE in Figure 16. It is seen that the dielectric loss factor has also increased with the time of thermal overload. This results from the increase of the amorphous phase in the insulation.

To confirm the last conclusion, the crystallinity degree of polyethylene insulation was checked. The results obtained are presented in Figure 17. The crystallinity degree of thermoplastic polyethylene is decreasing with the thermal aging time. Figure 18 shows that the dielectric loss factor for different degrees of crystallinity decreases as the crystallinity degree increases.

Dielectric Strength and Crystallinity Degree

The dielectric strength of thermoplastic polyethylene presented in Figure 19 was measured in a needle-plane gap [9]. The results present the average value obtained of 10 tested samples. The specimen was placed between plane and steel needle electrodes of 5 μm point radius. The needle electrodes were driven to a depth of 2 mm by means of a micrometer screw. The voltage was increased at a rate of 2 kV per minute until breakdown. The samples of 80 mm diameter and 5 mm thickness were aged at temperatures of 80°C and 90°C, respectively.

The measurements of the dielectric strength and crystallinity degree were carried out on aged sample at 40 to 50 hours. The dielectric strength after 200 hours aging decreased approximately 25%. The crystallin-

ity degree, at the same time, changed from 55% to 45%. Aging of 90°C had a much stronger influence on decrease of breakdown voltage than aging at 80°C.

Results of previous research [15] showed that the dielectric strength of the amorphous phase of polyethylene is several times lower than the dielectric strength of crystalline phase. Therefore, the probability of tree inception and faster extension of tree channels is much higher in the amorphous phase.

Induction Time of Trees Inception

The analysis of the observed phenomena shows that the induction period to initiate an electrical tree is related to many factors. However, a hypothesis could be put forward, that the internal structure (the morphology structure, amorphous phase and voids) of the polyethylene will have the main influence on its duration. The question, whether partial discharges are appearing during the induction period, is still under discussion, i.e., whether the partial discharges do not exist or that they are so small to be measured with the actual available instruments. Towards the end of the induction period, the partial discharges will start, and the electrical treeing will appear, whose channels extend with time. The trees are caused by the interaction between high energy electrons accelerated by the electric field and the end radicals of the polyethylene. Thus, the extension and the shape of the trees depend on the morphology structure of the polyethylene.

Shape of Trees in Crystalline and Amorphous Polyethylene

Often, during the technological process of polyethylene cable insulation, the temperature of the cooling water is not kept exactly as described in individual regions of the cooling process of the insulation. If the cooling process of the extruded polyethylene is too fast, it leads to arresting the crystallization, and a large amount of areas with amorphous phase will arise. The second case, when the larger volume of amorphous phase created as described above, corresponds to thermal overload of the polyethylene cables in service.

The experiment was carried out to compare the trees' development in amorphous and partially crystallined polyethylene. Two types of cable insulation samples were prepared. All samples were heated to a temperature of approximately 115°C.

The first set of samples was then placed in molten nitrogen. This was done to obtain the amorphous phase in the entire volume of the sample.

The second set of samples was cooled over a 24-hour period down to the surrounding temperature, which was 18°C. In these samples, recrystallization occurred, and the measured crystalline phase reached 60% to 70% of the volume.

Later, the samples were tested with the needle method. During the needle test, traces of trees were created. Two types of trees were found. In insulation with large contents of the crystalline phase, the trees were of branch or bush shape. The channels were long with a small number of branches of significant length. At the tip of the needle started a few main channels, usually from one to five, which lengthened during the experiment. From the main channels spread small side channels of much smaller diameter and length. Sometimes, the side channel had increased rapidly through very fast elongation and, therefore, created a new main channel with new side channels.

The tree branches in amorphous polyethylene were very thin. At the beginning of their development, they propagated in the manner noted for crystalline polyethylene. Later, it was impossible to observe the individual phases of increasing the successively arising channels. This process has occurred in an avalanche manner. The branches quickly started to increase in number but then remaining in a nearly spherical shape.

Velocity of Tree Growth

The initiation and the development of treeing in polyethylene power cable insulation leads within a short time to the destruction of the insulation system. The velocity of growth of the treeing in polyethylene depends

upon many factors, such as the magnitude and time of applied voltage, temperature, type of chemical components and the morphology structure of the polyethylene. One of the most influencing factors is the morphology structure. Therefore, the influence of morphology structure on the development of treeing was studied

The parameter which characterizes the resistance of polyethylene insulation to partial discharges is the velocity of the tree propagation in the tested material. Measurements of this parameter have been made in cable insulation specimens of crystalline and of amorphous polyethylene. The specimens were prepared in the manner described above.

The measured velocity of the tree growth was found to be 15-20 μm/min for amorphous polyethylene. The amorphous polyethylene is less resistant to the penetration of the treeing. The average velocity in amorphous polyethylene is 22 μm/min and is approximately 30% higher than the corresponding velocity propagation in polyethylene with large content of crystalline phase. Figure 20 presents the length of tree formation as function of time of voltage application.

The tree channel is caused by the initial gas discharge. Inside the tree channel exists a gas pressure which controls the subsequent partial discharge, which in turn influences the tree channel extension. Thus, the velocity of extension of the tree channel depends on the gas pressure in a tree channel and is a result of a complicated process of the polyethylene decomposition.

The existing voids and micro-voids or other micro-defects in polyethylene [10, 11, 13, 16] increase the velocity of the tree extension, and they also have some influence on the shape of the tree channels.

Propagation of Tree Branches

The polyethylene specimens with tree channels were etched to observe the tree channel path development in crystalline structure. Figure 21 shows the tree channel path at a boundary of three spherulites. No single case was found where the treeing channel would pass through the spherulite.

The studies of treeing growth in different types of polyethylene showed that treeing branches develop in the amorphous phase of insulation, leaving the structure of spherulites unchanged. When the treeing branches meet a spherulite, they tend to circumvent and bypass it and continue to develop in amorphous phase between two neighboring spherulites.

During the partial discharges, the amorphous phase, a material with lower melting temperature, is more prone to erosion. The crystalline phase is melting at a higher temperature. Therefore, the probability that the crystalline phase will be damaged by partial discharges is much lower.

Figure 22 shows typical tree channels in semi-crystalline polyethylene. In all etched specimens, the tree channels proceeded in the spaces between the spherulites.

CONCLUSIONS

Electrical and spectrographic measurements have been made of various characteristics of PE aged using various aging factors. The results show correlation between aging time and electrical parameter degradation and a correlation between aging time and the infrared absorption spectra. This performance implies relationships between measurable changes in infrared absorption spectra and electrical degradation due to aging. Specifically, the data obtained for both polyethylene sheet and foil samples imply the following:

1. Aging caused by multifactor stresses causes a degradation of electrical properties such as dielectric loss factor and breakdown strength.

2. The degradation in electrical properties is associated with an increase in infrared absorption in the 1720 cm^{-1} band.

3. There are correlations between changes in dielectric loss factor and electrical breakdown strength and changes observed in infrared absorption spectra.

4. The analysis of absorption spectra is useful for studying the aging mechanism and the effect of multistresses on dielectric properties.

5. The change of crystallinity degree is not unambiguously related to an increase or decrease in breakdown strength of polyethylene.

6. During a thermal overload of thermoplastic polyethylene cable insulation, the polyethylene undergoes morphology structure change. The changes of morphology structure cause changes in electrical properties.

7. Changes in thermoplastic polyethylene structure are exhibited through creation of amorphous phase regions within the insulation. The increase of the amorphous phase in the material causes a deterioration of the electrical properties, demonstrated by the increase of the dielectric loss factor, the decrease of electric breakdown and the resistance to partial discharges.

8. The treeing which results from partial discharges develops only within the amorphous phase. Thus, increase of the amorphous phase within the insulation's volume leads to a faster treeing process.

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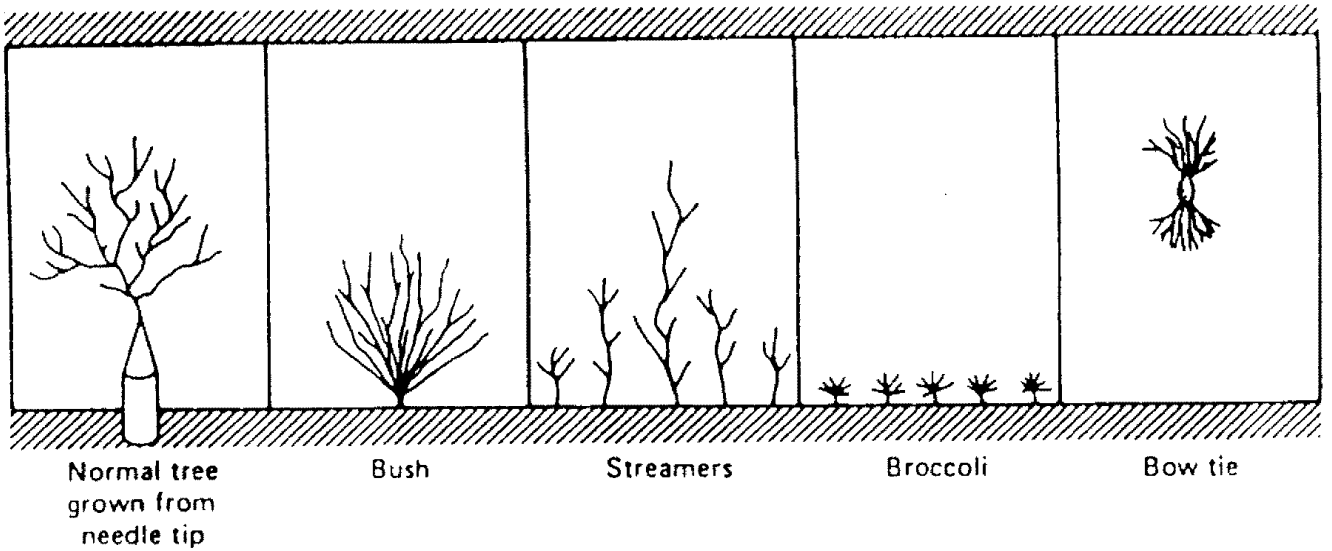
Fig. 1.   Classification of trees in polymers
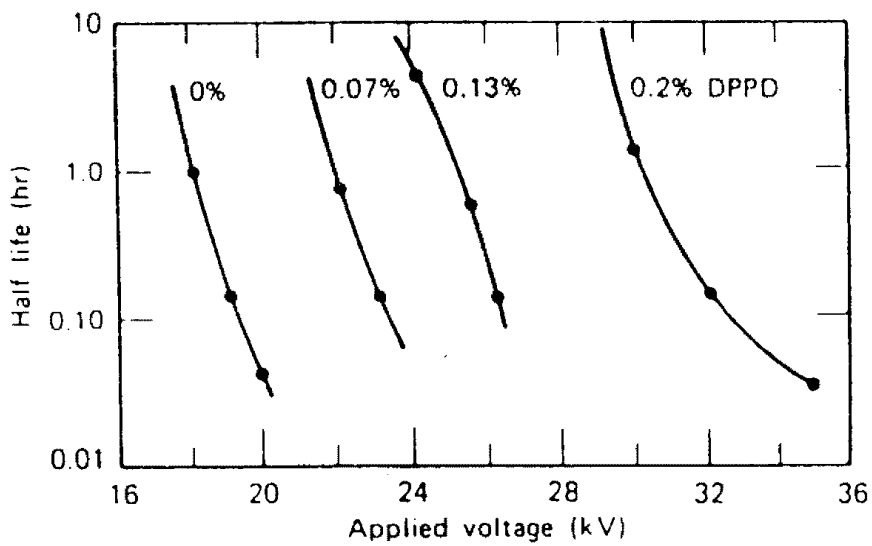
Fig. 2.   Voltage life curves of needle test polyethylene samples containing varying
amounts of DPPD staining anti–oxidant.
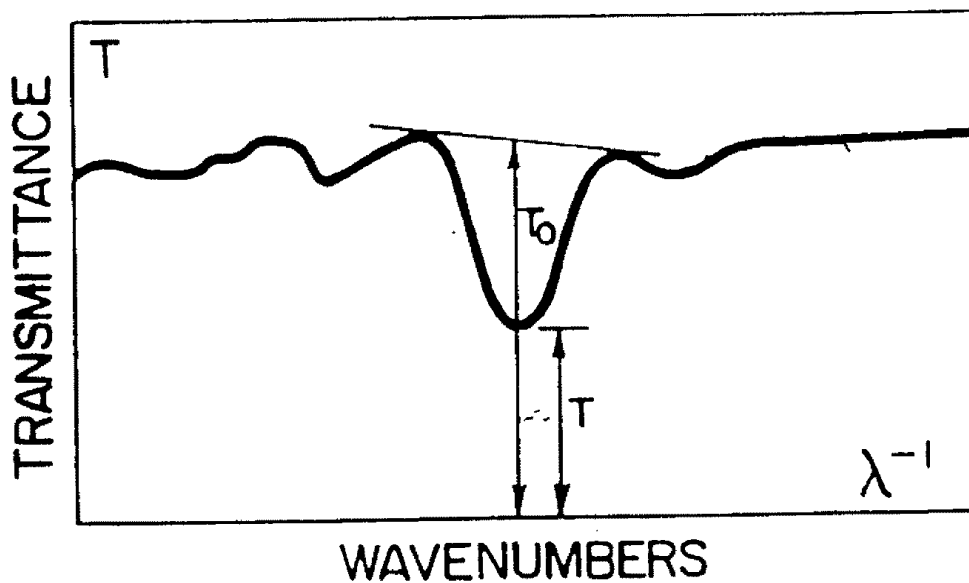
Fig. 3.   Definition of T and T_0 for determining coefficient of absorption.
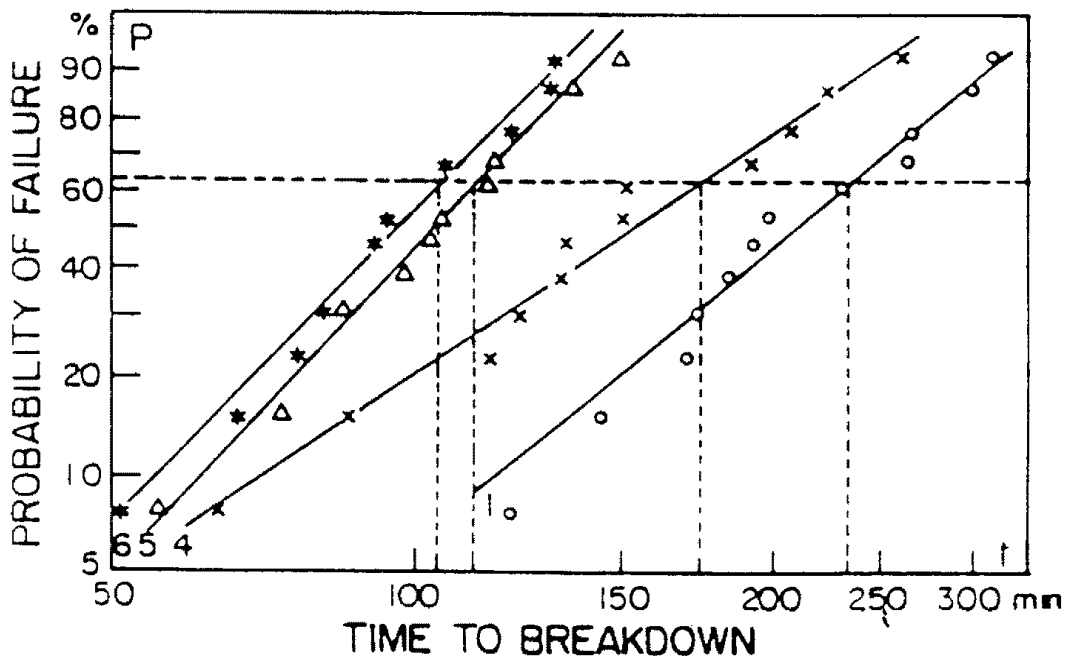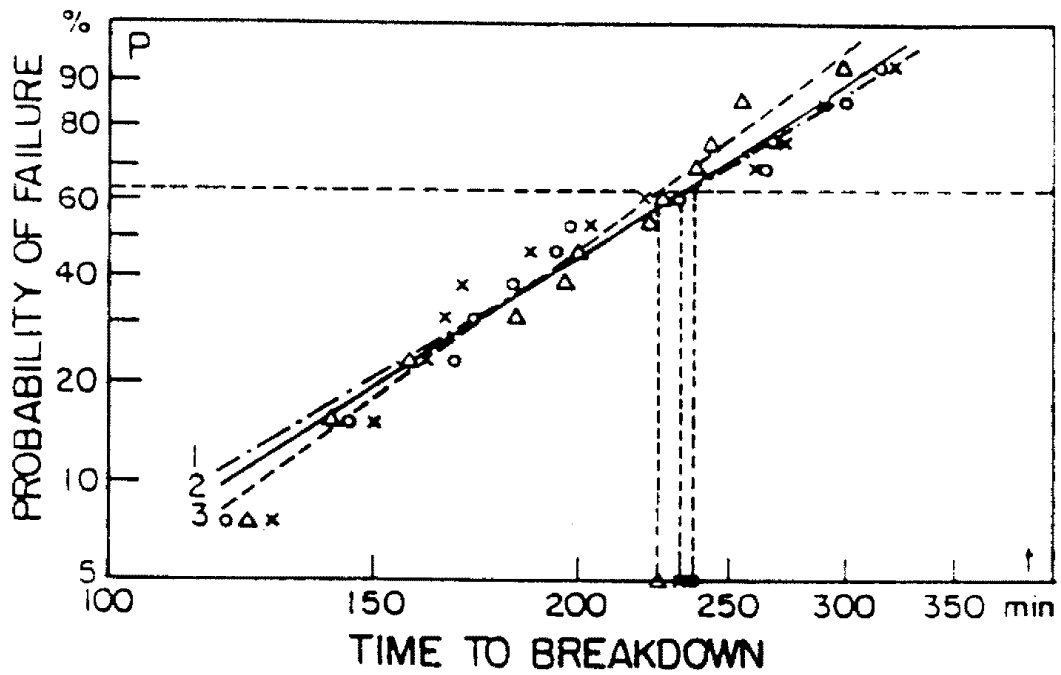
Fig. 4.   Weibull function distributions of time to breakdown of polyethylene
subjected to the needle test:

1 – unaged samples, 2 – aged by alternating voltage,
3 – aged by direct voltage, 4 – aged thermally in air,
5 – aged thermal-chemically (5% NaCl) solution,
6 – voltage-water aged.
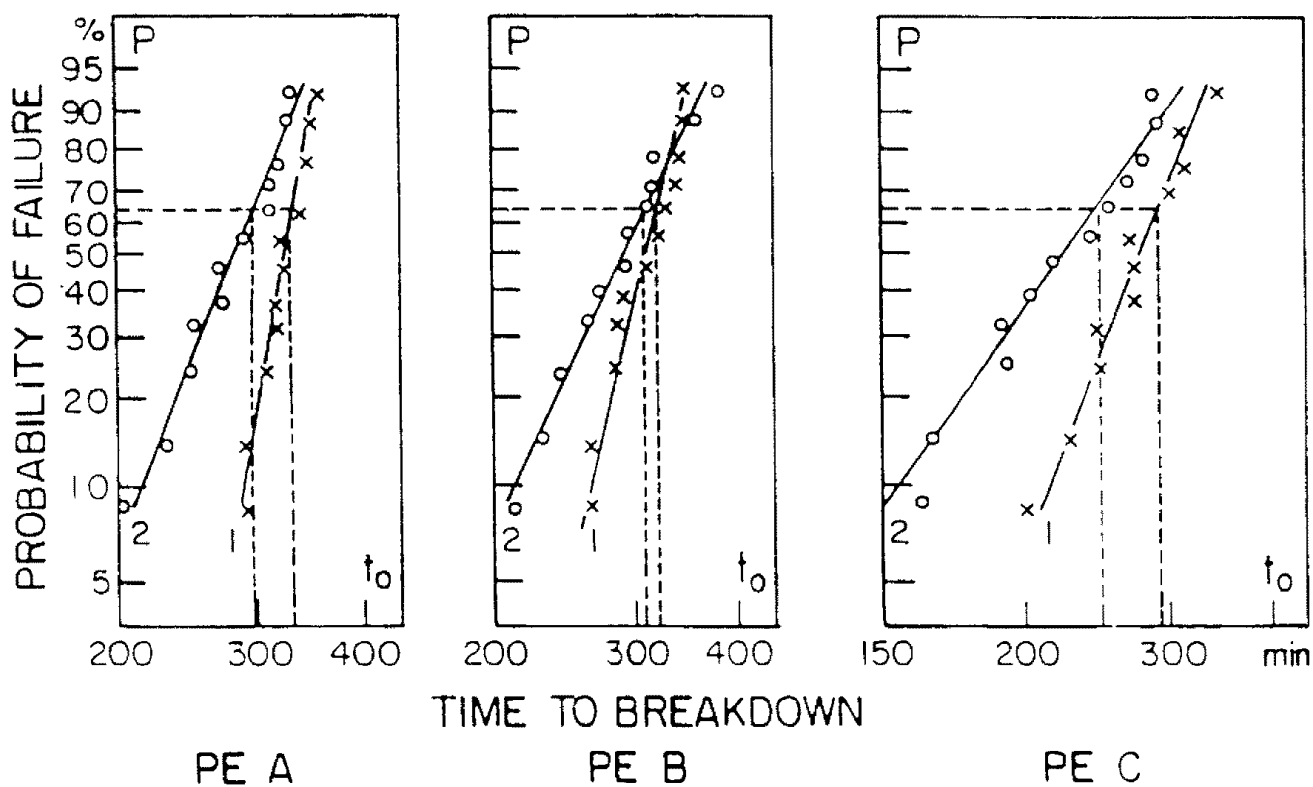
Fig. 5. Distribution functions of time to breakdown of polyethylene A (PE A), B (PE B), and C (PE C): 1 – unaged samples, 2 – voltage-water aged for 168 h.
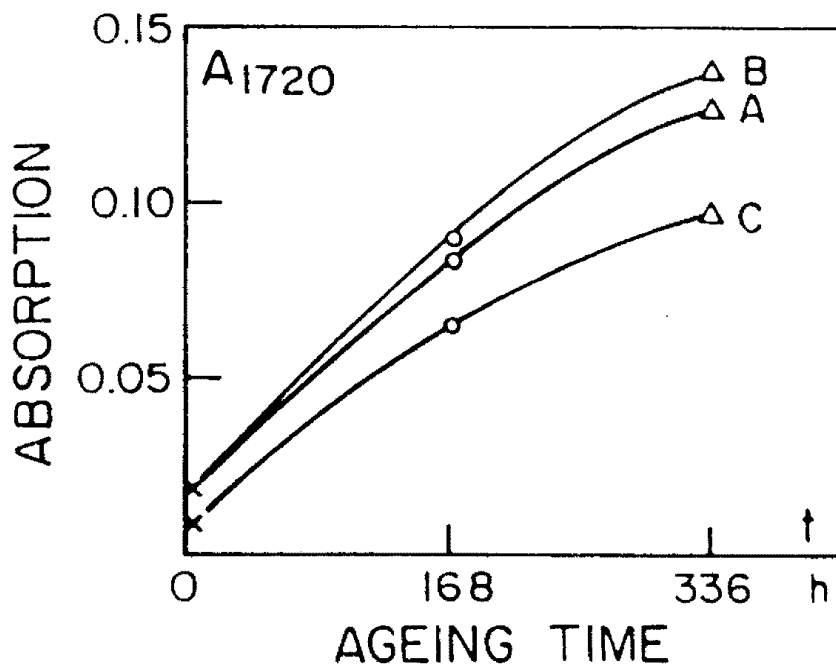
Fig. 6. Infrared absorption of the 1720 cm^{-1} band, as a function of aging time for A, B, C types of PE, voltage-water aged.
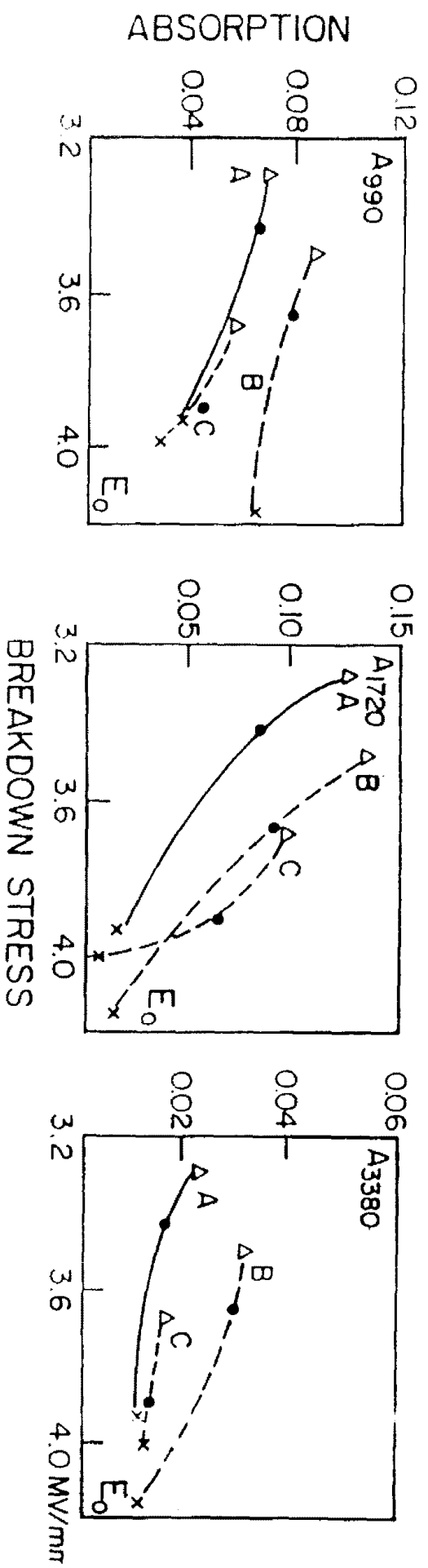
ABSORPTION

Fig. 7. Dependence of the 3380, 1720 and 990 cm⁻¹ bands absorption and electric stress at breakdown for
A, B, C types (see Fig. 3) of PE determined by needle test. Unaged samples (x), samples treated
with electric stress and water for 168 (•) and 336 (Δ) h.

BREAKDOWN STRESS
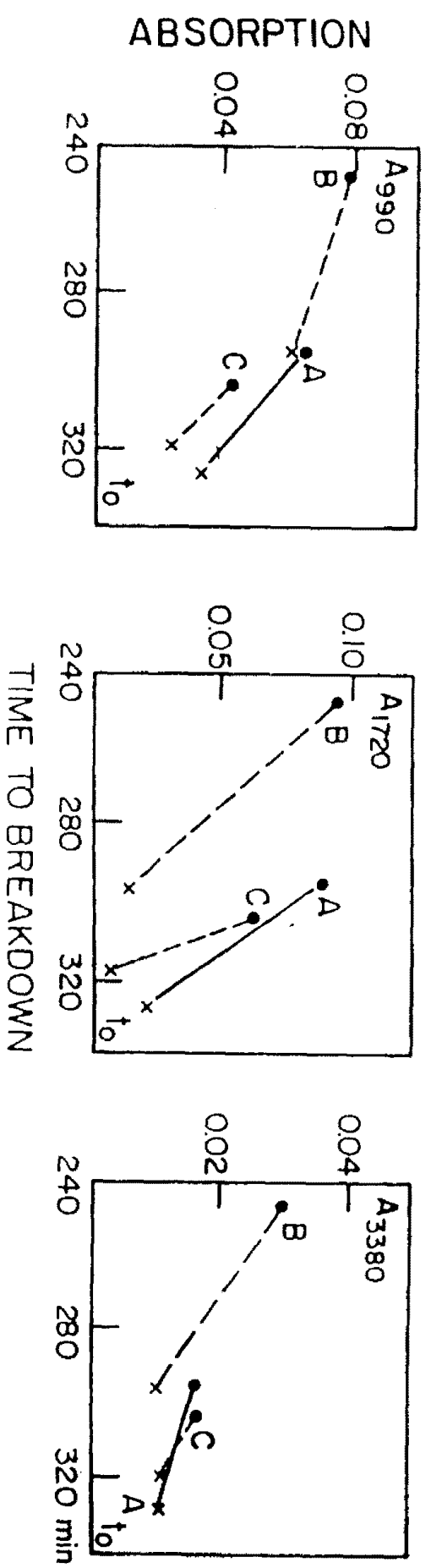
ABSORPTION

Fig. 8. Absorption of the 3380, 1720 and 990 cm⁻¹ bands as a function of time to breakdown for A, B, C
types of PE.

TIME TO BREAKDOWN

— 19 —
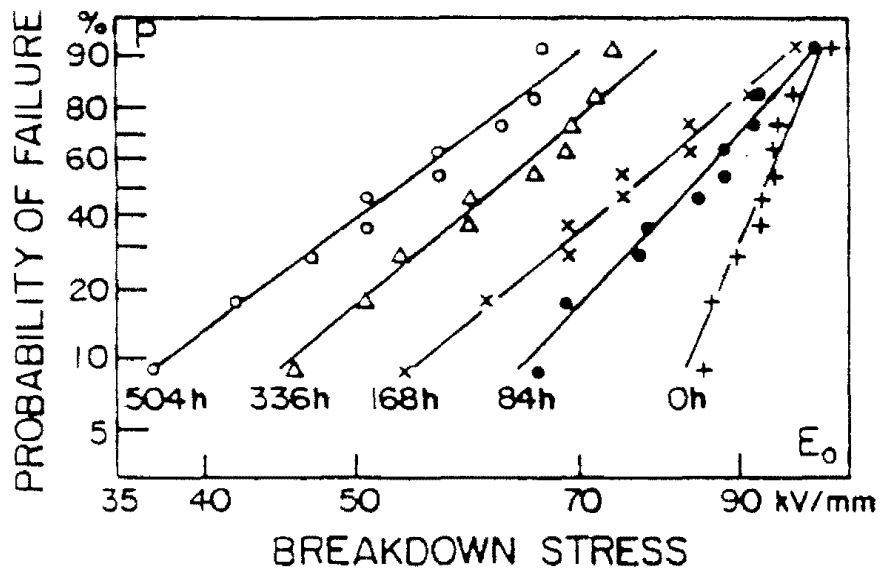
Fig. 9. Distribution functions of breakdown stress of foil,
aged for different time by UV radiation.
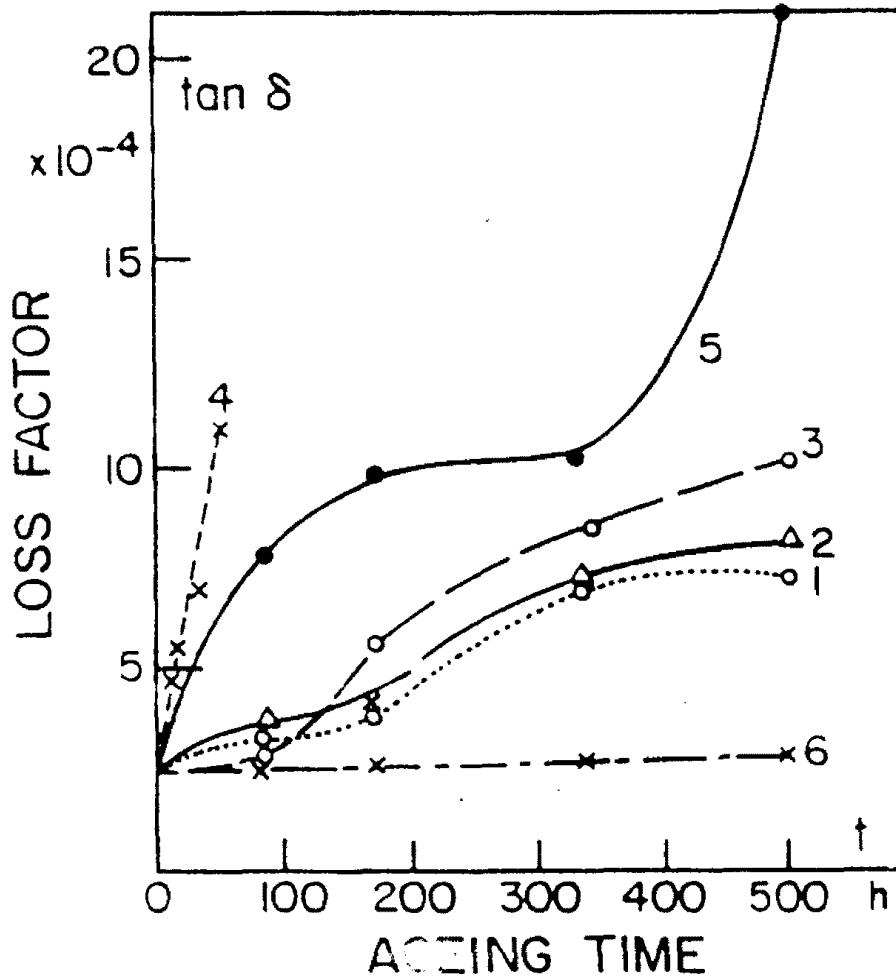
Fig. 10. Relation between the dielectric loss factor
and aging time of the PE foil: 1 – thermal–water
aging, 2 – thermal–chemical aging, 3 – voltage–
water aging, 4 – partial discharge aging, 5 – UV
radiation aging, 6 – aged under normal conditions.
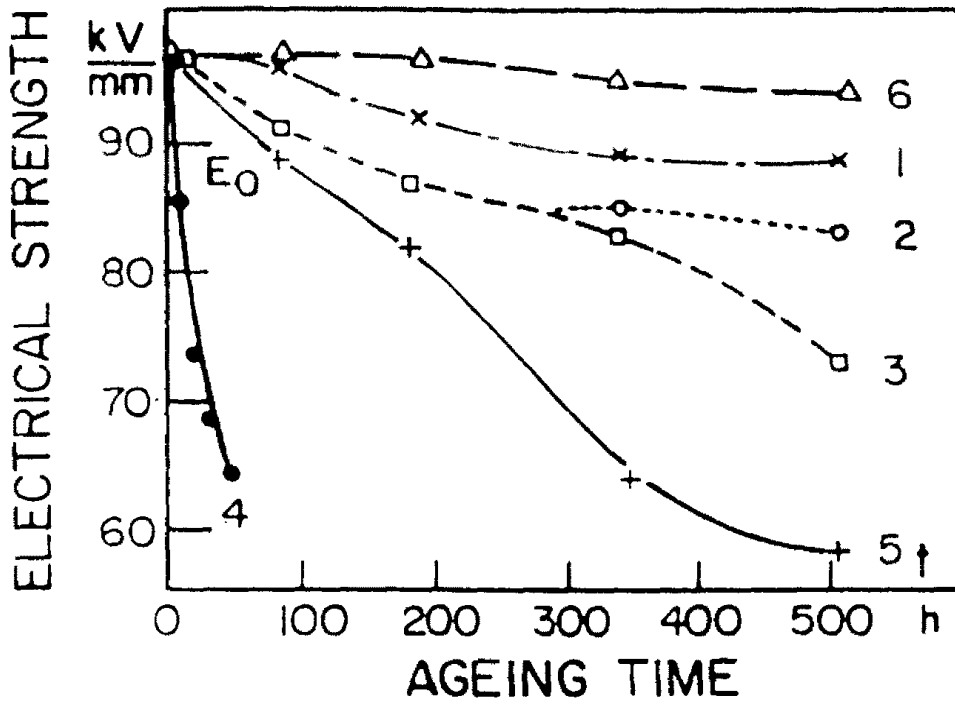
Fig. 11. Relation between dielectric strength of PE foil and
aging time for different types of aging:
1 – thermal–water, 2 – thermal–chemical,
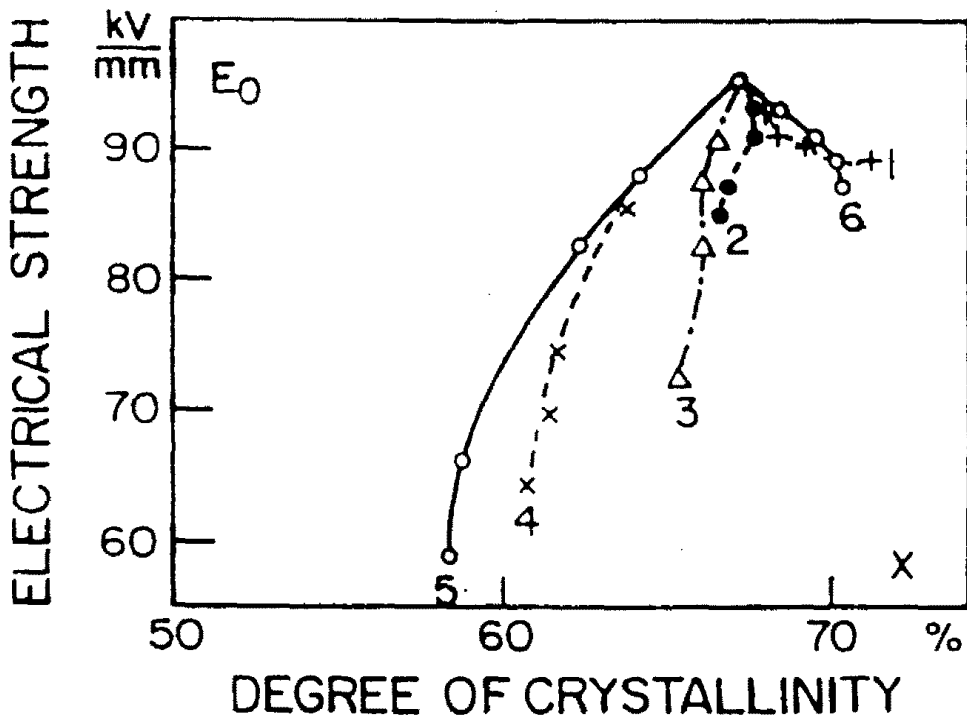3 – voltage–water, 4 – partial discharges aged,
5 – UV radiation, 6 – under normal conditions.
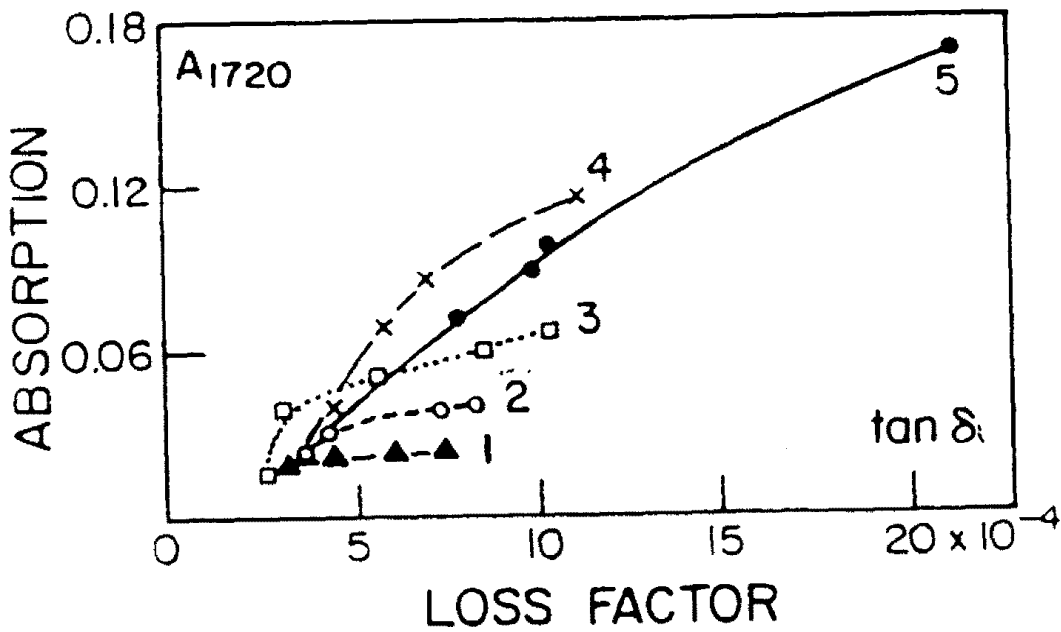
Fig. 12. Relation between absorption of the carbonyl bands
and the dielectric loss factor of PE foil for dif-
ferent types of aging: 1 – thermal–water, 2 – thermal–
chemical, 3 – voltage–water, 4 – partial discharges,
5 – UV radiation
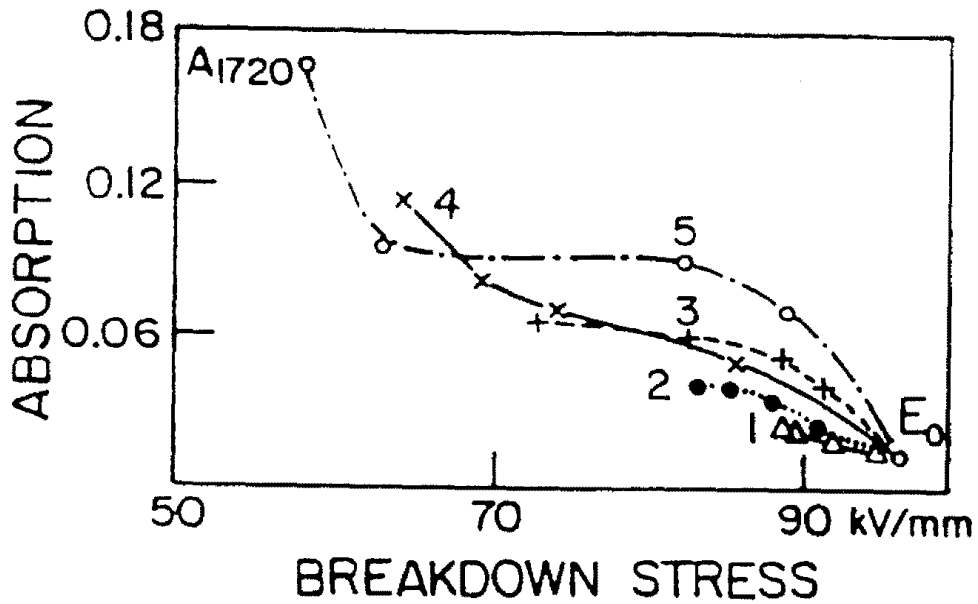
Fig. 13. Relation between absorption of the carbonyl bands and the electric strength of PE foil for different types of aging:
1 – thermal-water, 2 – thermal-chemical, 3 – voltage-water, 4 – partial discharges, 5 – UV radiation

Fig. 14. Relation between electric strength and degree of crystallinity of PE foil for different types of aging: 1 – thermal-water, 2 – thermal-chemical, 3 – voltage-water, 4 – partial discharges, 5 – UV radiation, 6 – under normal conditions

Fig. 15. Morphology structure of polyethylene cable insulation aged at elevated temperature with enlarged regions of amorphous phase (arrows are showing regions of amorphous phase)
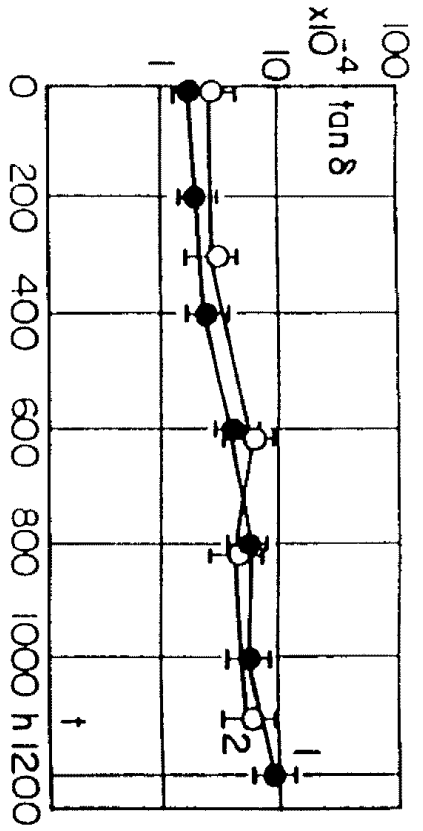

Fig. 16. Dielectric loss factor as function of thermal overload time
1 – polyethylene type 1
2 – polyethylene type 2

Fig. 17. Crystallinity degree as function of thermal overload time
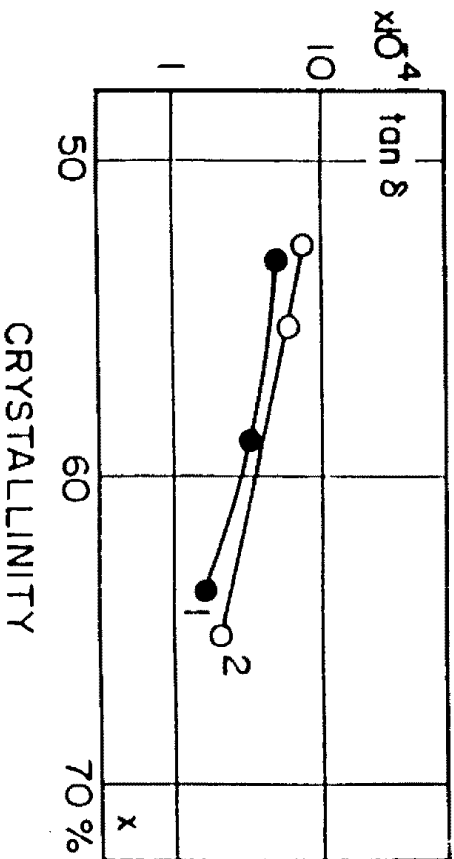1 – polyethylene type 1
2 – polyethylene type 2

Fig. 18. Dependence of dielectric loss factor upon the crystallinity degree
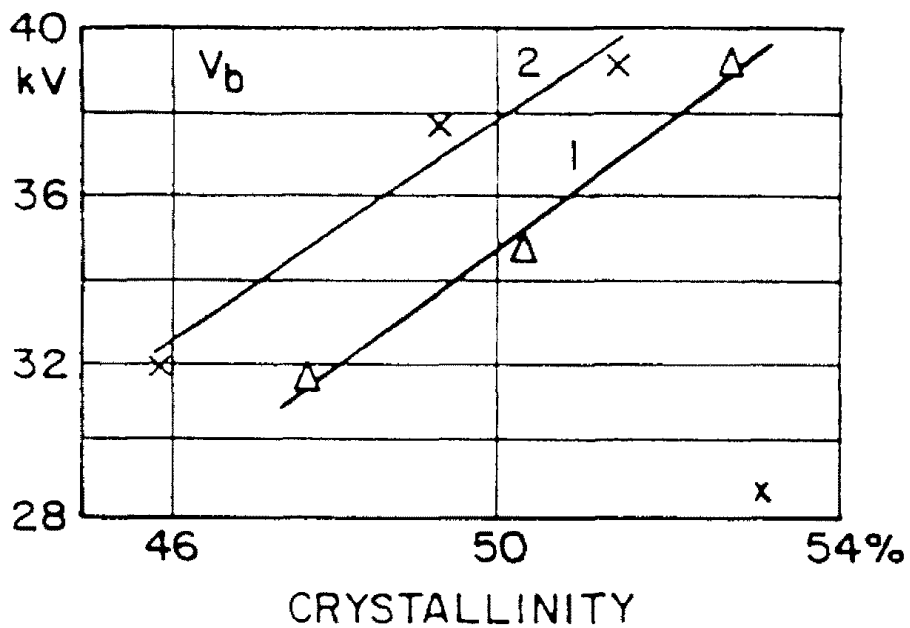1 – polyethylene type 1
2 – polyethylene type 2

Fig. 19. Variation of dielectric strength with degree of crystallinity
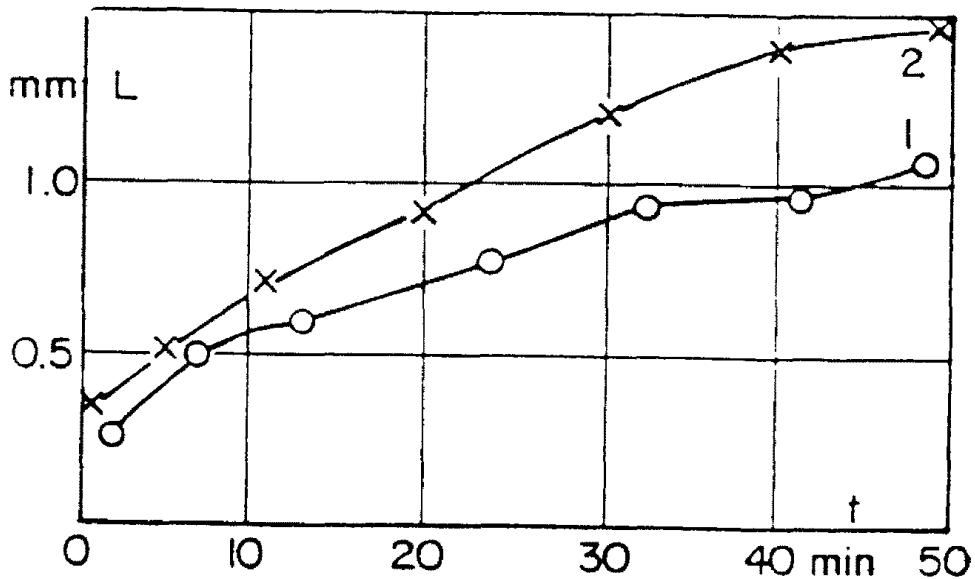 1 - thermal aged at 90°C
  2 - thermal aged at 80°C

Fig. 20. Length of tree propagation as function of voltage application time
  1 - normal thermoplastic polyethylene
  2 - thermoplastic polyethylene with large amount of amorphous phase

Fig. 21. Three channel path at boundary of spherulites in thermoplastic polyethylene cable insulation

Fig. 22. Tree channels in spaces between the spherulites in polyethylene of 60-70% crystallinity degree