

## THE PRESSURE WAVE PROPAGATION METHOD FOR THE ANALYSIS OF INSULATING MATERIALS: APPLICATION TO LDPE USED IN HV CABLES

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### ABSTRACT

In the field of electrical insulation, polymers are progressively replacing formerly used materials. When such an insulator is subjected to a voltage gradient, an internal space charge and a polarization develop, eventually reducing the apparent electrical strength of the material. We have applied the pressure wave propagation (PWP) method to the study of HV insulators, such as low density polyethylene (LDPE) used for HV cables. The spatial charge distribution is successively measured in samples subjected to various processes. The influence of additives in the development of a space charge under electric stress is directly shown. We have also observed that the nature of the electrode-insulator interface strongly affects the charge distribution throughout the insulator, and that charge injection from these interfaces occurs at much lower applied voltage gradients than previously assumed. The evolution in time of the charge distribution in samples submitted to voltage gradients, at 20 and 70°C, is studied. These results show that an equilibrium is reached, from which the life expectancy of materials and cables can be evaluated.

### INTRODUCTION

Synthetic materials are more and more used in the field of electrical insulation. Polymers are progressively replacing formerly used materials such as impregnated paper. Among them, polyethylene resins are of particular importance as regards HV cables or insulators for electrical apparatus. Although these materials are already widely used in ac application, various problems occur when they are subjected to dc voltage gradients; many are still unresolved.

When an insulator is submitted to a HV gradient, several phenomena may occur: internal space charges move under the influence of the applied electric field, dipoles are partly oriented along the direction of this field, ionic dissociation can take place and, depending on the nature of the electrode-insulator interfaces, charges can be transferred at these interfaces. In some regions, the resulting electric field may exceed the applied one, so that the apparent electrical strength can be reduced.

It is clear that a better understanding of the interface phenomena and of the transport properties of insulating materials would be of great interest for the analysis of these effects. Some years ago, a non

ambiguous method appeared to be the direct measurement of the space charge and polarization distributions within the samples. It can be made using various experimental techniques [1] and particularly the pressure-wave-propagation (PWP) method, which has already been applied to various types of electrets. This method is nondestructive and is very suitable to study the evolution of the charge distribution within a sample submitted to various physical treatments [2]. Its application to the analysis of the processes occurring in HV insulators under electric stress is of great interest.

First, the influence of the chemical formulation of the materials on their transport properties can be directly studied. This can help to define basic materials to be used for the manufacture of HV cables.

Second, the phenomena related to the conductor-insulator interfaces can be analyzed by comparing the charge distribution obtained after polarizing samples with various electrodes.

Third, evolution with time of the internal field in structures under stress may allow predictions on the life expectancy of materials and cables.

In this paper, we present an investigation of the above mentioned phenomena in low density polyethylene (LDPE), using laser-induced pressure waves.

### PRINCIPLE OF THE PWP METHOD

The principle of the PWP method [3-5] can be described as follows: consider a sample, in a plane-parallel geometry, with contacting electrodes on both surfaces. At time  $t=0$ , a short risetime pressure wave is applied uniformly to one of its surfaces. The wavefront penetrates into the sample and propagates with the velocity of sound  $v$ . The resulting displacement of the space charges contained in the compressed region produces a variation of the induced charges on the electrodes, which depends on the space charge distribution and on the pressure profile  $p(z, t)$ .

It has been shown that if the sample is kept in open-circuit conditions, the voltage appearing between the electrodes during the propagation of the wavefront through the sample is given by:

$$V(t) = \chi G(\epsilon_r) \int_0^{z_f} E(z, 0) p(z, t) dz \quad (1)$$

where  $\chi$  is the compressibility of the material,  $z_f=vt$  is the position of the wavefront, and  $E(z, 0)$  is the electric field distribution before the penetration of the wave,  $G(\epsilon_r)$  is a function of the relative permittivity  $\epsilon_r$  containing the dependence of  $\epsilon_r$  with pressure. If the sample is short-circuited, the current flowing in the external circuit is given by:

$$I(t) = \chi C_0 G(\epsilon_r) \int_0^{z_f} E(z, 0) \frac{\partial}{\partial t} p(z, t) dz \quad (2)$$

where  $C_0$  is the unperturbed sample capacitance.

Therefore, if the profile of the pressure wave  $p(z, t)$  is known, the electric field distribution in the sample can be derived from the time variations of the open-circuit voltage or short-circuit current during the propagation of the wave.

The details of the numerical analysis leading to the electric field distribution are given in [6]. In order to obtain the charge distribution using Poisson's equation, the calculated values of the electric field are smoothed before derivation.

When the pressure pulse duration is short as compared to the transit time through the sample, the short-circuit current measured during the propagation gives directly the charge distribution [7]. This is not valid during the penetration of the pulse in the sample. Moreover, this qualitative approach, which is very useful when dealing with thin polymer films, is not applicable to thick dielectric materials such as those involved in HV insulation.

Indeed, in this case the insulators used are generally a few mm thick and the pressure wave is distorted during its propagation, because of the imperfect elastic properties of the material.

The importance of this effect in thick LDPE samples is illustrated in the following, and the distortion of the pressure pulse resulting from attenuation and dispersion effects can be taken into account [6].

### EXPERIMENTAL

The samples which were prepared and supplied by the Silec Company, were made of LDPE used for HV cables, either "pure" or containing 0.06% wt diphenyl paraphenylene diamine (DPPD), a commonly used additive. These samples were 2 mm thick plates. One side was covered with a "semiconducting" electrode similar to that used in HV cable technology. Such an electrode is made by doping the LDPE material with increasing graphite concentration. On the other side, three types of electrodes were used. Either thin films made of 5 nm of chromium and 30 nm of gold were vacuum deposited, or a thin layer of conductive silicone grease was painted on the surface. In a third situation, the second electrode was also a semiconducting one, so that the sample had a symmetrical geometry.

The samples were polarized by applying HV between the electrodes, during increasing periods of time, either at room temperature or at 70°C. The following results were obtained for voltage gradients ranging from 1.5 to 45 kV/mm.

After each period the charge distribution was measured using the PWP method. In the experiments performed at 70°C, the samples were cooled down to room temperature before testing. In some cases, the discharge in short-circuit conditions at 70°C was also studied.

A description of the experimental set up used to measure field and charge distributions can be found elsewhere [1,6].

In order to create a short rise time pressure wave, the semiconducting electrode was exposed to short duration (3 ns) laser pulses, emitted by a Nd/YAG laser. The pressure generated at the surface of the electrode propagates through it and is efficiently transmitted to the sample because of the good acoustic impedance match between the various layers.

In order to determine the deformation of the pressure wave during its propagation through the sample, the pressure profiles transmitted at the rear surface of the front electrode, and after propagation through LDPE samples of increasing thicknesses, were measured using a piezoelectric quartz transducer. The amplitude of the pressure waves transmitted through 1 and 2 mm thick samples are plotted on Fig. 1 on a scale relative to the peak value of the entering pressure pulse. After propagation through 2 mm, the rise time of the wave increases from 25 to 70 ns, whereas the peak amplitude is reduced in a 2/5 ratio. This evolution of the pressure profile has been taken into account as indicated in Ref. [6] and the corresponding spatial resolution is 50  $\mu$ m for a 2 mm thick sample. From an extrapolation of these results, we conclude that the PWP method can be applied to samples as thick as 10 mm, with a spatial resolution of the order of 0.3 mm. If necessary, this resolution could be largely improved by lowering the temperature during the measurement of the field distribution, thus reducing the attenuation and dispersion of the pressure wave.

### RESULTS AND DISCUSSION

#### *Presentation of the results*

Each type of physical treatment was applied to several samples, which showed similar behavior with some minor differences.

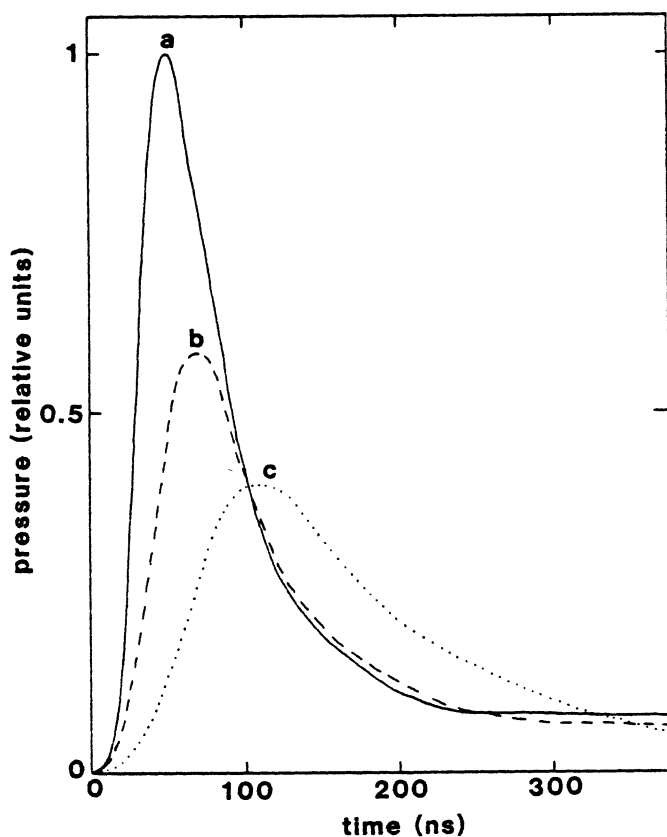


Fig. 1: pressure profiles plotted on a logarithmic scale (a) entering a sample, (b) after propagation through a 1 mm thick LDPE plate, and (c) after propagation through a 2 mm thick LDPE plate.

When an evolution in time is studied, at each step the open-circuit voltage and short-circuit current are measured during the propagation of a pressure pulse through the sample. The observation of the short-circuit current gives a qualitative knowledge of the charge distribution except during the penetration of the pressure pulse, that is to say, during the first 200 ns. Such results are shown on Figs. 4a and 5.

The open-circuit conditions are used, together with the pressure profiles shown in Fig. 1, to calculate the electric field and charge distribution from Eq. (1). Spatial charge distributions obtained in this way are shown on Figs. 2, 3, 4b and 6. On all Figures the polarity of the applied field is indicated by the position of the positive electrode.

#### *Influence of the chemical composition of the material*

A comparison has been made of the charge distributions found in samples made of either pure or DPPD doped LDPE, after application of a voltage gradient under the same conditions. The samples had a semiconducting electrode on one side and a conductive silicone grease on the other side. A 45 kV/mm voltage gradient was applied, with the positive polarity on the semiconducting electrode, either during 72 h at 20°C, or during 16 h at 70°C. These durations were chosen so as to reach an equilibrium.

Then the remaining charge distributions were measured and are shown on Fig. 2a and 2b.

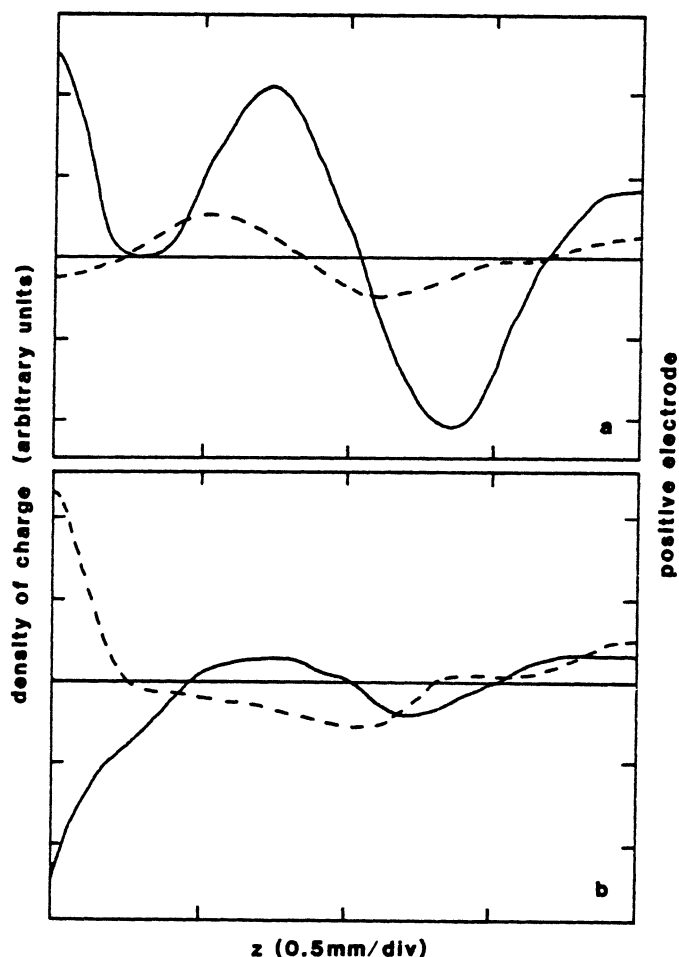


Fig. 2: Comparison between the charge distribution obtained in pure LDPE (full line) and in DPPD doped LDPE (dotted line), after application of 45 kV/mm stress (a) during 16 h at 70°C, (b) during 72 h at 20°C.

Strong charge injection from the semiconducting electrode and internal charge migration or dipole orientation in the bulk can be observed on all samples. Charge injection from the conductive silicone grease negative electrode is also present in some samples. The most striking feature is the difference in the amplitudes of the charge densities and corresponding field distributions, in pure and doped materials, especially at elevated temperature.

For pure LDPE, the maximum value of the electric field reaches 5% of the previously applied field at 20°C, and is as large as 30% of this field at 70°C. In some regions of the samples, this field has the same polarity as the applied one, so that the resulting field under stress exceeds the applied field, reducing the apparent electrical strength of the material. For DPPD doped samples, the maximum value of the field does not exceed 5% of the applied one, at both temperatures.

Consequently this very direct observation shows that this additive, which is primarily used as an antioxidant agent, also improves the electrical strength of LDPE, by preventing the accumulation of large charge densities in the material under stress.

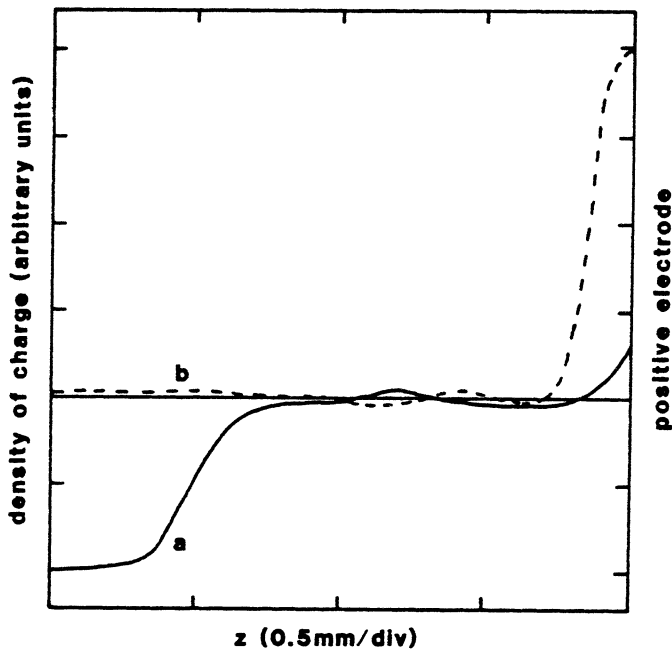


Fig. 3: Comparison between the stabilized charge distributions obtained in two pure LDPE samples with different electrodes, after application of 5 kV/mm stress, during 100 h, at 20°C. (a) evaporated electrode, (b) conductive grease electrode.

The same analysis applied to materials of various chemical compositions will permit to define materials with improved HV insulation performances.

#### Conductor-insulator interfaces

In order to study the influence of the conductor-insulator interfaces, we have observed the evolution in time of the charge distributions in pure LDPE samples submitted to a voltage gradient, for the three types of interfaces described above.

We show on Fig. 3 the stabilized charge distributions in two samples in which the rear electrode was either an evaporated electrode or a conductive silicone grease electrode, after application of a 5 kV/mm voltage gradient during 100 h, at 20°C.

On Fig. 4, the evolution in time of the charge distribution in a sample polarized at 70°C, with a 5 kV/mm applied voltage gradient, is presented. The rear electrode in this case was an evaporated electrode. From these data, and from other data not presented here, the following observations were made.

Charge injection from the electrodes occurs even for low applied electric fields (1.5 kV/mm), for the three types of electrode-insulator interfaces, at 20°C and at 70°C. At room temperature, this process is the dominant one, since charges are present only near the electrodes, with the same polarity as that of the adjacent electrode.

The injection depends strongly on the nature of the interfaces, as illustrated on Fig. 3.

The behavior of one interface depends also on the polarity of the applied field.

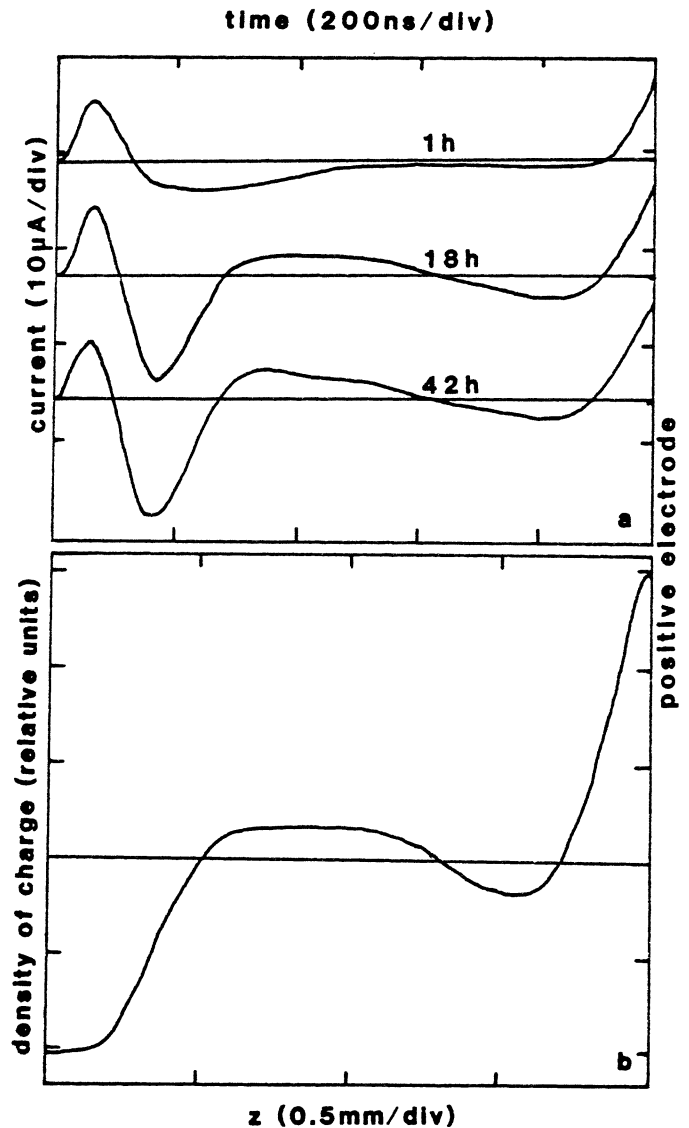


Fig. 4: (a) Evolution of the charge distribution in a pure LDPE sample after application of a 5 kV/mm stress, at 70°C. The rear electrode is an evaporated Cr-Au electrode. (b) Calculated charge distribution corresponding to a polarization time of 42 h.

The nature of both interfaces influences the equilibrium charge distribution not only near the electrodes, but also throughout the sample.

As shown on Fig. 4, charge injection is already present at 70°C after 1 h, at 5 kV/mm. The same observation was made for a field as low as 1.5 kV/mm. After a polarization time of a few hours, internal charge migration or dipole orientation is also observed in the bulk of the sample. This can be explained by the increased mobility of charges at elevated temperature and by an increase of charge transfer at the interfaces. It can be noticed that the internal migration of charges enhances the field near the interface and tends to increase the injection process, whereas the injected charges have the reverse effect, leading to a stabilization.

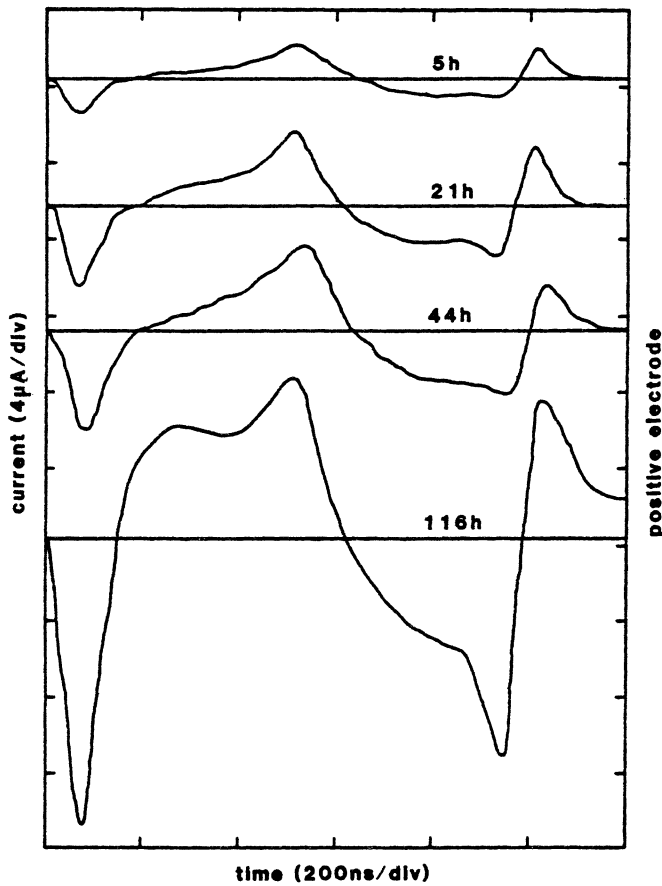


Fig. 5: Evolution in time of the charge distribution, as represented by the short-circuit current, in a sample submitted to a voltage gradient of 10 kV/mm, at 20°C. The transit time through the sample is 1 µs.

Evolution in time of an insulating structure

Because many parameters are involved in the build-up of a charge distribution in an insulator under stress, further experiments were made in a very simple structure, as close as possible to that of a HV cable. The samples were made of pure LDPE, and had identical semi-conducting electrodes on both surfaces. This symmetrical geometry enables the measurement of the charge distribution for the two opposite directions of propagation of the pressure wave. These two measurements made on the same sample are in very good agreement and they lead to a more accurate determination of the charge distribution near the electrodes.

The evolution in time of the charge distribution, indicated by the short-circuit current, in a sample submitted to a voltage gradient of 10 kV/mm, at 20°C, is shown on Fig. 5. The calculated charge distribution corresponding to a polarization time of 116 h is given by curve C of Fig. 6.

It can be seen that a charge distribution which remains when the applied field is turned off is already present after a few hours. The same observation was made for a field as low as 1.5 kV/mm. Even for such short times, charge injection from the semiconducting electrodes and charge migration or dipole orientation in the bulk of the sample are observed.

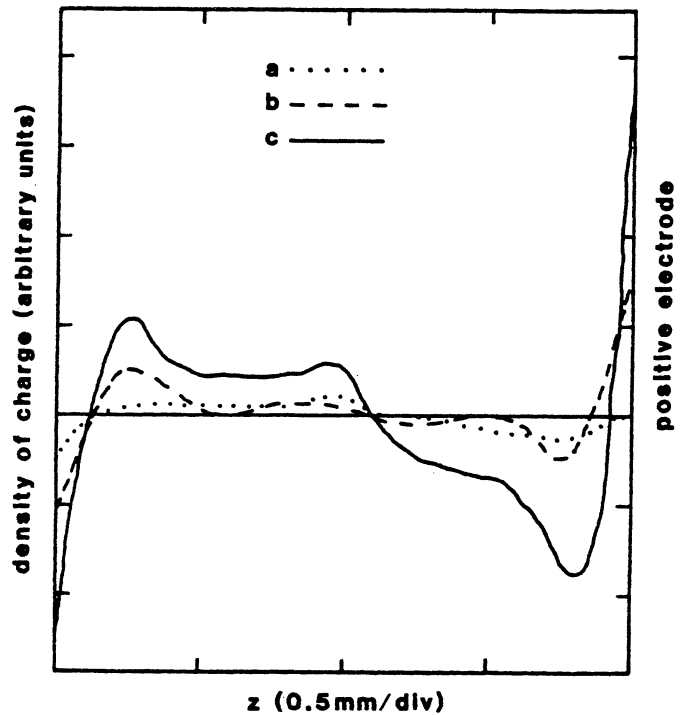


Fig. 6: Comparison between the stabilized charge distributions obtained under various electrical stresses, at 20°C, in pure LDPE samples. (a) 5 kV/mm, (b) 7.5 kV/mm, (c) 10 kV/mm.

For long polarization times, a stabilization of the charge distribution occurs. On Fig. 6, the stabilized distributions corresponding to 5, 7.5, and 10 kV/mm applied fields, at room temperature, are given by curves a, b, and c respectively. The time after which a stabilization is observed decreases as the applied field increases. For instance, this time is approximately equal to 200 h at 5 kV/mm, 150 h at 7.5 kV/mm and 110 h at 10 kV/mm.

The distribution of the electric field in the samples, after polarization, was also calculated. The maximum value of this field was found to be 100 V/mm for an applied voltage gradient of 5 kV/mm and 600 V/mm at 10 kV/mm, which corresponds respectively to 2 and 6% of the applied field.

Similar experiments made at 70°C show that the maximum value of the field associated with the equilibrium is in the range of 20 to 40% of the applied field, and is reached after 40 h.

It is interesting to note that the same results were obtained in pure LDPE polarized at 45 kV/mm, with one of the electrodes made of conductive silicone grease.

This indicates that for a given material, the maximum value of the electric field is essentially governed by the applied voltage gradient and the temperature.

The above results show that in some regions of the insulator the resulting field may exceed the applied field, by as much as 140% at 70°C, or 105% at 20°C. Consequently, the electrical strength of the insulator can be significantly reduced in applications in which it is submitted to a HV gradient during long times. Thus the observation of the evolution of the electric field in an

insulating structure under stress gives an estimation of its life expectancy.

#### CONCLUSION

In this paper, we have shown that the PWP method can be used successfully for the analysis of insulating materials in structures similar to those encountered in various electrotechnical applications. Taking into account the deformation of the pressure wave during its propagation, the method can be applied to dielectric samples of thicknesses comparable to those used in HV cables.

The direct observation of the processes which take place under electrical stress show that the method will help to define such parameters as the composition of the chemical content of the materials involved, the nature of the electrodes to be used, taking into account the effect of the associated interfaces.

After application of a voltage gradient during a long time, we have observed that the field resulting from the generation of a charge distribution in the insulator may exceed the applied field significantly, reducing the apparent electrical strength.

As a diagnostic tool, the PWP method may allow predictions of the lifetime of materials under stress and eventually allow in-situ measurements for diagnostic purposes.

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