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BREAKDOWN STRENGTH OF IMPREGNATED POLYPROPYLENE FILMS AGED UNDER HIGH A.C. FILEDS

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Abstract—Electrical breakdown measurements were performed on polypropylene films impregnated with benzyltoluene and aged for different electric field values (100 $V_{rms}\ \mu m^{-1} < E_a < 300\ V_{rms}\ \mu m^{-1}$). Two films of 13.6 μm thickness were sandwiched between two plane electrodes and immersed in the liquid at 80°C and with substantial 0_2 concentrations. For this arrangement, the threshold voltage discharge (V_{th}) was *ca* 6–7 kVrms. The results show that for a given ageing time the films breakdown strength dependence on the ageing voltage V_a is different for $V_a < V_{th}$ and $V_a > V_{th}$. This result is a confirmation of the occurrence of electrical discharges in the capacitor for $V_a > V_{th}$. The area of degradated film is also different in both cases. For $V_a < V_{th}$ the region with lower breakdown strength is restricted to the part of the film sandwiched between the electrodes. For $V_a > V_{th}$ the region with lower breakdown voltage is larger (about 1 mm) than the electrode diameter (8 mm). This is the result of the radial propagation of discharges or streamers. The influence of an epoxy additive which is an effective stabiliser of impregnated PP films was also investigated. © 1997 Published by Elsevier Science Ltd

INTRODUCTION

During service, insulation of power capacitors, which usually consists of rough polypropylene films impregnated with an aromatic hydrocarbon liquid [1], withstands severe electrical, mechanical and thermal stresses. Its ageing may cause premature breakdown of the capacitor [2], as a consequence of the electrical discharges occurring in the liquid which is affected by high electric fields. Physiochemical analysis of aged dielectrics has shown that the degradation of the material is strongly heterogeneous: its breakdown voltage is lowered in only some points of the film [3].

In the absence of discharges, an epoxy additive, such as diglycyl-ether of bisphenol A (DGEBA), enhances the life-time of industrial capacitors [4] and greatly slows down the decrease with time of the dielectric characteristics of PP films [5]. The presence of the additive does not change the discharge onset voltage, but it favours the appearance of electrical discharges of higher energy [6].

This present paper analyses the behaviour of benzyltoluene impregnated PP films subjected to electric discharges, in capacitor models filled with an O_2 saturated liquid in order to accelerate ageing. After a sample has been subjected to high voltage stress for a few hours, we analyse the degradation products and try to correlate their presence with the discharge phenomenon. In particular we investigate whether the sites near the field enhancement zones are those for which the concentrations of these products is enhanced. We also pay particular attention to the breakdown voltage by which we mean the voltage at which the dielectric material loses its insulating properties. The effect of an additive on the dielectric strength (epoxy) is examined systematically. In this work, we have measured the life-time of a capacitor and the breakdown strength of electrically aged films. Chemical degradation was evaluated using FTIR spectroscopy.

EXPERIMENTAL

PP films 13.5 μm thick, rugged on both sides and employed in this experiment were provided by Bollore Technologies; they contained 95% PP isotactic and were impregnated with benzyltoluene (BT), which was obtained after two distillations of a commercial blend (M/DBT), consisting mainly of mono-benzyltoluene (BT), dibenzyltoluene (DBT) and tribenzyltoluene (TBT) [7]. This blend was treated on activated earth (attapulgus clay) and then filtered with a 0.2 μm porosity filter. The pressure in the distillation column was about 100 Pa, the temperature at the upper part of the column was 90°C. The electrical characteristics of this liquid were: relative permittivity, 2.4; resistivity $2 \times 10^{12}\ \Omega$ m; water content, *ca* 50 ppm.

Ageing was performed in a capacitor made of polytetrafluoroethylene (PTFE) shell containing two cylindrical duraluminium electrodes. Two PP films were sandwiched between the electrodes. The electrodes, carefully polished in order to eliminate asperities as much as possible were 8 mm in diameter. The capacitance of the capacitor, when filled with liquid, was about 30 ± 5 pF. In the experiments described here, the impregnant was oxygen-saturated (relative pressure of oxygen over the liquid surface = 0.15 MPa). This capacitor was tested using an

a.c. field of 50 Hz at 80°C. For these conditions ageing was accelerated.

The electrical discharge detection was carried out with a multichannel analyser described in Ref. [8]. The system determined the number of the sine wave for which the discharge occurred and its position within the cycle (a cycle was divided into 250 sections). The discharge detector had a sensitivity of 0.1 pC and its resolution was 320 μS. The high voltage on the capacitor was increased in steps of 300 V. The acquisition of data lasted for 5 min for each value of V_a (applied voltage).

The experimental cell was subjected to a high alternating voltage V_a. For $V_a > V_{th}$, discharges were recorded with their amplitude and their repetition rare. The detection apparatus consists of a parallel RLC circuit with an amplifier so that the apparent charge which crosses the external circuit due to the occurrence of a partial discharge into the cell is measured. First V_{th} is picked up, then a relevant number of discharges are recorded with their amplitudes and their positions within the cycle. After that the distribution of the numbers and the mean amplitudes of the discharges vs phase angle are deduced.

The life-time of the capacitor was defined as the past time between the application of a constant electrical field and the breakdown of the film.

The dielectric strength of the PP film, extracted from the capacitor after electrical ageing was measured at different sites, under d.c. voltage ramping conditions (500 V/S), at 20°C. The voltage was applied to the film placed between a plate electrode and a stainless steel sphere, 2 mm in diameter. The electrodes and the film were immersed in BT to avoid discharges before the film broke down [9]. More than 50 measurements were performed on each film. They were classified into three groups. The first group comprises the values relative to points exterior to the test capacitor ("outer ring" zone) $R < x < 5R/2$, the two other groups contain the values relative to points between the electrodes, one group consisting of the points situated at distances $R/2 < x < R$ ("outer ring" zone) and the other one of those points with $0 < x < R/2$ (central zone) with respect to the axis of the parallel-plate capacitor (R is the electrode radius) (Fig. 1). The average breakdown voltage is obtained by averaging the experimental results of the breakdown voltage.

The aged films were analysed using a Nicolet Fourier Transform Infrared spectrometer in combination with an I.R. Plan Spectra-Tech microscope in different zones.

RESULTS

On-set voltage of electrical discharges

Discharges in capacitor. Figure 2 shows the number of discharges in each of the 250 sections into which a single established sine wave was divided. The capacitor was filled with benzyltoluene (BT), for 6.2, 6.5 and 6.8 kV_{rms}, 50 Hz. No discharges were detected up to about 6.2 kV_{rms} (Fig. 2A). During the positive half cycle, for 6.5 kV_{rms} applied voltage, (Fig. 2B), a few discharges were recorded within the same section. These few positive discharges were imposed with external interface. During the positive and negative half cycle, for 6.8 kV_{rms} applied voltage (Fig. 2C), positive and negative discharges were detected within the same section. The discharges were detected again only when the applied voltage was raised above 6.5 kV_{rms}.

In the liquid, each discharge is composed of a first discharge followed by a lot of smaller discharges [10]. The first large discharge creates a small bubble near the edge of the electrode [11]. This first discharge gives rise to an electronic avalanche in the liquid, while the subsequent pulses originate from discharges in the gas bubble.

Applied voltage dependence of the breakdown voltage of electrically aged PP films. The breakdown voltage is shown in Fig. 3 of films aged for 4 hr under different electrical stresses at 80°C and 1.5 bars of O_2, in the capacitor. The line is a guide for the eye. For the samples subjected to voltage $V_a < 6$ kV_{rms}, no discharges were detected. The average value of the breakdown voltage V_b was found to be constant in the "outer ring" zone. For the "inner ring" zone it decreases linearly with increasing V_a. For an applied voltage $V_a > 6.5$ kV_{rms}, both positive and negative discharges were measured and the average breakdown voltage decreases steeply. The average breakdown voltage decreases significantly in the zones affected by the electric field and within a circular ring of 1 mm from the edges of the electrode system (electrode diameter 8 mm). This is the result of the radial propagation of discharges or streamers.

Degradation of PP film: FTIR analysis

A typical FTIR spectrum of a sample that was aged for 4 hr at 7.5 kV_{rms} ($E = 228$ kV_{rms}) is shown in Fig. 4. We observed new absorption peaks only in the electrically stressed part. The absorption peaks at 1647 cm^{-1} and 1260 cm^{-1} can be attributed to a benzophenone based product which results from the oxidation of BT [12]. This shows that the liquid is highly oxidized. The peak at 1076 cm^{-1} has already been observed in Ref. [9] and it was attributed to the disproportionation products of P$^{\cdot}$ radicals [13]. The band at 1018 cm^{-1} has been observed with other impregnation liquids [12]. We attribute this band to a cyclic compound.

The initiation step of the film degradation is essentially the reaction of free R$^{\cdot}$ radicals which extract a hydrogen atom from a tertiary carbon of the PPH macromolecule.

$$PP + R^{\cdot} \text{ ------> } PP^{\cdot} + RH.$$

We assure that these R$^{\cdot}$ radicals are formed by electric discharges. The origin of these R$^{\cdot}$ radicals is very probably the impregnant, or same major

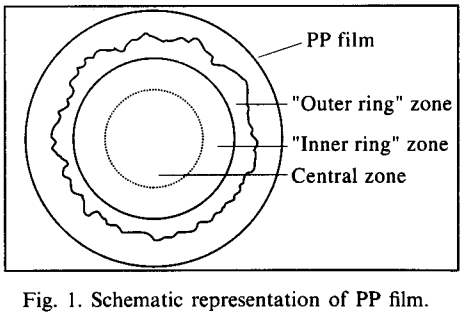


Fig. 1. Schematic representation of PP film.

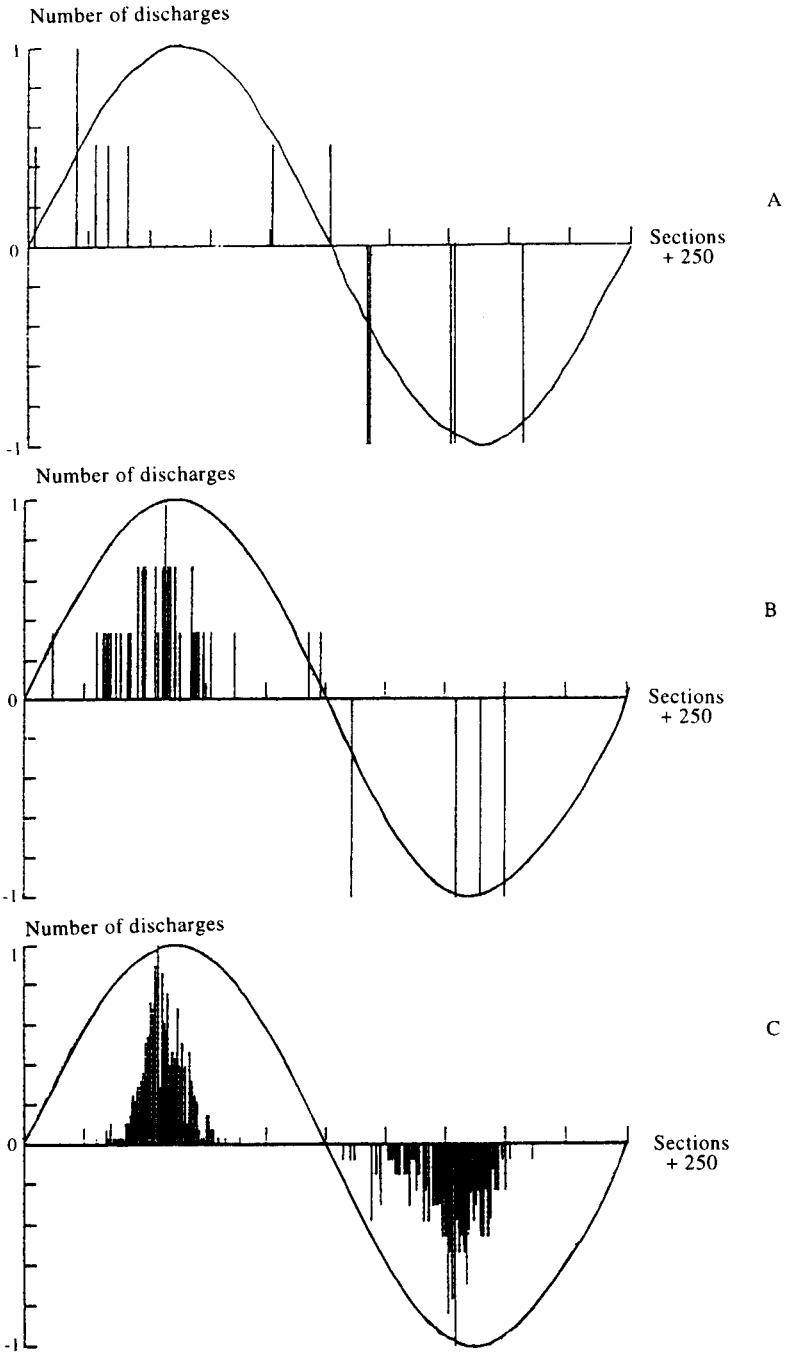


Fig. 2. Distributions of the relative number of discharges, as a function of the phase of capacitor subjected to a.c. voltage of various effective values. (A) $V_a = 6.2\ kV_{eff}$, (B) $V_a = 6.5\ kV_{eff}$, (C) $V_a = 6.8\ kV_{eff}$.

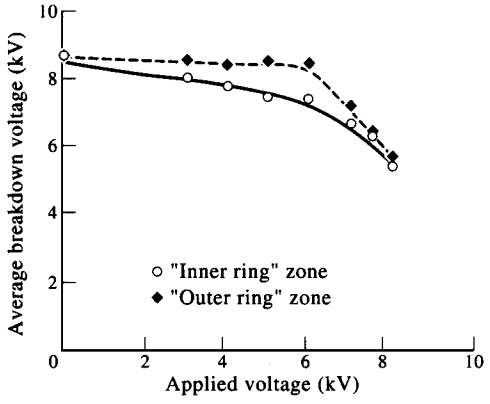


Fig. 3. Average breakdown voltage of a PP film aged for 4 hr as a function of the voltage applied to the capacitor.

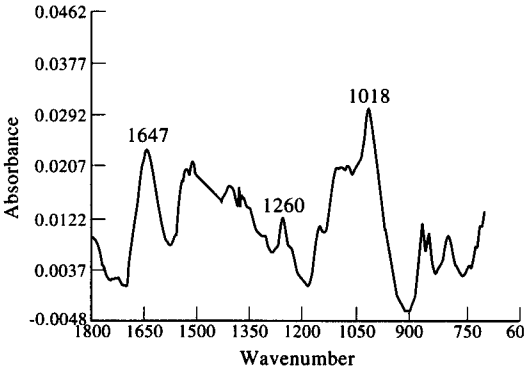
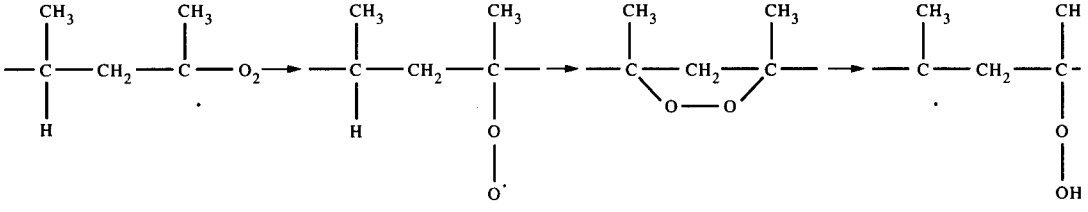
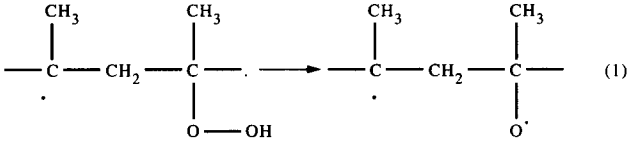


Fig. 4. FTIR spectra obtained as the difference betwe samples of films aged for 4 hr at 7.5 kV_{rms} and non-ag films.

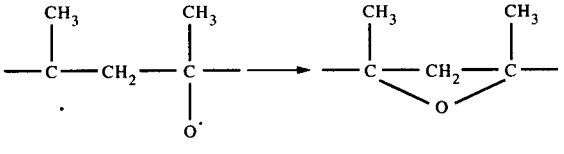
impurity such as H_2O or dissolved O_2. Due to the presence of a substantial oxygen concentration, the macroradicals PP· give peroxy radicals and then hydroperoxide.





(1)

The presence of a non polar aromatic liquid gives probably sufficient stability for radical (1) permitting intramolecular or intermolecular recombination [14].



Influence of the epoxy additive. We have measured the life-time of aged capacitors (Fig. 5). This life-time of electrically stressed capacitors decreased when DGEBA was added to BT. The life-time was reduced from *ca* 6 hr (in the absence of epoxy) to about 1 hr when the DGEBA concentration was > 1%.

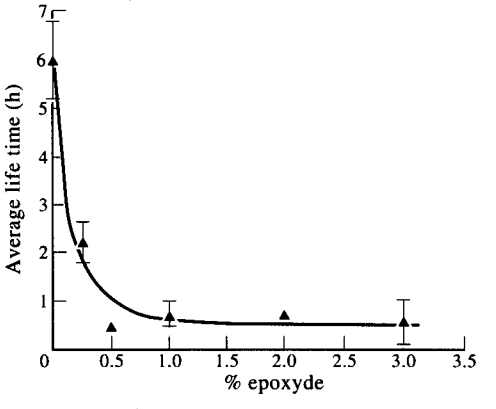


Fig. 5. Average life-time of capacitors aged at 7.5 kV_{rms} a 1.5 bar excess pressure of O_2, as a function of epoxy concentration.

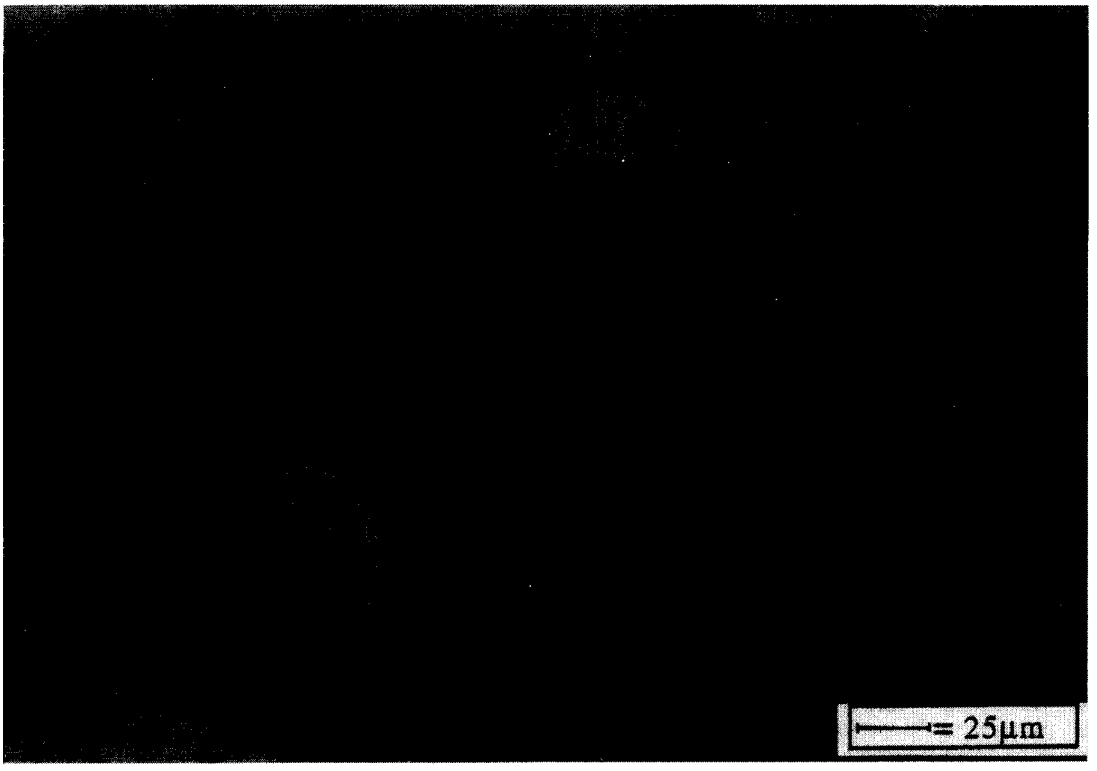


Fig. 7. Deposit on the surface of the film (impregnated with BT + 3% DGEBA) aged for 1 hr under 7.5 kV_{rms}.

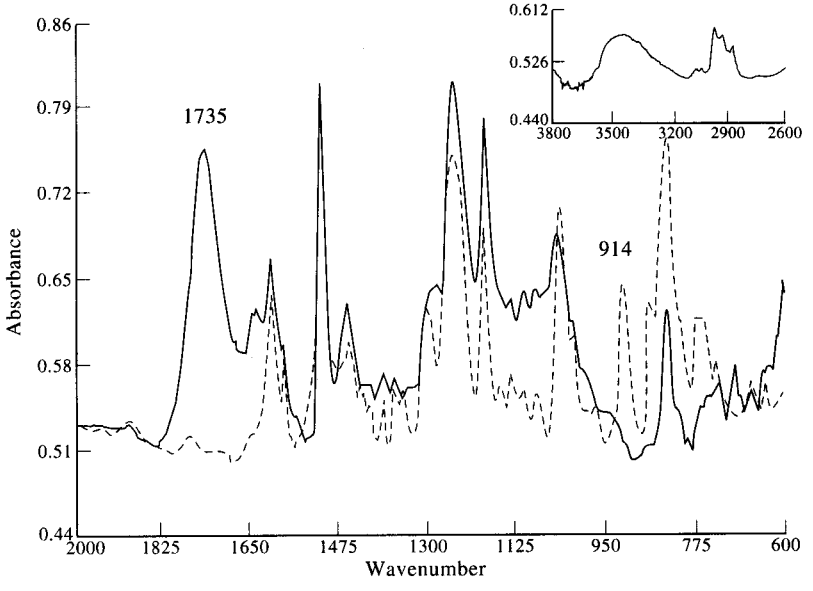


Fig. 8. FTIR spectrum: FTIR spectrum of a pellicule detached from a PP film, impregnated with BT + 3% DGEBA and aged for 1 hr at 7.5 kV_{rms}. FTIR spectrum of DGEBA.

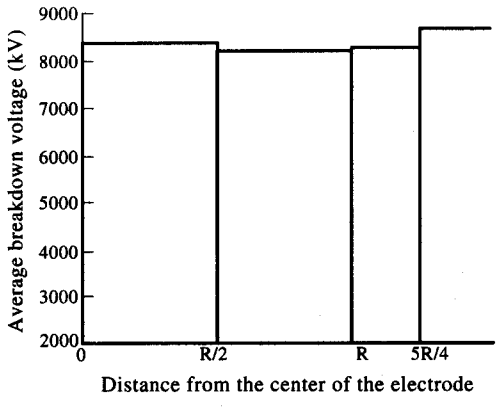


Fig. 6. Average breakdown voltage of PP film (impregnated with BT + 3% DGEBA) aged for 1 hr under 7.5 kV_{rms} at different distances from the centre of the electrode.

Moreover, we have measured the breakdown voltage of the films extracted from the capacitors (Fig. 6). The additive did not influence the dielectric strength of the film impregnated with BT + 3% DGEBA. The breakdown voltage measured at points situated in either the "outer ring" or the "inner ring" zones had the same value as for a new film.

With 3% DGEBA in the impregnant and the 1 hr ageing by an electric field of 250 V μm^{-1} at 80°C with O_2, the microscopic observation of PP film, (washed with cyclohexane), evidenced the pressure of a pellicular deposit on the surface of the film. The pellicule would be easily detached from the film (Fig. 7). The FTIR spectrum of this pellicule was similar to that of DGEBA epoxy (Fig. 8). However, this spectrum has three more absorption bands: one at 3400 cm^{-1} (OH), another one at 1735 cm^{-1} (aldehyde) and a large band between 1076 and 1150 cm^{-1} (ether). We notice the absence of absorptions bands at 914 and 862 cm^{-1}. These bands are characteristic of the epoxy functional group [15, 16]. The absence of the epoxy functional group indicates that epoxyde reacts chemically in the region of more active ageing by opening of its cycle, DGEBA may polymerize at the film. The pellicular deposit does not adhere to the film.

The oxidation evidenced by FTIR spectroscopy was that of epoxy and not of the PP film. The presence of DGEBA favoured the oxidization of the liquid and, consequently, the electrical resistivity decreased by a factor of 3 [5]. This favours the occurrence of electrical discharges of higher energy but does not change the discharge onset voltage [6]. It appears that the discharges facilitate the polymerization of DGEBA. This polymerization protects the PP film against uniform electrical degradation. However, a more intense discharge may quickly perforate the film.

CONCLUSION

In capacitors with BT impregnated PP, a good correlation was shown to exist between the threshold voltage discharge (V_{th}) and a change in the kinetics of the decrease of the breakdown voltage. We have shown that: For $V_a < 6\ kV_{rms}$, the phenomenon, which ages the capacitor, is a electrochemic phenomenon [17]. There was no degradation in the "inner ring" zone. For $V_a > 6.5\ kV_{rms}$, electrical discharges contribute to an accelerated ageing of the PP film insulation of a parallel plate capacitor. The average breakdown voltage decreases significantly in the zone affected by the electric field and in a circular ring of 1 mm from the edges of the electrode system.

The life-time of electrically stressed capacitors diminished when using an additive to the BT. This fact can be explained by electrical discharges with higher energy content which may quickly perforate the film.

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