

ANNEALING EFFECT ON THE CONDUCTIVITY OF XLPE INSULATION IN POWER CABLE



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ABSTRACT

Conductivity of XLPE insulation of power cables annealed at 90°C at temperatures between 50 and 97°C has been measured. In all cases there is an initial increase in conductivity, associated with the presence of crosslinking by products that are volatile and disappear after 20 days annealing approximately. At low temperatures (50°C), the conductivity decreases after long annealing times and its value stabilize approximately after 45 days annealing. This behavior can be associated with current limited by space charge. At high temperatures, conductivity after long annealing times is higher than the initial value. Obtained results are in good agreement with a hopping controlled mechanism. Infrared spectroscopy indicate that, during annealing, defects diffuse from the semiconducting shields into the XLPE. At lower field, ohmic conductivity is observed, associated with thermally generated carriers. Thermally stimulated depolarization technique (TSDC) is used to corroborate data obtained by conductivity measurements.

KEYWORDS

XLPE, conductivity, ARC, TSDC, FTIR, cable, insulation.

INTRODUCTION

Polyethylene (PE) is a non-polar polymer. It is found in semi-crystalline state at room temperature. In industrial applications, crosslinked PE (XLPE) is commonly used as an electrical insulator due to its outstanding insulating characteristics and its low price. It is well known that PE conductive properties are conditioned by its morphology [13] and that insulation degradation can be related to volume space charges [4]. Particularly, conduction and space charge formation in low-density PE were studied including the interval of temperatures of interest for cable diagnostics [5].

Current versus voltage (I-V) characteristics studied in LDPE by Stetter [6] at 42, 82 and 110°C, indicate that for low electric fields (E) an ohmic behaviour is observed; for higher electric fields, conduction can be interpreted on the basis of a quadratic law in E which depends on temperature and reveals the existence of traps. More recently, several authors have also observed that current-voltage characteristics in PE can be explained by the

theory of space-charge-limited currents model (SCLC) [7]. Nath et al. [8] apply the band theory to LDPE by developing a mathematical model based on the hopping of carriers which are injected into amorphous regions. They consider a SCLC and a process of charge-trapping at amorphous-crystalline boundaries. In addition, they suppose that the density of trapping centres is high enough so that the interaction between them results in an effective lowering of trap depth (Poole-Frenkel effect). In this latter model, the "distance between traps" parameter is introduced and it has a constant value. Even more recently, following the same research theme, it is concluded that electronic transport is bound to thermally activated hopping, which is assisted by electric field with a very low activation energy [9]. Recent studies of our research group have deal with conductivity in XLPE cable samples measured by the absorption resorption current method (ARC) [10]. This studies revealed that conductivity is highly influenced by the thermal treatment of the samples in the presence of the cable semiconducting shields (SC), and it is based on a hopping process.

Thermally stimulated depolarization current technique (TSDC) has been applied to XLPE cable electrets [11,12]. A broad current peak can be observed at 105°C, which has been associated with charge in the crystalline volume [11]. The combination of TSDC and infrared spectroscopy (IR) studies [12] proved that annealing procedures in MV cables lead to the diffusion of components from the SC screens into the XLPE insulation bulk. Moreover, the defects associated with diffused particles act as trapping centres of charge injected from the electrodes. This process is clearly promoted at temperatures above a critical temperature approximately equal to 80°C. Annealing at these temperatures reverses the polarity of TSDC spectra (from heteropolar to homopolar). Further annealing results in new current polarity inversion, and the current regains its heteropolar character [11].

The aim of the present work is to obtain further experimental results concerning conductivity in XLPE in order to better understand conductivity processes in this material. Following this purpose, several samples were annealed at $T_a=90^\circ\text{C}$ for different times t_a between 0 and 90 days and conductivity (σ), TSDC spectra and infrared spectroscopy (IR) was measured.

EXPERIMENTAL

Cable was supplied by General Cable S.A. and consisted of a cylindrical insulating XLPE layer with 7 and 13 mm of inner and outer radii, respectively. The samples had two 1 mm thick semiconducting layers (SM) in contact with the inner and external surfaces of XLPE. The insulating material was crosslinked mixing low density PE with a crosslinking agent (di-t-butyl peroxide) and heating it up to a temperature higher than 200°C to produce a vulcanization reaction. A new sample was used in each measurement in order to avoid the effects of previous measurements.

ARC measurements

Samples for absorption resorption current measurements (ARC) consisted in 7cm long sections cut from the cable. A 2mm guard electrodes were prepared at each end of cable sample in order to avoid the effect of surface conductivity and to avoid the dispersion of the field lines. Conductivity (σ) was determined by means of equation:

$$\sigma = \frac{\epsilon_0}{C_o U} (I_a(t) + I_r(t)) \quad [1]$$

where $C_o = 4.4$ pF is the geometrical capacitance of the sample (determined by a Hewlett-Packard impedance analyser model HP-4192a LF), $I_a(t)$ and $I_r(t)$ are the absorption and resorption currents (ARC), respectively, U is the voltage applied to each cable sample and ϵ_0 is the vacuum permittivity. The ARC method provides a convenient way to obtain σ because measurements require less time to be performed than with the usual current-voltage characteristics method since there is no need to reach a stationary current.

XLPE cable samples were placed in a measuring cell inside a Carbolyte type PF60 oven. A K-type thermocouple (Keithley 6517-TP) placed inside the insulation of another identical sample (which was located close to the sample under test) was used to measure the sample temperature. The DC voltage source of a Keithley 6517 A electrometer was used to polarize the samples. To measure the absorption current a voltage of 1 kV (corresponding to a mean field of 0.22 MV/m) was applied to the inner electrode of the samples and the outer electrode was grounded through the aforementioned electrometer. After a period of time, the inner electrode was switched to ground and the resorption current was recorded. The charging period (absorption current) was about the double of the discharging period (resorption current), 2000 and 1000 s respectively, so that Eq. (1) can be used for conductivity calculations as a good approximation.

TSDC and FTIR measurements

Samples for TSDC measurements consisted in 7 cm long sections cut from the cable. The semiconducting layers were used as electrodes. To avoid shortcircuits, the external semiconducting layer was partially removed from the sample ends. This left a 2 cm wide semiconducting strip centered in the sample. The cable core of twisted

aluminum wires was used to make contact with the inner semiconducting layer. The contact with the outer semiconducting layer was made by an adjustable metallic clamp.

The experimental setup for TSDC consists of a Heraeus forced air oven controlled by a Eurotherm 902 PID temperature programmer. Inside, two identical cable samples were placed close together in parallel position. One of them was used to host the temperature probe inside the insulating layer, and TSDC measurements were performed on the other one. To polarize the sample, the cable core was positively biased with a Brandenburg 807R (3-30kV) potential source whereas the outer electrode was grounded. In this way, potentials up to 20kV can be applied to the sample. Once the polarization stage was finished, the external semiconducting layer was connected to a Keithley 6514 electrometer and the cable core was grounded.

Sample polarization was performed by two different methods. In the first method (NIW method), the potential V_p is applied, during a cooling ramp, at a polarization temperature T_p and removed when the sample reaches a temperature T_f in the same cooling ramp. In the second polarization method (WP method) the polarization is performed isothermally at T_p and the electric field is switched off after a time t_p , just before the sample cooling begins. In all cases the heating rate was 2.5 °C/min whereas the cooling rate was 1°C/min.

Fourier transform infrared (FTIR) measurements were performed with a Nicolet 510M spectrometer on 6x6mm XLPE sheets. Samples 300 μ m thick were cut directly from the cable by mechanical methods.

RESULTS AND DISCUSSION

Conduction mechanisms

Stationary conductivity (σ) of cable samples measured by ARC at temperatures (T_m) between 50°C and 97°C are plotted in figures 1 and 2 as a function of annealing time (t_a) at 90°C.

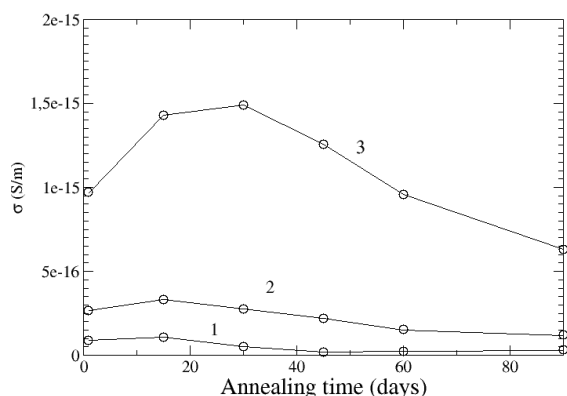


Figure 1. Conductivity versus annealing time (1: $T_m=50^\circ\text{C}$, 2: $T_m=60^\circ\text{C}$, 3: $T_m=72^\circ\text{C}$)

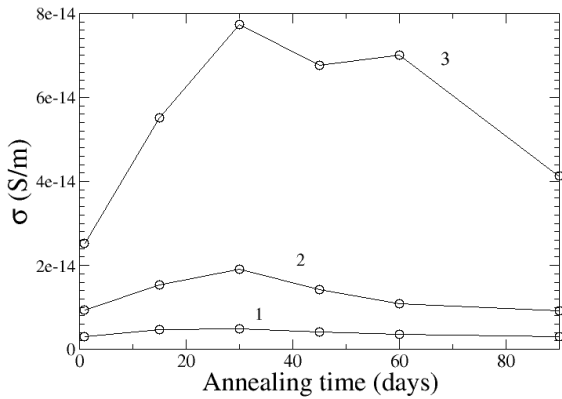


Figure 2. Conductivity versus annealing time (1: $T_m=78^\circ\text{C}$, 2: $T_m=90^\circ\text{C}$, 3: $T_m=97^\circ\text{C}$)

All curves show a maximum in conductivity that appears for t_a around 15 days at the lower measuring temperatures (50 and 60°C , figure 1). This maximum becomes more noticeable when T_m increases, and shifts towards higher annealing times, up to 30 days when $T_m=97^\circ\text{C}$ (figure 2).

Besides this behaviour, sample conductivity after long annealing show a differentiated behaviour depending on measuring temperature. For low T_m (50, 60 and 70°C , figure 1), σ for $t_a=90$ days is lower than the initial value, however this long term decrease is less noticeable as T_m increases. Samples measured at 78, 90 and 97°C (figure 2) show an increase in conductivity after long t_a , that almost double the initial value for $T_m=97^\circ\text{C}$.

The initial increase in σ , observed in all cases with a maximum for annealing times around 20 days, is probably due to the presence and diffusion through the isolation of crosslinking by products. This fact generates internal charge and trapping centers that enhance σ . However, these products are known to be volatile and disappear for long annealing times, developing the observed maximum.

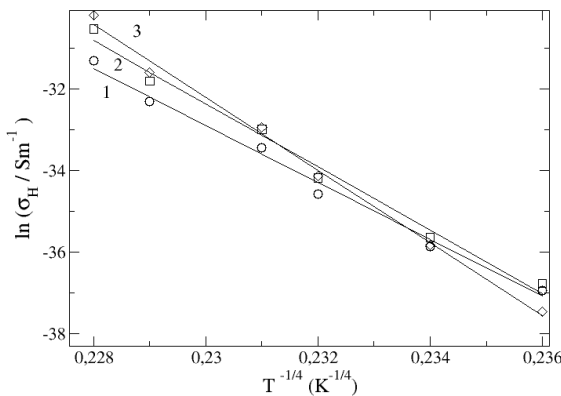


Figure 3. Mott plot of $\ln(\sigma_H)$ versus $T_m^{-1/4}$ (1: $t_a=1$ day, 2: $t_a=15$ days, 3: $t_a=30$ days)

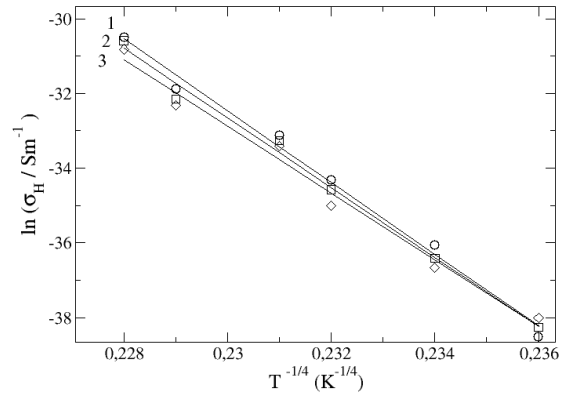


Figure 4. Mott plot of $\ln(\sigma_H)$ versus $T_m^{-1/4}$ (1: $t_a=45$ days, 2: $t_a=60$ days, 3: $t_a=90$ days)

We have represented in figures 3 and 4 as a Mott plot ($\ln(\sigma_H)$ versus $T^{-1/4}$) the contribution to conductivity of the hopping mechanism between traps in the cable samples, σ_H , where $\sigma_H = \sigma - \sigma_{EX}$, and σ and σ_{EX} are the total conductivity and the contribution to conductivity by extended states respectively. A value of $\sigma_{EX}=1.65 \cdot 10^{-17} \text{S/m}$ has been used according to previous works [10].

A linear behaviour of $\ln(\sigma_H)$ versus $T^{-1/4}$ can be estimated from these figures, which implies that the transport mechanism is basically via thermally assisted hopping conduction [13]. Hill [14] also found a similar relation between conductivity and annealing temperature, by considering a field-assisted hopping conductivity at the limit of low electric fields, which is evidently one of the important experimental conditions observed in our present work. This good agreement between data and the aforementioned law is more evident for long annealing times.

Assuming that conductivity in XLPE is associated with a hopping mechanism, as the Mott plot suggests (figures 3 and 4), we can adequately explain the observed change in σ after long annealing times as follows; In the hopping mechanism, the energy difference and the distance between traps should modulate the whole process. If we take into account a possible diffusion of components from the semiconducting shields into the XLPE bulk (as previous works suggest [12], annealing should promote an increase in conductivity because of the increase in traps density. This phenomena can explain the increase in σ observed in long annealed samples measured at the higher temperatures (figure 2). At low temperatures, current is limited by space charge (SCLC). The mechanism for SCLC is the formation of a layer of defects close to the SM. This layer results from the assumed diffusion of defects from the SM during annealing. It becomes ionized and blocks the current progressively until saturation, generating the observed decrease in σ for long t_a (figure 1).

To corroborate this assumption, Fourier Transform

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Infrared measurements (FTIR) were performed at different depths in the XLPE isolation of as received cable and cable annealed 3 hours at 140°C. We will focus our analysis of FTIR results in the peak corresponding to 3300cm⁻¹, identified in previous works as a peak associated with a component not present originally in XLPE, that diffuses from the SC [12]. Figures 5 and 6 show the measured intensity of this peak in four layers cut from the isolation surface down to 1mm depth (layer 1: XLPE surface in contact with the SC shield, layer 4: XLPE at 1mm depth).

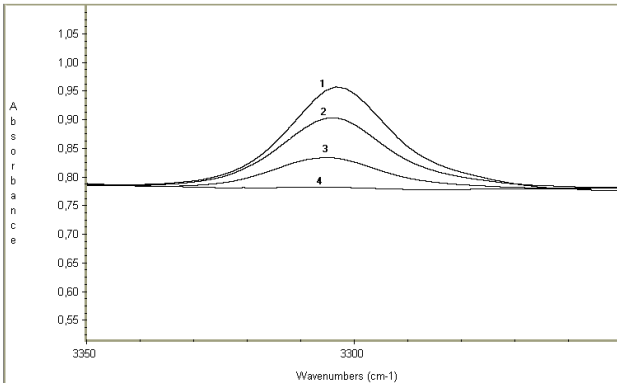


Figure 5. FTIR of as received cable XLPE at different depths (1: surface, 2: 0.25mm 3: 0.5mm 4: 0.75mm)

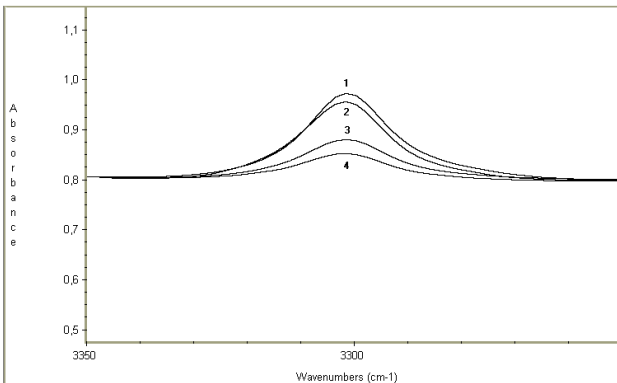


Figure 6. FTIR of cable XLPE annealed 3 hours at 140°C at different depths (1: surface, 2: 0.25mm 3: 0.5mm 4: 0.75mm)

We can observe in these results that these impurities are already present in the XLPE from the beginning (i/e, in the as received cable, figure 5). This initial diffusion occurs during cable manufacturing where temperatures as high as 200°C can be reached. With annealing (figure 6), these components diffuse into the XLPE bulk from the surface, as indicate the increase in peak height at the deeper layers. A continuous injection of new impurities occurs from the SC shield, remaining their concentration almost constant at the surface of the XLPE .

Figure 7 shows the peak obtained in the same sample of figure 6 after further annealing at 140°C but now removing the SC external shield. We can observe a decrease in the relative intensity of the peak in all layers.

This fact indicates that the component responsible of it, certainly, is diffused from the external SC.

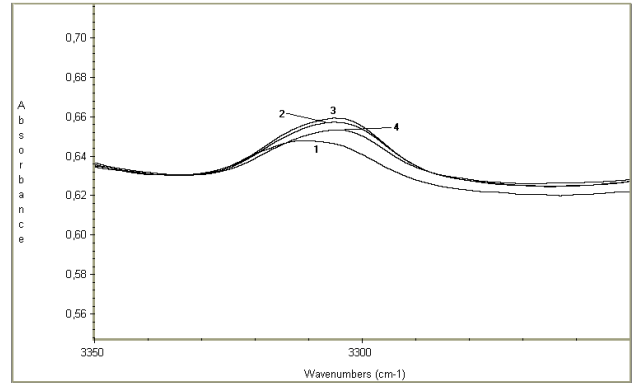


Figure 7. FTIR of cable XLPE annealed 3 hours at 140°C with SC and 4 days without SC, at different depths (1: surface, 2: 0.25mm 3: 0.5mm 4: 0.75mm)

TSDC and I-V results

TSDC is a complementary technique that can be used to study injection and charge trapping in dielectrics. Figure 8 shows the TSDC spectrum obtained by the NIW method (see experimental section for details) of a sample annealed for 2 hours at 140°C. We can observe an homopolar peak at 105 °C. The homopolar peaks are associated in this kind of measurements to charge injected from the electrodes and trapped in the dielectric. It is remarkable as well that only after annealing of the sample, the homopolar response appears. This fact indicates that impurities diffused from the SC into the XLPE during annealing (as show FTIR results) act as trapping centres during the polarization process.

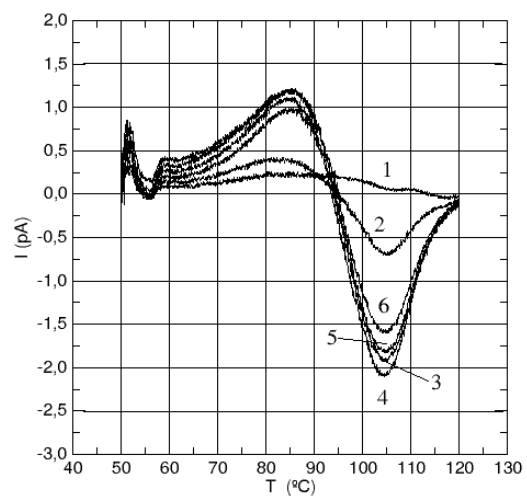


Figure 8. NIW-TSDC of cable annealed 2 hours at 120°C ($T_p=100^\circ\text{C}$, $V_p=10\text{kV}$, T_f : 1.- 90°C, 2.- 85°C, 3.- 75°C, 4.- 70°C, 5.- 60°C, 6.- 50°C)

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A noticeable fact is that this homopolar response appears only when the polarization field is 'on' below 87°C. In the cooling ramp, the material starts from the melt state and is approximately between 90 and 85°C when significant crystallization starts. This result indicates that charge trapping occurs probably in the amorphous-crystal interphases.

Figures 9 and 10 show the TSDC spectra obtained by the WP method (see experimental section for details) for different polarization fields, in as received cable samples (figure 9) and after 3 days annealing at 140°C (figure 10).

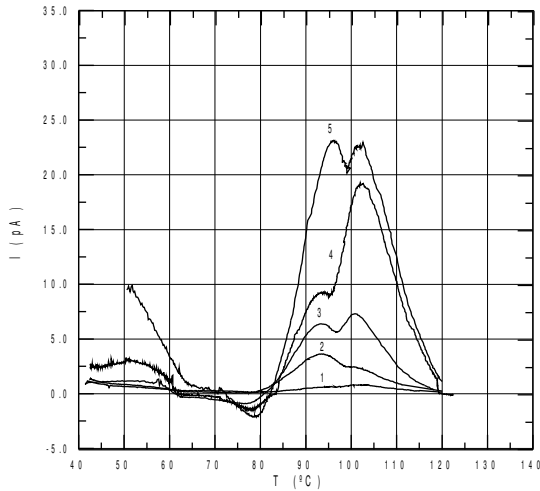


Figure 9. WP-TSDC of as received cable ($T_p=45^\circ\text{C}$, $t_p=5\text{min}$, V_p : 1.- 4kV, 2.- 8kV, 3.- 12kV, 4.- 16kV, 5.- 24kV)

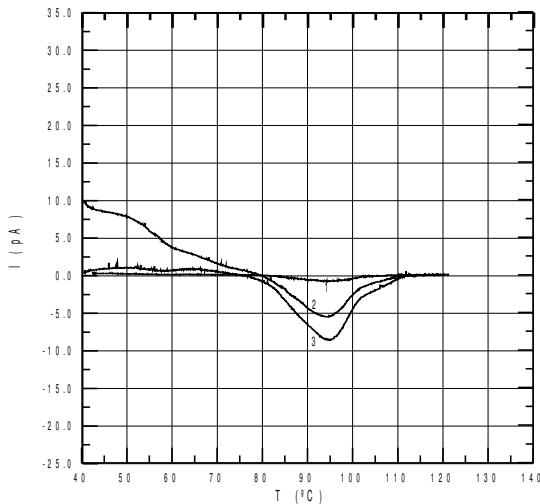


Figure 10. WP-TSDC of cable annealed 3 days at 140°C ($T_p=40^\circ\text{C}$, $t_p=5\text{min}$, V_p : 1.- 8kV, 2.- 16kV, 3.- 24kV)

We can observe again that annealing (and the subsequent diffusion of components from the SC layers into the XLPE) is necessary to obtain the homopolar peak. What is more remarkable in these measurements

is the fact that for polarization potentials of 8kV the homopolar response is scarcely appreciable. This result suggests that there is a polarization field threshold, below which injection of charge from the electrode does not take place. To analyse this possibility, intensity-voltage (IV) curves were measured in the 1kV-10kV range. Figure 11 shows the obtained results, where a clear change in behaviour is obtained for voltages around 6.5kV. Below this value ohmic conductivity is observed, associated with thermally generated carriers. Above 8kV injection of new carriers occurs from the electrode and an important increase in conductivity is obtained.

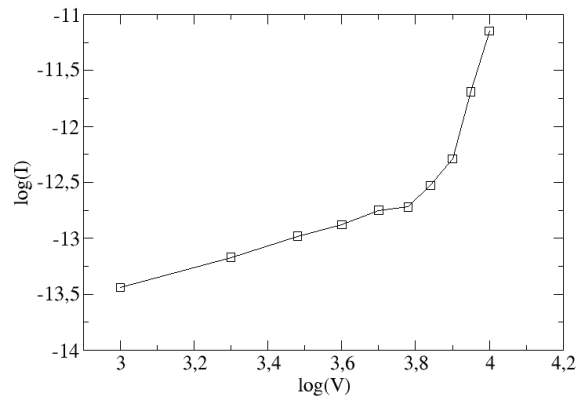


Figure 11. $\log(I)$ versus $\log(V)$ of cable annealed 5 days at 120°C.

CONCLUSIONS

Conductivity in XLPE cable insulation is associated with a hopping mechanism, so that the energy difference and the distance between traps modulate the process, and a temperature dependence given by Mott's law results. Injection of impurities from the SC shields into the XLPE occurs during the manufacturing process, and continuously during annealing at 90°C (working temperature of cables). These defects act as trapping centres of charge injected from the electrodes. At low temperatures, current is limited by space charge (SCLC) by the formation of an ionized layer, close to the surface, that blocks the current progressively until saturation. There is a polarization field threshold, around 6.5kV, below which injection of charge from the electrode does not take place.

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