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Assessment of a Charge Transport Model for LDPE through Conduction Current Measurement

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Abstract

Conduction current measurements have been widely used to characterize charge transport behavior in insulating materials. However, the interpretation of transport mechanisms and more generally of non-linear processes from current measurements alone is not straightforward. For this reason, space charge measurements, on the one hand, and models of charge transport encompassing charge generation, trapping and transport have been developed. The completeness and accuracy of a model can be assessed only if a substantial range of stress conditions, being field and temperature for the current topics, is available. The purpose of this communication is to enrich the investigation of low density polyethylene - LDPE insulation material characteristic using conduction current measurement. Measurements were conducted on 250 μm thick LDPE samples, for DC fields in the range 2 to 50 kV/mm and for temperatures from 20 to 70°C. Experimental data, i.e. transient current in charge/discharge and quasi-steady state currents are compared to the prediction of a bipolar transport model that has been developed over the last years and fitted to the case of LDPE. The deviation of model results is substantial, with essentially an overestimation of the non-linearity of the current-field dependence. These differences are discussed along with prospects from improving the model. Aside from these modelling approaches, we show that thermal preconditioning of samples appears to be influential in the measured apparent conductivity.

Keywords: LDPE, conduction current, charge transport

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1. Introduction

Investigation on polyethylene material as electrical insulating material receives significant attention as its demand increases, especially since polyethylene is more and more used in high voltage DC cables application. Current understandings regarding charge transport and mechanisms related to space charge will benefit for reaching better performance and reliable HVDC insulation systems. Low Density Polyethylene (LDPE) as part of polyethylene group is the main concern in this paper.

LDPE charge transport characterization by conduction current has been conducted in various researches. Charging mechanism characteristics by means of threshold representation [1, 2] on space charge features provides one way to describe its character. Comparison between polymers was also conducted, as LDPE vs HDPE – i.e. high density PE [2], LDPE vs. LDPE + Antioxidant vs. XLPE – i.e. crosslinked PE [3], and XLPE vs EPDM, i.e. rubber with ethylene-propylene-diene monomer [4]. The purpose of this paper is to enrich study regarding LDPE charge transport by presenting measurement results on charging and discharging currents and comparing results with an already available model of conduction based on bipolar charge generation and transport. The model has been parameterized and refined over the years and encompass charge injection, charge transport and charge recombination [5, 6, 7]. Its optimization is based on experimental results relevant to charging/discharging current, space charge measurements and electroluminescence.

Along these objectives, preconditioning factors that influence the measurement and how modelling reacts to it are also investigated. Indeed, variations in preconditioning is considered as time elapsed before measurement once the sample is set to a given temperature, or previously applied electrical stress in the course of measurements. This could explain variations observed in output results. The model that has been developed can indeed integrate to some extent this thermo-electrical history.

2. Experimental Procedure

2.1. Conduction Current Measurements

LDPE material was considered for the conducted investigation. LDPE without antioxidant, provided by Borealis, was chosen. For the measurement, LDPE pellets were first press-moulded to be prepared as plaque specimen. Plaque sample was processed at 140 ℃ under a pressure of 3 bars for 20 minutes. Completed samples are disks of 8 cm in diameter with 250 ± 10 μm in thickness. Kapton was used as template and pressing layer during press moulding, the template was arranged to create plaques of 250 μm thickness. For ensuring measurement contact, each sample was provided with gold electrodes by sputtering, the gold layer has 5 cm in diameter and 30nm in thickness. A silicone layer was laid at the periphery of the electrode to avoid edge effects.

Several samples were prepared to be tested in different thermal preconditioning procedure: no thermal preconditioning, 1-hour, <52 hours, and >52 hours thermal conditioning.

Conduction current measurements were registered in air at 3 different temperatures (30, 50, 70° C) and 13 values of the applied electric fields (2, 4, 6, 8, 10, 13, 16, 19, 22, 25, 30, 40, 50 kV/mm). The sample was clamped between two brass electrodes with polished surface. The current was recorded through a Keithley 617 ammeter with a 2 s dwelling time under charging state for 3 hours, and discharging state for 1 hour.

Extracted quantities mainly are transient currents and quasi steady state current, which will be derived as current density and conductivity. Current density from transient current measurement provides information related to conduction mechanism, current density is deduced by the following equation:

$$J(t) = \frac{I(t)}{A} \tag{1}$$

where I(t) is the measured current and A the area of electrode (20 cm²). Conductivity value of the insulation is calculated using the following equation:

$$\sigma = \frac{J_\infty}{E_0} \tag{2}$$

where J_∞ is the steady state current density, E_0 the applied field. In this work, the current values utilized in current density equation are quasi steady state current values which were taken during the last 800 s of the 10800 s measurement time of charging current measurement.

2.2. Model

The model features bipolar transport and trapping of electrons and holes. The model was created to fit experimental measurement of current, space charge, thermos-stimulated currents, electroluminescence, etc. [6, 8]. Fig. 1 below illustrates the schematic representation of the model for LDPE [5]. It is a two levels model for each kind of carriers, defining so 4 kinds of species: mobile and trapped electrons and same for holes.

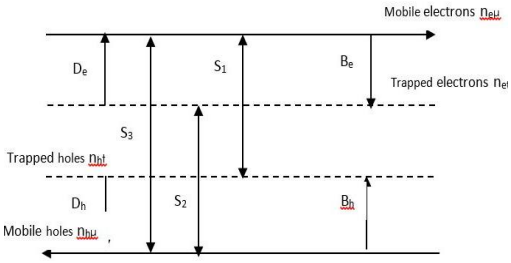

Fig. 1 Physical model schematic

The set of equations constituting the model is common to transport models in dielectric media, being liquids, solids or gases:

- Transport of electrons and holes, neglecting diffusion:

$$j_e(x) = \mu_e n_{e\mu}(x) E(x) \tag{3}$$

$$j_h(x) = \mu_h n_{h\mu}(x) E(x) \tag{4}$$

where μ_e is the electron mobility, μ_h the hole mobility, $n_{e\mu}$ the mobile electron density, $n_{h\mu}$ the mobile hole density, E the electric field, and x the spatial coordinate.

- Poisson's equation:

$$\frac{\partial E(x)}{\partial x} = \frac{\rho(x)}{\varepsilon} \tag{5}$$

where ε is the dielectric permittivity, ρ the net charge density.

- The conservation equation, meaning that local variations of density of given specie are due to transport or to variation as a source:

$$\frac{\partial n_i(t)}{\partial t} + \frac{\partial j_i(x)}{\partial x} = s_i \tag{6}$$

where s encompasses the source terms (i.e. trapping, detrapping, and recombination process).

Those source terms have for example the following form for mobile electrons:

$$s_1 = -S_1 n_{ht} n_{e\mu} - S_3 n_{h\mu} n_{e\mu} - B_e n_{e\mu}\left(1 - \frac{n_{et}}{n_{0et}}\right) + v.exp\left(\frac{w_{tre}}{k_b T}\right) n_{et} \tag{7}$$

S_i is the recombination coefficient, B_e the trapping coefficient for electrons and B_h the trapping coefficient for holes. Densities of trapped holes and electrons are stated with n_{et} and n_{ht}, while maximal trap densities of electrons and holes are stated by n_{0et} and n_{0ht}. w_{tre} is the detrapping barrier height.

Modeling of charge injection during applied voltage at each electrode is expressed with the following equation, for electrons as an example:

$$J_e(0) = AT^2 exp\left(-\frac{w_e}{kT}\right) \left[exp\left(\frac{e}{kT}\sqrt{\frac{eE(0)}{4\pi\varepsilon}}\right) - 1\right] \tag{8}$$

Equation for charge extraction at the other side is written as follows:

$$J_e(d) = \mu_e n_{e\mu}(d) E(d) \tag{9}$$

The total current density through the material which incorporates the electrons and holes current density follows:

$$J(t) = \frac{1}{D}\int_0^D (J_e(x,t) + J_h(x,t))dx \tag{10}$$

Latest refinements incorporated into the model concern the use of Langevin-type recombination, where the recombination coefficients are function of the mobility of the carriers [8]. The mobility is a constant effective mobility that already takes into account the possible trapping and detrapping of charges into shallow traps.

3. Results and Discussion

3.1. Transient Current Measurements

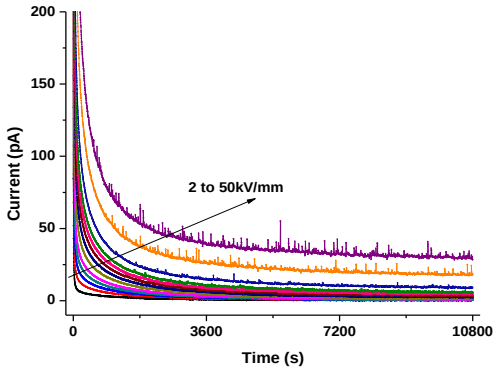

Fig. 2 Charging current transient in 250 µm thick LDPE plaque measured at 30°C for 13 different values of the applied field ranging from 2 to 30kV/mm (cf. §2.1)

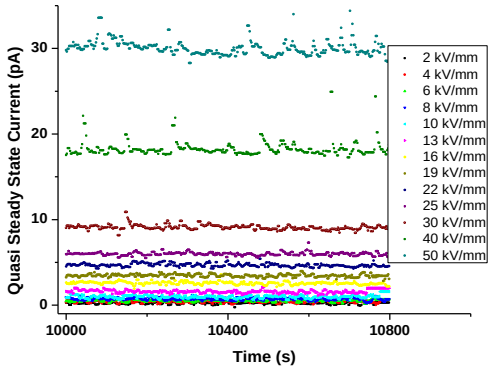

Fig. 3 Quasi steady-state charging current in 250 µm thick LDPE plaque measured at 30°C (long time data of Fig. 2)

Transient current measurements were realized on LDPE and examples of the results obtained at 30°C are shown in Fig. 2 and Fig. 3. Fig. 2 depicts a quickly reducing current magnitude toward a steady state. Fig. 3 focuses on the longer time region in which current have been averaged for plotting the characteristics. The current appears indeed steady at this scale. In Fig.

2 and 3, some noise is detected for high field steps possibly due to some micro-discharges in the high voltage range (all measurements were realized in air at atmospheric pressure). However this noise does not have substantial impact on estimated conductivity. We shall see later on that the present charge time configuration (3 hours) is not sufficient to achieve steady state.

3.2. Precondition Effect on Measurement

Substantial change in time in the conductivity has been reported recently depending on pre-annealing time of LDPE and crosslinked polyethylene (XLPE) by H. Ghorbani [9]. Indeed, aside from the apparent decrease in conductivity as a function of stressing time measured at 50°C, there was also a decrease in conductivity with the pre-storage time at 50°C before the measurements. Similarly, Montanari et al. [3] reported on a decrease in the transient currents measured at room temperature when LDPE or XLPE samples have been previously thermally annealed for 90h at 50°C [3]. The current decrease was all over the measurement time of 3h. As measurements realized here are relatively long (50h per temperature step) when realizing consecutively the all set of polarization/depolarization steps, there can be an evolution of the conductivity due to this conditioning.

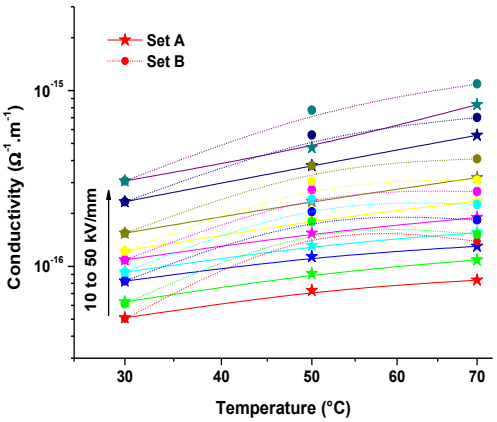

Fig. 4 Comparison of conductivity with varying preconditioning of LDPE plaque sample under various applied field.

Set A was conducted continuously for the 3 temperatures with same sample (Sample 1), while Set B was conducted with a different fresh sample for each temperature value.

Quasi steady current values plotted in Fig. 4 were obtained for the following cases:

(a) Set A: same sample stressed successively in the different field steps and different temperatures (30, then 50 and 70°C);

(b) Set B: one different sample for the different temperature levels.

In all the results, the apparent conductivity for the fresh sample is higher than the one for the previously stressed. Quantitatively the difference between the two steps is a drop of the conductivity by about 30 to 50% after pre-stressing. The variation is about the same for 50 and 70°C. The trends are consistent with the previously reported results. However, the situation is a bit more complicated in the present case compared to results of H. Ghorbani as here the history concerns both the thermal and electrical conditioning: both are likely to decrease conductivity for different reasons:

(a) electrical pre-stressing may generate space charge into the insulation, e.g. close to the injecting electrode: this will act as counter-field for further charge injection and is a process that can explain the decay in time of the current. If trapped charges are stable, the memory effect will be generated owing to the pre-existing charge. A transport model might anticipate such features.

(b) thermal pre-stressing can induce drying/outgassing of sample if some residues are present, and/or change of the morphology as crystallinity. Substantial changes of crystallinity were reported H. Ghorbani [9] for the long term testing at 50°C on LDPE and XLPE. Crystallinity of LDPE increases as heat treatment time lengthened. This can in turn alter the electrical response of the material. One way to distinguish morphological vs. residue effects would be to probe again samples one exposed to ambient conditions.

3.3. Current vs. field characteristics

Several works have reported on the threshold of current vs. field for various specimens such as: XLPE, rubber, HDPE, and LDPE [2-4]. The current density is plotted as a function of field following this 'threshold' representation – i.e. log-log plot in Fig. 5. Samples did not undergo thermal preconditioning before measurements apart from the stabilization time at the set tem-

perature. It can be seen that the J-E curves of sample 2 (50° C) and sample 3 (70° C) starts unlike temperature-field characteristics of polyethylene. Indeed, the current tend to drop while increasing the applied field, which is an unexpected behavior. The effect is not observed for the measurement at 30℃. The explanation for the effect is most probably the one described above, i.e. a conditioning effect at the measurement temperature: as measurement at each voltage level requires 4 h time, these conditioning effects can be significant. Beyond a field of 10kV/mm, the curves for the different temperatures have similar shapes.

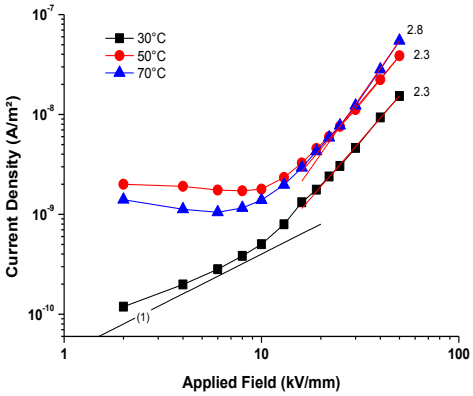

Fig. 5 Current density log-log plot for LDPE plaque specimens. Electrical threshold are defined by applying fitting lines

The slope of J-E plot as in Fig. 5 should define whether ohmic conduction or ionic or space charge limited conduction [10] take place in the transport processes. Current density plot in this work shows variation of the slope as field increases. Notably for 30℃ data, the characteristic changes from a nearly ohmic regime (slope close to 1) to highly non-linear regime with a slope estimated to 2.3. The threshold takes place at about 13 kV/mm where charge transport behavior of LDPE changes. For higher temperature, it is difficult to decide if the threshold varies owing to the evolution in time of the response of the material.

4. Comparison to Model Outputs

4.1. Model Results

Fig. 6 shows a comparison between experimental and simulated current transients obtained at 20℃. It must be stressed here that the model has been applied with the currently available data set of coefficients, see [5], best fitted to measurements at room temperature, but without any attempt of later optimization of the parameters.

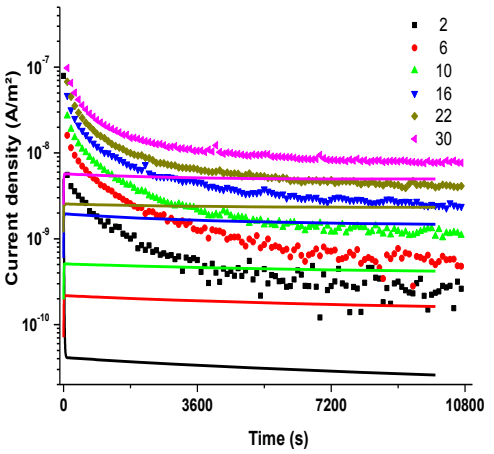

Fig. 6 Transient current density vs. time: comparison between actual measurements symbols and simulation (solid lines) at 20° C for fields of 2, 6, 10, 16, 22 and 30 kV/mm as shown in the legend

Results for the lower voltage are relatively noisy owing to the fact that the average current is small, of the order of 0.2 pA in average at long time. It can be seen from Fig. 6 that at measurement time longer than 2000 s, charge transport characteristics appear differently between lower field and higher field.

Simulation result at 20 ℃ shows increasing transient current as applied voltage rises. Results from the model reveal a steep transient in the first minutes followed by a slower transient over 1 h for the step at 2kV/mm and this step is not so pronounced for higher fields. The steeper decay is due to the fact that an initial density of charges is supposed to be present in the material and this was to cope with experimental electroluminescence measurements [5]. These pre-existing charges move under the effect of the applied field. The slower decay in current results from the injection at the electrode, followed by transport and trapping of the both type of carriers. For the other steps in field, the preexisting charge is that computed along the depolarization stages following the previous steps. At this stage of the model, orientation polarization processes are not included: they could be present and at the origin of the decay in the experimental current.

4.2. Discussion

As stated above, the transient part of the charging current has several reasons for being not reproduced, notably the fact that the orientation polarization contributions are not included in the model. This has been done recently in the case of poly (ethylene naphthalate), a polymer known for having strong dipolar response. Impedance spectroscopy data available in the frequency domain have been fitted to known relaxation functions according to identified relaxation processes. Then it was converted to the time domain and this orientation polarization contribution could be treated separately from the transport aspects. The translation to the case of LDPE is not straightforward, as it is a non-polar material and therefore polarization if any has to be related to polar residues as oxidized groups for example. Second, the weak magnitude of such processes would make it tricky to analyze in the frequency domain for conversion in the time domain. So, we are currently not in position to explicitly dissociate orientation polarization from space charge processes in LDPE. This, all the more that efforts in parameterizing the model at short time have been put more on electroluminescence features – reflecting charge recombination processes than on transient current.

The behavior at long time should in principle fit more directly to the experimental. However, comparing between experiment and simulation, the difference almost reaches one decade in quasi steady state part for 2 kV/mm. With increasing field, the difference tends to be less, but still is by a factor 2 for 30kV/mm. So, on the all, the model tend to over-estimate the non-linearity of the response of the material. This is so while the rough material for making films is the same as that used for preparing samples on which the model is based. One could argue on the necessity of refining the model such as integrating polarization and using the latest developments in the physical hypotheses in it [8]. However, we would like to make the point on the experimental features. The main differences, regarding current measurement results are that previous experiments [5] were achieved in dry atmosphere instead of air. Although polarization was 3 h, the selected field values for long polarization protocols were much coarser with data at 10, 40, 60 and 80kV/mm. Presently, the first source of inconsistency to be fixed is the difference in experimental results regarding conductivity data, which were an order of magnitude higher in [7-5]. One possible route is the method of preparation of the films: Ghorbani [9] showed that the nature of the films used as cover layer may indeed have a great influence on conductivity values.

These results point on the carefulness to be given in the preparation of samples and measurements on insulating materials, and more generally on the definition of the system that we intend to probe and model. Electrode nature and processing conditions constitute full field of potential discrepancy between experimental results.

5. Conclusions

Our purpose in this paper was to assess the robustness of the outputs of a charge transport model by comparing the model predictions to a set of experimental data obtained at various fields and temperatures on LDPE. Current-voltage characteristics reveal once more that a threshold at around 10kV/mm define a change in conduction mechanisms beyond which conductivity is clearly non-linear. Experimental data obtained at 20 °C have revealed a substantial deviation from results expected from the transport model. Perhaps one of the first conclusions is that one to be extremely careful in defining the system, i.e. material, processing, electroding, conditioning and measurement conditions as they may greatly impact the results. Second, there is interesting memory or pre-conditioning effects to control and understand. Part of it is of pure electrical nature, as previous charging effects on a given characteristic. In principle, if the model is complete, it should predict charge storage and subsequent impact on transport. A more difficult case to handle is thermal preconditioning effects, which are revealed here through a decrease of the measured current, but that would demand further investigation as its origin can be multiple, resorting to physical evolution of the structure or to moieties evacuation.

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