A NEW APPROACH TO THE MODELLING OF THE CONDUCTION PROCESSES IN A WATER TREE DEGRADED LDPE

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INTRODUCTION

A Debye-type conduction model based on the dielectric relaxation of independent dipoles for polyethylene was presented in [1]. A more realistic new approach to the modelling of these processes is proposed in this work. It is based on the assumption of interaction between dipoles following the "Many-Body Universal Model of Dielectric Relaxation [2,3,4]. We have tried to validate the proposed model in a LDPE affected by different levels of water tree degradation. The model is founded on several hypotheses: one of them takes as a basis the variations of permittivity with the degree of water treeing damage and others take into account the microscopic structure of the material. With these simplifying hypotheses the proposed model can account for the larger part of our experimental results. The application of the "Many Body Universal Model of Dielectric Relaxation" implies that for a long enough measuring time, a "slow down" process in polarization is activated: it is due to the mechanism of local redistribution of energy (flip-flop transitions) which results in a power dependence for polarization current, with a negative exponent whose absolute value is $m \in (0,2)$. Because of the mathematical complexity of this theory, we propose in this paper a modification of the model of non-interacting dipoles presented in [1] in order to take into account interactions. With the aid of the new model we have tried to explain the variations of parameters in the power current law (best-fitted dependence for the polarization current measurements) as a function of (a) the degree of degradation by water trees and (b) the polarization voltage. Finally, a discussion about the validation of the model and his range of applicability is performed.

MODEL

The mechanism of dipole orientation often is considered as responsible for polarization currents in solid dielectrics. Nevertheless, in a dielectric material in which no important dipolar groups are present, polarization currents could be attributed to ion jumps between equilibrium nearby-positions. Both mechanisms can be explained by means of a unique simplified model in which a charged particle can jump between two close positions separated by a potential energy barrier –two potential well– [5,2]. For this model and considering a plane-plane sample for relatively low applied electric fields, the variation of local polarization P with time is given by,

$$\frac{\partial P(x,t)}{\partial t} = -\alpha P(x,t) + \frac{\alpha b(x)}{kT} E(x,t) \qquad (1)$$

where E(x,t) is the electric field, α is proportional to the probability per unit time that one ion changes its equilibrium position in the absence of applied electric field and due to the thermal energy,

$$\alpha = 2\nu \exp\left(-\frac{W}{kT}\right) \qquad (2)$$

where ν is the vibration frequency of ions inside their wells, W is the height of the potential energy barrier which separates the two respective well sites and b(x) is given by,

$$b(x) = p^2 n(x) \qquad (4)$$

where the concentration of ions is represented by n(x) and the dipolar moment associated to one ion is represented by p. From (1) and considering that the contribution of ohmic conduction is irrelevant, we get a simplified expression for the current density,

$$J(t) = \varepsilon_0 \frac{\partial E(x,t)}{\partial t} + \frac{\partial P(x,t)}{\partial t} \qquad (4)$$

We will study the dielectric response to a step voltage given by,

$$V(t) = V_{DC} \cdot u(t) \qquad (5)$$

Due to the difficulty for the interpretation of the physical meaning in the coefficients of the power current response supplied by the "Many Body" theory –especially from an experimental/applied point of view–, we propose a simplified alternative approach: departing from the non-interacting dipoles model, we introduce a simple mathematical algorithm that accounts for dipole interactions. This new model also predicts a power current law for the response of the dielectric material (similar dependence of the experimental Curie-von Schweidler law, $I(t)=I_0 t^{-m}$) but this time, coefficients $\{I_0, m\}$ can be readily determined as easy

functions of the microscopic/macroscopic parameters of the material. Dipole interactions will be described by an "effective" potential energy barrier which grows with the measuring time t as,

$$W(t) = W_0 + \Delta W(t) \tag{6}$$

where W_0 is the potential energy barrier of the non-interacting dipoles model. Consequently, it implies that parameter α of the new model must depend on time t. Such a time dependence must be a decreasing one and its initial value must coincide with that of the non-interacting model, α_0. We propose next expression for $\alpha(t)$,

$$\alpha(t) = \frac{\alpha_0}{1 + \dfrac{t}{\tau}} \tag{7}$$

Homogeneous material

In the case of a homogeneous material, all the variables in equation (1) are independent from position, x. Furthermore, for t > 0 electric field is independent from time and is given by $E = V_{DC}/L$, then equation (1) becomes,

$$\frac{dP(t)}{dt} = -\alpha(t)\bigl(P(t) - P_s\bigr) \tag{8}$$

where,

$$P_s = \frac{bV_{DC}}{kTL} \tag{9}$$

By integration of (8) and making use of (7) and (4) we get,

$$J(t) = \frac{V_{DC}}{L}\varepsilon_o\delta(t) + \alpha_o P_s\left(1 + \frac{t}{\tau}\right)^{-\alpha_o\tau - 1} \tag{10}$$

Finally, if t >> τ , current is

$$I(t) = I_o\, t^{-m} \tag{11}$$

where,

$$\text{(i)}\; m = 1 + \alpha_o\tau = 1 + 2\nu\tau\exp\left(-\frac{W_0}{kT}\right) \tag{12}$$

$$\text{(ii)}\; I_o = S\alpha_o P_s\tau^m = S\alpha_0\frac{bV_{DC}}{kTL}\tau^m$$

which means that we get expressions that directly connect the microscopic/macroscopic variables of our LDPE sample with the actual values of our measured experimental current parameters $\{I_0, m\}$ [6].

Inhomogeneous material

In the case of a water tree degraded LDPE –inhomogeneous material– we cannot obtain , in general, an analytical solution from (1). Consequently we used numerical methods for the calculation and simulation of polarization, P(x,t), and polarization current, I(t). The hypotheses used for the numerical calculations are:
1. Parameter $\alpha(t)$ is independent of position, x.
2. Parameter b(x) which is related to the permittivity of the material depends on the position as,

$$b(x) = \begin{cases} b_{max} - nx, & x \in \left[0, \dfrac{g}{100}L\right] \\[2ex] b_{PE}, & x \in \left[\dfrac{g}{100}L, L\right] \end{cases} \tag{13}$$

where g represents the percentage value of degraded material, b_{max} is the value of b(x) that corresponds to a maximum relative permittivity whose actual value was chosen as 3.7, b_{PE} is the value of b in the non-degraded region, whose relative permittivity was chosen as 2.3 and n is the slope of the assumed linear dependence. The related permittivity values are taken from real permittivity measurements performed on polyethylene specimens degraded by water trees [7]. We also took into account for our calculations the relationship between the macroscopic variable relative permittivity, ε_r, and the microscopic parameter $b = np^2$,

$$\varepsilon_r(x) = 1 + \frac{b(x)}{\varepsilon_0 kT} \tag{14}$$

RESULTS

Experimental results

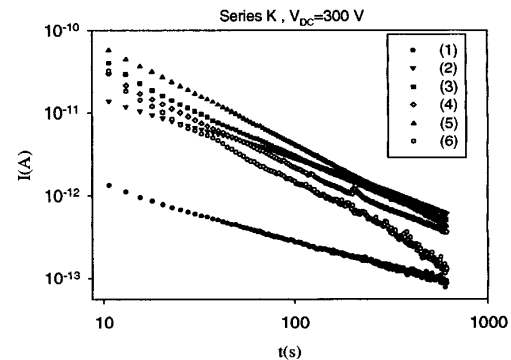In Fig.1 we show the results of polarization current

Figure 1. Polarization current measurements for laboratory LDPE plane-plane specimens (Series K), a polarization voltage of $V_{DC} = 300$ V and different water tree degraded width percentages, g: (1) 0%, (2) 14.8%, (3) 22.3%, (4) 34%, (5) 56.3%, (6) 70.7%.

measurements [6] performed on LDPE laboratory specimens of plane-plane geometry, with quasi-uniform layers of water tree degradation (g≡degraded width percentage). Polarization curves of Fig.1 were adjusted to the experimental Curie-von Schweidler law, $I(t)=I_0t^{-m}$, and their corresponding best fitted parameters are summarized in Table 1.

Table 1. Best fitted parameters $\{I_0,m\}$ for polarization curves of Fig.1 as a function of their corresponding degraded width percentage, g, and period of accelerated ageing, t_{env}.

t_{env}(h)	g(%)	I_0(A)	m
0	0	$6.251 \cdot 10^{-12}$	0.666
15	14.8	$1.1477 \cdot 10^{-10}$	0.8106
34	22.3	$4.1352 \cdot 10^{-10}$	1.0566
64	34	$3.5889 \cdot 10^{-10}$	1.0893
146	56.3	$1.2905 \cdot 10^{-9}$	1.2491
250	70.7	$6.1215 \cdot 10^{-10}$	1.3001

Another study of polarization current measurement was performed in new (Series M) and aged (Series J, g=63%) samples when varying the polarization voltage V_{DC} from 100 V up to 1000 V. Their corresponding $\{I_0,m\}$ parameters are summarized in Table 2. From them, the significant trend to saturation in exponent m, for both new (increasing variation) and aged (decreasing variation) LDPE samples, is highlighted in Fig.2.

Table 2. Best fitted parameters $\{I_0,m\}$ for polarization curves as a function of polarization voltage, V_{DC}.

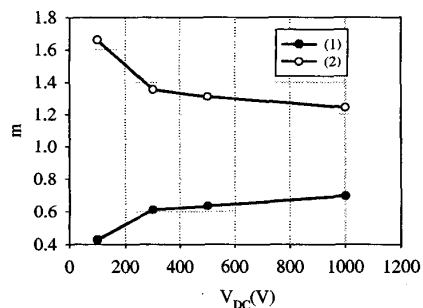| V_{DC}(V)	I_0(A)		m	
	Series M	Series J	Series M	Series J
100	$4.7826 \cdot 10^{-13}$	$1.3465 \cdot 10^{-9}$	0.42736	1.6603
300	$2.5357 \cdot 10^{-12}$	$1.6406 \cdot 10^{-9}$	0.61002	1.3546
500	$4.0321 \cdot 10^{-12}$	$2.4369 \cdot 10^{-9}$	0.63351	1.3102
1000	$1.0702 \cdot 10^{-11}$	$4.0661 \cdot 10^{-9}$	0.69758	1.2419

Figure 2. Detail of the tendency to saturation in the evolution of the exponent m in new (1) and aged (2) LDPE samples as a function of polarization voltage.

Simulation results

Experimental results are interpreted from the proposed model of interacting dipoles by using equation (11). Nevertheless, for small water-tree degraded layers, an interpretation of the exponent parameter m of experimental polarization curves is not possible as m<1. According to equation (12i), this model can only explain the current curves corresponding to a higher level of degradation which gives experimental exponents m greater than unity (m>1): i.e. the experimental results of Fig.1 that are related to ageing times: t_{env}= 34 h, 64 h, 146 h, 250 h. From numerical integration, we can obtain the values of the characteristic microscopic parameters of our conduction modelling (τ, α_0, W_0) as a function of the level of degradation in the insulating material, as shown in Table 3. Fig.3 is an example of the degree of adjustment between the current results I(t), obtained from numerical integration (curve (2)) and from the curve fitting (curve (1)) to the experimental data corresponding to a sample of degraded width g=22.3% (t_{env}= 34 h).

Table 3. Microscopic parameters from numerical integration (τ, α_0, W_0) as a function of the degree of water tree degradation, g.

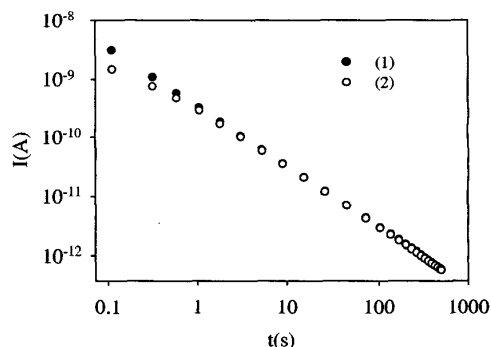| g(%)	τ	α_0	W_0
22.3	0.14	0.124	0.768
34	0.12	0.37	0.74
56.3	0.52	0.454	0.735
70.7	0.27	1.0385	0.713

Figure 3. Current results from numerical simulation (2) and from the linear curve fitting of experimental data (1) for a LDPE treed sample of degraded width, g=22.3% and microscopic parameters: τ=0.14, α_0=0.124, W_0=0.768.

From Table 3 we observe an increase for the value of α_0 with degradation which corresponds to a diminution in the potential energy barrier, W_0.

DISCUSSION

Evolution of I(t) with degradation

The diminution of energy barrier W_0 with degradation is a consequence of the increasing facility in the movement of charge carriers, which is due to the destruction of the internal structure of the polymer known as "water treeing" [8,9]. The increase of α_0 as an effect of the diminution of W_0 is given by equation (2).

Then, if equations (12) are supposed to be valid in a qualitative sense for the case of a water treed material, the increase of α_0 explains the increasing dependence of I_0 and m with degradation (Fig.1, Table 1 and Table 2). Concerning numerical calculations, there was a good agreement between measurements and simulation results of polarization currents in aged samples for ageing periods: 34 h, 64 h, 146 h and 250 h.

Evolution of I(t) with V_{DC}

(I_0) From equation (12ii) we consider that, basically, parameter I_0 is linearly-dependent on V_{DC} (i.e. dependencies on V_{DC} through parameters α_0 and τ are supposed to be less important or irrelevant). Consequently I_0 should increase with V_{DC} both in aged samples —in a qualitative sense— and in new samples —in an analytical sense–, as we found in our current measurements (see Table 2).

(m) New samples.- From equation (12i) we can explain the enhanced values of m as V_{DC} grows (Table 2, Fig.2) if we show the physical reason for the decrease of W_0 with V_{DC}: a higher applied voltage, V_{DC}, produces a higher internal energy of the system which means again an increased facility of movement inside the atomic-molecular system (both for the charge carrier jumps and for the rotations of molecular chains that constitute the polymer). The resultant effect is "just as if" the effective two-wells energy barrier , W_0, had a diminution. Naturally and furthermore, the superposition of the curve representing the two wells potential energy to the curve of the applied potential energy (application of V_{DC}) is equivalent to a physical diminution in the height of the potential energy barrier. Aged samples.- We should emphasize the fact that the more degraded the water treed LDPE, the more heterogeneous it becomes and a larger number of possible polarization mechanisms it acquires, i.e., the degraded LDPE can adopt a whole spectrum of possible different energy barriers, W_{0i}, one for each type of atomic-molecular species involved and/or process of dielectric relaxation. Such different mechanisms are available due to the state of degradation of the polymer but they will only be effective mechanisms —they will be activated– depending on the level of polarization voltage that is applied for each experience. If a higher DC voltage is applied, new polarization mechanisms with a greater height, W_{0i}, are activated, which implies from equations (2,12i) that the corresponding jumping frequencies $\alpha_{0i}=2v\exp(-W_{0i}/kT)$ are smaller in these processes and then that the decreasing slopes in the log-log representation, m_i, are smaller. The incorporation of those new mechanisms results in an average slope, m, that diminishes as V_{DC} grows (see Table 2, Fig.2). Although the process of diminution of a particular barrier height with the applied voltage described for new samples also applies to aged samples, it is clear that the effect of creation of new barriers in aged samples must be a predominant one.

CONCLUSIONS

A new conduction model that takes into account dipole interactions is presented from a modification of the two-wells Debye model. The proposed model provides easy analytical expressions for the dependencies of the experimental Curie-von Schweidler parameters on both microscopic and macroscopic variables of the LDPE plane-plane samples under test. From the evolution of these variables with water tree degradation width, g, and polarization voltage, V_{DC}, we could simulate polarization currents that had a good deal of accord to measurements in accelerated-aged LDPE specimens.

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