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# Charge Transport Model Considering the Presence of Ions in an XLPE Containing By-products

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**Abstract**— Chemical residues, generally resulting from material preparation, exist in the volume of polymer insulations and impact their dielectric properties. This is particularly the case in XLPE, where by-products from cross-linking and antioxidants exist. In the presence of an electric field, these chemical species can dissociate or ionize, giving rise to ionic charges. Like electronic charges, ions contribute to conduction and to space charge accumulation in the insulation under DC stress. However, it is difficult experimentally to distinguish the role of electronic and ionic species. In this paper, we simulate the impact of the presence of ions by modeling their generation and transport in an XLPE containing dissociable by-products. Ionic phenomena are combined with electronic ones in a bipolar charge transport model, assuming that ionic charges result from dissociation of neutral species in the insulation volume. The model is described, and simulation results are presented and discussed as regards the impact of the presence of ionic species on space charge and current.

**Keywords**—charge transport modeling, space charge, cross-linked polyethylene, ions generation, ions transport

## I. INTRODUCTION

Polymeric materials, especially cross-linked polyethylene (XLPE), are increasingly used as electrical insulation in power cables, due to their suitable electrical, thermal and mechanical properties, and the advantages they bring in terms of maintenance, manufacturing process, investment cost and environmental impact. However, the generation and accumulation of space charges in polymeric insulations under DC electric field is a risk for HVDC extruded cable systems in service conditions. These charges distort the electric field and can locally increase electrical stress in the insulation and damage it, leading to premature aging of the material and possible dielectric breakdown [1, 2]. The origin of these charges can be from injection processes at the electrodes, *i.e.* at the insulation/semiconductor interfaces, or internal generation mechanisms such as the dissociation or ionization of neutral species in the insulation. Ionic species originating from the dissociation of cross-linking by-products are known to be responsible for the accumulation of heterocharges in XLPE under DC stress [3]. The effects of ions on space charge distribution and current can be observed experimentally, but it is difficult in this way to differentiate electronic and ionic species or to know the type of ions and their quantity, as space charge measurement techniques only provide access to a net density of charges. Charge transport models, intended to help predict and understand the contribution of ionic charges in addition to that of electronic charges, unfortunately often fail to take into account the presence of ions in these materials. Some authors have proposed modified versions of the two-level of traps bipolar charge transport model to account for ionic species in XLPE [4, 5, 6]. Some of them [5, 6], treat ionic

charges analogously to electronic charges, *i.e.* they consider that ions can move, trap, de-trap and recombine in the dielectric as for electrons and holes.

In this work, ion generation and transport are modeled, combining with electronic phenomena, using a bipolar charge transport model which assumes that shallow traps are distributed over a single energy level, while deep traps follow an exponential distribution [7]. We assume that electronic charges are generated by injection at the electrodes and that ionic charges originate from the dissociation of neutral species in the insulation volume. The model is solved using a finite element method, and has been developed for plate of non-degassed XLPE insulation, *i.e.* containing by-products. Our objective in this paper is not, however, to reproduce the experimental measurement results, but rather to initiate work aimed at introducing the contribution of ionic species into this bipolar charge transport model and, at the same time, to show, on the basis of simulation results, the importance of ions on the electrical behavior of polymer insulations.

## II. MODEL DESCRIPTION

### A. Description of ion generation and transport

The bipolar charge transport model (BCTM) used has already been published recently in the literature for electronic charges, *i.e.* for electrons and holes [7], and will not be detailed in the present paper. In addition to electronic charges, the model also accounts for ionic species, positive  $A^+$  and negative  $B^-$ . We assume that there exist in the material neutral molecules, dissociated (before the application of the electric field) and/or dissociable (under the action of an electric field). Hence, considered ions  $A^+$  and  $B^-$  result from the dissociation of an initially neutral molecule  $AB$ . The density of molecules  $AB$  and ions  $A^+$  and  $B^-$  is driven by the following equation [4]:



where  $k_d$  and  $k_r$  are the dissociation and recombination coefficients, respectively. These two coefficients are neither equal, nor of the same unit ( $k_d$  is expressed in  $s^{-1}$  whereas  $k_r$  is in  $m^3.s^{-1}$ ). We have made the assumption that a thermodynamic equilibrium is reached before any experiment, and therefore, before any application of an electric field, there is the same density of ions  $A^+$  and  $B^-$ , uniformly distributed throughout the dielectric thickness. This means that the initial net charge density is null before any voltage is applied to the material. We consider that the initial density of neutral molecules, noted  $[AB]_0$ , is much greater than the initial density of ions, noted  $C_0 = [A^+]_0 = [B^-]_0$ . This would be consistent with results obtained for cumyl alcohol, a cross-linking by-product of XLPE, for which  $[AB]_0$  would be of the order of  $10^6$  C/m<sup>3</sup> whereas  $C_0$  is around 1 to 10 C/m<sup>3</sup> [8]. We

also consider that throughout the experiment, no additional ion or neutral molecule is created, apart from the ions  $A^+$  and  $B^-$  resulting from this dissociation process and their recombination into molecules  $AB$ . Recombination between ion and electronic charge is neglected.

Since thermodynamic equilibrium is initially assumed to be reached, we define an equilibrium constant (also called the dissociation constant) for this state, which is function of the density of species at equilibrium, according to the expression:

$$K_{eq,diss} = \frac{[A^+]_0[B^-]_0}{[AB]_0} \quad (2)$$

The application of an electric field to the material can cause ions  $A^+$  and  $B^-$  to move within the material with a mobility that we assume to be constant in this present paper for each ionic species, even if in reality, ion mobility depends on temperature, applied electric field and the density of these ions [9]. Positive and negative ions can also diffuse within the insulation under the effect of a concentration gradient. The neutral molecules, on the other hand, do not move under the action of an electric field, but can diffuse due to concentration gradient in the material. Thus, when an electric field is applied to the material, the density of ions and neutral molecules is no longer uniform across the thickness of the material and also varies over time. We introduce a second reaction variable, noted  $Q_{diss}$ , which, compared to the equilibrium constant  $K_{eq,diss}$ , determines the direction of the reaction in equation (1). This reaction variable depends on space and time. At any point in the insulation thickness and at any time, it is written:

$$Q_{diss} = \frac{[A^+][B^-]}{[AB]} \quad (3)$$

It is important to note that, as  $Q_{diss}$  varies along the dielectric thickness, the direction of the reaction will not be homogeneous in the material. At a given instant  $t$ , there may be dissociation in a certain region, while in another region the reaction goes in the direction of recombination. Finally, due to their large size (compared to that of electrons), we consider that all ionic charges as well as the neutral molecules cannot be extracted from the material.

### B. Model equations

In our BCTM, considering electronic and ionic species, the space and time dependent equations to solve are the following Poisson and transport-continuity equations:

$$\nabla \cdot (\epsilon_0 \epsilon_r E) = (+qn_{n\mu} + qn_{ht} + qn_{A^+} - qn_{e\mu} - qn_{et} - qn_{B^-}) = \rho \quad (4)$$

$$\frac{\partial n_a}{\partial t} + \nabla \cdot (n_a \mu_a E - D_{diff,a} \nabla n_a) = s_a \quad (5)$$

where  $n_a$  is the charge density (in  $m^{-3}$ ), the index  $a$  defines the type of charge: mobile electron ( $e\mu$ ), trapped electron ( $et$ ), mobile hole ( $h\mu$ ), trapped hole ( $ht$ ), ions ( $A^+$ ) or ( $B^-$ ).  $\mu_a$  and  $D_{diff,a}$  are respectively the mobility (in  $m^2 \cdot V^{-1} \cdot s^{-1}$ ) and the diffusion coefficient (in  $m^2 \cdot s^{-1}$ ) for each kind of charge, being electronic or ionic.  $E$  refers to the electric field (in  $V \cdot m^{-1}$ ) and  $\rho$  to the net charge density (in  $C \cdot m^{-3}$ ).  $q$  is the elementary charge (in  $C$ ),  $\epsilon_0$  the vacuum permittivity and  $\epsilon_r$  the relative permittivity of the material (equal to 2.3 for XLPE).  $s_a$  designates the source term for each charge carrier.

The mobility expression for electronic charges, assumed to be of the hopping type, is given in [7], and that for ionic carriers is considered constant. The diffusion coefficient  $D_{diff,a}$  for electronic and ionic charges is related to their respective mobility by Einstein's relation :

$$D_{diff,a} = \frac{k_B T}{q} \mu_a \quad (6)$$

where  $T$  is the temperature (in K). As for neutral molecules  $AB$ , their diffusion coefficient is expressed in the form [10]:

$$D_{diff,AB} = \frac{2 \times D_{diff,A^+} \times D_{diff,B^-}}{D_{diff,A^+} + D_{diff,B^-}} \mu_a \quad (7)$$

The source terms  $s_a$  model all the physical mechanisms taken into account but not related to transport (trapping, detrapping, recombination). The source terms for electronic charges have already been described in the literature [7]. For positive and negative ions, and neutral molecules, the source terms depend on the direction of the reaction (dissociation or recombination), and so on the reaction variable  $Q_{diss}$  :

- If  $Q_{diss} < K_{eq,diss}$  (dissociation), the source terms are expressed in the form:

$$s_{A^+} = +k_d n_{AB} ; s_{B^-} = +k_d n_{AB} ; s_{AB} = -k_d n_{AB} \quad (8)$$

with  $k_d$  the dissociation coefficient, assumed to be constant.

- If  $Q_{diss} > K_{eq,diss}$  (recombination), the source terms are of the form:

$$s_{A^+} = -k_r n_{A^+} n_{B^-} ; s_{B^-} = -k_r n_{A^+} n_{B^-} ; s_{AB} = +k_r n_{A^+} n_{B^-} \quad (9)$$

with  $k_r$  the recombination coefficient of ions, of the form:

$$k_r = \frac{q(\mu_{A^+} + \mu_{B^-})}{\epsilon_0 \epsilon_r} \quad (10)$$

### C. Model implementation

The model is developed and solved using the finite element method with COMSOL Multiphysics®. The physical module "Transport of Diluted Species" (TDS) is chosen to solve the transport-continuity equations (5) for the different species (electronic, ionic and neutral) taken into account. The mathematical module "Poisson Equation" (POEQ) is used to model the Poisson equation (4) and to couple it to the transport-continuity equations. For the time integration, the "Backward Differentiation Formula" (BDF) solver is selected. The sample has a thickness of 200  $\mu m$  and is meshed into 2000 finite elements of non-uniform length, the regions close to the electrodes being more refined.

## III. SIMULATION RESULTS

In the present paper, all simulations are performed on a non-degassed cross-linked polyethylene (XLPE), containing ionic species. The material is submitted to a DC voltage of 6 kV (equal to an applied electric field of 30  $kV \cdot mm^{-1}$ ), at a temperature of 25  $^\circ C$ , during 24 h. Values of the model parameters used for the simulations are reported in Table I. After simulation, a post-processing of the data has been performed and consists in adding the influence charges (*i.e.* capacitive charge and image charges) at each electrode.

TABLE I. MODEL PARAMETERS USED FOR THE SIMULATIONS.

Parameters	Notation	Value	Units
Injection barrier heights	$w_{i(e)}$ for electrons	1.23	eV
	$w_{i(h)}$ for holes	1.16	eV
Hopping barrier heights	$w_{hop(e)}$ for electrons	0.30	eV
	$w_{hop(h)}$ for holes	0.15	eV
Inter-shallow traps distance	$\lambda_{(e)}$ for electrons	$3 \times 10^{-9}$	m
	$\lambda_{(h)}$ for holes	$5 \times 10^{-9}$	m
Minimum depth of deep traps	$\Delta_{min(e)}$ for electrons	0.38	eV
	$\Delta_{min(h)}$ for holes	0.35	eV
Maximum depth of deep traps	$\Delta_{max(e)}$ for electrons	1.50	eV
	$\Delta_{max(h)}$ for holes	1.35	eV
Pre-exponential factors	$N'_{(e)}$ for electrons	$5 \times 10^{42}$	$\text{m}^{-3} \cdot \text{J}^{-1}$
	$N'_{(h)}$ for holes	$3 \times 10^{42}$	$\text{m}^{-3} \cdot \text{J}^{-1}$
Shape parameters	$T_{0(e)}$ for electrons	1400	K
	$T_{0(h)}$ for holes	1300	K
Adjustment factors for recombination	$r_1 (e\mu ht)$	0.8	-
	$r_2 (et h\mu)$	0.1	-
	$r_3 (e\mu h\mu)$	1	-
Initial density	$N_0$ for electronic charges	0.5	$\text{C} \cdot \text{m}^{-3}$
	$C_0$ for ionic charges	0.5 - 5	$\text{C} \cdot \text{m}^{-3}$
	$[AB]_0$ for neutral molecules	100	$\text{C} \cdot \text{m}^{-3}$
Constant ion mobility	$\mu_{A^+}$ for positive ions	$10^{-14} - 10^{-18}$	$\text{m}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$
	$\mu_{B^-}$ for negative ions	$10^{-14} - 10^{-18}$	$\text{m}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$
Dissociation coefficient	$k_d$ , concerning ions and neutral molecules	$10^{-5}$	$\text{s}^{-1}$

### A. Reference case : results for electronic transport only

Simulations have first been performed for electronic charges only, without taking ions into account. The net space charge density as a function of time and space is mapped in Fig. 1. The charge density profile shows an injection of electronic charges at both electrodes. Positive charges are relatively dominant in the material and have a higher apparent mobility than negative charges. However, the dynamics of negative charges shows a progressive penetration in the material volume over time, which tends to compensate the dominance of positive charges. Fig. 2 shows the evolution of simulated current density as a function of time. The current decreases, most significantly at short time, then tends to stabilize with time. After 24 h, the steady state is almost reached, the current density is quasi constant.

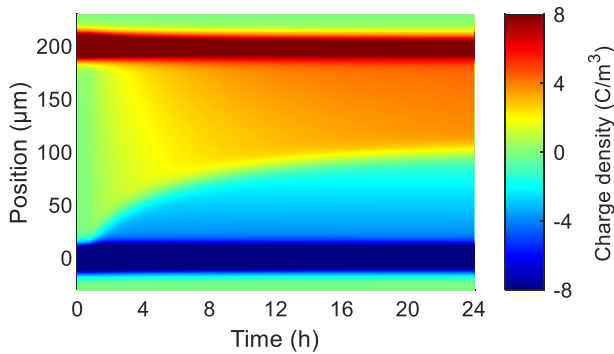


Fig.1 Simulated net charge density as a function of time and space when no ions are accounted for, for a 200  $\mu\text{m}$ -thick XLPE polarized at 30 kV/mm during 24h at 25°C.

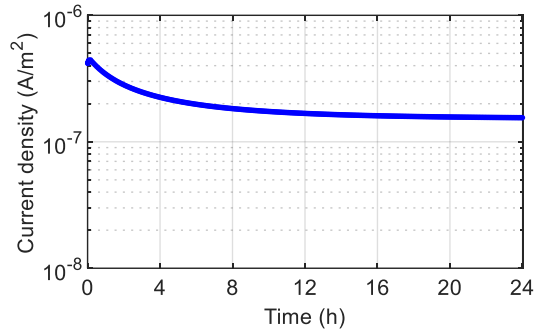


Fig.2 Simulated current density vs. time when no ions are accounted for, for a 200  $\mu\text{m}$ -thick XLPE polarized at 30 kV/mm during 24h at 25°C.

### B. Results for ion transport only

Simulations have then been performed with the assumption that only ions are present in the material. In this way, we can observe the impact of ion generation and transport on space charge distribution and current evolution, without this impact being disturbed or hidden by the presence of electronic charges. Fig. 3 and Fig. 4 show, respectively, the spatio-temporal evolution of the simulated net charge density and the temporal evolution of the simulated current density. For this simulation, we have used the parameters for ions reported in Table I, fixing the initial ion density at 2  $\text{C} \cdot \text{m}^{-3}$ , positive ion mobility at  $10^{-14} \text{m}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$  and negative ion mobility at  $10^{-15} \text{m}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$  (we therefore consider that positive ions are more mobile than negative ions). Charge density cartography reveals above all the known behavior of ions, which is the accumulation of heterocharges close to the electrodes. As the mobility of positive ions is chosen to be greater than that of negative ions, we might expect to see more positive than negative charges wandering around the material, but the profile “visibly” shows the opposite. This specific behavior is explained by the fact that positive ions, generated inside the material, move more rapidly towards the cathode, where they will accumulate, leaving behind negative ions. These negative ions, less mobile, will also move towards the anode, and accumulate. The time-dependent evolution of current density, plotted in Fig. 4, shows an increase in current with time in the presence of ionic species in the material. After 24 hours of experiment, the steady state is not reached, and the current continues to vary. This could be due to the fact that ions, once generated in the volume of the material, transport towards the counter-electrodes, or to the fact that diffusion of ions and/or neutral molecules is very important in view of the strong concentration gradients of these species in the material.

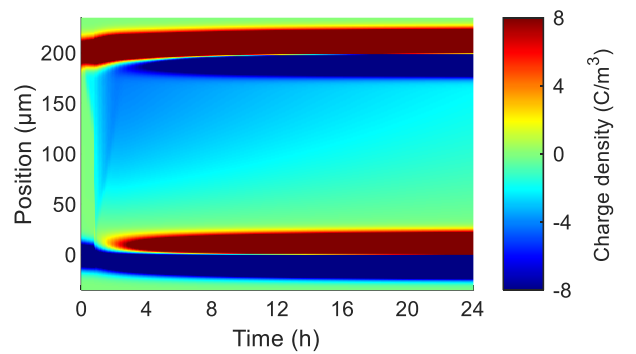


Fig.3 Simulated net charge density as a function of time and space when ions only are accounted for, for a 200  $\mu\text{m}$ -thick XLPE polarized at 30 kV/mm during 24h at 25°C.

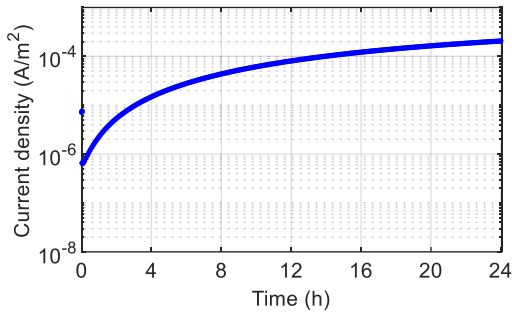


Fig.4 Simulated current density vs. time when ions only are accounted for, for a 200  $\mu\text{m}$ -thick XLPE polarized at 30 kV/mm during 24h at 25°C.

### C. Results for combined ion and electronic transport

Simulations were performed under more realistic conditions, taking into account both electronic charges and ions in the material. Simulations, whose results for net charge density and current are presented in Fig. 5 and Fig. 6, were carried out in the case where the parameter values for the mobility of positive and negative ions are respectively set to  $10^{-14}$  and  $10^{-18}$   $\text{m}^2\cdot\text{V}^{-1}\cdot\text{s}^{-1}$ , so as to have quasi immobile negative ions and mobile positive ions. The initial ion density,  $C_0$ , is still fixed at  $2 \text{ C}\cdot\text{m}^{-3}$ . The parameter values for electronic charges used are identical to those in the reference case (only electronic transport). The same protocol is used as for the cases in the previous sections (200  $\mu\text{m}$ -thick XLPE polarized at 30 kV/mm during 24 h at 25 °C).

The simulation results obtained in this case combining ionic and electronic charges, compared with the reference cases with electronic charges only, show the importance of the impact of the presence of ions in the material, ranging from the accumulation of heterocharges to a continuous increase in current, via less and less uniform charge distributions, *i.e.* with strong concentration gradients that become established in the thickness of the dielectric, and this, as a function of ion mobility values, but also of their density initially present in the material.

## IV. CONCLUSIONS

In this contribution, ion generation and transport are modeled and combined with electronic phenomena in a bipolar charge transport model with the aim of giving a boost to the modeling of ion-related phenomena in solid organic dielectrics, but also to confirm, on the basis of our model simulation results, the importance of the impact of the presence of ions on the electrical behavior of these insulating materials. Introducing ionic species into the model and taking their transport into account shows that the presence of ions in the material has a significant impact on the behavior of space charges and on the evolution of the current in the material, and therefore on the distribution of the electric field within the material. We have made the assumption that the only source of generation of positive and negative ions is the dissociation of neutral molecules initially present in the material. However, in reality, other sources of ionic charge generation exist, such as ionization, *i.e.* the creation of a positive ion and an electron from a neutral molecule, or species diffusion from semiconductors in contact with the insulation. These ion generation mechanisms can be considered and modeled in future work, as well as the dependence of ion mobility on temperature, electric field and ion concentration.

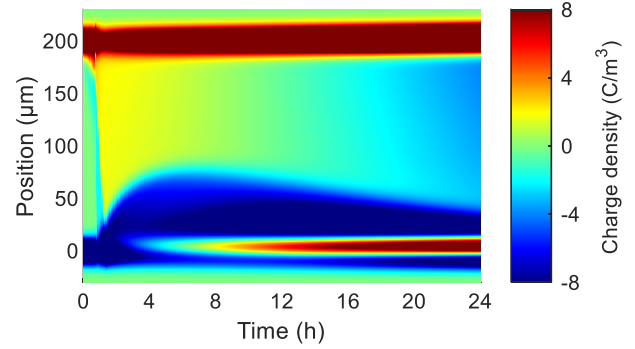


Fig.5 Simulated net charge density as a function of time and space when ions and electronic charges are accounted for. Mobility of positive and negative ions are respectively :  $10^{-14}$  and  $10^{-18}$   $\text{m}^2\cdot\text{V}^{-1}\cdot\text{s}^{-1}$ .

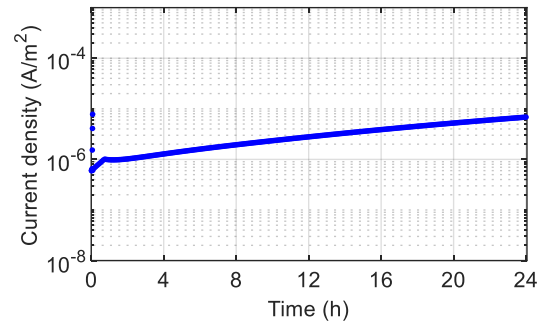


Fig.6 Simulated current density vs. time when ions and electronic charges are accounted for. Mobility of positive and negative ions are respectively :  $10^{-14}$  and  $10^{-18}$   $\text{m}^2\cdot\text{V}^{-1}\cdot\text{s}^{-1}$ .

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