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This is the final peer-reviewed author's accepted manuscript (postprint) of the following publication:

Published Version:

Suraci S.V., Spinazzola C., Fabiani D. (2022). Analysis on the impact of additives on space charge behavior of thermally aged XLPE plaques. Institute of Electrical and Electronics Engineers Inc. [10.1109/CEIDP55452.2022.9985288].

Availability:

This version is available at: <https://hdl.handle.net/11585/918791> since: 2023-02-28

Published:

DOI: <http://doi.org/10.1109/CEIDP55452.2022.9985288>

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This is the final peer-reviewed accepted manuscript of:

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In: 2022 IEEE Conference on Electrical Insulation and Dielectric Phenomena (CEIDP), Denver, CO, USA, 2022, pp. 41-44

The final published version is available online at:

<https://doi.org/10.1109/CEIDP55452.2022.9985288>

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Analysis on the impact of additives on space charge behavior of thermally aged XLPE plaques

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Abstract- This article investigates the space charge properties of XLPE-based materials characterized by different concentration and types of additives and fillers inside the polymeric compound. Materials were aged under three different temperatures (87 °C, 110 °C and 130 °C) for 24, 18 and 12 months, respectively. Space charge profiles of both unaged and aged materials were obtained through the Pulsed Electro Acoustic (PEA) method. Additives and fillers are proven to significantly impact the space charge behavior of the insulating material both in the unaged and aged states. The impact of antioxidants, together with their kinetics under thermal aging conditions, is analyzed and claims an effective containment of the degradation kinetics, keeping the accumulated space charge to low values.

I. INTRODUCTION

Crosslinked polyethylene (XLPE) is widely used as primary insulation for most of current extruded power cables [1]–[5], thanks to its excellent insulation properties along with good mechanical and thermal characteristics. Depending on the voltage to be insulated, XLPE materials must address various requirements e.g., stability over time, high dielectric strength [1]. In point of fact, high voltage (HV) insulation systems require high purity materials in order to contain space charge accumulation to very low values. Thus, the concentration of additives in these insulating materials is very low.

Indeed, additives and fillers are demonstrated to possibly act as trapping centers of accumulated charges, causing electrical field distortions, whose effect can lead to cable failure in the case of high electrical fields.

On the other hand, due to the significantly lower electric fields involved, low voltage (LV) systems do not require the same purity. For this reason, it is possible to fill the base polymer with various types of additives (e.g., plasticizers, antioxidants, flame retardants) according to the application requirements. It is estimated that the concentration of additives and fillers inside insulation systems of LV cables may reach 60%_{w/w} [6], [7]. As a consequence, the presence of these species in such high concentration impacts the polymer properties and their evolution with aging [8]–[10].

Antioxidants (AOs) are the most important ones since they ensure protection against oxidation. Hence, they efficiently delay the degradation mechanisms incepted by the

environmental stresses, guaranteeing sufficiently long operational time [11]–[13].

Flame retardants are present in high concentrations inside LV cables (~40%_{w/w}). They act as smoke suppressant releasing water after an endothermic chemical reaction.

In this paper, the authors investigate the impact of the additives on the space charge accumulation under accelerated thermal aging conditions, with the final objective of using space charge measurements as a technique to evaluate the degradation state of cable insulation.

II. EXPERIMENTAL SETUP

A. Materials and aging

Three different XLPE-based compounds, characterized by an increasing concentration of additives and fillers, are investigated in this article. Namely:

1. Plain XLPE (no additives)
2. XLPE+1 phr of phenol-based AO+1 phr of thioether-based AO
3. XLPE+1 phr of phenol-based AO+1 phr of thioether-based AO+50 phr of flame retardants (ATH).

The entire set of materials were subjected to accelerated aging under three different temperatures. Aging properties are reported in Table 1.

TABLE I
ACCELERATED AGING CONDITIONS

Aging temperature	Withdrawal time	Maximum aging time
87 °C	3,450 h	17,300 h
110 °C	2,600 h	12,960 h
130 °C	2,520 h	8,640h

B. Pulsed Electro-Acoustic (PEA) setup

Space charge properties were measured through the Pulsed Electro-Acoustic (PEA) method. In PEA, the sample is subjected to two coexistent applied voltages: a DC voltage V_0 and a pulsed voltage $V_p(t)$. The former allows the injection of charges in the tested material, while the pulsed voltage causes an acoustic pressure wave due to its interaction with charged particles. This wave is finally transduced into a voltage signal by a piezoelectric sensor. The trend of this signal in time is then related to the charge distribution in space by known parameters, such as the sound velocity on the specimen, its thickness, and the applied electric field [10 - 11].

In this work the applied DC electric field was set equal to 10 kV/mm at room temperature, in a shielded environment. The volt on phase lasted 10,800 s while the volt off phase lasted 3,600 s.

The value of total absolute stored charge density in the bulk at a chosen depolarization time is given by (1) [9-10]:

$$q_s(E, t_d) = \frac{1}{l} \int_0^l |q(x, E, t_d)| dx \quad (1)$$

where l is insulation thickness and t_d is the depolarization time, $q(x, E, t_d)$ is the space charge profile detected at time t_d .

The maximum value of q_s , namely Q_{max} , can be calculated at the beginning of depolarization once the poling electrode charge has been dissipated ($t_d=5s$).

The apparent trap-controlled mobility approximate equation is again reported in [9 - 10] and given by:

$$\mu(T) = \frac{\varepsilon}{q_{max}^2(t)} \cdot \frac{dq(t)}{dt} \quad (2)$$

where ε is the dielectric permittivity of the considered medium and $dq(t)/dt$ is the slope of the depolarization curve at time t .

Finally, the trap depth is derived by the approximated equation [10-11]:

$$\Delta U = kT \ln \left(\mu \frac{kT}{veR^2} \right) \quad (3)$$

where ΔU_i is the difference between two energy levels (trap depth), v is the attempt frequency, k is the Boltzmann constant, T is temperature, μ is the trap mobility, R is the mean distance between localized states ($5 \cdot 10^{-7}$ m), e is the electron charge ($1.6 \cdot 10^{-19}$ C).

III. RESULTS AND DISCUSSION

A. Total absolute space charge density

By applying Eq. 1, it is possible to derive the values of total absolute space charge density for Material #1 (Fig. 1.a), Material #2 (Fig. 1.b) and Material #3 (Fig. 1.c).

Some data relevant to samples aged at 130°C are not here reported because it was not possible to test them due to the excessive degradation these samples faced during the aging period.

From Fig. 1, it is possible to see that the total absolute space charge density generally increases with aging exposure. Nonetheless, the increasing trend is different depending on the polymer characteristics, such as the presence of additives.

Focusing on the values related to the unaged materials, one can notice that Material 1 and 2 depict very low values of space charge density (<0.1 C/m³). On the contrary, the introduction of ATH fillers in high concentration causes the raise of the electrical property value up to ~ 0.4 C/m³. This was expected for two main reasons. On one hand, the concentration of antioxidants (Material #2) is very little, so that negligible variations from the additive-free polymer (Material #1) may be recorded. On the other hand, the high concentration of ATH, causes the formation of wide interfaces due to the

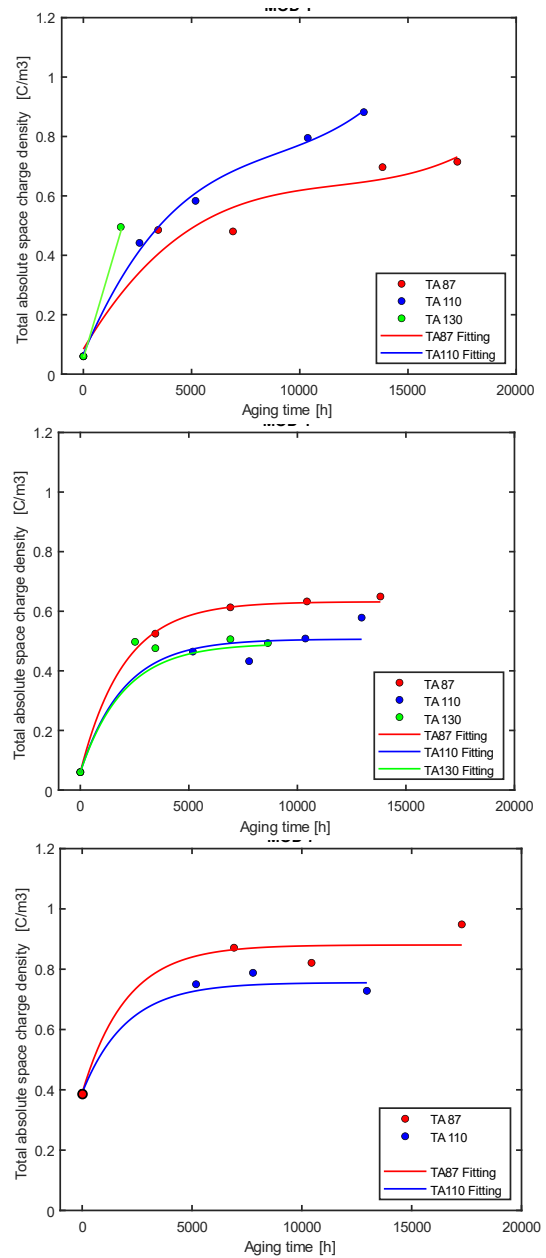


Fig. 1. Total absolute space charge density for (a) plain XLPE, (b) XLPE+AOs, (c) XLPE+AOs+ATH for the three aging conditions.

agglomeration of these species [6], increasing the amount of accumulated charge [16].

With reference to the response of the materials with aging, plain XLPE (Material #1) exhibits the biggest increase throughout the considered aging periods. The drawn trending curves suggest a continuous raise of the property also for long aging times. In particular, as the aging temperature increases, the increasing trend becomes steeper. This is relatable to the chemical degradation kinetics occurring at the different aging temperatures. As known, temperature catalyzes the degradation reactions according to the well-known oxidation reaction scheme [17]. Thus, at equal aging time, the concentration of degradation species is supposed to be higher in the case of higher temperature aging, as 130°C, in this case.

The products arising from the degradation reactions e.g., oxidized species may act as deep trap sites, as it will be discussed in Section III.B and enhance the accumulation of space charge in the specimen. In particular, as the concentration of these species increases, the total absolute space charge density is expected to raise as confirmed by Figure 1.a.

The introduction of antioxidants (Materials #2 and #3) significantly modifies the total absolute space charge density values with aging. Antioxidants hinder the degradation reactions involving oxygen, so that the concentration of oxidized products is very limited. This keeps the space charge density to low values as seen in Figures 1.c and 1.d.

During the first aging period, the charge quantity abruptly increases by $\sim 0.4 \text{ C/m}^3$. Further aging causes a stabilization of charge values, which reach a plateau at lower values than the ones obtained with reference to Material #1.

Referring to Material #3, Figure 1.c exhibits an accumulated space charge trend with aging very similar to the one discussed for Material #2. This behavior implies that the evolution of the total absolute space charge density with aging is ruled by the antioxidant kinetics e.g., consumption and conversion. Nonetheless, the values of space charge density are shifted upwards due to the presence of ATH, as mentioned above about the unaged materials.

Finally, it is worth noting that as the aging temperature increases, the plateau values are lower. This behavior is unexpected due to the faster degradation kinetics already described for harsher aging conditions. Nonetheless a possible reason for that may be found in the efficiency of the chemical reactions involving the antioxidants. Being thermally activated, the antioxidant degradation reactions may be more efficient in the case of higher temperatures as seen in [18], keeping the space charge accumulation evolution to lower values.

B. Trap depths

Figure 2 reports the values of trap depth as a function of aging times and conditions for the three tested materials. For all the materials, the variation of trap depth is very limited and substantially invariant from the first aging period on, regardless the aging condition. Indeed, one can notice a little initial increase of trap depths for Materials #1 and #2, probably due to the introduction of new degradation species e.g., oxidized chains or antioxidant products (in the case of Material #2), characterized by higher trap depths than the ones related to the unaged samples. It is worth noting that the number of the degradation species, and the corresponding traps, increase due to the oxidative phenomena taking place during aging. As a result, the increase of the total absolute space charge density (Figure 1.a and 1.b) may be explained by the increasing in number of trap sites depicting the same depths (Figure 2.a and 2.b).

On the contrary, Material #3 shows an initial higher value of trap depth ($\sim 0.85 \text{ eV}$) which is not modified by aging stresses. This behavior may be explained by the ATH filler properties. On one hand, as already discussed in the previous section, these fillers may be considered as deep traps due to their chemical formulation and tendency to agglomerate leading to the

formation of interfaces. On the other hand, they are inorganic species, hence, their degradation due to aging stresses may be considered as negligible with respect to the main polymer lattice. Thus, it may be assumed that the trap depths registered in Figure 2.c may be related to the ATH fillers, whose trap depth, being significantly high, hides the one coming from the oxidation products. Nonetheless, the number of shallower traps coming from degradation mechanisms increases with aging and it leads to the increase of the total absolute space charge density (Figure 1.c).

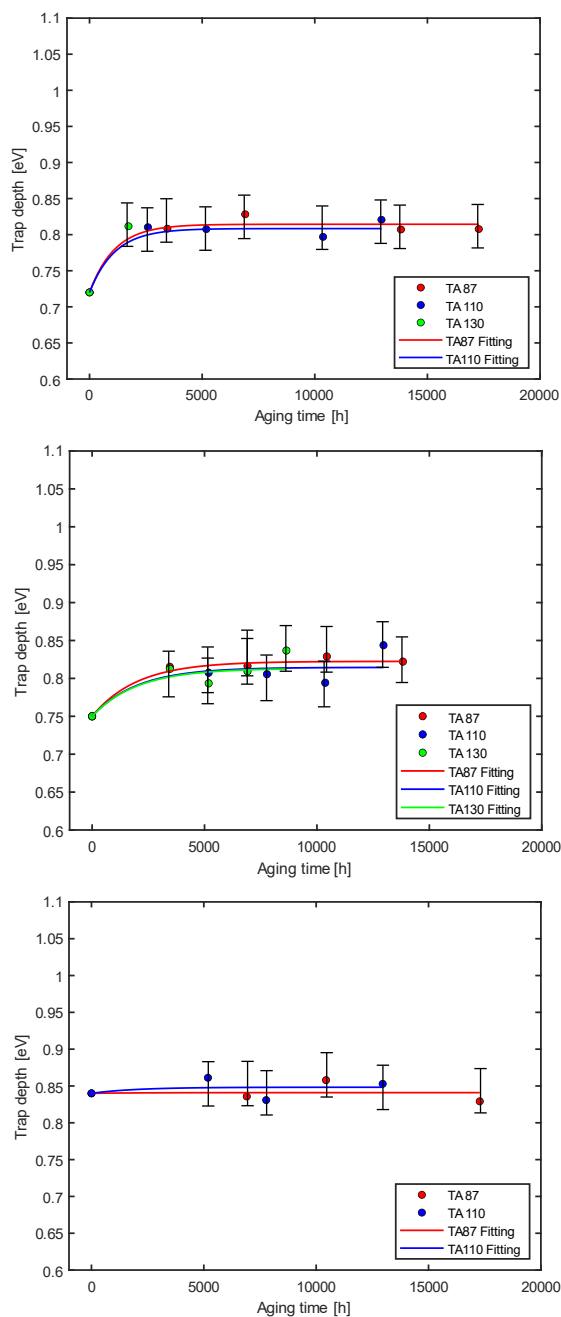


Fig. 2. Trap depth values for (a) plain XLPE, (b) XLPE+AOs, (c) XLPE+AOs+ATH for the three aging conditions.

IV. CONCLUSIONS

In this article, the space charge characteristics, and their evolution with aging of XLPE-based materials was presented and discussed.

The XLPE without any additive showed very good space charge properties i.e., low space charge density and low trap depth but the absence of antioxidants caused a huge increase of space charge properties during aging due to degradation mechanisms. The introduction of antioxidants caused a slightly worsening of the initial space charge characteristics e.g., higher space charge density. Nonetheless, it ensured acceptable values of the same quantities throughout all the aging periods and conditions considered, suggesting a beneficial action of antioxidants into containing the increase of space charge accumulation due to degradation phenomena.

Furthermore, the analysis of different aging temperatures allowed the evaluation of the effectiveness of the antioxidant additives in the prevention of aging degradation phenomena. It was demonstrated that a higher aging temperature would result into a more effective antioxidative property of the additive. Consequently, in the case of higher aging temperatures, the creation of degradation species is hindered by the more efficient antioxidants. As a consequence, the trap sites are limited in quantity and the corresponding total absolute space charge density is kept to lower values than the one of softer aging conditions.

Finally, ATH fillers, mainly used as flame retardants, influenced the polymer performance with aging by increasing both the total absolute space charge density and trap depths. Nonetheless, it was verified that the aging mechanisms for these materials are led by antioxidant kinetics thanks to the comparison between the material with just antioxidants (Material #2) and the full formulation (Material #3). Moreover, trap depth of ATH fillers resulted to be higher than the one of both base material and degradation species created during aging and it is supposed to hide the trap depth related to the degradation species.

Future work on this topic will include the analysis and modelling of other electrical properties e.g., conductivity and permittivity of the same materials.

ACKNOWLEDGMENT



The project leading to this application has received funding from the Euratom research and training programme 2014-2018 under grant agreement No 755183.

This publication reflects only the authors' view and the European Commission is not responsible for any use that may be made of the information it contains.

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