Control of Water Tree Length and Density in Cable Insulation Polyethylenes

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Abstract- **The goal of this paper is to study the influence of the chemical crosslinking of polyethylene on the water tree initiation and propagation in polymer insulation. For this, the water tree resistances of crosslinked and thermoplastic low density polyethylene were compared. Three types of crosslinked polyethylene systems were evaluated: one containing only peroxide and the other two having, beside peroxide, a tree retarding additive system. The results were compared with those obtained on their thermoplastic correspondents.**

The data show that there are differences in both the water tree length and density that can be ascribed to the polyethylene systems. However, only differences in the water tree density could be ascribed to the material form (thermoplastic or crosslinked). The observed results are consistent with differences, on microscopic level, in permittivities and local breakdown strengths.

I. Introduction

Crosslinked polyethylene (XLPE) has been extensively used in the last years in underground cables, replacing paper oil which was previously used for cable insulation. The main reason for the preference of this material over low density polyethylene (LDPE), is that by introducing crosslinks, the thermal and dimensional stabilities are improved, without affecting the electrical properties of the insulation [1,2]. When choosing the insulating material for power cables, especially at 6-36 kV, i.e. medium voltage (MV), besides its mechanical resistance and electrical performance, the water tree resistance is very important, XLPE having a superior service reliability compared to LDPE at MV [1,2,3].

In some cable constructions, the insulation material is exposed to water and this, in combination with the electrical stress, will cause water filled tree-like structures to grow. Thus water trees are produced [4]. These trees degrade the dielectric properties of the insulation (reducing breakdown strength and increasing dielectric loss) which then limits the service life length of the cable [1]. A question that is not fully resolved is whether the crosslinks themselves play a role in water treeing process in addition to them providing the high temperature operability of the cable. This paper describes the extension of previous studies to *material systems* that are much closer to those used in practice.

Our previous work introduced crosslinks by irradiation of polyethylene [5]. That analysis could not find any difference in the water tree growth. Thus, it might be concluded that crosslinks created in the solid phase do not lead by themselves to a tree retarding network.

The most common technique used for cable manufacturing is not irradiation but chemical (peroxide) crosslinking [6]. This is why it was of interest to perform a similar work using samples of this type. In addition to the crosslinks, this approach will add contributions from crosslinking by-products (acetophenone, etc.) and a morphology established in the melt phase. The transient effects of crosslinking by-products are well established [1].

II. Experimental

The water treeing tests were performed on samples of three *model material systems* which are designated A, B and C. Their composition can be characterised as:

A: thermoplastic LDPE sample;
XLA: sample A crosslinked with peroxide;
B: thermoplastic LDPE sample containing a non-polymeric (i.e. mobile) water tree retarding additive;
XLB: sample B crosslinked with peroxide;
C: thermoplastic LDPE sample containing a synergistic water tree retardant package;
XLC: sample C crosslinked with peroxide.

These systems were especially prepared in the laboratory for these studies.

The preparation of the thermoplastic (TP) samples A, B and C is carried out as presented in [4]. The crosslinked (XL) samples were prepared by melt pressing 20 min at 200 ^{o}C at 200 bars. The samples were cooled to room temperature, still under pressure, by a cooling rate of 15 ^{o}C/min. After crosslinking all plaques were degassed at 70 ^{o}C for 72 h to remove the crosslinking by-products. Thermoplastic samples were also treated to give a similar thermal, and hence morphological, history.

Water trees were grown in cells assembled by attaching the sample to be analyzed on a polyethylene tube (Fig. 1).
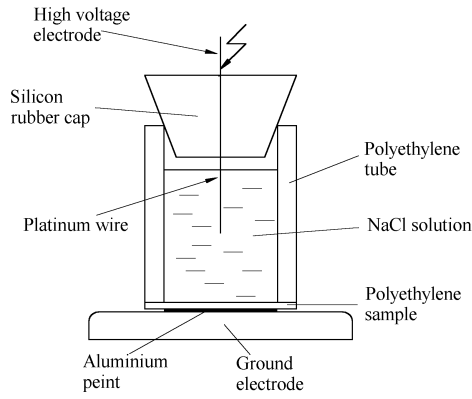
Fig. 1. Cell used to grow water trees

The initiation sites for water trees were created by pressing a sheet of abrasive paper (P240 grit 50 micron defect size) on one face of the sample, for 2 min at 50 MPa, at room temperature. This approach provided many potential initiation sites on the surface, but, as the preparation was carried out cold, the local morphology is unmodified. The trees were grown in 10 samples of each type, using a 0.1 mol/l NaCl solution at an electric field of 4 kV/mm, 5 kHz, for 25 hours, at room temperature (Fig. 1).

After ageing, the samples were dyed in order to facilitate the measurements of water tree lengths and number. A rhodamine solution at 60 °C was used for this treatment. Three 200 μm slices were microtomed from each sample and optically examined (Fig. 2). The trees are characterized in two ways:

Length - The lengths of all water trees from each slice were measured (Fig. 2). The average length L_a for each slice was used to determine the average water tree length L_k for each of the 10 samples.

Density – The number of trees in each slice were counted in the same manner as used for the tree lengths. These data provided the average water tree density (D_k).
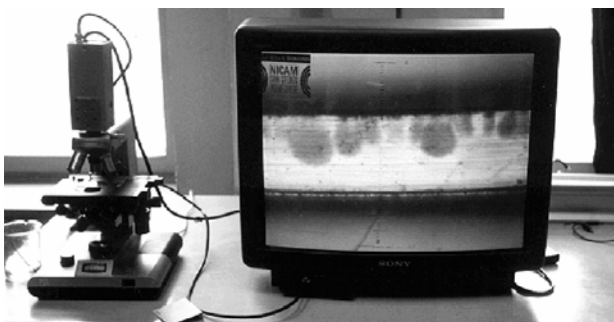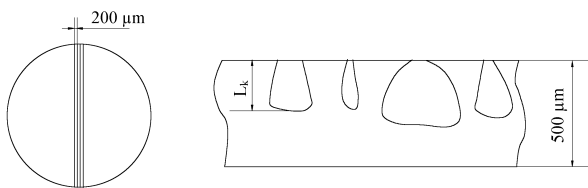

Fig. 2. Upper – diagrammatic representation of water trees. Lower - setup used to measure water tree lengths and water tree densities

III. RESULTS

Fig. 3 shows the 10 slice average water tree lengths (L_k) for the 6 materials evaluated in this study.
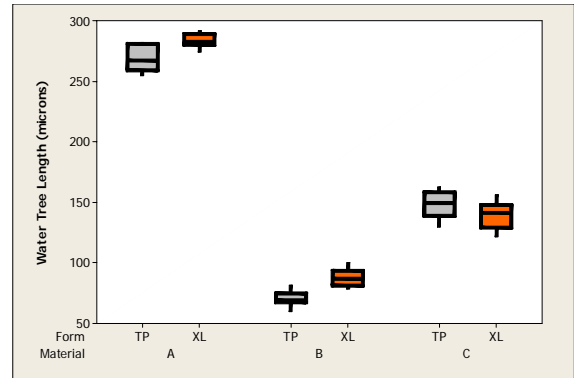

Fig. 3. Mean water tree length (L_k) data, represented in a box & whisker plot – the boxes enclose 50% of the data, the whiskers 100%, the central lines represent the medians.

The data are represented here in a non parametric (box and whisker) format. There is a clear separation of the data (see the boxes) for the main *model* systems (A, B, C) and for some of the *forms* (TP and XL).

The important issue to be determined is the statistical significance of the differences. We have chosen to accomplish this by using a multi-factor analysis of variance technique (MF–ANOVA), the data are adequately fitted by the Gaussian probability distribution to enable us to use this approach. The outcome of this analysis is shown in Table I.

The low numerical values of the **P Statistic** in Table I shows some clear statistical differences. We can be more certain (lowest P) that the material choice has an effect on the water tree length. Whereas we are less certain that *form* (TP or XL) has an effect, however a P statistic of 0.006 still shows a significant difference.

The optical system is also able to determine the number of water trees that have been initiated on the surface. Cursory examination shows that water trees are not initiated from every surface defect, and there are large differences between the samples. The water tree density data were also analyzed using the Gaussian distribution and Table II shows the mean densities. The results of the analyses for both the water tree lengths and the water tree densities are brought together in Fig. 4.

TABLE I
MF – ANOVA FOR WATER TREE LENGTH DATA: FORM – THERMOPLASTIC OR CROSSLINKED, MATERIAL - A, B, C

Source	Degrees of Freedom	Sums of Squares	F Ratio	**P Statistic**
Form (TP or XL)	1	1050	8.1	**0.006**
Material (A, B or C)	2	411827	1592	**0.000**
Error	56	7241		
Total	59	420118		

TABLE II
WATER TREE DENSITIES

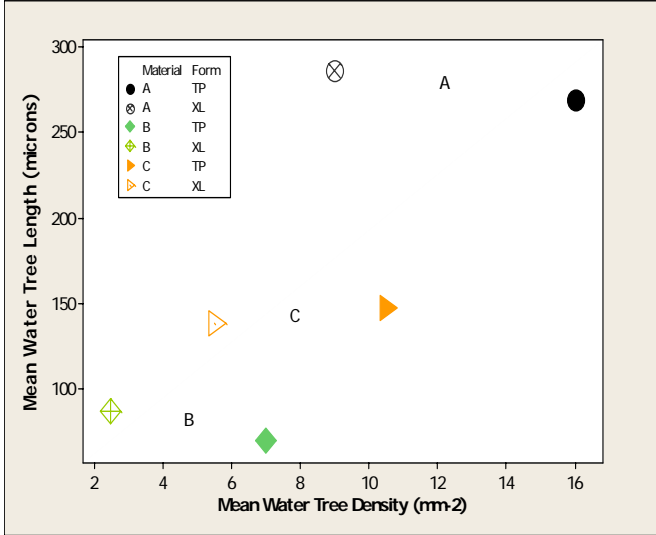| Non-crosslinked samples	Mean Water Tree density [mm^{-2}]	Crosslinked samples	Mean Water Tree density [mm^{-2}]
A	16	XLA	9
B	7	XLB	2.5
C	10.5	XLC	5.5

Fig. 4. Relationship between mean water tree density and mean length for the selected *model material systems* and *form*.

Inspection of the means show that there are both large and significant differences in the water tree density for both of the experimental factors studied here *form* and *material system*.

IV. DISCUSSION

The effects of the factors *material system* (A, B, C) and *form* (TP or XL) are summarized in Table III.

In practical applications of cables the main interest is the time T_w for the trees to grow sufficiently so that the dielectric is weakened such that failures by electrical trees are much more likely. The time T_w will have two components:

- The initiation time T_i, investigated by means of water tree density;
- The time for the water trees to grow <u>once</u> they are initiated – T_g, investigated by means of water tree length measured after a fixed time.

TABLE III
THE EFFECT MATERIAL SYSTEM AND FORM ON WATER TREE GROWTH

	Material System	Form
Length	Large	Small
Density	Large	Large

A. Water Tree Length

The results of Fig. 3 and Table I lead us to conclude that once initiated, water trees grow at a similar speed (same lengths in these fixed time studies) in both thermoplastic and crosslinked systems. This is not so surprising if the following arguments are considered:

- Trees grow, predominantly in the amorphous regions of the polymer,
- The permanent network crosslinks are located within the amorphous regions,
- The morphology within the amorphous region is likely to be very similar between both thermoplastic (TP) and crosslinked (XL) forms since it is approximately 300 carbon atoms between the crosslinks.

Thus, on a nanometer scale it might be equally easy to perform the necessary deformations of the entangled chains to build the narrow tree channels in both *forms*.

It is well accepted that the growth of the crystallites, as the polymer cools from the melt, will commence in areas with no crosslinks and will also tend to exclude the additives. This will tend to concentrate the additives within the amorphous regions; a form of the "zone refining" (a method common in solid state physics of purifying a solid by passing it through an induction heater; this causes impurities to remain in the molten or non crystalline sections). Thus the observed material effect will be most likely due to steric or dielectric effects of the additives operating on the 10 – 100 nanometer range. One effect that can account for the observed features is the heightened differences in permittivities of the amorphous and crystalline material regions. This is due to the polar nature of the additives in *model* systems B and C. The raised permittivities in the amorphous regions of B and C will result in lower local stresses in these regions. The compensating stress will be raised in the crystallites; however it is much more difficult to grow trees in this region. Consequently the energy available for deforming the polymer chains in the amorphous regions is very much reduced: thus shorter trees are grown in different *material systems*.

B. Water Tree Density

The effects on tree density of the *model material systems* (A, B, C) and the *form* (thermoplastic and crosslinked) are clear and striking. The best explanation for these density effects is the modification of the tree inception processes, especially considering that not all of the defects on the surface lead to water tree inception. In the cases studied here, the *material system* and *form* combine to reduce the likelihood of inception. In fact the similarity in tree lengths strongly suggests that this is a spatial effect rather than an inception time effect. In the latter case we would expect to find a wide scatter in the tree length data, and not standard deviations of 5 to 10% of the means. In this case we can interpret the data to show that there are fewer sites that initiate water trees for the XLB and XLC combinations than for TP A as an example (Fig. 4).

One initial postulate was that the crosslinking affects the local hardness, making it more difficult to produce surface defects. Although crosslinking does reduce the crystallinity, this change is only a few percent, which is too low for density differences on the dimension scale of the sandpaper used to produce the defects. In addition, mechanical testing shows that the presence of additives in the amorphous regions tends to facilitate chain movement (sometimes referred to as *reptation*, - the diffusive snake-like motion of a polymer along its tube of constraints, is the basic mechanism of long-time relaxation in polymer solutions and melts). Thus, we would expect systems B and C to be worse performers than A; however this is not the case. Thus we must conclude that the initial postulate is unlikely.

The most probable explanation is based on the spatial variation of the probability that a defective location will actually start to propagate a tree. The local probability will depend upon the local electrical parameters: breakdown strength E_B and stress E. As we have previously described the presence of additives in systems B and C will increase the permittivity and thus decrease the stress in the amorphous regions; this makes tree inception less likely in B and C than in A. However this element alone would predict that we would not have a crosslinking effect, which we clearly do have. Thus we believe that the crosslinks modify the local stress strain properties and toughness properties. These in turn will, via the electrokinetic considerations described by Lewis [7], increase the local critical strength E_c for crack initiation:

$$E_c = \left[\frac{2 \Im Y}{k \varepsilon^2 c} \right]^{1/4}, \qquad (1)$$

where $\Im$ is the fracture energy and Y the Young's Modulus at the nanometer scale and c is the half length of the initiatory crack.

Thus if, as we know, the crosslinks increase the Young's Modulus in the amorphous region then the local breakdown strength will be increased. The increasing breakdown strength will make tree inception less probable, thereby reducing the water tree density.

It is worth commenting that once the tree has started to grow a range of other phenomena will begin to operate (discharges, additional stress enhancements, etc.) upon which the mechanical properties will have little impact. This would still be consistent with the small effects of crosslinking on the tree growth rates (Section IV.B).

V. CONCLUSIONS

This work has shown that both crosslinking using organic peroxides and the material system (water tree retardant additives) affect the time for water tree development in cable insulations. The time depends upon initiation and growth ($T_w = T_i + T_g$).

The *material system* affects <u>both</u> the initiation (T_i) and the growth (T_g). *Material systems* B and C have significantly longer times (T_w) to grow large water trees than A.

The *form* (TP and XL) only affects the time T_w through its action on the initiation of the water trees (T_i). Crosslinking has only a small, though significant, effect on the growth (T_g) of large water trees once they are initiated: times are lowered for material A but increased for B and C.

The longest times for water tree growth, thereby implying highest cable reliability, come from the crosslinked versions of materials. There is also a significant effect of *material system*. Thus the performance of the crosslinked systems B and C is significantly superior to the LDPE system A.

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