EFFECT OF CROSS-LINKING ON THE ELECTRICAL PROPERTIES OF LDPE AND ITS LIGHTNING IMPULSE AGEING CHARACTERISTICS

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Abstract: Cross-linked polyethylene (XLPE) is commonly used within high voltage cable insulation. It has improved thermal and mechanical resistance compared to normal low density polyethylene (LDPE). However, the cross-linking process may also vary the electrical characteristics of the material. This paper investigates changes in electrical properties of one type of LDPE before and after cross-linking. The effective lightning resistance is also considered, as the application of repetitive lightning impulse over-voltages can be a factor in insulation material ageing of high voltage cables. The material was cross-linked using trigonox-145 peroxide with controlled concentration. Samples were moulded to have a Rogowski profile and gold coated to make sure that they are evenly electrically stressed. Obtained results show that there are reductions in both space charge injection and the permittivity of the material after it is cross-linked. The breakdown strength of the material was also improved. However, the samples studied are more susceptible to ageing due to lightning impulses.

1 INTRODUCTION

Over last two decades, oil impregnated paper cable insulation has been replaced by polymeric insulation material. Cross-linked polyethylene (XLPE) is used within high voltage cable insulation. It has improved thermal and mechanical resistance compared to normal low density polyethylene (LDPE). LDPE can be cross-linked using peroxides at elevated temperatures. However, the cross-linking process may also vary the electrical characteristics of the material due to the physical change in material itself and also the accumulation of residual by-products [1]. Impulse over-voltages are very common phenomena in electric power systems. A switching impulse is created by a switching surge or local fault while a lightning impulse is due to lightning strike to an overhead line. The application of repetitive lightning impulses can lead to accelerated ageing of extruded polymeric cables [2-4]. The results show that there may well be a reduction in electric field strength of the insulation of a power cable that experiences a lot of lightning impulse over-voltages. The effect of a lightning impulse is dependent on the molecular structure of material. This paper highlights some changes in electrical properties of LDPE before and after crosslinking by peroxide. The analysis of the behaviour of the materials that experience lightning impulses will give a better understanding of the impulse ageing process. In this work, moulded discs with a Rogowski profile have been manufactured in both LDPE and XLPE. The samples then have been electrically aged using standard lightning impulses. A real-time software based monitoring tool has been designed to control the impulse wave-shape and monitor the ageing process. The electrical properties of materials have been considered by breakdown measurements, dielectric spectroscopy and space charge measurements.

2 MATERIALS USED AND SAMPLE PREPARATION

The material used is the low density polyethylene LD100 from Exxon Mobil Chemical for blown film applications with a melting temperature from 160°C to 180°C. The material has MFI of 2g/10min (ASTM D 1238) and a transition temperature of 103°C. This is a low density (0.923g/mm^3) branched polyethylene. Trigonox145-E85 is the mixture of 85% 2,5-Dimethyl-2,5-di(tert-butylperoxy)hexyne-3 and mineral oil (**Figure 1**) from AkzoNobel . The mixture is liquid at room temperature and was stored in the fridge. It can be used as the initiator for the crosslinking process of polyethylene. This peroxide is more stable with increasing temperature than Dicumyl peroxide (DCP). The half-life of trigonox145 at 150°C is about 45 minutes and less than 1 minute at 200°C where as the half-life of DCP is 1 minute at 170°C. The LD100 pellets were soaked with the Trigonox145 to create cross-linkable polyethylene (trigonoxLD100). This was performed under room temperature for 5 hours and followed by an hour for drying out at 70°C in an oven. This process results in a concentration of 1.1% trigonox145 (XL1). Under 70°C for 5 hours, the process results in a 3.3% trigonox145 concentration (XL3). The trigonoxLD100 was safely heated at 150°C for 10 minutes before the pressure was applied (normally 1.5 to 2 tons) for about 1minute. After releasing the pressure, the mould was moved to another press at 200°C to cross-link sample for 10 more minutes (this ensures more than 70% of gel-content would be formed). The decomposition products formed during the cross-linking (**Figure 1**) are gaseous so no pressure was applied. After the cross-linking reaction, the mould was quench cooled using tap water and the sample released from the mould. The XL1 sample was made from LD100 with 1% trigonox addition and crosslinked for 5 min at 200°C. For LDPE, the pellets were just melted at 180°C before a pressure of 2 tons was applied. The mould was then quenched cooled with tap water.

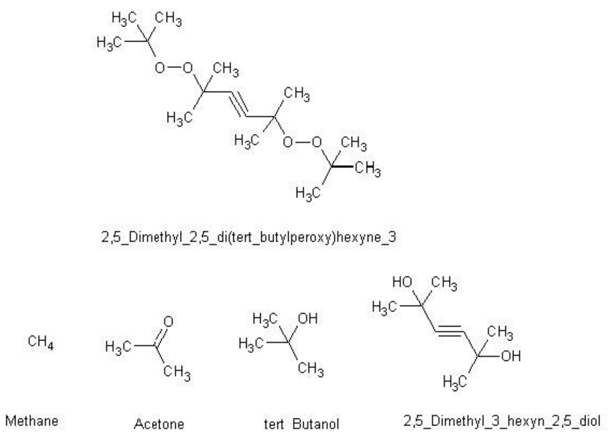

Figure 1: Trigonox-145 and cross-linking by-products

The moulded samples have the shape shown in **Figure 2**. The top surface of the sample follows a Rogowski profile, while the bottom surface is flat. Samples for breakdown and dielectric spectroscopy were gold sputter coated to improve the sample-electrode contact. The used for space charge measurement were left uncoated to prevent flashover.

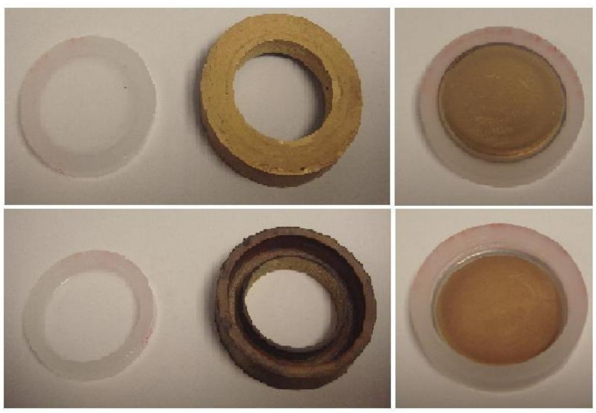

Figure 2: Samples and mask for coating

3 EXPERIMENTAL PROCEDURES

The samples after moulding experience an impulse ageing process. A set of 3000 negative lightning impulses with front/tail times of 1.2/40μs respectively were applied to create a peak electrical field of 85kV/mm across the samples. The ageing process is controlled by a Labview program which manages both the lightning impulse parameter calculation and ensures safety if breakdown occurs. Samples were aged in silicon oil under room temperature. An external optical trigger was implemented to provide a fixed impulse generation rate. Approximately 33 impulses were generated each minute. Samples after impulse ageing were used in either breakdown measurements, dielectric spectroscopy or space charge measurements. Sample breakdown strength was measured using a step up transformer which generates AC voltages with a ramp rate of 100V/s [5]. Dielectric spectroscopies of samples were measured using the Solartron system including 1296 dielectric interface and 1260 impedance analyser. The measurements were taken over the frequency range from 10^{-1} to 10^{6}Hz. A pulse electro-acoustic (PEA) experiment was used to measure the space charge profile [6]. Samples were stressed with an electric field of 34kV/mm. The average charge density (without sign) in the material during charge decay was measured and results curve fitted using a power law.

4 RESULTS

The breakdown strengths of aged and un-aged LDPE samples show no significant difference from a statistical point of view (**Figure 3**). The 90% confidence regions overlap over the whole spectrum. The mean value for breakdown strength of virgin LDPE is about 98.8kV/mm, whereas after 3000 lightning impulses the aged LDPE mean value is 98.4kV/mm. Therefore, it can be said that LD100 is not sensitive to lightning impulses.

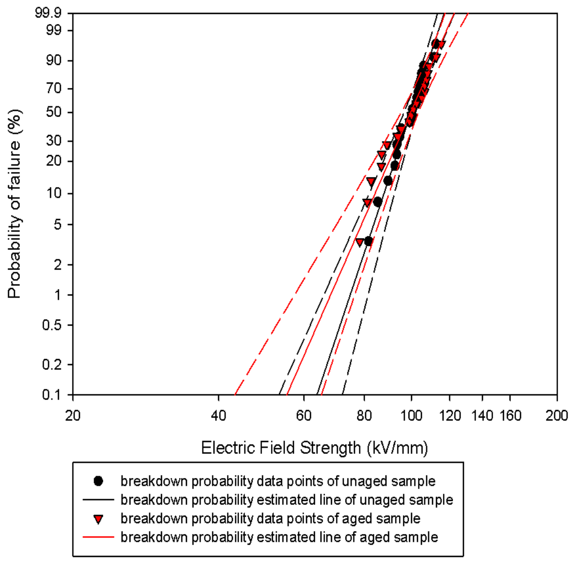

Figure 3: Breakdown strength of LDPE

The XL3 has the same LD100 as the base material but provides a much better performance with a mean breakdown strength of 129kV/mm (**Figure**

4). It suggests that the added peroxide trigonox-145 or the cross-linking process improves the electrical strength. However, the material becomes more sensitive to lightning impulse ageing as the aged samples have lower mean breakdown strength (121kV/mm).

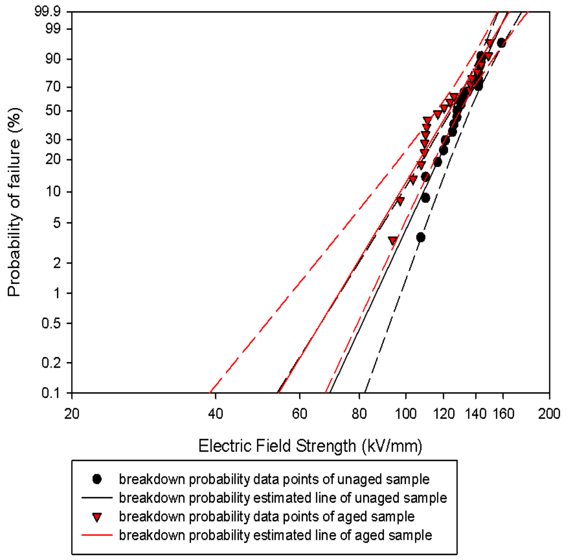

Figure 4: Breakdown strength of XL3

The obtained results for dielectric spectroscopy of the samples are shown in **Figure 5** and **6**. For LDPE (**Figure 5**), the loss factor of the aged sample is higher than the un-aged sample over a certain frequency range. However, there is no clear difference at power frequencies which were used for the breakdown measurements. The real parts of the relative permittivity are also constant with no difference between samples. The measured value of real relative permittivity is about 2.6, this is high compared to normal LDPE (about 2.3-2.4). This may be the consequence of the quenching operation during sample preparation, the smaller molecular weight and the presence of more branches on the main chains. All these lead to a less dense structure and molecules can move more easily under the application of an electrical field. The insensitivity of impulse ageing of this LDPE may be because of the disorder in the amorphous regions after quenching, which dominates the properties of the material and overshadows any effect of impulse ageing. After cross-linking by peroxide, the real part of permittivity reduces from 2.6 to 2.3 (**Figure 6**). The cross-linking limits the movement of chains by linking chains to create a bigger structure. The slope of loss factor in the low frequency region of XL3 is a bit steeper as the conduction of by-products may contribute to losses. The losses for aged XL3 are very much the same as the virgin sample except at higher values of the low frequency region. Therefore, ionic conduction can occur and increase losses. The real part of the permittivity increases after 3000 lightning impulses. With no appearance of oxidation, this increase could be due to chain scission and molecules can then move more easily within the bulk.

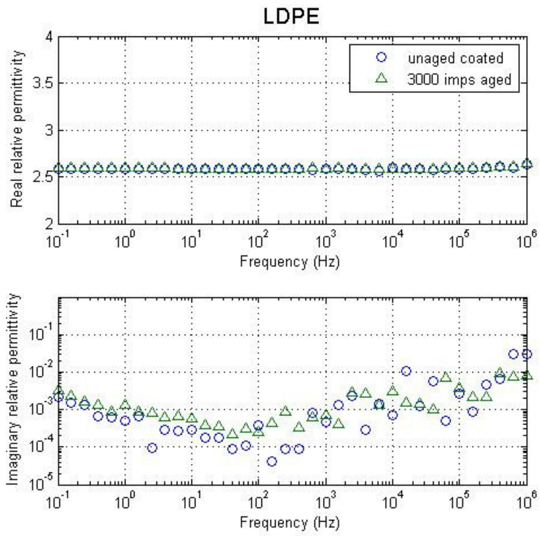

Figure 5: Dielectric spectroscopy of LDPE

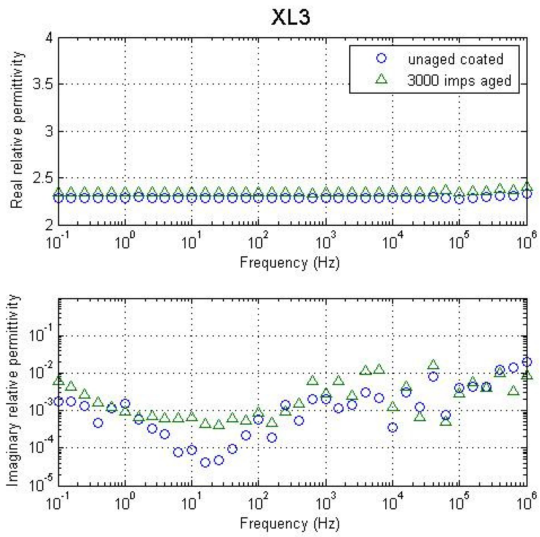

Figure 6: Dielectric spectroscopy of XL3

Figure 7 and **Figure 8** show the space charge profiles of LDPE samples. There is a significant amount of positive hetero-charges formed in the LDPE near the cathode. Previous work showed similar formation of hetero-charges in LDPE if the material is quenched cooled [7].The obtained results indicate that hetero-charge forms within about 10 minutes. After that, the charge profile remains constant for the next 50 minutes. The charge gradually decays after the applied field is removed. After an hour, a lot of charges still exist in the bulk. The lightning impulse aged LDPE shows a similar charge profile pattern as the virgin LDPE. The difference in peak charges for two samples can be ignored. The amount of charge in aged and virgin samples after 60 minutes poling is the same but the charges in the aged sample decay more slowly (**Figure 12**)

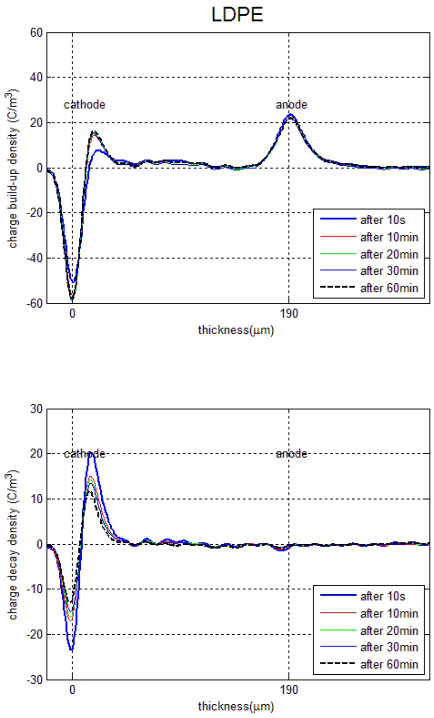

Figure 7: Space charge profile of LDPE

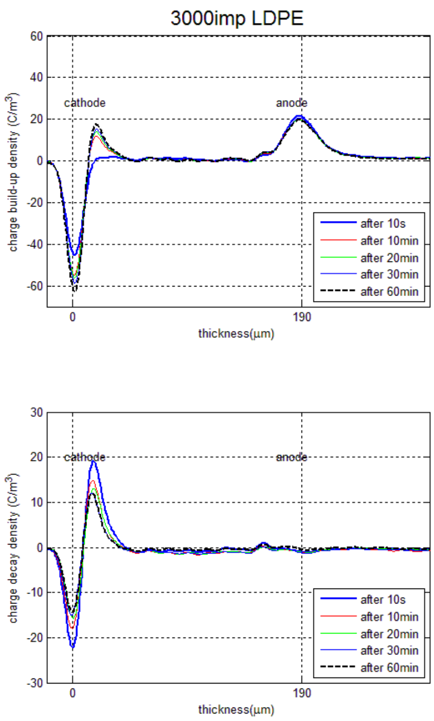

Figure 8: Space charge profile of impulse aged LDPE

Charge profiles of XL1 and XL3 are shown in **Figure 9** and **10**. The result shows bipolar charge injection takes place with significant positive space charge in XL1. The positive charges are attracted to the cathode but limited by the negative charges from cathode. With the same electrical field stress of 34kV/mm as used with LDPE samples, very little positive charge formed near the cathode in the bulk of XL3 sample. A small amount of negative charge was observed near the anode. As shown in **Figure 12**, the increase in amount of peroxide content for crosslinking from 0 to 3% reduces charge accumulation in XLPE but the rate of charge decay also decreases. It suggests that the effect of crosslinking process includes both changes of morphology and existence of by-products in the bulk of the material.

There is a difference in the space charge patterns of the impulse aged XL3 and virgin XL3. Positive charge formation is more consistent and there is a deeper penetration into the bulk of the impulse aged sample (**Figure 11**). The shape of bulk charge is similar to results reported previously for impulse aged HDPE [5]. The amounts of charge formation are quite small for both virgin and impulse aged XL3. The charges in virgin XL3 are concentrated near to the electrodes where the charges spread through the whole sample. The average total charge in **Figure 12** shows that there is a larger amount of charge in the impulse aged XL3 and this charge decays slower than virgin XL3.

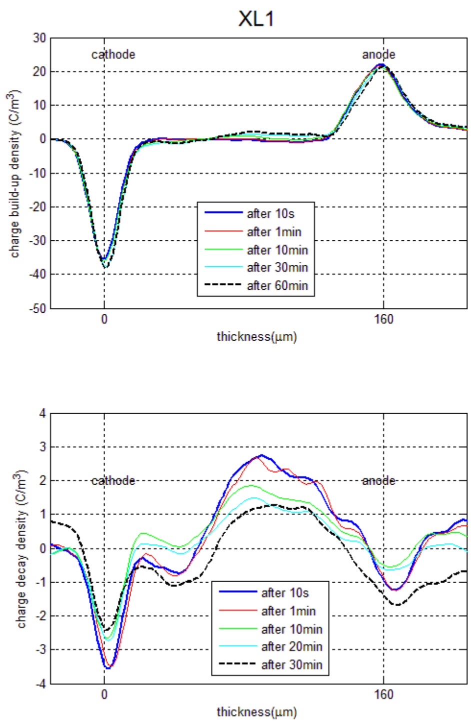

Figure 9: Space charge profile of XL1

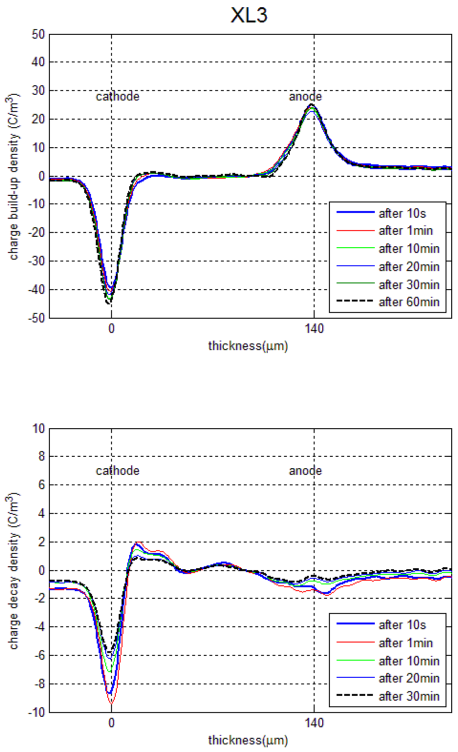

Figure 10: Space charge profile of XL3

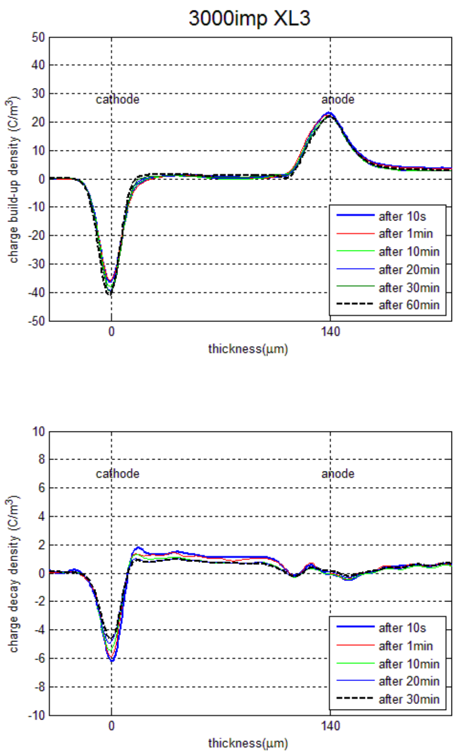

Figure 11: Space charge profile of impulse aged XL3

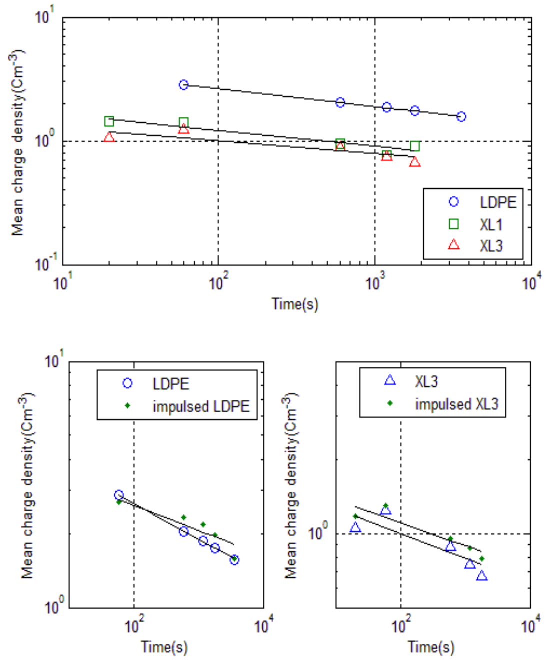

Figure 12: Average charge densities of samples

5 DISCUSSIONS

It is thought that the effect of lightning impulses may be to produce hot electrons with high energy that enter the material bulk. These electrons collide with molecules of the sample damaging molecular bonds and disturbing the crystalline structure of material. The homogeneity of the material is reduced and this leads to a reduction in breakdown strength of the material. With quench cooled LDPE, the crystallinity of material is low. The small contribution to increasing the amorphous region after impulse ageing can only lead to a small change in breakdown strength. The space charge results for LDPE show that the same amount of charge was obtained in the case of the impulse aged sample but the charges decay with a slightly lower rate. Therefore, the effect of impulse ageing seems to be negligible.

The peroxide crosslinking process changes the morphology of quenched cooled LDPE and consequently lead to increased breakdown strength of the material. The chains are linked together, molecular size increases and the mobility of molecules is reduced. The effect of impulse ageing may cause bond breaking and ionization. It may lead to the increase in ionic conduction and subsequently interfacial polarization. The movement of molecules under an electric field may also become easier as the molecule weights could be reduced after the ageing process.

6 CONCLUSIONS

It has been shown that the XLPE formed using trigonox-145 exhibits a higher electrical strength and lower space charge formation compared to quench cooled LDPE. However, the XLPE seems to be more sensitive to the effect of lightning impulse ageing. The impulse aged XLPE has a reduced breakdown strength, higher losses due to conduction and more charge injection under a DC applied voltage.

7 ACKNOWLEDGMENTS

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8 REFERENCES

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