

Influence of Thermal Treatment and Residues on Space Charge Accumulation in XLPE for DC Power Cable Application

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ABSTRACT

The effects of cross-linking by-products (residues) quantity and thermal treatment on space charge accumulation and decay in manufacturer modified XLPEs for dc power cable application have been investigated using the pulsed electro-acoustic technique. The threshold stress for space charge generation among the modified and reference XLPEs during voltage-ramping was found to show considerable variation and to depend upon the material and the amount of residue present. However, the modified XLPE material was found to exhibit a higher threshold for space charge accumulation than the reference XLPE whatever the conditions. De-gassed samples were found to exhibit the highest threshold stress, with that of the modified de-gassed XLPE accumulating no space charge at all even after 24 h stressing at 70 kV. In general heterocharge regions were formed when the residues were present and homocharge or no charge was formed when the residues were removed by degassing. Differences were also found in the space charge decay following short-circuit (volts-off), with the decay of heterocharge being rapid, whereas that of homocharge being slow. The charge accumulations have been tentatively explained by the mechanisms of ion-pair separation when residues are present, and interfacial injection when residues are absent. Decay of the heterocharge is governed by ion-pair displacement just as the generation and will have a similar time scale. In contrast homocharge decay will be governed by charge de-trapping and extend to time scales well beyond that of injection for charge in the deepest traps.

Index Terms - Space charge, cross-linking by-products; XLPE, dc power cable, PEA.

1 INTRODUCTION

CROSS-LINKED polyethylene (XLPE) is extensively used for the bulk insulation of underground high voltage power cables due to its high dielectric strength and electrical resistivity in combination with some excellent physical properties such as resistance to cracking and moisture penetration [1,2]. However, its good dielectric properties may bring some unwanted drawbacks to its performance under certain conditions. For instance, the low charge carrier mobility and charge trapping within the polymer give rise to space charge, resulting in localised electric stress enhancement. This is particularly true in the case where XLPE

is used as dc power cable insulation; the formation of space charge may distort the electric field distribution throughout the cable insulation thickness. Such localised electric stress enhancement may lead to the premature failure of the cable insulation at stresses well below the anticipated or designed values [3].

For these reasons, space charge measurement has attracted the attention of researchers over the world in the past two decades and there is a large research base concentrated on the mechanism of space charge formation, migration and accumulation in film samples. It has generally been concluded that such space charge may result from electron (and hole) injection from the electrodes and electric field assisted ionization of the impurities in the material or from an inhomogeneous polarization [4-10]. With the success of space

charge measurement on the film samples, attempts had also been made to apply the same techniques (mostly the PEA) to extruded polymer insulated power cables to study the space charge behaviour in cable geometry [11-13].

However, research on cable geometry is time consuming and costly because of the difficulty of prototype cable manufacture, thus many experiments still need to be carried out on plaque samples for the purpose of space charge suppression research in modified insulating materials. The planar sample for the research has been pressure moulded but the material was prepared in the same way as the cable insulation manufacturing process (i.e. material blending, melting, curing and afterwards conditioning) in order to reflect the effect of material processing and subsequent treatment (conditioning) on charging characteristics. These samples were made of differently modified XLPEs and were studied using the pulsed electroacoustic (PEA) method [14, 15]. They have a thickness of 3 mm (including embedded semiconducting electrodes) sufficient to reduce the surface effect that may be observed in thin film samples [16]. A relatively high voltage has therefore to be applied across the sample in order to obtain an adequate stress for space charge formation. Inevitably, the attenuation and dispersion of the acoustic wave propagation through the thick sample is another concern in the precise measurement of the space charge distribution in this case. Relevant measures employed in the research to improve experiment accuracy are briefly described in the paper. The space charge results from these samples show a great variation in terms of the field threshold for charge generation under voltage ramping and charge accumulation over the stressing period, and also space charge decay after the removal of the external voltage. Two types of material have been investigated: a reference XLPE and a XLPE with the manufacturer's modifications. Each of the sample-types have been subjected to different degassing treatments to produce three forms of specimen, i.e. fresh (undegassed) samples, partially degassed with 0.5% residue samples (data supplied by industrial partner), and thoroughly degassed samples. All samples were studied in terms of space charge accumulation and decay. For comparison between XLPE that must necessarily contain cross-linking by-products and additives and a polyethylene that has no such chemical species present, thermal moulded low density polyethylene (LDPE) was also tested in the research.

2 EXPERIMENTAL

2.1 PEA SYSTEM FOR THICK PLAQUE SAMPLES

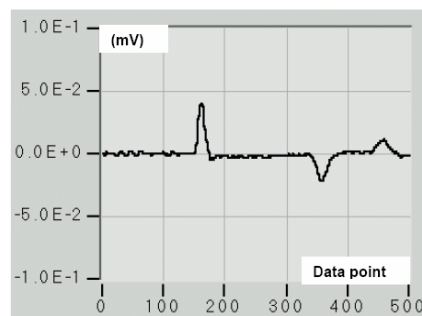
To satisfy the requirement of a relatively high stressing voltage across the sample, in addition to the application of a high voltage bushing on the high voltage electrode, an embedded electrode was used to supply adequate flashover distance along the sample's surface from the upper electrode (high voltage) to the ground electrode. The PEA system enables a dc voltage as high as 100 kV to be applied across the sample. Figure 1 shows the electrode system of the PEA used.

Acoustic wave propagation in the thick samples is significantly attenuated and dispersed due to the lossy and dispersive properties of the polymer. These features

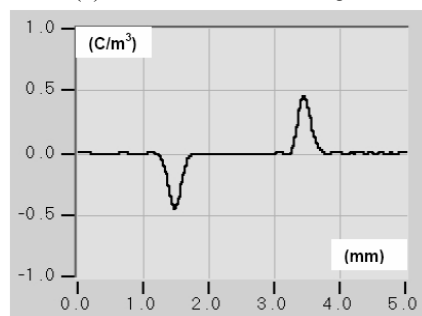
deleteriously affect the detecting sensitivity and spatial resolution [13,17]. A data compensation algorithm was developed in order to overcome the effects of attenuation and dispersion occurring in acoustic wave propagation. Figure 2a and 2b show an example of the space charge distribution at the calibration voltage before and after compensation for the acoustic signal propagation. The attenuation and dispersion of the acoustic wave is clearly observable in the relative magnitudes and widths of the peaks of the induced surface charges at the two electrodes in Figure 2a. The signal from the upper electrode (the peak to the right) appears lower and wider after the transmission through the sample thickness. With the uniform distribution of the electric stress throughout the planar sample, the induced capacitive charges at two electrodes should possess the same charge density and profile if no bulk space charge is present in the dielectric material. Following the application of compensation algorithm, nearly identical charge peaks on the two electrodes are recovered, as shown in Figure 2b [18].



Figure 1. Photograph of the PEA electrodes system for thick plaque samples.



(a) Raw data of calibration signal



(b) True space charge distribution

Figure 2. Space charge profile (in calibration) before and after attenuation and dispersion compensation

2.2 SAMPLES

The samples tested are a reference XLPE and XLPE materials purposely modified for HVDC power cable application. LDPE was also introduced in the research to compare its space charge characteristics with results of fully degassed XLPE samples. All of them have the same semiconductive electrodes made of cross-linked polyethylene loaded with carbon black. In order to understand the influence of the residue on the space charge formation, samples have been subjected to different thermal treatment and then tested with a variety of cross-linking by-product concentrations. Figure 3 illustrates a sample's dimensions and cross section.

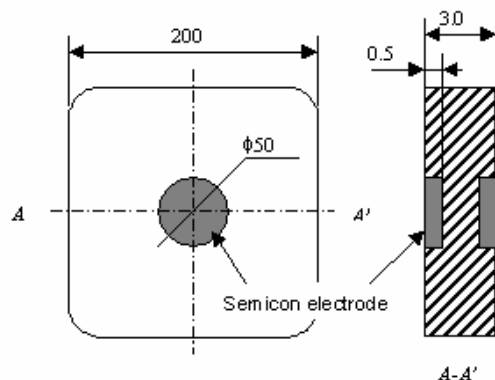


Figure 3. Sample dimensions (mm) and its cross section.

Apart from the flashover problem mentioned previously, several samples from the first batch failed during the voltage ramp procedure or the stressing period due to electrical breakdown occurring at the edge of the embedded semiconducting electrode. One sample was sectioned through the test region to check the attachment between the semiconducting electrode and the bulk XLPE, and the condition of the insulation, and the profile of the electrode. No significant defect or poor quality was observed in the insulation except for the sharp inner edge of the semiconductor. It is therefore believed that the main cause for the electrical failure of the sample is the field enhancement in the vicinity of the edge. A semiconducting electrode with a rounded edge was used in the subsequent samples.

Commercial confidentiality denied us knowledge of the composition of the modified XLPE and its processing procedure except for the thermal treatment (e.g. degassed for three days at 90 °C in oven).

2.3 EXPERIMENTAL PROCEDURE

Space charge measurements on each sample started with a voltage ramp test in which the applied voltage was stepped up from zero to 70 kV dc. The intention of this experiment is to find the threshold voltage above which a bulk charge starts to accumulate. Of course a time scale is implicated in this statement, so in fact the measured threshold field is that appropriate to short times, i.e. space charge developed on the time scale of the voltage ramp. Following ramp-up space charge measurements were carried out at regular intervals over the stressing time with the external voltage on or switched off just for the duration of the PEA measurement so as to remove the masking effect of the capacitive charge induced on the electrode. The space charge distribution in most of the samples

reached equilibrium within 24 hours and some of them even sooner. At the end of the stressing test a space charge decay test was conducted by recording the space charge profile at different time intervals after the removal of applied voltage.

3 EXPERIMENTAL RESULTS

3.1 RAMP VOLTAGE TEST

The voltage ramp test results of reference XLPE and modified XLPE samples are presented in Figure 4, in which the magnitude of the induced charge at the two electrodes is plotted against the externally applied voltage. To reduce the influence of the measuring time on the charge formation the measurement at each voltage step was carried out quickly (within 10 seconds), and only the peak heights of output voltage which are proportional to the induced charges at the upper and the ground electrodes were read directly from the oscilloscope. The readings are therefore expressed in the units of voltage. Due to the attenuation of the acoustic signal across the sample, the peak amplitude of the upper electrode (away from the transducer) is always lower than that from the ground electrode, as shown by curves labelled as the anode in the figure. The ramp voltage test was completed within about two and half minutes for each sample.

In Figure 4, the linear relationship between the external voltage and the induced charges at the electrodes in the degassed samples and the modified XLPE with 0.5% residue indicates that no space charge is generated in the bulk materials over the time of voltage ramping from 0 to 70 kV. In all the fresh samples and some partially degassed reference XLPE samples, space charge accumulation occurred in the bulk material after the voltage reached 50 kV, which could be observed from the deviation of the curve from its linear tendency shown by the dotted line.

3.2 SPACE CHARGE EVOLUTION OVER THE LONG-TERM FIELD STRESSING

Stressing tests up to 24 h were conducted on all samples, during which time the space charge distribution was measured at regular intervals and the results are presented below. In order to measure homocharge accurately, the measurement was also carried out at each time with the external voltage switched-off for the duration of the measurement. The charge profile titled "0 time" in each diagram stands for the result obtained soon after the voltage ramping up process finished.

3.2.1 REFERENCE XLPE SAMPLE

3.2.1.1 MEASUREMENT WITH VOLTAGE APPLIED

Figure 5 shows the space charge distribution for samples with different residue contents. The results of the undegassed sample and the partially degassed samples see Figure 5a and 5b, show similar quantities of heterocharge after about 1 hour of stressing. Therefore the influence of the amount of residue (i.e. residue content) on the charge distribution is not very significant. However the rate of charge accumulation is obviously affected by the amount of residue in the sample. Thus for example, quite considerable space charge had built up in the undegassed sample prior to constant stressing at 70 kV, whereas in the sample with 0.5% residue most of the

space charge formed in the first hour of constant voltage stressing after the voltage ramp. Due to flashover occurring along the surface of the undegassed sample no further data could be acquired after one hour of stressing as the test was

terminated. In the de-gassed sample, Figure 5c, no space charge is accumulated over 24 h at 70 kV even though it had already been stressed at 50 kV for 24 h before the voltage was stepped-up to 70 kV.

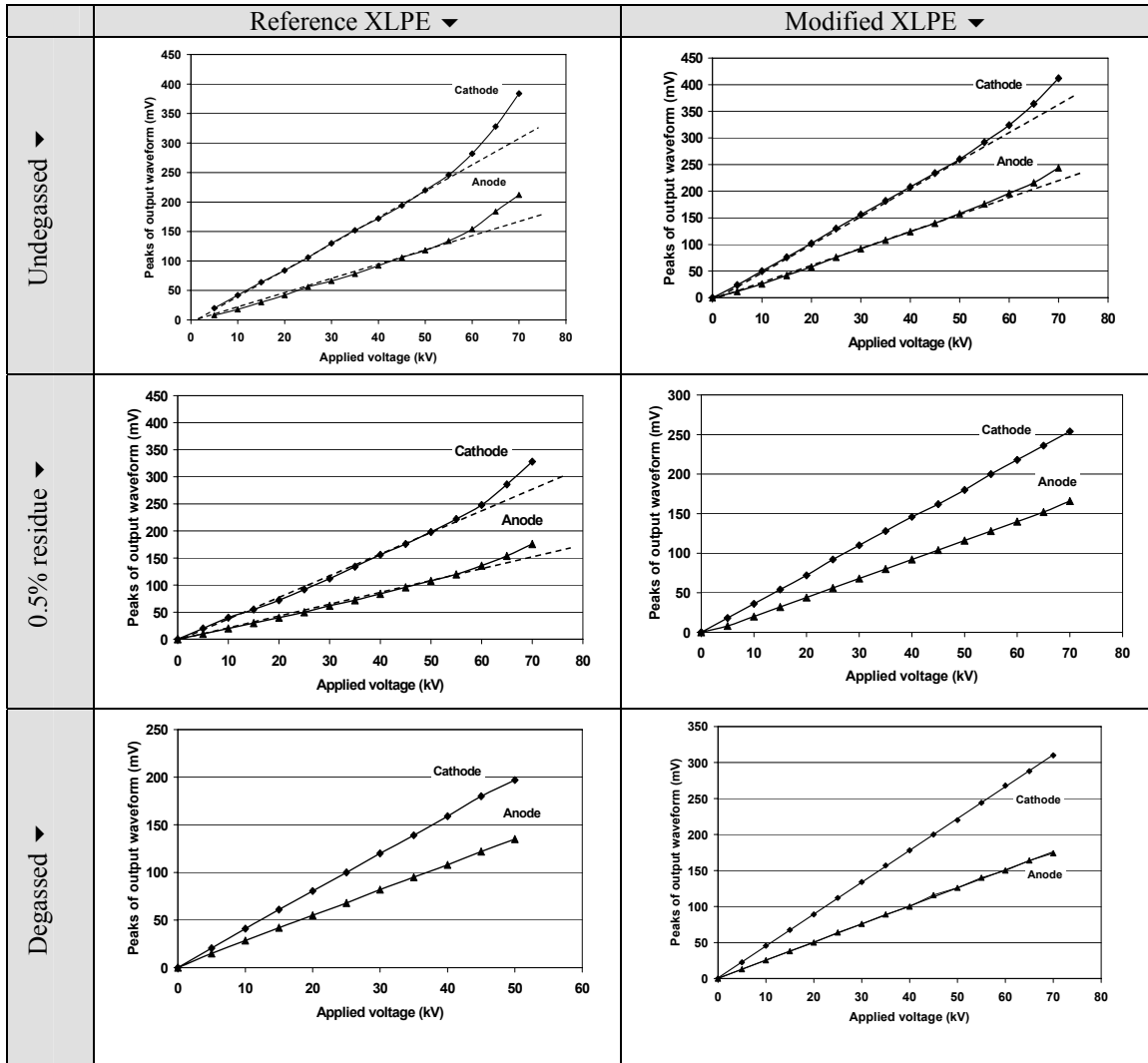


Figure 4. Relationship between the induced electrode charge and the external voltage.

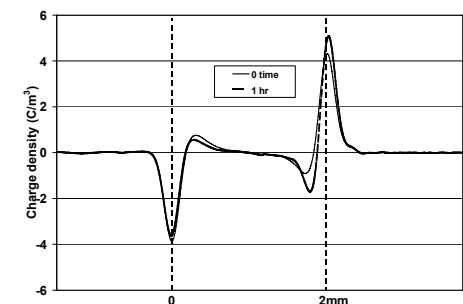
3.2.1.2 MEASUREMENT WITH TRANSIENT REMOVAL OF VOLTAGE

Space charge distributions were also measured at various times within the stressing term by transiently removing the external voltage for the duration of the measurement. This is intended to especially check for the presence of homocharge, which may otherwise be merged into the electrode charge due to the capacitive charge produced by the external voltage. The results are shown in Figure 6. In the case of degassed and partially degassed samples, heterocharges are dominant as seen in Figure 6a and Figure 6b. In the absence of the induced electrode charge due to the external stressing voltage, the homocharge in the degassed sample is small but clearly observed, as shown in Figure 6c.

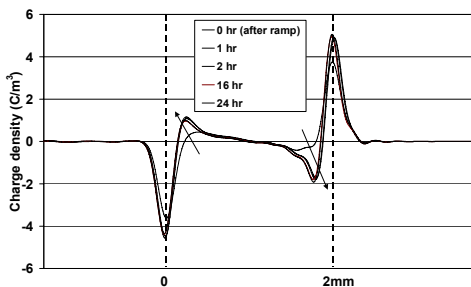
3.2.1.3 SPACE CHARGE DECAY

Space charge decay test were made on each sample following the permanent removal of the applied potential after 24 hours of stressing at 70 kV, and the results are shown in Figure 7. It must be pointed out that Figure 7a shows the retested results of the undegassed sample after the first flashover for which no decay test was carried out since the pulse voltage could not be applied across the sample.

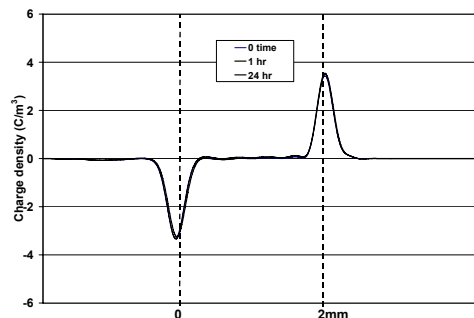
The undegassed sample and the sample with 0.5% residue have a heterocharge accumulation and show a fast space charge decay rate in comparison with that of the degassed sample, which exhibits homocharge regions that show almost no decay over 12 h.



(a) Undegassed reference XLPE.



(b) Partially degassed reference XLPE with 0.5% residue.



(c) Degassed reference XLPE.

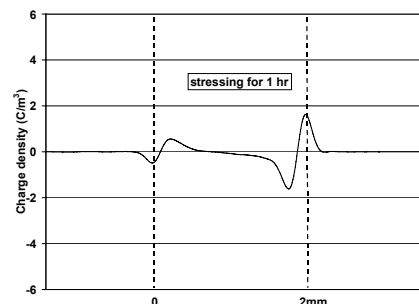
Figure 5. Space charge profiles in reference XLPE (measured with volts on).

3.2.2 MODIFIED XLPE FOR HVDC CABLE

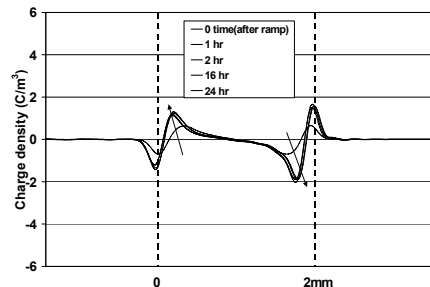
3.2.2.1 MEASUREMENT WITH VOLTAGE APPLIED

Figure 8 shows the space charge measurements in the modified XLPE (for use in HVDC power cable insulation) that have been made with the external voltage remaining applied.

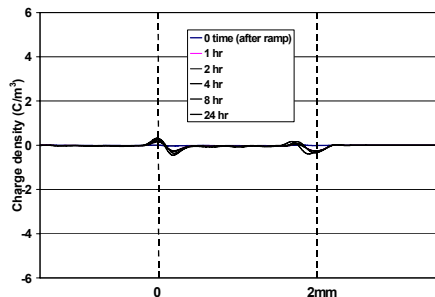
Heterocharge is generated relatively quickly in the undegassed sample see Figure 8a. After the fast heterocharge accumulation in the voltage ramp, the space charge distribution through the sample almost reached saturation within one hour. In the sample with 0.5% residue, the heterocharge accumulation progressed much more slowly and saturated in about 8 hours, in contrast to the behaviour of the unmodified XLPE. Figure 8b presents the space charge evolution in this sample over the whole stressing period. As with the unmodified XLPE though the final charge distributions in these two samples are very similar after 24 h ageing. The de-gassed modified XLPE material did not accumulate any space charge over 24 h of stressing just as was the case with the reference XLPE.



(a) Undegassed reference XLPE.



(b) Partially degassed reference XLPE with 0.5% residue.



(c) Degassed reference XLPE with homocharge accumulation.

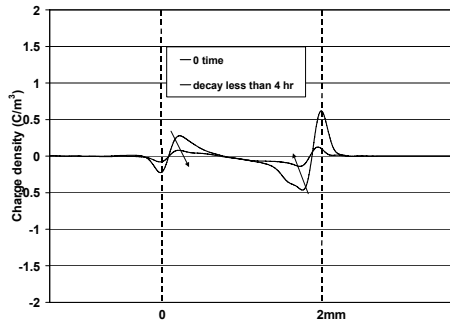
Figure 6. Space charge profiles in reference XLPE (with the transient removal of voltage during measurement).

3.2.2.2 MEASUREMENT WITH TRANSIENT REMOVAL OF VOLTAGE

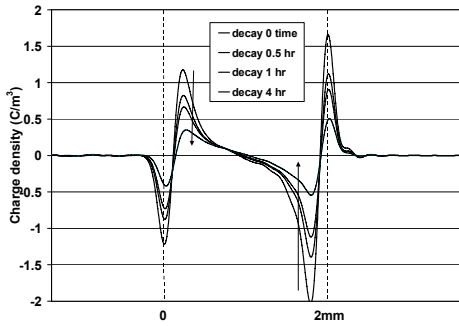
The results of the voltage-off measurements made during the stressing period are presented in Figure 9. The amount of heterocharge in the undegassed sample is higher than that in the sample with 0.5% residue. No space charge accumulation in the degassed sample can be seen in Figure 9c, where a straight base line is shown when the measurement was made after 24 hours of stressing. This is in contrast to the reference XLPE where a small amount of homocharge was observed.

3.2.2.3 SPACE CHARGE DECAY

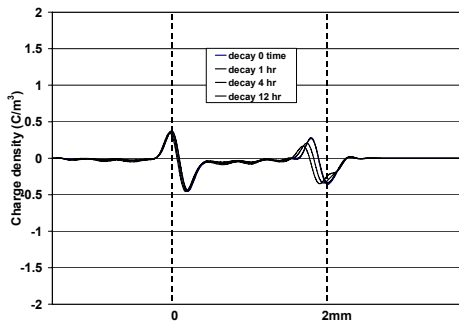
The space charge in the modified XLPE sample also possesses a fairly high decay speed similar to the behaviour of the undegassed reference sample. In Figure 10a, the heterocharge in the bulk material has decayed away within two hours after the electrodes were short-circuited. In contrast the sample with 0.5% of impurities displays a very slow decay rate as shown in Figure 10b, similar to the behaviour of the undegassed reference XLPE. No decay test was carried out in the degassed sample because of the absence of space charge.



(a) Undegassed reference XLPE.

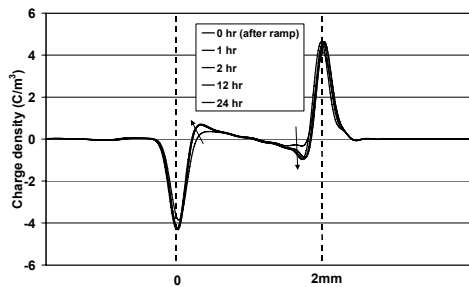


(b) Partially degassed reference XLPE with 0.5% residue.

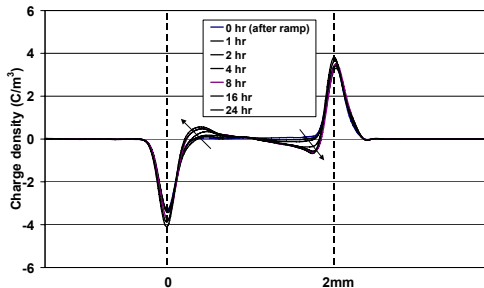


(c) Degassed reference XLPE.

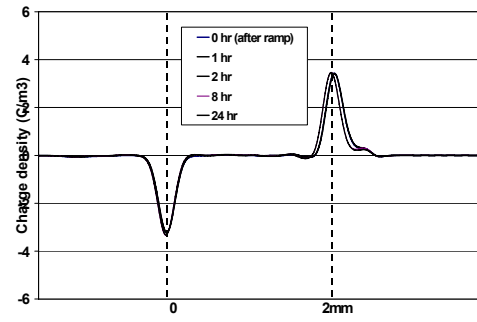
Figure 7. Space charge decay in reference XLPE.



(a) Undegassed modified XLPE.

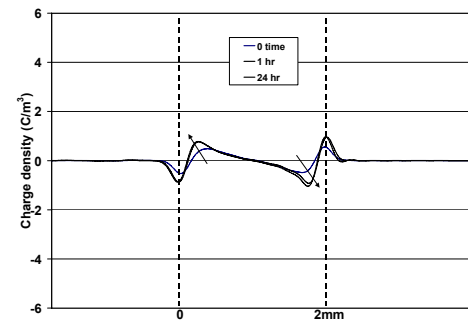


(b) Partially degassed modified XLPE with 0.5% residue.

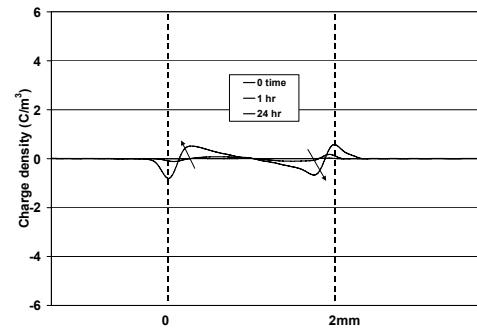


(c) Degassed modified XLPE

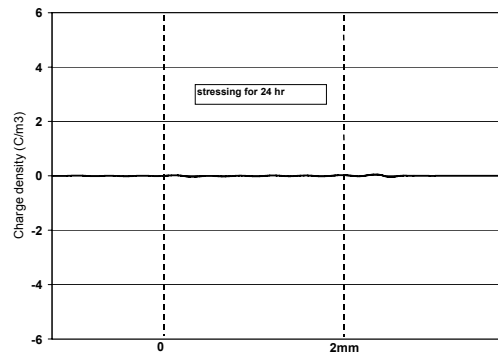
Figure 8. Space charge profiles in modified XLPE (measured with volts on).



(a) Undegassed modified XLPE.

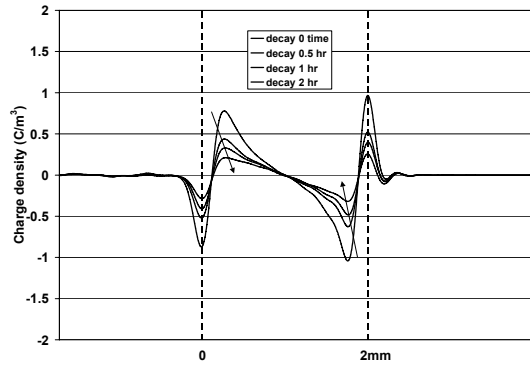


(b) Partially degassed modified XLPE with 0.5% residue.

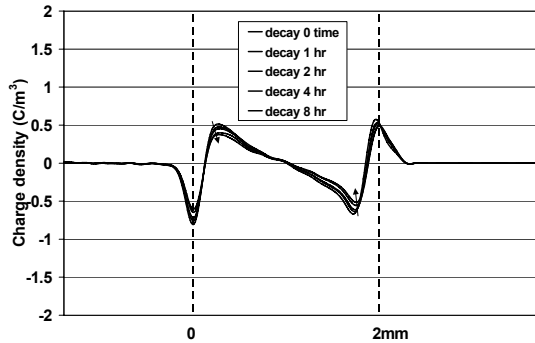


(c) Degassed modified XLPE.

Figure 9. Space charge profiles in modified XLPE (with transient removal voltage during measurement).



(a) Undegassed modified XLPE.

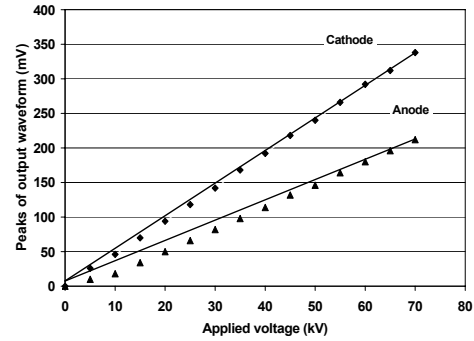


(b) Partially degassed modified XLPE with 0.5% residue.
Figure 10. Space charge decay in modified XLPE.

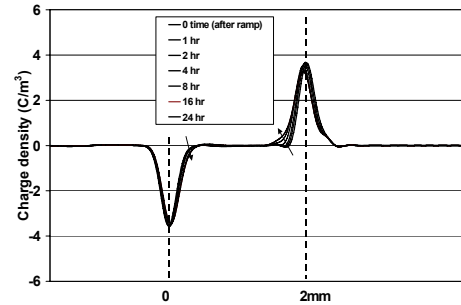
3.2.3 LDPE SAMPLE

The space charge distribution and its evolution were also studied for a low-density polyethylene (LDPE) sample with the same dimension as that of XLPE samples. The sample was prepared by thermal pressing, and no peroxide was added. This allows a comparison between XLPE that must necessarily contain cross-linking by-products and a polyethylene that has no such chemical species present.

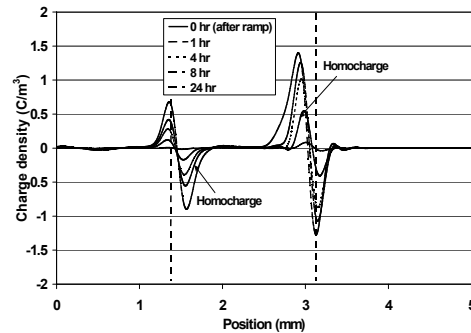
From the ramp test result shown in Figure 11a, the linear relationship between the induced charge on electrodes and the external voltage suggests that no space charge appears in the voltage stepping up process. Within the subsequent stressing time, homocharges gradually accumulated in the bulk insulation in the vicinity of the electrodes, as shown in Figure 11b and 11c. This is shown most clearly in Figure 11c where the measurements are taken with the external voltage transiently removed. As with the homocharges observed in the previous degassed XLPEs, the homocharge in this LDPE sample also showed a very slow decay rate, as illustrated in Figure 11d.



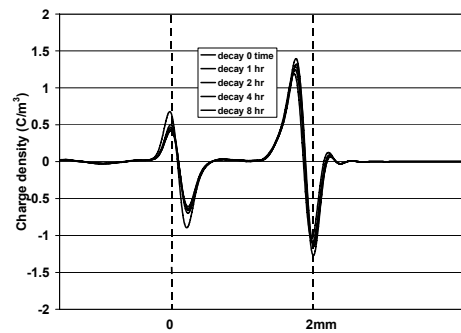
(a) Ramp test



(b) Homocharge accumulation just observable with volts on



(c) accumulation clearly revealed with transient removal of voltage



(c) Slow charge decay in LDPE

Figure 11. Space charge behaviour of LDPE sample.

4 DISCUSSIONS

4.1 THRESHOLD STRESS FOR FAST CHARGE FORMATION IN RAMP TEST

The ramp test results in Figure 4 indicate that the undegassed reference XLPE and modified XLPE samples have a heterocharge accumulation during the voltage ramp process. This can be seen by the extra induced charge on the electrodes as the external voltage increase, which are due to the enhancement of the interfacial stress by heterocharge. Heterocharge formation during the voltage ramp was also observed in the partially degassed XLPE reference sample in the same way. The threshold stress, above which space charge starts to generate, is determined by dividing the corresponding voltage by the sample's thickness. The specific threshold stress for space charge generation in each individual sample is listed in Table 1. For those specimens having no observed space charge during the voltage ramp test, the thresholds are described by the term "higher than" (>) the stress determined by the maximum stressing voltage, e.g. 70kV.

Table 1. Threshold stress (kV/mm) of space charge formation.

	Undegassed	0.5% residue	Degassed
Ref-XLPE	28	31	>40
Mod-XLPE1	33	>37	>38
Mod-XLPE2	>40	>35	>41
Mod-XLPE3	25	31	>42
LDPE		>42	

Obviously, in the undegassed samples, space charges are initiated at relatively low electric field strength in comparison with those of the corresponding partially degassed or degassed samples. It also seems that the modification to the material has achieved positive effects in terms of space charge suppression and generally raising the threshold field for space charge generation when the residues are still present. This even carries over to the de-gassed material, where no space charge appears in the modified XLPE in contrast to a small amount of homocharge for the reference XLPE.

Similar results have also been obtained for two other modified XLPE samples (XLPE2 and XLPE3 in Table 1) for HVDC application, which are not reported in the paper. It seems that when XLPE is subjected to thermal treatment to remove some of the volatile impurities the space charge accumulation can be suppressed to some extent or even ultimately stopped. For instance, the threshold stresses for space charge in the XLPE reference and purposely modified XLPE1 been increased from 28 kV/mm and 33kV/mm in the fresh samples (with maximum residue content) to the values of 31 kV/mm and higher than 37 kV/mm respectively in the sample with 0.5% residue. If they are further degassed to remove all the cross-linking byproducts (the degassed sample), no space charge at all may be formed in the voltage ramp process. As with all the degassed specimens, no space charge was observed in the LDPE sample, which contains no peroxide, during the voltage ramping from 0 to 70 kV.

4.2 SPACE CHARGE POLARITY AFTER 24HR STRESSING

From the above test results, it is evident that specimens with various material modification and thermal treatments (degassing) possess different space charge development speeds and final charge distributions. In particular, the polarity of the space charge region next to the electrodes depends upon the presence of cross-linking residues, with heterocharge being formed when the residues are present and homocharge or no charge when they are absent. Table 2 details the final space charge polarities of these samples.

The results presented in this table clearly indicate that the residue or the impurity, no matter how high its concentration, is responsible for the appearance of the heterocharge. In samples such as pure LDPE or those that have been degassed, it is very difficult to develop space charge under the electric stress applied in this research, except for the small homocharge accumulation in reference XLPE and LDPE sample. From the results of the degassed samples, one can therefore conclude that the modification to the XLPE material is favourable in suppressing space charge accumulation. Of course, the effect of the modification to XLPE on other space charge characteristics, such as charge accumulation rate and decay speed, and space charge amount are other considerations to be taken into account in the material selection process.

Table 2. Space charge polarity of samples after 24hr ageing.

	Undegassed	0.5% residue	Degassed
Ref-XLPE	Hetero	Hetero	Homo
Mod-XLPE1	Hetero	Hetero	Not observed
Mod-XLPE2	Hetero	Hetero	Not observed
Mod-XLPE3	Hetero	Hetero	Homo
LDPE		Homo	

Additionally, the fact that a longer time is required for the small homocharge to develop in the degassed sample or LDPE sample may suggest that homocharge formation is more difficult than heterocharge [19, 20]. The former case may need higher electric stress or longer time under the same electrode and dielectric arrangement to develop the same amount of space charge.

4.3 SPACE CHARGE ACCUMULATION RATE

The time for the space charge to reach its saturation (steady-state equilibrium) distribution is a practical method to describe the charge accumulation rate.

The results shown in Figure 12 reveal that the samples having residual impurities (which normally develop heterocharges as discussed in the previous section) normally have shorter space charge saturation times than the degassed samples in which homocharges appear. Among these samples, Reference XLPE and Mod-XLPE3 display quite similar charge accumulation features. In both of them the undegassed sample or that with 0.5% residue, reach the space charge saturation distribution within 1 hour, whereas development of homocharge in the degassed specimen was fairly slow.

As an exception, sample mod-XLPE2 shows a relatively slow space charge accumulation rate in the undegassed sample and in the sample with 0.5% residue. Similarly to the degassed

XLPE sample, thermally pressed LDPE also shows a very low space charge accumulation rate.

4.4 SPACE CHARGE DECAY RATE

Space charge profiles in the decay test presented in section 3 show a big variation associated with the materials and degassing conditions, which are summarised and discussed below.

First, in most cases the space charge decay speed corresponds to its accumulation speed, the faster the space charge accumulates, the faster it decays, or vice versa. Typical results which lead to this conclusion are presented in Figure 13 where modified XLPE1 and XLPE3 both with 0.5% residues are regarded as typical slow charging and fast charging examples respectively.

As shown in Figure 13, space charge accumulates very quickly in XLPE3 (with 0.5% residue) and almost saturates in one hour. Correspondingly the space charge also decayed very quickly when the applied voltage was removed. Only ~20% of the initially accumulated charge was left when the sample was short-circuited for 8 hours of discharge. In modified XLPE1 (0.5% residues), however, the space charge accumulated gradually and only reached its equilibrium about 15 hours after the stressing started. After the sample electrodes were short-circuited for 8 hours, there was still a considerable amount of charge residing in the sample. The space charge in the reference XLPE and other modified XLPE showed similar characteristics in the accumulation and decay process.

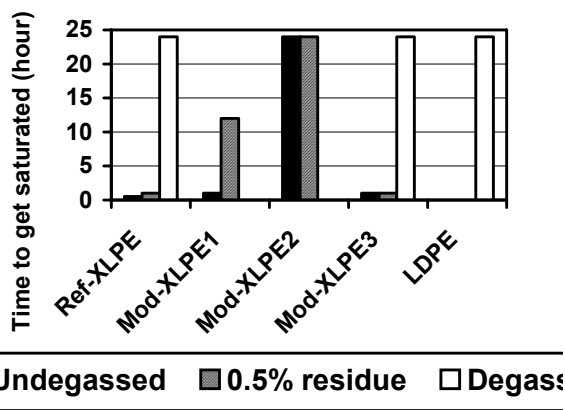


Figure 12. Saturation time of different samples

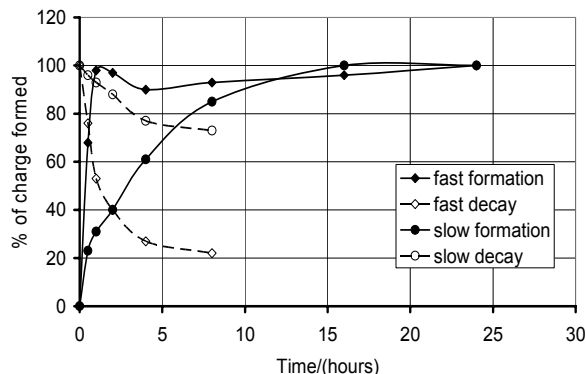


Figure 13. Space charge accumulation and decay rate for mod-XLPE1 (slow) and Mod-XLPE3 (fast).

Secondly, homocharge in the degassed samples decays much more slowly than heterocharge in the undegassed sample. As shown in Figure 14, heterocharge in the undegassed XLPE-1 has a very high decay rate. After 3 hours, the maximum charge density declined to about 20% of its original value. In contrast the homocharge accumulated in the degassed sample only reduced to about 90% of its initial value at 3 hours and thereafter remained relatively stable. The homocharge accumulation rate is also similarly slow, see section 4. The space charge evolution and decay in Figure 11c and 11d also show similar features for the homocharge observed in the LDPE sample.

The work of C. Zhang et al [21] on space charge decay in LDPE and its blend polymer suggested the existence of two de-trapping processes, which govern space charge decay with two time constants. The relationship between the total amount of charge (per unit area) Q and time t in decay may be expressed in equation of

$$Q = Ae^{-t/\tau}$$

where τ is time constant and A is a constant. However, for the homocharge and heterocharge dealt with the research, no similar phenomenon was observed. A check of apparent time constants of all samples gives a big range of variety from 2 hours to hundreds of hours for heterocharge and homocharge respectively. The understanding of space charge decay mechanism is expected to rely on the identification of charge carrier in different cases. From this point of view, the decay of different types of space charge (i.e. homo- and heterocharge) cannot be simply estimated by the local electric field due to space charge (existing driving field) in the short-circuited condition. A thorough discussion about the role of the space charge field in decay process has been made by two of authors (LAD and JCF) in [22] and they concluded that its effect is negligible compared to the de-trapping activation energy over the time of measurement. Good examples are those heterocharge and homocharge existing in partially degassed XLPE and additive-free LDPE. Their driving fields can be worked out from space charge distorted field profiles, shown in Figure 15b and Figure 15d respectively, as about 6kV/mm. However, their decay rates are very different. Similar results have been reported by Mizutani [23] in two types of LDPE (pressure and metallocene catalyst polymerised respectively). Both of them had almost same space charge distribution and induced electric field, but space charge in one sample showed a very high stability.

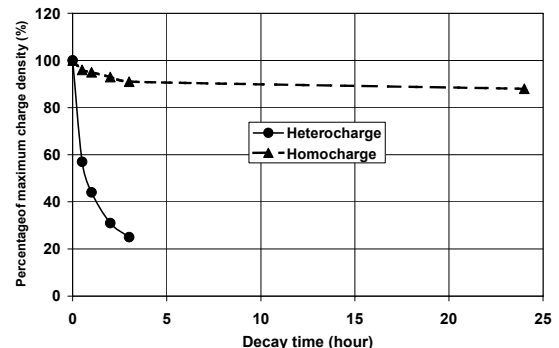


Figure 14. Comparison of homo and hetero charge decay speed.

It could be understood from the voltage ramping test in Figure 4 and homocharge saturation results in Figure 12 that homocharge generation and accumulation generally need a higher field and longer stressing time. As discussed afterwards in section 4.6.2, more energy and time beyond the scale of accumulation would be required to activate these trapped charges.

4.5 ELECTRIC FIELD DISTORTION DUE TO SPACE CHARGE

A consequence of space charge accumulation is the distortion of the external applied electric field, which results in localised field enhancement. The electric field distribution can be derived from the space charge profile by using Poisson's equation. It is easy to understand that heterocharge will result in interfacial stress enhancement and central stress reduction in the specimen, whereas the homocharge influences the electrical field in the opposite way. Figure 15 a-c show the electric field distribution distorted by space charge profiles shown in Figure 8 whilst Figure 15d displays the result when homocharge is present in LDPE.

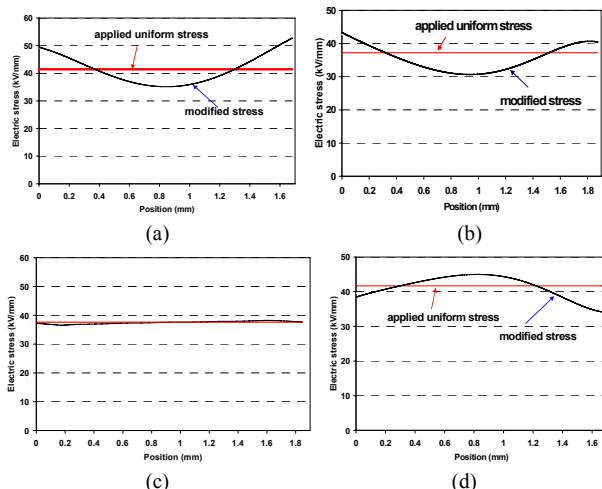


Figure 15. Electric field distribution modified by (a) heterocharge in fresh modified XLPE, (b) heterocharge in modified XLPE with 0.5% residues, (c) unobserved charge in fully degassed modified XLPE, (d) homocharge in LDPE.

4.6 CHARGE GENERATION MECHANISM

There are two possible origins for the generation of heterocharge in an insulating material under an applied potential difference: a) injection followed by transit of the sample to an electrode where the removal of charge carriers is slower than the transit rate, and b) the separation of existing (and field generated) charge species in the material. These candidates for the generation mechanism will be discussed separately.

4.6.1 INJECTION AND RAPID TRANSIT

It has been shown that in the absence of cross-linking residues homocharge regions are formed, so it is clear that charge injection does take place. So if this is the origin of the heterocharge the presence of residues would have to increase the transport rate of the charge carriers. Since the residues are volatile we could imagine them carrying the charge as molecular ions, a process that may be faster than charge

carriers become trapped and transit the sample via trap-to-trap transfer. We would also expect the extraction of charge from molecular ions located at the counter-electrode to be slow, so this mechanism may be a potential candidate. The decay of such heterocharge regions could be achieved in two ways, a charge neutralisation by injection of carriers of the opposite polarity or a return transit across the sample. The injection-neutralisation is likely to be rapid but the decay rate of the space charge regions is similar to its accumulation rate, which suggests an inverse process. However the return transit would take place in a low bulk field and so would be at a much slower rate to the forward transit.

4.6.2 SEPARATION OF EXISTING MOLECULAR ION SPECIES

In this case neutral chemical residues would have to be regarded as being in chemical equilibrium with their ionic components. In an electric field the ionic fraction, which does not have to be high, displaces with respect to one another. The time required to set up the heterocharge distribution is just the time required for ions to be separated by a Debye length, i.e. the separation distance for which the ions essentially become independent. This is governed by the dissociation rate of a net-neutral pair and would therefore be fast. The increased field for space charge generation in the modified material would be the result of an increased difficulty of ion pair separation in this material. In this mechanism the two heterocharge concentrations in the sample would add to zero. This is not quite the case for the results presented here, see Figures 7, but it has been shown that some charge injection take place in the de-gassed specimens and such an effect in addition to the charge separation would lead to the slight imbalance that is observed. Decay of the hetero space charge in this case would occur via displacement of the molecular ions to form neutral entities again. Since this does not require transit of the sample the bulk field has no real influence. The decay rate will be governed to a great extent by the return to chemical equilibrium in the space charge regions. Here the charge on the electrodes has to be considered. While the potential difference was applied this would be very large and hence able to maintain a large excess concentration of one species of molecular ion. When the potential is removed the electrode charge reduces instantaneously to that of the image charge, which is very much less. The amount of excess molecular ions that can be held against the chemical potential attempting to restore the locality to the equilibrium concentration of net-neutral molecular ions is much reduced. The rate of reduction will be determined by the rate of the chemical reaction, just as is the rate of generation. Because the image charge can hold a quantity of excess ions in its locality the return to a neutral region will not be complete in the same time that separation generates the heterocharge regions, but a large part of the excess charge will be removed in this time. Since the image charge also diminishes as the excess charge diminishes, the final part of the decay process will be a slower feedback process involving neutral-pair formation reducing the image charge that feeds back to allow further pair formation.

It would seem from the above considerations that a mechanism involving the residues in molecular ion-pair

separation is the most likely explanation of the results. In the absence of such residues, and when material modifications perhaps bind the residues chemically into the matrix, a small amount of charge injection will occur with the injected charges being rapidly trapped near to the injecting electrode. The decay of such charge will be governed by the de-trapping rate if neutralisation by counter-charge injection is not involved, which may extend beyond the time scale of the charge accumulation if the trap-depths are greater than the activation energy for injection. However injection of a neutralising species may well be the first stage of space charge decay and this will take place on the same time scale as the original space charge generation [24]. In this case there will be a considerable fraction of the space charge remaining that will decay on the time scale governed by the trap depths.

In the case of mod-XLPE1 examined here it would seem that such injection requires a higher electric field strength than that required by the reference XLPE. This implies that the modification has an effect on the electrode-polymer interface, since it is this that determines the injection features.

5 CONCLUSIONS

The space charge distributions generated in a batch of modified XLPEs and reference materials by an applied potential are presented and discussed, leading to the following conclusions.

Using PEA to measure space charge in thick plaque sample, which is prepared exactly following the production methods of polymer insulation of cables, could allow measurements on cable-type morphologies without having the difficulties associated with cable measurements.

Heterocharge is produced when cross-linking residues are present, whereas homocharge or no charge at all is found when the residues are absent. It is clear that the residues have a major effect upon the space charge distribution in all of the XLPE materials studied. The space charge decay occurs on a similar time scale to its generation. Heterocharge accumulates quickly and a large part of it decays just as quickly. Homocharge accumulates slowly and decays at least as slowly and possibly even slower for a large part of the space charge. These differences in time scale are found even though the space charge driving field for decay is the same (initially $\sim 6\text{kV/mm}$) in both cases. Therefore, the charge decay mechanism and rate need to be carefully considered in terms of the type of charge carrier, polymer physical and chemical structures, as well as from driving electric field built up by space charge itself.

Significant differences have been observed between the different XLPEs and the reference materials. The modified XLPE materials have been shown to raise the applied field required for space charge formation compared to the reference XLPE, even in the presence of cross-linking residues. This gives a higher working field for which the material would not develop space charge field distortion. The amount of space charge generated in the as-received undegassed state is slightly smaller than that for the reference XLPE under the same conditions, but there is still a considerable amount. Thus the modification does not strongly affect the amount of

heterocharge produced in the steady state, but increases the applied field at which it can be produced. A partial reduction of the quantity of residues, however leads to a suppression of space charge accumulation in the modified XLPE compared to the reference XLPE.

REFERENCES

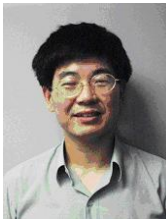
- [1] R. Arrighi, "From Impregnated Paper to Polymeric Insulating Material in Power Cables", IEEE Trans. Electr. Insul., Vol. 21, pp.7-18, 1986.
- [2] T. L. Hanley, R. P. Burford, R. J. Fleming and K. W. Barber, "A General Review of Polymeric Insulation for Use in HVDC Cables", IEEE Electr. Insul. Mag., Vol. 19, No. 1, pp. 13-24, 2003.
- [3] T. Tanaka and A. Greenwood, *Advanced Power Cable Technology*, Vol. 1, Ch. 2.6, CRC Press inc. USA, 1983.
- [4] J. Lewiner, "Evolution of Experimental Techniques for the Study of the Electrical Properties of Insulating Materials", IEEE Trans. Electr. Insul., Vol. 21, pp.351-360, 1986.
- [5] T. Mizutani, "Space Charge Measurement Techniques and Space Charge in Polyethylene", IEEE Trans. Dielectr. Electr. Insul., Vol.1, pp.923-933, 1994.
- [6] G. M. Sessler, "Charge Distribution and Transport in Polymers", IEEE Trans. Dielectr. Electr. Insul., Vol.4, pp.614-628, 1997.
- [7] Y. Li and T. Takada, "Experimental Observation of Charge Transport and Injection in XLPE at Polarity Reversal", J. Phys. D: Appl. Phys. Vol.25, pp. 704-716, 1992.
- [8] T. Doi, Y. Tanaka and T. Takada, "Measurement of Space Charge Distribution in Acetophenone Coated Low-density Polyethylene", IEEE conf. electr. Insul. Dielectr. Phenomena (CEIDP), pp.32-35, 1997.
- [9] Y. Tanaka, Y. LI, T. Takada and M. Ikeda, "Space Charge Distribution in Low-density Polyethylene with Charge-injection Suppression Layers", J. Phys. D: Appl. Phys., Vol.28, pp.1232-1238, 1995.
- [10] K. S. Suh, J. H. Koo, S. H. Lee, J. K. Park and T. Takada, "Effects of Sample Preparation Conditions and Short Chains on Space Charge Formation in LDPE", IEEE Trans. Dielectr. Electr. Insul., Vol.3, pp.153-160, 1996.
- [11] N. Hozumi, H. Suzuki, T. Okamoto, K. Watanabe and A. Watanabe, "Direct Observation of Time-dependent Space Charge Profiles in XLPE Cable under High Electric Fields", IEEE Trans. Dielectr. Electr. Insul., Vol. 1, pp.1068-1076, 1994.
- [12] X. Wang, D. Tu, Y. Tanaka, T. Muronaka, T. Takada, C. Shinoda and T. Hashizumi, "Space Charge in XLPE Power Cable under dc Electrical Stress and Heat Treatment", IEEE Trans. Dielectr. Electr. Insul., Vol.2, pp.467-474, 1995.
- [13] M. Fu and G. Chen, "Space charge measurement in polymer insulated power cables using flat ground electrode PEA system", IEE Proc. Sci. Meas. Technol. Vol. 150, No. 2, pp.89-96, 2003.
- [14] T. Takada and T. Sakai, "Measurement of Electric Field at a Dielectric/Electrode Interface Using an Acoustic Transducer Technique", IEEE Trans. Electr. Insul., Vol. 18, pp. 619-628, 1983.
- [15] T. Maeno, T. Futami, H. Kushibe, T. Takada and C. M. Cooke, "Measurement of Spatial Charge Distribution in Thick Dielectrics Using the Pulsed Electroacoustic Method", IEEE Trans. Electr. Insul., Vol. 23, pp.433-439, 1988.
- [16] G. Chen, T. Y. Tay, A. E. Davies, Y. Tanaka and Takada, "Electrodes and charge injection in low-density polyethylene using the pulsed electroacoustic technique", IEEE Trans. Dielectr. Electr. Insul., Vol. 8, pp. 867-873, 2001.
- [17] Y. Li, M. Aihara, K. Murata, Y. Tanaka and T. Takada, "Space Charge Measurement in Thick Dielectric Materials by Pulsed Electroacoustic Method", Rev. Sci. Instrum, Vol. 66, pp.3909-3916, 1995.
- [18] M. Fu, *Space Charge Measurement in Polymer Insulated Power Cables Using the PEA Method*, PhD thesis, University of Southampton, UK, 2002.
- [19] T. Tanaka and A. Greenwood, *Advanced power cable technology*, Volume II, CRC Press Inc., USA, 1983.
- [20] B. Sanden, E. Ildstad and R. Hegergerg, "Space Charge Accumulation and Conduction Current in XLPE Insulation", 7th Inter. Conf. on Dielectric Materials, Measurements and Applications, Bath, UK, 1996.

- [21] C. Zhang, T. Mizutani, K. Kaneko and M. Ishioka, "Decay of Space Charge in LDPE and Its Blend Polymer", *J. Phys. D: Appl. Phys.* 35, pp.1875-1879, 2002.
- [22] L. A. Dissado, V. Griseri, W. Peasgood, E. S. Cooper, K. Fukunaga and J. C. Fothergill, "Decay of Space Charge in a Glassy Epoxy Resin following Voltage Removal", *IEEE Trans. Dielectr. Electr. Insul.*, Vol. 13, pp.903-916, 2006.
- [23] T. Mizutani, H. Semi and K. Kaneko, "Space Charge Behavior in Low-Density Polyethylene", *IEEE Trans. Dielectr. Electr. Insul.*, Vol.7, pp.503-507, 2000.
- [24] S. Mitsumoto, M. Fu, L. A. Dissado and J. C. Fothergill "Short time interval decay measurement of space charge in epoxy resin" *IEEEJ Trans. on FM*, Vol .126, No.4, pp.260 – 261, 2006



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