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The Influence of Material Modification and Residues on Space Charge Accumulation in XLPE for DC Cable Application

M. Fu
Department of Engineering
University of Leicester, Leicester, LE1 7RH, UK

G. Chen
School of Electronics and Computer Science
University of Southampton
Southampton, SO17 1JB, UK

L. Dissado and **J. Fothergill**
Department of Engineering
University of Leicester, Leicester, LE1 7RH, UK

ABSTRACT

Effects of cross-linking by-products (residues) of polyethylene on space charge accumulation and decay are investigated in the paper using the pulsed electro-acoustic technique. Space charge profiles have shown a great variation both in the charge initiation over the voltage ramping up process and later on long term stressing and decay (volts off) among the samples subjected to different conditioning, which results in diverse residues content (fresh, 0.5% residue and thoroughly degassed). The results show that by-products of cross-linking or the residual impurities play a key role in the space charge accumulation in cross-linked polyethylene. On the removal of impurities by degassing, small homocharge was built up in the vicinity of the electrode. It is concluded that space charge accumulation is governed by the charge injection through dielectric/electrode interface when sample is thoroughly degassed.

Index Terms - Space charge; Cross-linking by-products; XLPE and PEA.

1 INTRODUCTION

Cross-linked polyethylene (XLPE) as the most prominent polymeric material is being 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 of using XLPE as dc power cable insulation; the formation of space charge may distort the electric field distribution along 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 in the extreme situation [3].

For these reasons, space charge measurement has fascinated the attention of researchers over the world in past two or three decades and a numerous research has been concentrated on the mechanism of space charge formation, migration and accumulation in film samples. It is generally concluded that such space charge may result from electrons (and holes) 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) in 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 the prototype cable manufacture, thus many experiments still need to be carried out on plaque samples on the purpose of space charge suppression research in the modified insulating materials. The planar sample material for this research is exactly prepared in cable insulation manufacturing process which intends to well reflect the effect of material processing and afterwards treatment (conditioning) on charging characteristics. In this research, a batch of thick plaque samples made of differently

amended XLPEs is experimentally studied using the pulsed electroacoustic (PEA) method [14,15]. As the sample made for this research is usually thicker in order to reduce the surface effect observed in the thin film sample [16]. A relatively high voltage has therefore to be applied across the sample to obtain an adequate stress for the 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 briefed in the paper. The space charge results among these samples have shown a great variation in the terms of charge initiation in the voltage ramping process, charge accumulation over the stressing period and decay after the removal of external voltage. The samples subjected to different degassing treatment i.e. at the conditions of fresh (undegassed), partially degassed (0.5% residue) and thoroughly degassed were also studied in terms of space charge accumulation and decay.

2 EXPERIMENTAL

2.1 PEA SYSTEM FOR THICK PLAQUE SAMPLES

To satisfy the requirement of a relatively high stressing voltage across the sample, besides the application of voltage bushing on the electrodes system, an adequate flashover distance along sample's surface is also essential from the upper electrode (high voltage) to the ground electrode. Thus a bigger sample in size is required in the test. This makes it difficult to maintain an even contact between the sample and the electrode insulator surfaces. Some air gaps may inevitably form between them. As a result, the flashover voltage across the sample is significantly reduced. At the beginning of the test, this problem frequently occurred through this air gap path over the surfaces of sample at a voltage about 50kV. The problem was successfully solved by smearing silicone grease around the embedded semiconducting electrode of the sample. The new PEA system enables a dc voltage as high as 100kV to be applied across the sample. Photo in figure 1 shows the electrode system of PEA used in the research.

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

Acoustic wave propagation in the thick samples is significantly attenuated and dispersed due to the lossy and dispersive properties of the polymer. As a result, these features will wreck the detecting sensitivity and spatial resolution [13, 17]. A data compensation algorithm was developed in the research to get rid of the effect of attenuation and dispersion occurred in acoustic wave propagation. Figure 2 (a) and (b) 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 indicated in the peaks of the induced surface charges at the two electrodes in figure 2 (a). The signal from the upper electrode (the peak to the right hand) 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 if no bulk space charge presents in the volume of the dielectric material, like the profile shown in figure 2 (b), two surface charges have the same density and distribution.

(a) Raw data of calibration signal

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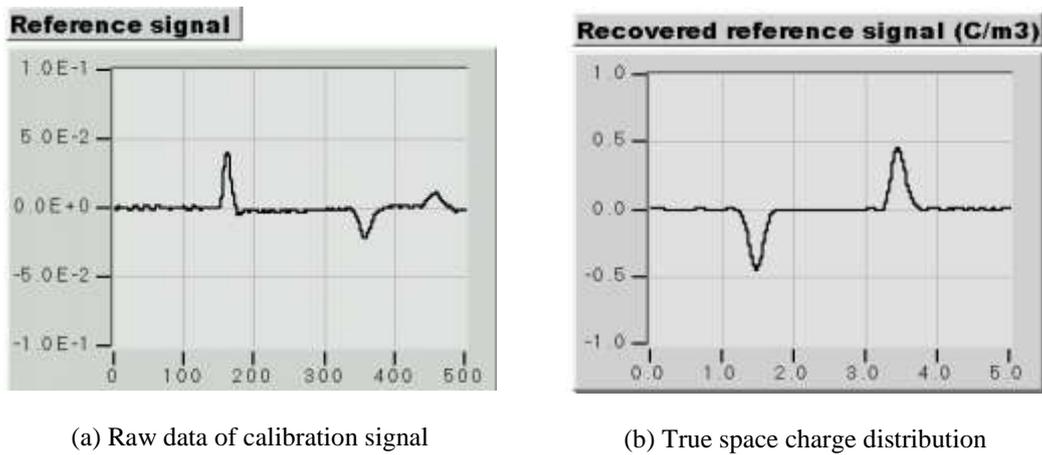
Figure 2. Space charge profile (in calibration) before and after attenuation and dispersion compensation

2.2 SAMPLES

The samples tested in the research are reference XLPE and modified XLPE. All of them are of same semiconductive electrodes being made of crossing-linked polyethylene 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 concentration. Figure 3 illustrates a sample's dimensions and cross section.
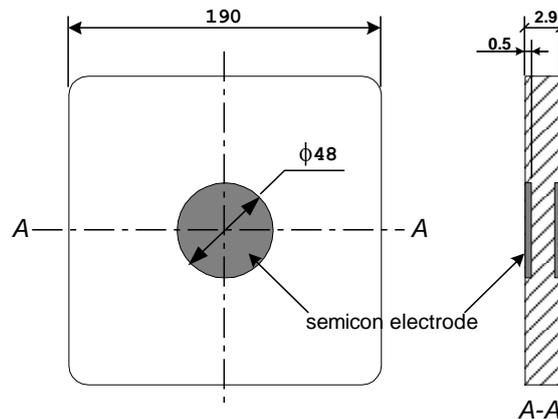
Figure 3. Sample dimension and its cross section

Apart from the flashover problem mentioned previously, several samples from the first batch failed due to electrical breakdown occurring at the edge of the embedded semiconducting electrode in the voltage ramp procedure or the stressing period. One more 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. This is believed to be the main reason for the electrical failure of the sample due to the rise of field in the vicinity of the edge. A semiconducting electrode with a rounded edge was then suggested 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.

2.3 EXPERIMENTAL PROCEDURE

Space charge measurements on each sample started with voltage ramp test in which applied voltage was stepping up from low to the required value of 70kV. The intention of this experiment is to find the threshold voltage above which a bulk charge starts to accumulate. The space charge measurement was then carried out at regular interval over the stressing time with external voltage on and off. The measurement under external voltage off is for good display in the case of homocharge by removing capacitive charge on electrodes. The space charge distribution in most of the samples reached its equilibrium within 24 hours, and some of them even sooner. At the end of the stressing test, the 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 are presented in Figure 4, in which the magnitude of the induced charge at the two electrodes against the externally applied voltage is plotted. To get rid of the influence of the stressing time on the charge formation, the measurement at each voltage step was carried out quickly (within 10 seconds), only the peak heights of output voltage which are proportional to the induce charges at the upper and the ground electrodes were read directly from the oscilloscope. The readings are therefore expressed in the unit 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 by anode in the figure.
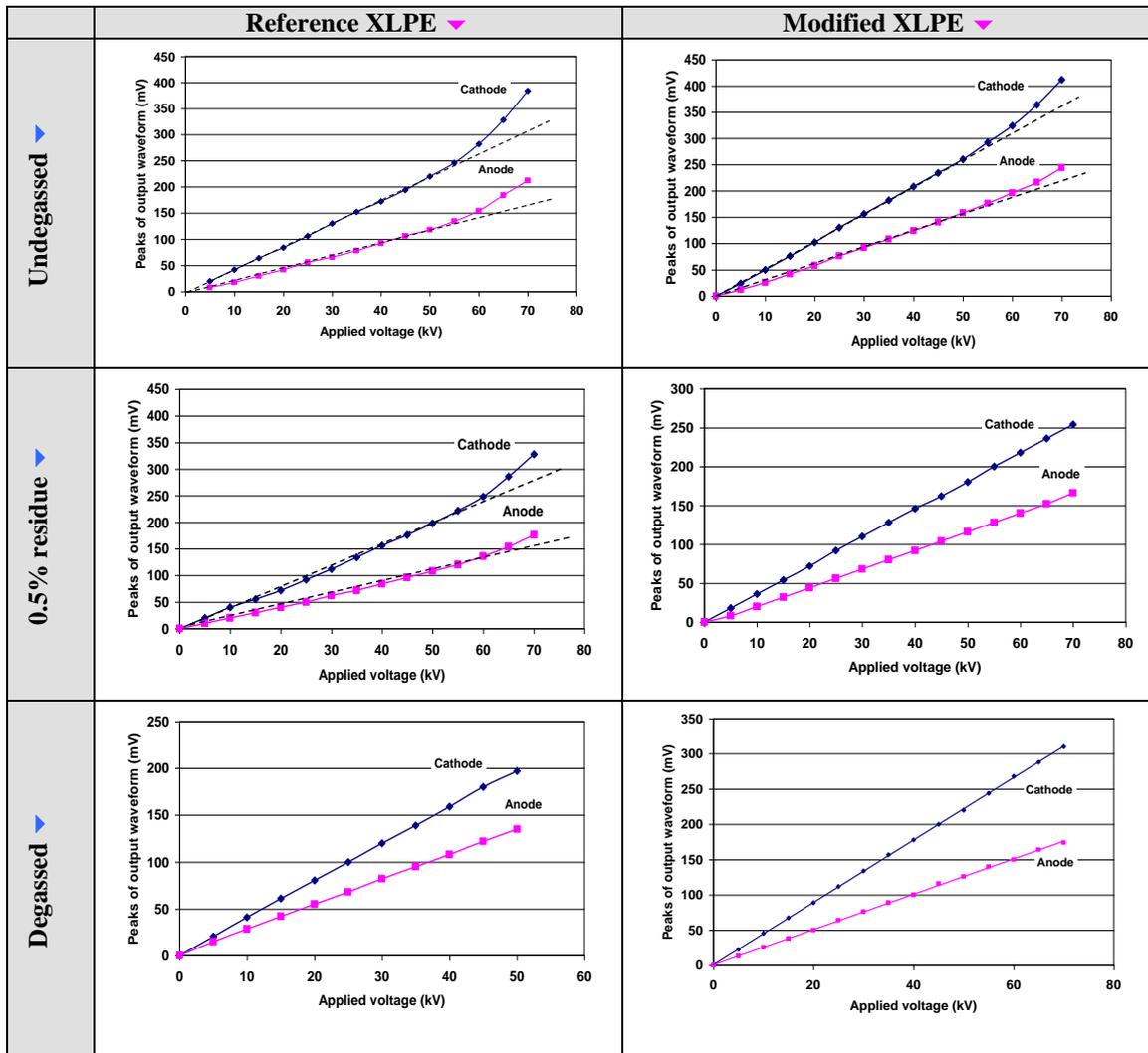
Figure 4. Relationship between the induced capacitive charge and the external voltage

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

3.2 SPACE CHARGE EVOLUTION OVER THE LONG-TERM FIELD STRESSING

Stressing test as long as 24 hours was conducted on all samples, during which space charge distribution was measured at regular intervals and the results are presented below. In order to clearly display the space charge in the case as homocharge, measurement was also carried out at each time with external voltage off. The charge profile titled "0 time" in each diagram stands for the result obtained soon after the voltage ramping up process.

3.2.1 REFERENCE XLPE SAMPLE

Volts on

Figure 4 shows the space charge distribution and progression among the samples with different residue contents. From the results of the undegassed sample and the partially degassed samples, it is seen that the influence of the residue content on the charge distribution is not very significant. In Figure 5 (a) and (b), two samples almost have similar space charge distributions after 1 hour stressing, while the charge accumulation speed is obviously affected by the residue in the sample. In the ramp test of the undegassed sample, a quite considerable charge had built up, whereas in the sample with 0.5% residue, most of the charge formed in the following one hour stressing after the voltage ramp. Due to the flashover occurring along the surface of the undegassed sample, no further data after one hour of ageing could be available as the test was terminated. As shown in Figure 5 (c), no space charge is clearly shown in the degassed sample although it had already been stressed at 50kV for 24 hours before the voltage was stepped to 70kV.
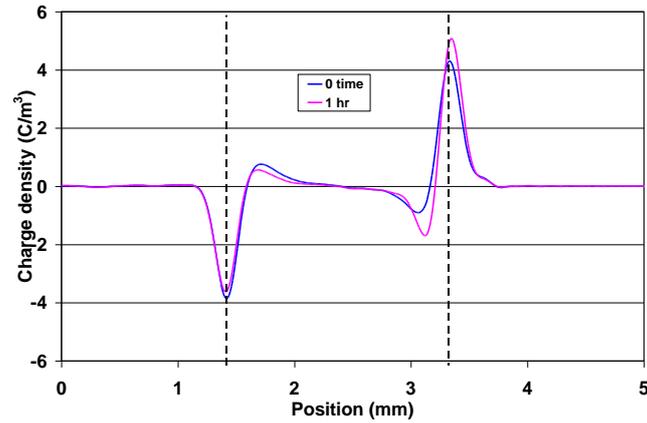
(a) Undegassed sample
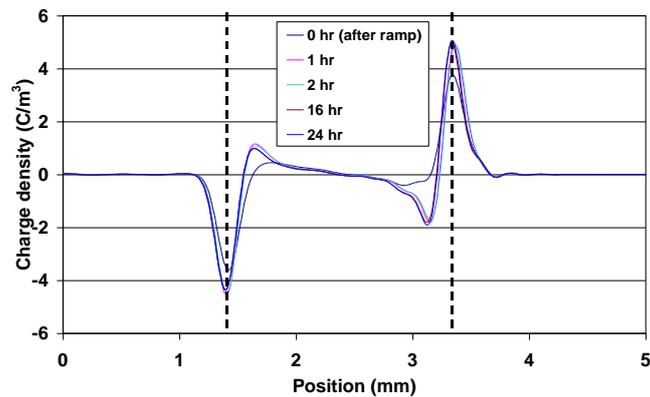
(b) Partially degassed sample with 0.5% residue
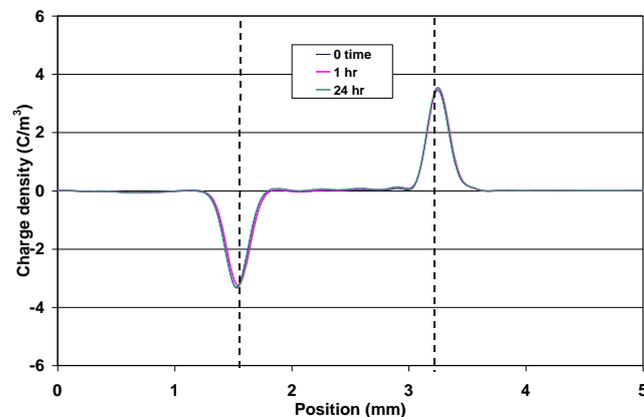
(c) Degassed sample

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

Volts off

Space charge distributions were also measured at the different times over the ageing term after removal of the external voltage to check the homocharge, which may be merged into the surface charge due to the external voltage. The results are shown in Figure 6. In the absence of the induced surface charge due to the external stressing voltage, the homocharge in the degassed sample is clearly observed, as shown in Figure 6 (c).
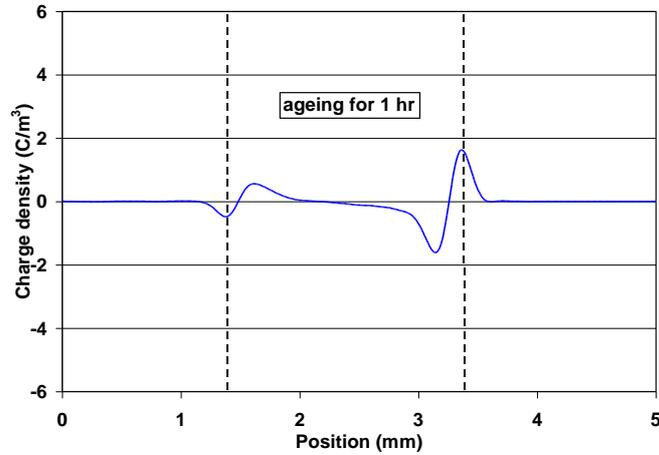
(a) Undegassed sample
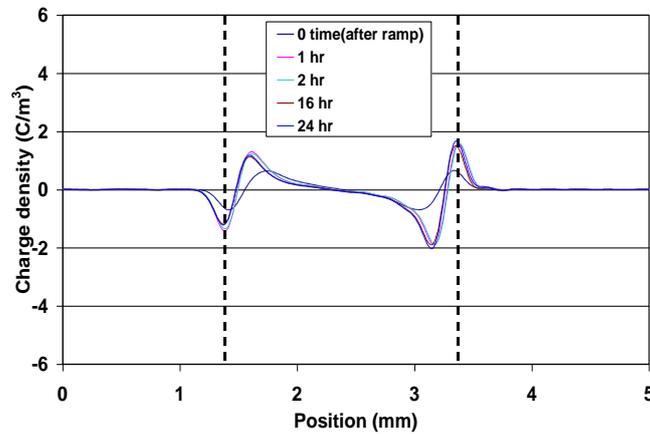
(b) Partially degassed sample with 0.5% residue
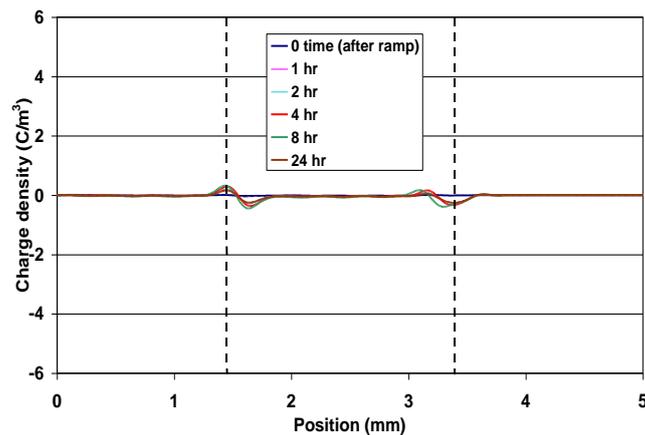
(c) Degassed sample with homocharge accumulation

**Figure 6.** Space charge profiles in reference XLPE (volts off)

Charge decay

Space charge measurements on each sample were followed by space charge decay tests, and the results are shown in Figure 7. It must be pointed out that Figure 7 (a) shows the retested results of the undegassed sample after the first flashover in which no decay test was carried out since the pulse voltage could not be applied across the sample.
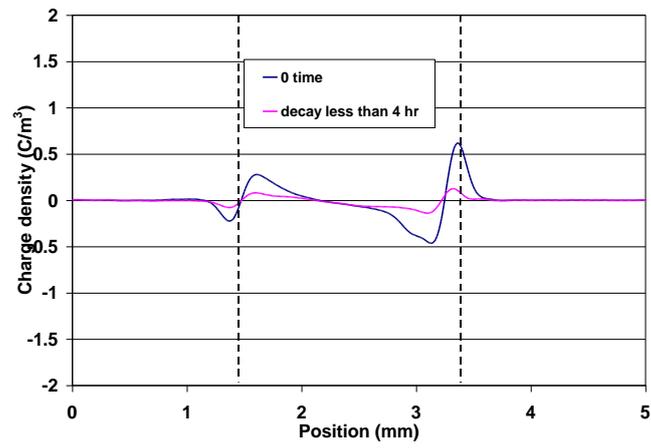
(a) Undegassed sample
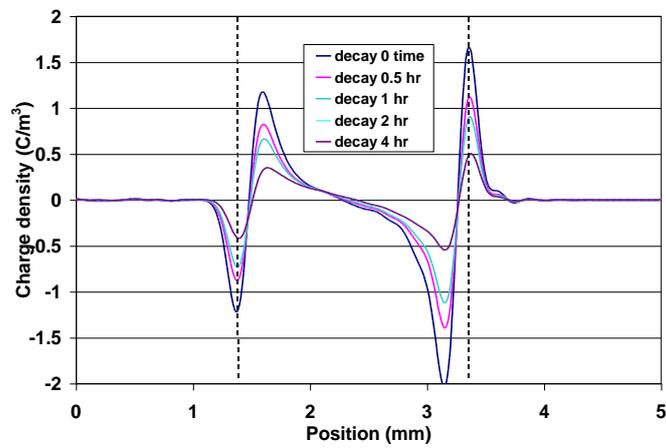
(b) Partially degassed sample with0.5% residue
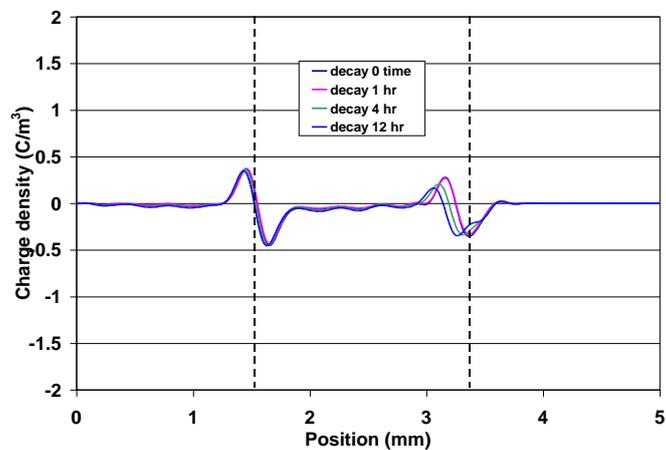
(c) Degassed sample

**Figure 7.** Space charge decay in reference XLPE

As the undegassed sample and the sample with 0.5% residue have a heterocharge accumulation, they possess a fast space charge decay speed in comparison with that of the degassed sample with a homocharge.

### 3.2.2 MODIFIED XLPE FOR HVDC CABLE

Volts on

Space charge measurements in modified XLPE for HVDC with the external voltage applied are shown in Figure 8.
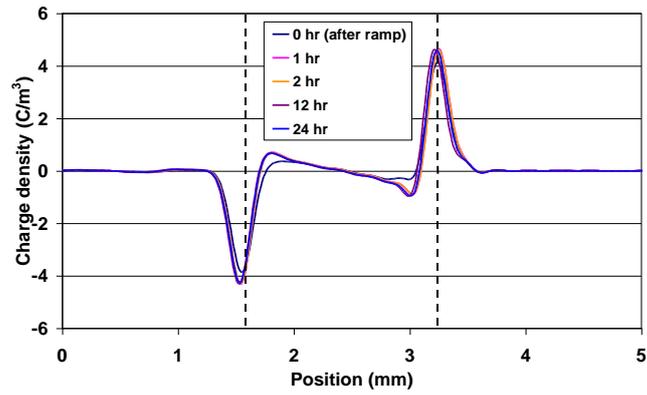
(a) Undegassed sample
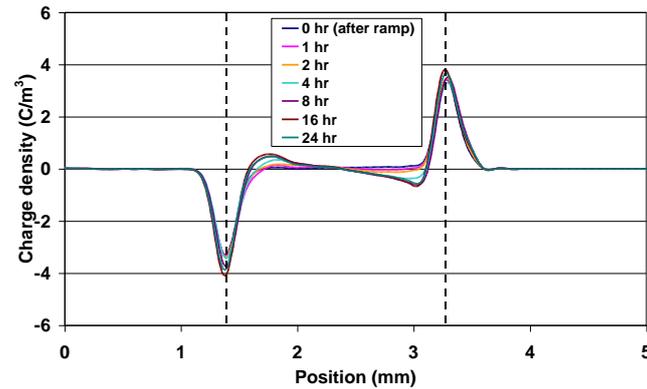

(b) Partially degassed sample with 0.5% residue
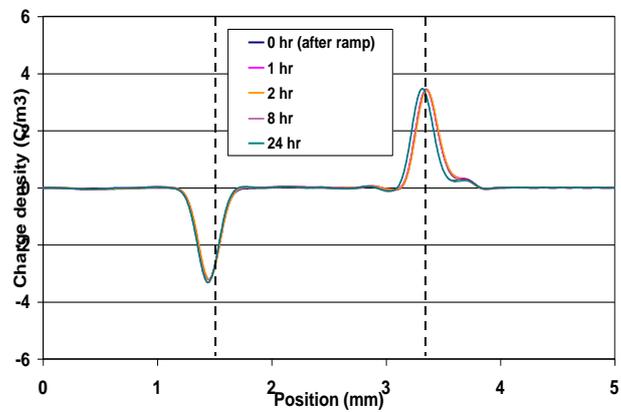
(c) Degassed sample

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

Without exception, the undegassed sample possesses a relatively fast charge generation speed, as shown in Figure 8 (a). After the fast heterocharge accumulation in the voltage ramp, the space charge distribution through the sample almost reached saturation within the one hour in the ageing term. In the sample with 0.5% residue, the space charge accumulation slowly progressed and saturated in about 8 hours. Figure 8(b) presents the space charge evolution in this sample over the whole ageing. The final charge distributions between these two samples are very comparable after 24 hours ageing. Unlike in the P3 material under the degassed condition, no space charge was observed in the P28 material over the whole ageing time.

Volts off

The results of voltage off are presented in Figure 9. The amount of charge in the undegassed sample is higher than that in the sample with 0.5% residue. No space charge accumulation in the degassed sample is visibly presented in Figure 9 (c), where a straight base line is shown when the voltage was removed after 24 hour ageing.
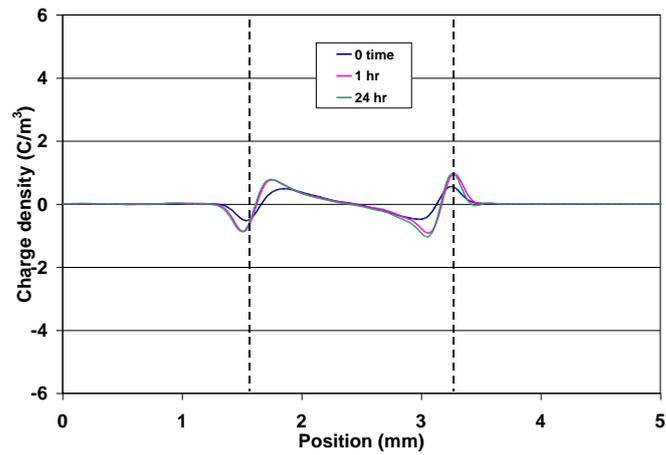
(a) Undegassed sample
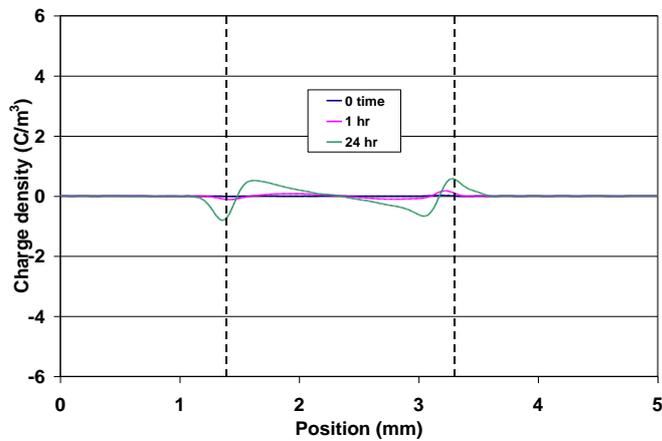
(b) Partially degassed sample with 0.5% residue
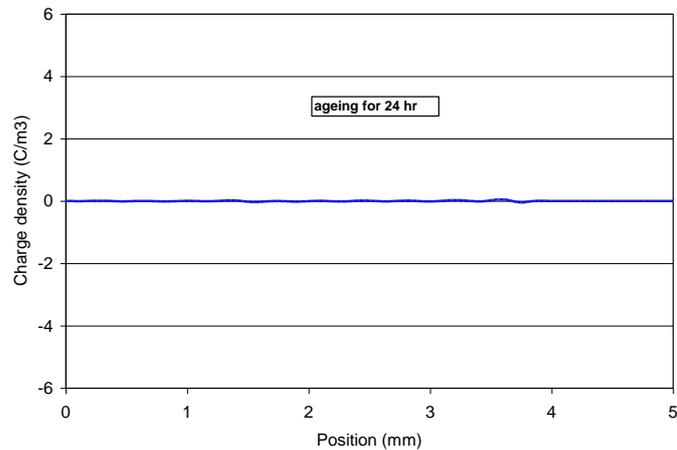
(c) Degassed sample

Figure 9 Space charge profiles in modified XLPE (volts off)

Space charge decay

Similar to the undegassed reference sample, the space charge in modified sample also possesses a fairly high decay speed. In Figure 10 (a), the massive heterocharge in the bulk material has died off within two hours after the electrodes were short-circuited, whereas the sample with 0.5% of impurities displays a very slow decay rate as shown in Figure 10(b). No decay test was carried out in the degassed sample because of the absence of the charge.
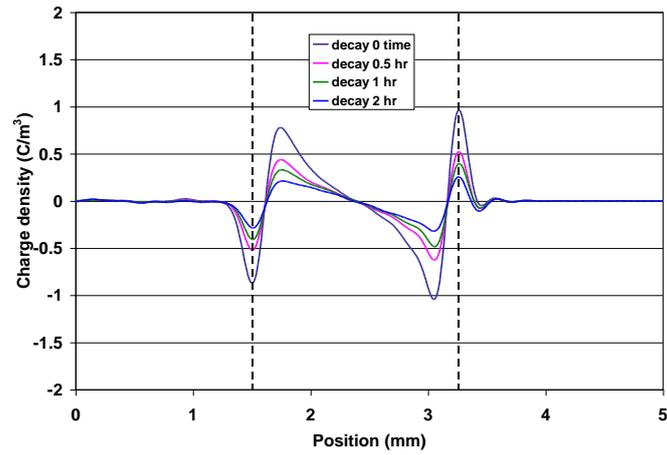
(a) Undegassed sample
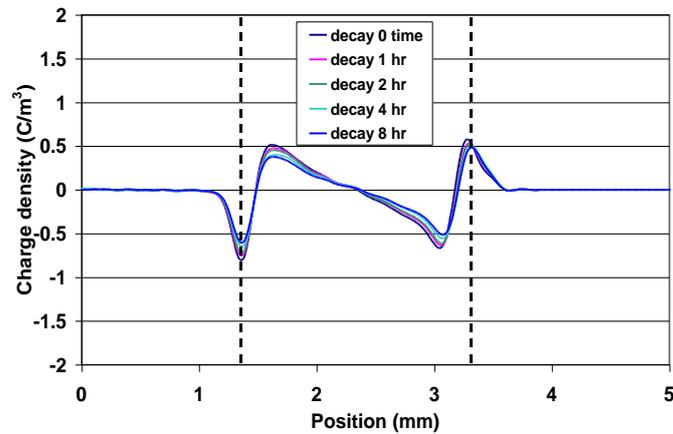
(b) Partially degassed sample with 0.5% residue

Figure 10. Space charge decay of sample P28

3.2.3 LDPE SAMPLE

For the comparison, the space charge distribution and its evolution were also studied on 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.

From the ramp test result shown in Figure 11 (a), the linear relationship between the induced charge on electrodes and the external voltage suggests no space charge appearing in the voltage stepping up process. Within the following stressing time, homocharges gradually accumulated in the bulk insulation at the vicinities of electrode, as shown in Figure 11 (b) and (c), and the latter one shows more clearly in the situation of external voltage removal. Like all the homocharges observed in the previous degassed XLPEs, the homocharge in this LDPE sample also showed a very slow decay speed, as illustrated in Figure 11 (d).
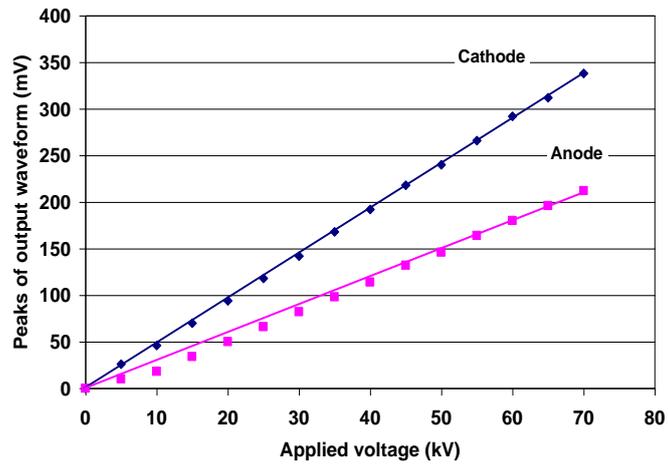
(a) Ramp test
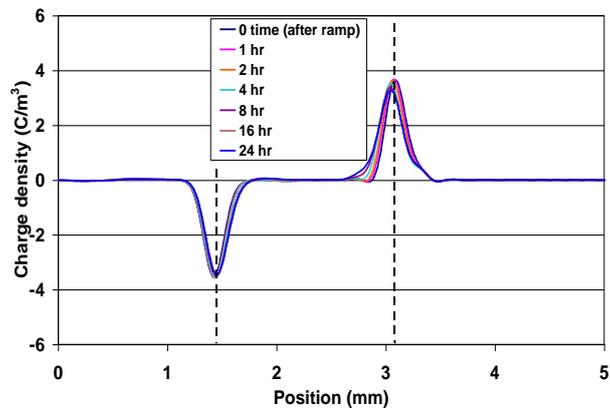
(b) Space charge profile (volts on)
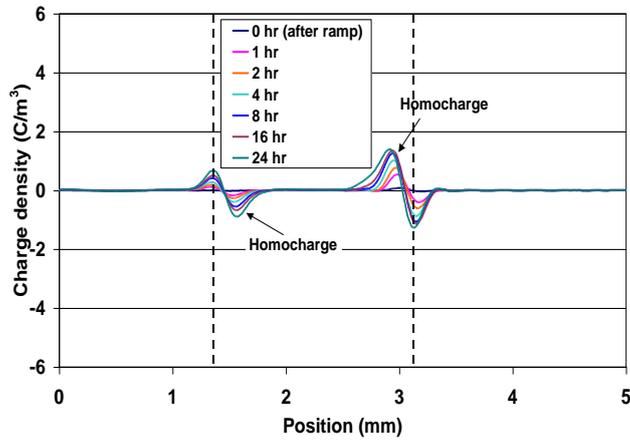
(c) Space charge profile (volts off)
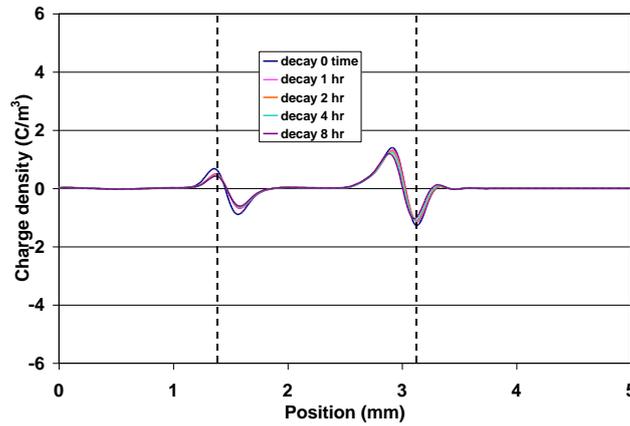
(d) Space charge decay
Figure 11. Space charge behaviour of LDPE sample

4 DISCUSSIONS

4.1 THRESHOLD STRESS FOR THE FAST CHARGE FORMATION IN RAMP TEST

The ramp test results in Figure 3 of the relationship between the induced charges at the electrodes and the external voltages indicate that the undegassed reference XLPE and modified XLPE samples have a heterocharge accumulation during the voltage ramp process. This fact is presented in the acceleration of tendency of induced charge on electrode with the increase of external voltage due to the enhancement of interfacial stress by heterocharge. The heterocharge formation during the voltage ramp in partially degassed XLPE reference sample was also observed 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 of each individual sample for the space charge initiation is listed in Table 1. For those specimens having no observed charge in 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

	Un-degassed	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 a relatively low electric field in comparison with those of the corresponding partially degassed or degassed samples. To assess the space charge behaviour at this stage, it seems that the modification to the material has achieved positive effects in term of space charge suppression.

Similar results have also been obtained in other two modified XLPE samples for HVDC application, which are not reported in the paper. It seems that the same XLPEs are subjected thermal treatment to remove 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 XLPE reference and purposely modified XLPE been increased from 28kV/mm and 33kV/mm in the fresh samples (with maximum residue content) to the values of 31 kV/mm and higher than 37kV/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 quickly formed in the voltage ramp process. Like all the degassed specimens, no space charge was observed in the LDPE sample during the voltage ramping from 0 to 70kV, because it is impurities free.

From the above results, it could be preliminarily concluded that the ionization of the cross-linking residue in the XLPE insulation may take place at a relatively low electric field. This may be the main contributor to the fast space charge at low stress. The conclusion on the other hand reveals the leading role of impurities in governing space charge accumulation in the bulk material.

4.2 SPACE CHARGE POLARITY AFTER 24HR AGEING

From the above test results, it is evident that specimens with various modification and thermal treatments (degassing) possess different space charge development speeds and final charge distributions. In particular, samples with and without

residues may have space charge of opposite polarities in the volume of the material adjacent to the electrode. Table 2 details the final space charge polarities of these samples.

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		

The results presented in this table clearly indicate that the residue or the impurity, no matter how high its content, is the main source of the heterocharge. In samples like pure LDPE or those having been degassed, it is very difficult to develop heterocharge 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 easily conclude that the modification to material is favourable in suppressing the space charge accumulation. Of course, the effect of the modification to XLPE on other space charge characteristics, such as charge building up and decay speed, and charge amount are other considerations to be taken into account in the material selection process.

Additionally, the fact of the longer time requirement for the small homocharge to develop in the degassed sample or LDPE sample may suggest that homocharge formation is more difficult than heterocharge [18,19]. The former case may need higher electric stress or longer time under the same electrode and dielectrics arrangement to develop same amount of space charge.

4.3 SPACE CHARGE BUILDING UP SPEED

The time for the space charge to reach its saturation is a practicable method to describe the charge building up speed.

The results shown in Figure 12 reveal that for the samples having residual impurities (which normally develop heterocharges as discussed in the previous section) 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 at the undegassed or with 0.5% residue condition, the space charge could reach its 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 building up speed in the undegassed sample and in the sample with 0.5% residue. Similar to the degassed sample, thermally pressed LDPE has also shown a very low charge accumulation speed.
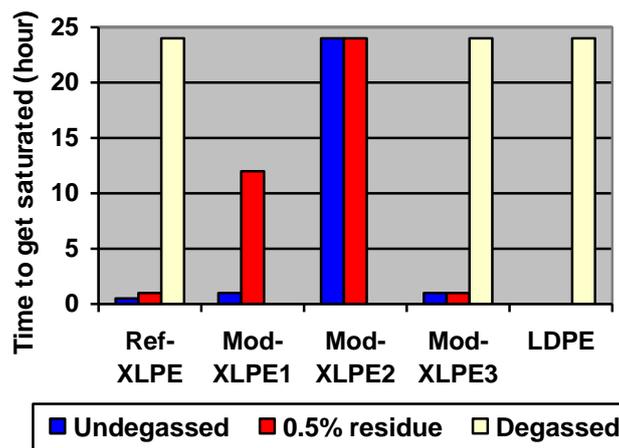


Figure 12. Saturation time of different samples

4.4 SPACE CHARGE DECAY RATE

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

First, in most cases the space charge decay speed corresponds to its building up 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 are regarded as typical fast charging and slow charging examples respectively.
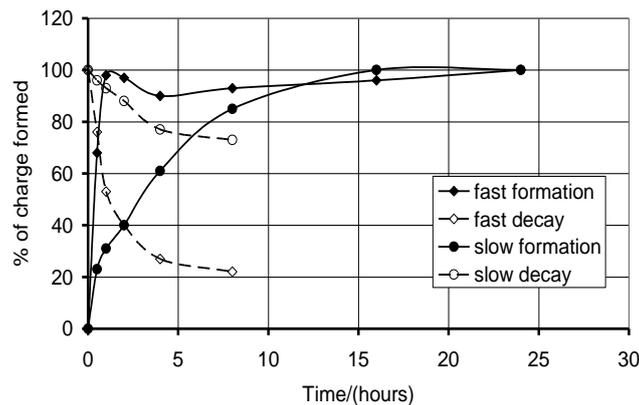

Figure 13. Space charge accumulation rate in accordance with decay rate

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

Second, homocharge in the degassed sample decays much more slowly than heterocharge in the undegassed sample. As shown in Figure 14, heterocharge in the undegassed XLPE-1 had a very high decay rate. After 3 hours, the maximum charge density declined to about 20% of its original value, while the homocharge accumulated in the degassed sample was relatively stable. Of course, if we refer back to the section 4, we know that homocharge also presented a very slow building up speed.

Space charge evolution and decay in Figure 11(c) and (d) also show the same features for the LDPE sample.
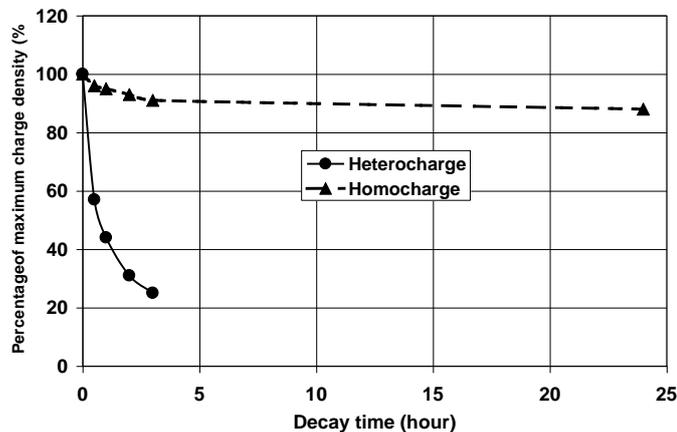
Figure 14. Comparison of homo and hetero charge decay speed

For the degassed sample, the removal of residue means that the homocharge is mainly contributed by electron injection at the cathode and extraction at the anode and it accumulates because of the mobility of electrons in the material is low. The homocharge due the trapped electrons and the left holes is more stable and as a consequence the charge will decay slowly if the external voltage is removed.

5 Conclusions

A brief introduction to the application of the PEA system for space charge measurements in thick plaque sample is given in the paper. Actual results of space charge distributions from a batch of modified XLPEs and reference materials are presented and discussed. Based on the research, the following conclusions may be drawn.

Firstly, the PEA system introduced here is ideal for measuring space charge in thick plaque samples which can be prepared exactly following the production of the polymeric cable. It can be viewed as a necessary method for studying and selecting materials for polymer insulated dc power cables.

Secondly, space charge results have revealed significant differences between the different XLPEs and the reference materials. They have proved the effectiveness of the modification to the XLPE for space charge suppression in dc insulation systems. Amongst these modified XLPEs, none of them displayed any space charge formation when stressed at ~40kV/mm for 24 hours after being subjected to the degassing treatment.

Thirdly, the by-products of cross-linking or the residual impurities play an important role in the space charge accumulation in XLPE. The modification to the material has reduced the space charge accumulation to a certain extent in comparison with the reference XLPE in the fresh condition, but a considerable amount of charge was still generated. When they were partially degassed leaving 0.5% of residue in the sample, the space charge density was impressively reduced. In the fully degassed sample, no space charge appeared in the testing condition. In fully degassed reference XLPE sample, a small homocharge was built up in the vicinity of the electrode. At this stage, space charge accumulation is governed by the charge injection from the electrode.

Finally, the space charge accumulation speed corresponds well with the decay speed. In other words, the faster the space charge accumulates, the quicker it decays. It is also found that homocharge in degassed material decays much more slowly than the heterocharge in the same undegassed material.

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