

This is the accepted manuscript, which has been accepted by IEEE for publication.

© 2012 IEEE. Personal use of this material is permitted. Permission from IEEE must be obtained for all other uses, in any current or future media, including reprinting/republishing this material for advertising or promotional purposes, creating new collective works, for resale or redistribution to servers or lists, or reuse of any copyrighted component of this work in other works. The full reference is:

**‘Effect of Electrical and Thermal stressing on Charge Traps in XLPE Cable Insulation’**

A. Tzimas, S. M. Rowland, L. A. Dissado

IEEE Transactions on Dielectrics and Electrical Insulation, Vol. 19, Issue 6, pp. 2145-2154, 2012.

DOI: [10.1109/TDEI.2012.6396975](https://doi.org/10.1109/TDEI.2012.6396975)

# Effect of Electrical and Thermal Stressing on Charge Traps in XLPE Cable Insulation

**Antonios Tzimas, Simon M. Rowland**

University of Manchester  
School of Electrical and Electronic Engineering  
Manchester, M13 9PL, UK

and **Leonard A. Dissado**

University of Leicester  
Department of Engineering  
Leicester, LE1 7RH, UK

## ABSTRACT

Insulation peelings were taken from a single 90 kV ac transmission cable whose segments had been stressed for between ~8000 and 10 000 h either at elevated temperature (363 K) or field 19.5 kV/mm (rms) (T= 293 K). Control peelings were taken from a cable segment that had experienced no stressing. The various peelings were subjected to an endurance test at 70 kV/mm (rms) and T = 363 K, which demonstrated that only those peelings that had experienced thermal stressing had a reduced lifetime compared to the set of control peelings. Space charge measurements made prior to the endurance test showed only negative space charge in all types of peelings and an analysis of the decay dynamics in the control set showed that it resided in two trap distributions at 0.8 to 0.9 eV and 1 to ~1.5 eV. Peelings from a second endurance test (55 kV/mm (rms), T = 363 K) were suspended when the test was terminated at 6088 h. Space charge measurements showed that these samples exhibited positive as well as negative space charge with the positive charge being ~60% of the total. The space charge decay dynamics also showed two trap distributions, ~0.65eV to 0.85 eV and 1eV to ~1.3 eV, with both positive and negative charge in each energy range. Detailed differences in the trap energy range and density in both distributions were found between peelings with different cable histories, and those differences that correlate with a reduction of endurance life are identified. A tentative explanation in terms of anti-oxidant consumption and trap generation by chemical reaction is proffered.

Index Terms — XLPE, ageing, cable insulation, space charge decay, charge traps, positive traps, negative traps, electrical ageing, endurance test.

## 1 INTRODUCTION

BETWEEN 1998 and 2002 the European Union, sponsored a research program code-named ARTEMIS [1] that aimed to investigate insulation ageing in 90 kV ac transmission cables insulated by cross-linked polyethylene (XLPE). Two cable manufacturers were involved to separately produce a High Voltage (HV) XLPE cable as a model for Extra High Voltage Systems (EHV) using the same special batch of super clean polymer provided by just one material supplier. Segments of each cable manufactured were stressed separately, at elevated fields, at elevated temperature, and a combination of these, for a period of time up to 10 000 h. A segment from each cable manufacturer was also kept unstressed as a control. Sections of cable were removed from stress at intermediate times and

subjected to tests. In some cases the cable was peeled and, after conditioning to remove volatile cross-linking by-products [2], the 150  $\mu\text{m}$  thick peelings were tested. In [3] ac-endurance tests were described that had been carried out on peelings from the cable of one manufacturer at a standard temperature of 363 K and field of 70 kV/mm (rms). These tests showed that when the peeling was from cable segments with a stressing pre-history as cable insulation that lasted for at least 5000h and included a thermal component their endurance life was reduced in comparison with peelings from the unstressed segment of cable. This provided direct evidence that high temperature during the cable stressing had aged the insulation [3]. In each set of samples one or more were suspended prior to breakdown so that other tests could be carried out. A similar procedure was applied to endurance tests carried out at 363 K and 55 kV/mm (rms), where a number of peelings survived for 6088 h when the test was terminated. Previous studies of space

charge behavior in cable peelings have shown it to be sensitive to past cable stressing [4] and subsequent endurance stressing [5, 6]. These un-failed peelings therefore provide a means of assessing the contention [7] that ageing results in changes in the density and energy distribution of the charge traps. This was done by measuring the space charge decay following voltage removal in the suspended samples, all of which had experienced the same time on the endurance test under the same conditions, and thus differed only in their stressing pre-history as cable insulation. Analysis of the space charge decay dynamics allows quantitative estimates of the trap charge density and their energy to be obtained [8, 9]. A comparison with the space charge behavior measured before the endurance test on peelings with the same cable pre-history showed that the endurance test itself had modified the samples to allow positive space charge accumulation where none had previously been observed. Furthermore it was found that there were differences in the trap distributions between peelings from the unstressed cable segment and those from cable segments that had been electrically and thermally stressed, and thus that the different cable pre-stressing had modified the trap distributions in XLPE in different ways. The origin of these changes and their relevance for the reduction in the endurance life of the peelings from the thermally stressed cable segment is discussed.

## 2 MATERIAL HISTORY

### 2.1 SAMPLE HANDLING

In the ARTEMIS program [2], one material supplier provided two cable manufactures with the same batch of XLPE resin in order to make model cables for Extra High-Voltage (EHV) with an insulation thickness of 14 mm corresponding to a 90 kV cable construction, as shown in Figure 1a. A segment from each cable was stressed under one of three conditions: a) thermal stress of 363 °K, b) electrical stress of a mean field of 19.5 kV/mm(rms) at approximately 293 K, and c) an electrothermal stress of 28 kV/mm(rms) and a temperature of 363 K. An additional segment was set aside and stored as an unstressed control. At various times a small section was cut off the segments and used for a range of tests. Table 1 shows the range of conditions and stress durations. The insulation of part of the section was peeled (peeling thickness of 150 µm) with a specially designed cryotome. The peelings were rolled into tapes with a width of 8cm as is shown in Figure 1b and were distributed to the rest of the ARTEMIS partners inside polyethylene-bags completely enclosed within aluminum bags, for investigation. From then onwards the tapes were stored at a temperature around 278 K in order to prevent any further changes. The properties of the peelings were then investigated using samples taken from between 2 mm to 4 mm (or as close as possible to those margins) from the inner semiconductive screen. Figure 2 shows on a rough scale the sequence in time that each process took place. Before carrying out any experiments, the samples were conditioned [10] (see Figure 2) so as to remove any volatile chemicals that are known to influence the behavior of XLPE in an undefined manner [2].

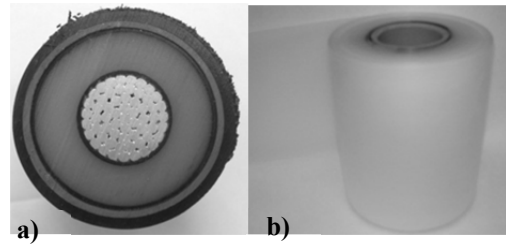


Figure 1. a) Section of the cable stressed during the ARTEMIS program and b) a tape roll after the insulation was peeled.

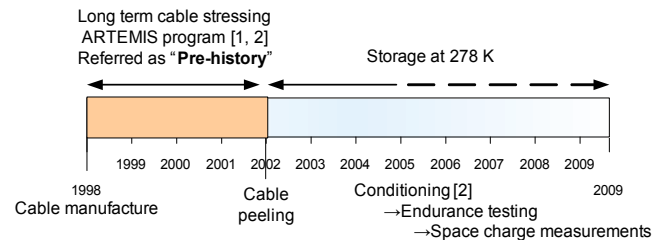


Figure 2. Time line of materials used.

### 2.2 SAMPLES USED

Table 1 presents all the materials (150 µm peelings) with their cable stressing histories that were used in the endurance tests described previously [3]. The samples are denoted by an acronym according to their stressing pre-history as a cable, i.e. US indicates peelings from the Un-Stressed control cable, while ETS indicates Electro-Thermally-Stressed and TS indicates thermally aged cable peelings respectively. The small suffixes “a” and “b” in the sample notation indicate the manufacturer that made the cable from which the peeling was taken. SA indicates peelings from a service aged cable of the same design that were included in the endurance test for comparison.

Table 1. Materials used for the endurance tests described in [3] and their pre-history as a cable. The highlighted materials are used for the current paper’s analysis.

Ref. name	Endurance Test	Pre-history of the material as a cable		
		ac stress (kV/mm)	Temp. (K)	Time (h)
US_a	A <sub>2</sub>	-	293	-
US_b	A <sub>2</sub> , B	-	293	-
E_S1a	A <sub>2</sub>	19.5	293	5000
E_S2a	A <sub>4</sub> , B	19.5	293	7747
E_S1b	A <sub>4</sub>	19.5	293	5581
E_S2b	A <sub>2</sub>	12.5	293	5000
_TS1a	A <sub>1,3</sub>	0	363	5000
_TS2a	A <sub>3</sub> , B	0	363	10000
ETS1a	A <sub>1</sub>	19.5	363	5000
ETS2a	A <sub>1,3</sub> , B	28	363	6000
ETS2a	A <sub>4</sub>	25	363	6000
ETS3a	A <sub>2</sub>	28	363	3000
ETS4a	A <sub>1</sub>	12.5	363	5000
SA	B	SERVICE	SERVICE	>18 YEARS

The samples used to carry out the de-trapping analysis in this study are highlighted in the Table 1 and are all from cable manufacturer ‘a’. Thus the only difference between the peelings lies in the stressing histories of these samples. The

electrical (E\_S2a) and thermal (\_TS2a) stressed samples have been stressed as a cable during the ARTEMIS programme, see Table 1 for details, whereas the US\_a samples did not experience any stress at that time. All the samples highlighted (including the US\_a peelings) subsequently experienced the stress conditions of endurance test B, see section 2.3, for 6088 hours without breaking down.

The peelings were thermally conditioned prior to the endurance tests, see Figure 1. The conditioning occurred at 323 K at ambient pressure for 48 hours and was aimed at removing volatile cross-linking by-products so that the tests could be carried on sample peelings with a standardized chemical composition [2, 10].

**2.3 ENDURANCE TEST**

Two protocols were used for the ac (50 Hz) endurance tests of the peelings described in [3]:

- A. 70 kV/mm (rms) at 363K,
- B. 55 kV/mm (rms) at 363K.

Results from endurance test A [3] are shown in Figure 3, where the sample notation is given in Table 1. This test was stopped after three samples had failed leaving one sample from each group in an un-failed state. The endurance time of these samples is marked on the plot and the samples are denoted as ‘suspended’. In all cases but one the US\_a and E\_S2a samples lie beyond the confidence limits for samples that have a thermal component in their stressing histories [3].

The endurance test B [3] involved five sets of peelings with very distinct cable pre-histories in the ARTEMIS program. These are peelings from: an unstressed, US\_a, an electrically, E\_S2a, a thermally, \_TS2a, an electro-thermally, ETS2a, and a service stressed, SA cable. After 6088 hours of the endurance test B conditions:

- no failure of unstressed material, US\_a had occurred,
- one failure of the electrically stressed material, E\_S2a had occurred and,
- two failures had occurred in the sets of thermally, \_TS2a, electro-thermally, ETS2a, and service stressed materials.

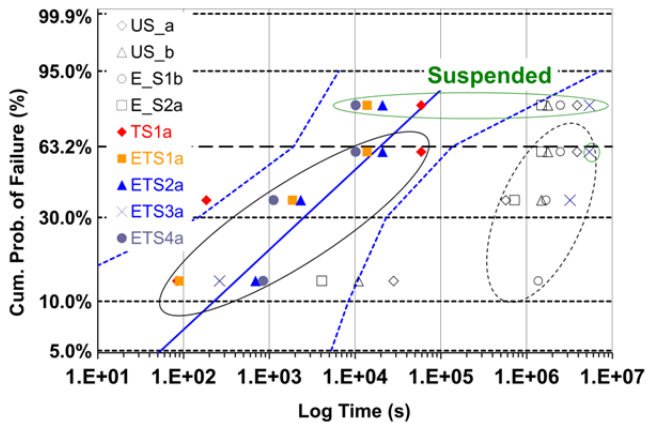


Figure 3. Weibull plot of Endurance test A life data taken from [3]. The solid dark line circle groups the samples with, and the dashed line circle groups the ones without any, thermal stressing pre-history as a cable. This test was suspended after the 3<sup>rd</sup> failure.

It should be noted that the results of both endurance test A and B follow the same trend, which is that materials with a thermal stressing component as a cable fail earlier than the material without thermal stressing component [3].

Endurance test B was suspended after 6088 h and sample peelings from the test representing the three main material stresses; unstressed, US\_a, electrical, E\_S2a, and thermal, \_TS2a, were investigated for their space charge decay characteristics and used in the subsequent charge trap analysis. The space charge measurements were obtained using the Pulsed-Electro-Acoustic (PEA) technique [9, 11].

**3 TOOLS USED FOR THE ANALYSIS**

**3.1 PULSED-ELECTRO-ACOUSTIC TECHNIQUE PROTOCOL**

The HV electrode is a carbon-black loaded semiconductive material and the ground electrode aluminum. A drop of silicone oil was placed between the electrodes and the sample to ensure good acoustic propagation. Previous work [9] on untested peelings from the unstressed cable has shown that the negative charge observed required 24 h to equilibrate between two trap ranges by transferring some of the charge residing in the shallower traps after 2h of poling to the deeper trap range, Figure 4. For this reason the poling time in the current experiments was chosen to be 24 hours for a field of 47 kV/mm at ambient temperature (~300 K).

**3.2 SPACE CHARGE DE-TRAPPING ANALYSIS**

All types of sample show similar space charge profiles, containing mainly negative space charge prior to the endurance tests. Figure 5 shows the profile from the US\_a peeling as a typical example of the profile before any endurance test stressing. Similarly Figure 6 shows the space charge profiles for the three peelings obtained after 6088 hours under the endurance test B [2]. In contrast to the measurements prior to the endurance test both positive

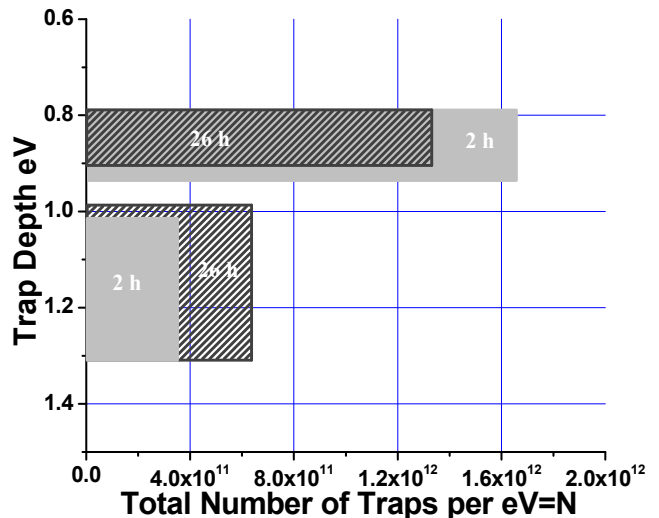


Figure 4. Distribution of space charge among traps from [9].

and negative space charge can be clearly observed in Figure 6. Almost all the positive charge adjacent to the anode of the US\_a sample is removed within 1200 minutes, whereas only a relatively small part of the negative space charge has decayed by this time. On the other hand the samples with electrical, E\_S2a, and thermal, \_TS2a, stressing histories retain both negative and positive charge for longer, suggesting the occupation of deeper traps.

Although the time-dependence of space charge decay may be governed by neutralizing electrode injection when the space charge generated electrode field is high enough [12, 13], at times greater than ~ 10 s when the space charge density is 10 C/m<sup>3</sup> or less it can usually be taken to be controlled by the occupied traps [8, 14], and hence can be used to determine their energy range and density [8, 9, 15, 16]. The model described in [8, 9] will be used for the current analysis. In this model it is assumed that space charge is trapped in a set of states that have a constant number, N per eV, covering a finite range of energies Δ<sub>max</sub> to Δ<sub>min</sub>. The time-dependence of the total charge (Q(t)) is determined by the progressive release of charge from deeper and deeper traps, and is given by;

$$Q(t) \approx a - Q(0)kT/[\Delta_{max} - \Delta_{min}]\ln(t) = a - eNkT\ln(t) \quad (1)$$

in the range of time given by,

$$\exp(\Delta_{min}/kT) < vt < \exp((\Delta_{max}/kT)) \quad (2)$$

Here  $\nu$  is the attempt-to-escape frequency given by

$$\nu = kT/h \quad (3)$$

Multiple ranges of traps with different values of Δ<sub>max</sub> and Δ<sub>min</sub>, as in Figure 4, are handled by determining a value for Q(0) (the total charge in a given set of traps) for each set and adding the contributions together at a given time to give the total charge observed at that time. In this way it is possible to handle a trap distribution that is not uniform in density per eV over its whole energy range. In our initial analysis we investigate the decay of the total charge by adding all the charge together independent of polarity using the following equation:

$$Q_{tot}(t) = A \int_{x_1}^{x_0} |\rho(x,t)| dx \quad (4)$$

A subsequent analysis will investigate the decay of space charge of each polarity separately. The integral in equation (4) is confined by the limits x<sub>0</sub>, x<sub>1</sub> that are defined at the interface between the electrodes and the sample, see Figure 6 where the space charge decay profiles are presented and the electrode area is represented with the two rectangular shapes. The location of the interface between sample and electrode has

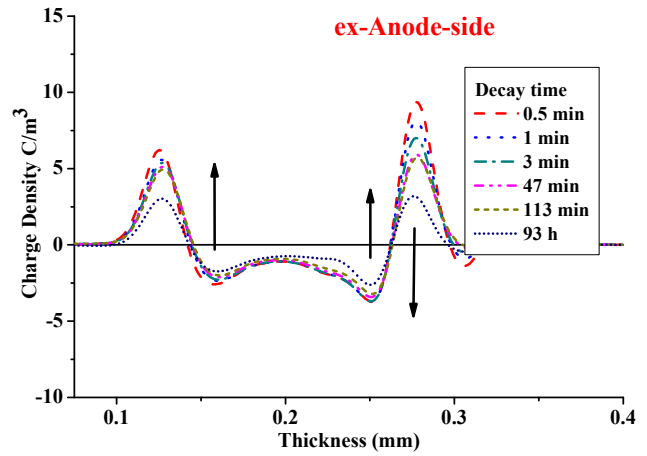


Figure 5. Space charge decay profiles of the unstressed sample, US\_a, before any endurance test stressing [5, 9].

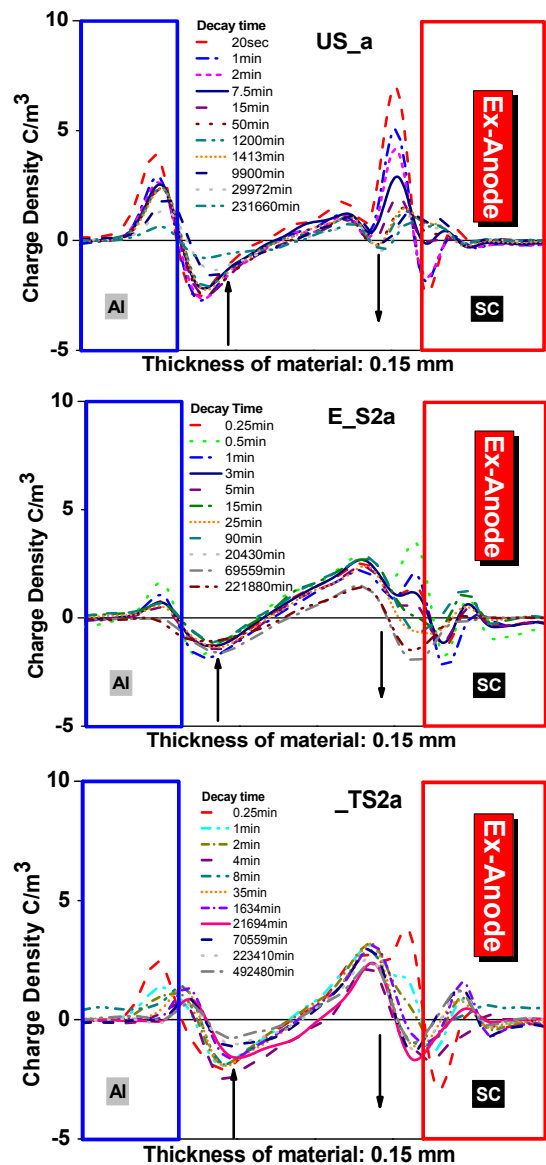


Figure 6. Space charge decay profiles of the unstressed sample, US\_a, electrically pre-stressed sample, E\_S2a, and thermally pre-stressed sample, \_TS2a after 6088 hours of endurance test B stressing.

been pin-pointed at the edge of the negative and positive electrode peaks of the charge profile once the voltage had been applied, i.e. at zero minutes of voltage application. The measured decay of the total space charge estimated from equation (4) for the three samples under investigation is given by the symbols plotted in Figure 7.

### 4 RESULTS AND ANALYSIS

The time dependence of the space charge decay for the three different peelings as well as the fitting of equation (1) to each set of data is shown in Figure 7, with the data represented by the symbols and the fits by the continuous lines. It can be seen that the space charge decay shows two features: a rapid decay that is completed by about ten minutes corresponding to one energy range of traps; and a long time decay starting around  $10^3$  min corresponding to traps at deeper energies. The fitting parameters can be found by determining the  $Q(0)$ , the time of onset and end of the logarithmic regions, and the slope of the logarithmic region separately for each trap distribution [9,17]. The results are given in Table 2, and a schematic diagram of this trap analysis is presented in Figure 8.

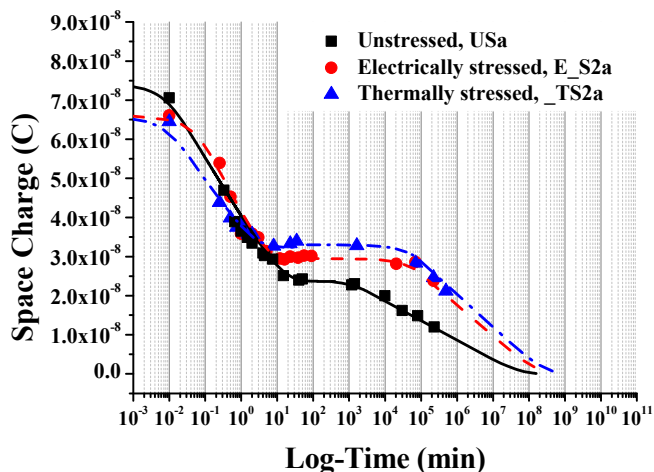


Figure 7. Decay over time of total space charge estimated from equation (4). The symbols give the measured values and the trap distributions fitted using equation (1) are shown by the lines.

Table 2. Parameters defining the trap distributions.

Parameter		Material		
		US_a	E_S2a	_TS2a
Shallow traps	Q(0) (C)	$5.0 \times 10^{-8}$	$3.7 \times 10^{-8}$	$3.33 \times 10^{-8}$
	$\Delta_{min}$ (eV)	0.64	0.69	0.64
	$\Delta_{max}$ (eV)	0.84	0.81	0.79
	N (eV) <sup>-1</sup>	$1.58 \times 10^{12}$	$1.92 \times 10^{12}$	$1.38 \times 10^{12}$
Deep traps	Q(0) (C)	$2.40 \times 10^{-8}$	$2.95 \times 10^{-8}$	$3.30 \times 10^{-8}$
	$\Delta_{min}$ (eV)	0.95	1.05	1.05
	$\Delta_{max}$ (eV)	1.23	1.26	1.27
	N in (eV) <sup>-1</sup>	$5.33 \times 10^{11}$	$8.74 \times 10^{11}$	$9.17 \times 10^{11}$

The energy distribution of space charge traps according to Table 2 as represented in Figure 8 shows that most (67%) of the charge in the unstressed sample occupies the shallower

trap range ~0.65 to 0.85 eV. In the electrically and thermally pre-stressed samples the space charge is more evenly spread between the two trap ranges.

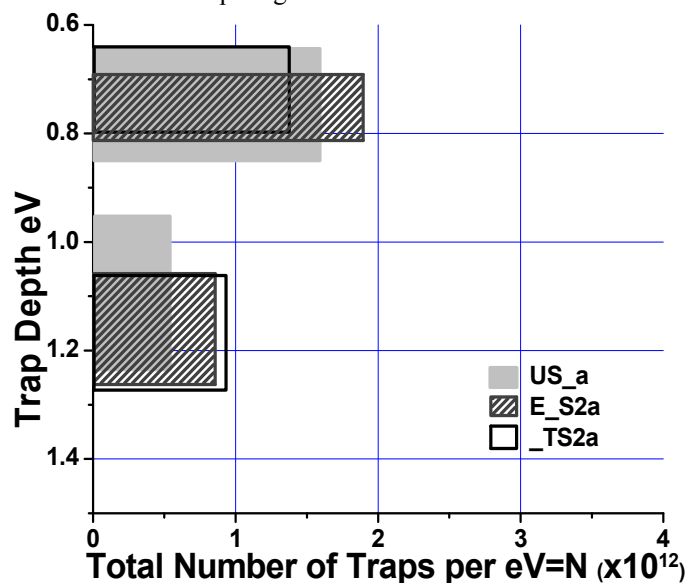


Figure 8. Energy distributions of space charge traps according to Table 2.

#### 4.1 CHARGE TRAP POLARITY ANALYSIS

The space charge decay presented in Figure 7 and its corresponding analysis, Figure 8, makes no distinction between positive and negative space charge. We have therefore analyzed separately the decay of the positive and negative space charge regions. The negative and positive space charge decay plots of the three samples under study are presented in Figure 9. The space charge data of the negative and positive polarity charge is calculated using equation (4) where the limits are confined within the electrodes surrounding negative or positive charge. Equation (1) has been again used to fit the data points, with the fit given by the continuous lines in Figure 9, and estimate the trap ranges that correspond to the positive and negative charge polarities, see Table 3. We have presented this analysis in Figures 10 and 11 in the form of schematic plots of trap energies, with the fitted values given in Table 3. Estimates of the value of  $Q(0)$  for the ‘shallow’ trap distribution is difficult and hence the trap number per eV should be taken as an approximate indication of magnitude rather than an exact value.

The first thing that can be noted is that the space charge decay before ten minutes observed in Figure 7 is contributed by both negative and positive space charge, though the contribution of the negative charge is less than that of the positive space charge. The energy range of the negative charge shallow traps (0.64-0.79 eV) is similar to that found in the US material prior to the endurance test (Figure 4) and lies within the range found for the positive charge shallow traps (0.65-0.85 eV). This means that any shallow trapped negative charge residing under the positive space charge (i.e. in the same location but with smaller concentration) would not be observed as it would decay during the same time as the shallow trapped positive charge. However in all three materials the decay of shallow trapped positive charge reveals extra deep trapped negative charge at a time of about 10 min

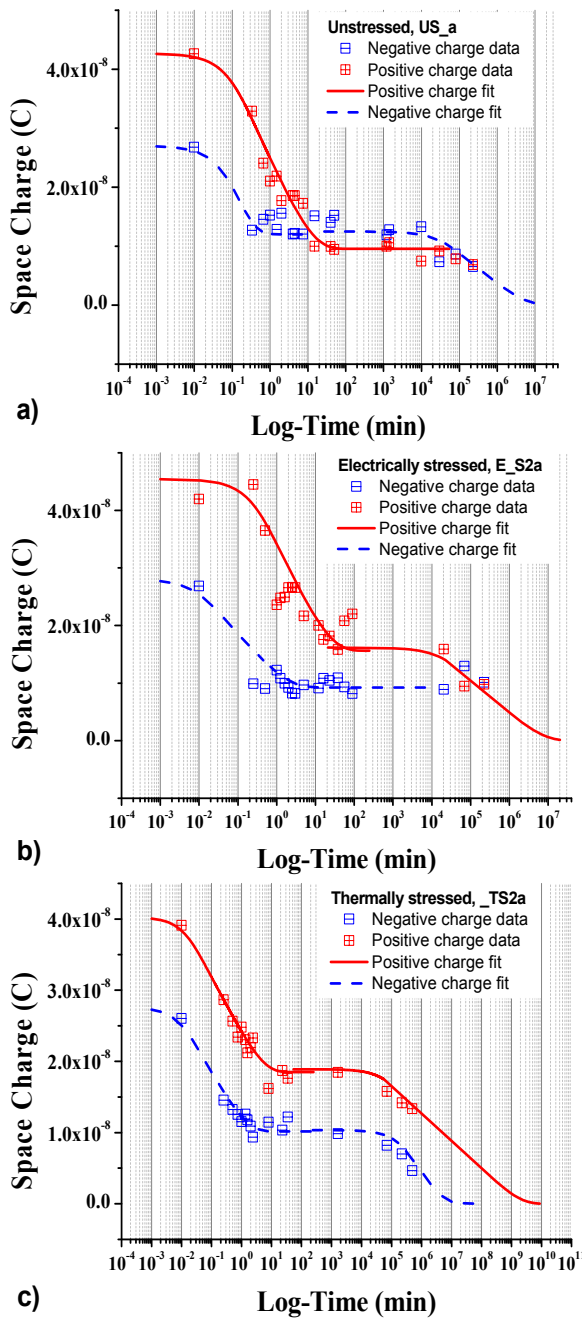


Figure 9. Space charge decay for negative and positive charge: a) unstressed sample, US\_a, b) electrically pre-stressed sample, E\_S2a, and c) thermally pre-stressed sample, \_TS2a.

as shown by small increases at this time. This is most noticeable in the US\_a material. The fitting has made no allowance for these variations, but instead takes a line through them dictated by the constant regions and regions of continuous decay.

The behavior of the deep trapped charge is different in the three materials. In the US\_a material the deep trapped negative charge starts to decay around  $10^3$  min corresponding to the decay in total charge at this time observed in Figure 7. This negative charge resides in deep traps of a similar energy range to that found in the US\_a material prior to the endurance test (Figure 3). The positive charge in the deep traps in the

Table 3. Parameters defining the separate trap distributions of positive and negative charge.

Parameter	Material						
	US_a		E_S2a		_TS2a		
Shallow	Polarity	(+)	(-)	(+)	(-)	(+)	(-)
	$Q(0) \times 10^{-8}$ (C)	3.31	1.5	2.98	1.87	2.17	1.74
	$\Delta_{min}$ (eV)	0.70	0.70	0.72	0.64	0.65	0.64
	$\Delta_{max}$ (eV)	0.84	0.73	0.85	0.79	0.81	0.78
	$N \times 10^{12}$ (eV) <sup>-1</sup>	1.51	3.66	1.48	0.798	0.837	0.768
Deep	$Q(0) \times 10^{-8}$ (C)	0.955	1.25	1.61	0.923	1.89	1.03
	$\Delta_{min}$ (eV)	>1.1	1.03	1.02	>1.08	1.04	1.08
	$\Delta_{max}$ (eV)	NA	1.17	1.18	NA	1.32	1.16
	$N$ in $\times 10^{11}$ (eV) <sup>-1</sup>	NA	5.62	6.27	NA	4.17	7.48

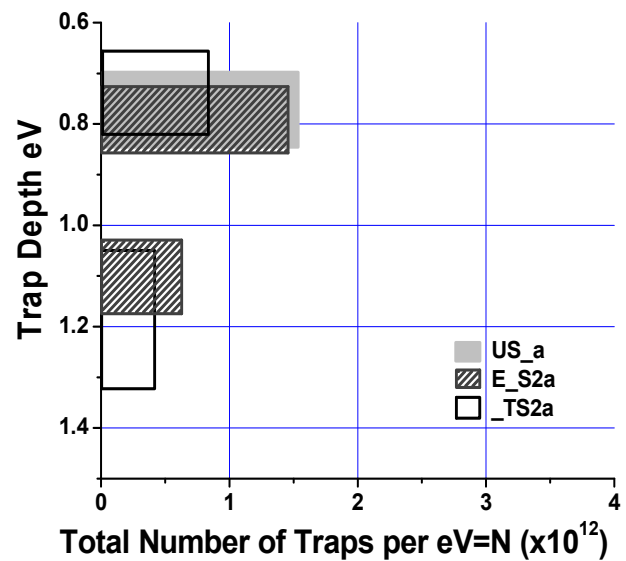


Figure 10. Positive trap range distribution according to Table 3.

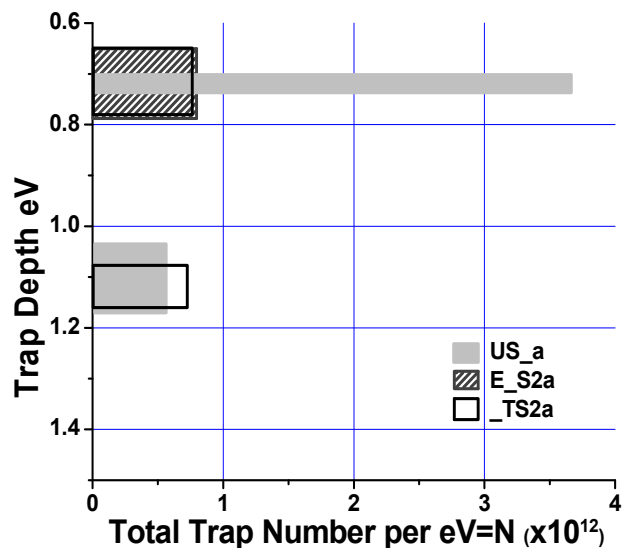


Figure 11. Negative trap range distribution according to Table 3.

US\_a material is less than the negative charge (~60%) and has not decayed by  $2 \times 10^5$  min. This means that the decay of the total space charge at long times seen in Figure 7 for the US\_a material is contributed by the removal of negative space charge. In contrast the deep trapped positive space charge is bigger than the negative space charge in the E\_S2a (~1.8:1) and \_TS2a (~1.8:1) materials. In the E\_S2a material the deep trapped positive space charge contributes the decay seen after  $10^4$  min in Figure 7, with the estimated trap depth range shown in Figure 10. In the \_TS2a material the deep trapped negative and positive space charge starts to decay at roughly the same time ( $\sim 3 \times 10^5$  min), as can be seen from the plot of total charge, Figure 7. This means that the charge in the \_TS2a material is trapped deeper than  $\sim 1.05$  eV. These results show two things. In the first instance the endurance stressing has introduced both deep and shallow positive charge traps. In the second instance the different cable history of samples prior to the endurance test has resulted in differences in the charge trap density and energy range.

## 5 DISCUSSION

Prior to any ac endurance test stressing [9] the peelings show only negative space charge that is trapped in two energy ranges 0.8-0.92 and 1-1.3 eV. Both of these ranges are essentially deep traps, nevertheless in order to differentiate them we have referred to them as “shallow” and “deep” traps respectively. The lack of evidence for positive space charge before the endurance test [4, 5, 9], indicates that either no positive carriers are injected, or such positive carriers that are injected are not trapped or trapped at depths shallower than our time range can detect [8,13]. Since the conditions and protocol for space charge measurement are the same before and after the endurance test the observation of positive space charge after the test means the traps in which it is found to lie (similar in energy range to those of the negative charge see Figure 10 and Table 3) must have been introduced by the severe conditions (high electrical stress (55 and 70 kV/mm<sub>rms</sub>) and high thermal stress (90 °C)) of the endurance test itself. Although the charge traps observed have to be associated with chemical features [18] it is not clear what their nature is. Spectroscopic studies [19, 20] have shown that some portion of the cross-linking by-products, acetophenone and cumyl alcohol leach out from the cable during stressing although much is retained in non-permeable voids [19] or attached to the polymer chains. At the same time acrylate solvent species enter the cable from its semicon layers. These species together with the by-product  $\alpha$ -methylstyrene are known to act as charge traps [18] but the thermal conditioning of the XLPE peelings in the Artemis programme [1] was designed to remove such volatile species [2], however a spectroscopic analysis has shown [20] that some portion of their concentration is retained after such conditioning. These species may be the origin of the charge traps, particularly

prior to the endurance test. In [18] it was calculated that both acetophenone and  $\alpha$ -methyl styrene could act as a trap for electrons at deep energy levels ( $\geq 1$ eV), whereas the binding energy of an electron to cumyl alcohol was only  $\sim 0.3$  eV and hence too small to be a candidate for the negative charge traps observed here. The negative traps prior to the endurance test could therefore originate with the residual concentrations of acetophenone or  $\alpha$ -methyl styrene. The binding energy of holes (i.e. hole trap depth) of acetophenone is only ( $\sim 0.3$  eV) [18] and thus it cannot function as the trap for positive charge observed here. A possible candidate for the positive charge traps is  $\alpha$ -methyl styrene, which traps both positive and negative charges at depths (binding energies) of ( $\sim 1-1.5$  eV) [18]. The positive traps could also be related to acrylates that are known to migrate into the cable from the semicon layers, especially under thermal stressing [19, 20]. An additional possibility for the generation of positive traps is chemical modification such as oxidation and possibly consumption of the antioxidant [21, 22] which may have taken place under thermal stressing and possibly also under the influence of the inward migration of acrylate species. It should be noted that the trap depths calculated may not be the binding energy of the charge to a molecule of a group that is part of the polymer. The charges may be strongly bound to a small molecule, e.g. the acrylate solvent for the cable semicon that has penetrated the cable [19, 20]. Hence the estimated trap depth may be the activation energy required for the molecular ion to transport out of the polymer. Such an interpretation would associate the magnitude of the deep trapped charge with the amount of such molecular species that was retained in the polymer at the stage when the space charge measurements were made. The measurements made before the endurance test indicate that for all samples the chemical species retained after the conditioning was only able to trap negative charge and so is likely to be the acetophenone retained by the samples in non-permeable voids [19, 20], although the existence of some positive charge trapped in the same location as the negative charge and with trap depths lying within the negative charge energy range cannot be entirely ruled out.

The different cable history of the samples results in differences in the trap distributions after the endurance test. In terms of shallow and deep trapped charge the following have been observed:

- a) **Shallow Trapped Charge:** in all samples the amount of positive shallow trapped charge is greater than the amount of shallow trapped negative charge; the ratio is greatest in the unstressed sample (US\_a) and smallest in the thermally pre-stressed sample (\_TS2a). The amount of negative shallow trapped charge is much the same for all samples and probably corresponds to the acetophenone retained after the conditioning. In contrast, the amount of shallow



trapped positive charge follows the sequence  $US\_a > E\_S2a > \_TS2a$ . The shallow positive charge traps must be generated during the endurance test and the results show that the  $US\_a$  sample is the most susceptible to their generation. They must then be associated with chemicals more of which have been retained in the  $US\_a$  sample than in the  $E\_S2a$  or  $\_TS2a$  sample. Since these chemicals do not trap positive charge prior to the endurance test, the traps must then be the consequence of their reaction or consumption during the endurance test. The “shallow” trapped component of the charge therefore would only correlate with the effect of cable thermal stressing on reducing the endurance life [9] (see Figure 3), if the reduction of the shallow positive charge traps were indicative of differences between the samples in the extent to which life preserving chemical species, such as anti-oxidant, were removed or consumed during the endurance test. The thermally stressed cable sample would then have to be regarded as the most susceptible to such consumption.

- b) **Deep Trapped Charge:** in the  $US\_a$  sample there is more deep trapped negative charge than positive charge and the highest filled trap in this range has a depth of  $\sim 1\text{eV}$  allowing the deep trapped negative charge to start to decay at  $10^4$  min. On the other hand, there is more deep trapped positive charge than negative charge in the  $ES\_2a$  and  $\_TS2a$  samples. In both samples  $ES\_2a$  and  $\_TS2a$  the deep trapped positive charge starts to decay at  $\sim 10^4$  min corresponding to a highest filled trap with a depth of  $\sim 1\text{eV}$ . The deep trapped negative charge in  $ES\_2a$  sample has not decayed by  $2 \times 10^5$  min, indicating that in these cases the trap depth has to be greater than  $1.1\text{eV}$ . The amount of deep trapped negative charge in the  $US\_a$  sample is about 30% higher than it is in either  $ES\_2a$  or  $\_TS2a$ , which have almost the same amount of deep trapped negative charge. This would be consistent with an interpretation of the deep trapped negative charge as due to residual acetophenone [19,20], which acts as traps for electrons [18]. The decay of the deep trapped negative charge in the  $US\_a$  sample would then be associated with the reduction of the acetophenone ion concentration to the level retained in the  $ES\_2a$  and  $\_TS2a$  samples after migration and reaction during Artemis cable stressing [19].

The significant features here are the amount of deep-trapped positive charge and the times at which the deep trapped charge is removed. In the case of the  $US\_a$  sample there is very little deep-trapped positive charge compared to the others and it does not decay during our measurements. This indicates that the  $US\_a$  sample is not very susceptible to the processes that develop the deep positive traps during the endurance test, and that such traps as are generated are deep or relate to ionic

species that are very difficult to remove, perhaps bound to the polymer.

Both the  $E\_S2a$  and  $\_TS2a$  samples contain about 70-100% more deep trapped positive charge than the  $US\_a$  sample, indicating that these samples are more susceptible to the generation of such traps during the endurance test than are the  $US\_a$  ones. The specific feature of the  $\_TS2a$  sample that is different to the other two samples is the retention of both positive and negative deep trapped charge to  $10^5$  min. The ability of the  $\_TS2a$  sample to retain deep trapped positive charge whereas the  $ES\_2a$  sample starts to lose this charge at  $\sim 10^4$  min could be due to changes introduced in the XLPE during cable stressing. For example taking into account that both the  $ES\_2a$  and the  $\_TS2a$  peelings were from cable segments that allowed the ingress of acrylate species over time [19, 20], we could envisage that in the case of the thermal stressing ( $\_TS2a$  peelings) such species may react with the polymer chain to yield species attached to the chain that bind positive charge [23–25]. Such chemical reactions would be favoured in the TS material where consumption of anti-oxidant is likely to have occurred. Since chemically bound species are not able to migrate the activation energy for the removal of positive charge from these centers would be given by the binding energy ( $> 1.0$  eV). In the electrically stressed cable segment ( $ES\_2a$  peeling), the acrylate species would be likely to be present but without attaching to the chain, so they can still migrate from the material. This picture would indicate that the reduced endurance life of the  $\_TS2a$  sample is caused by a sequence of events:

- a) Ingress of acrylates species from the cable semicon, possibly as field-driven ions;
- b) Thermal consumption of anti-oxidant and chemical binding of species to the chain;
- c) Binding of positive charge to the attached chemical entity during the endurance test;
- d) Binding of negative charge to residual acetophenone during the endurance test;
- e) Reaction between negative molecular ions (perhaps trapped acetophenone) and positive chemical centers bound to the polymer chain.

The apparently vexed question as to whether space charge causes ageing or is the consequence of ageing [7] cannot be answered by these results. All that we have done is show that charge traps are modified, in quantity, in energy range, and in type, during electro-thermal stressing. Only some of these changes seem to lead to a reduction in insulation life. Since the electrical stressing is that of an ac-field we would expect the actual amount of space charge not to be significant in modifying the endurance life, but rather the amount of charge that is retained from cycle to cycle that consequently increases the injection currents [26]. Since the greatest effect on endurance life is experienced by peelings with a thermally stressed history [3], it would seem likely that the synergistic process

leading to failure follows the lines: thermal degradation and anti-oxidant consumption from high temperatures leading to deep positive trap generation that increases the ac injection currents [26], which in turn is responsible for further trap generation and anti-oxidant consumption.

Electrostatic stresses are not likely to be important in the present circumstances. This is not necessarily the case for dc ageing, and in either case once some space charge is accumulated itself will influence any injection currents and further degradation, i.e. the ageing is and must be a synergistic process.

## 6 CONCLUSION

Our results show that either AC electrical or thermal stressing modifies the charge trap distribution in XLPE insulation, with the high electro-thermal stress conditions of the endurance life test introducing positive charge traps that were not present before the test. Different cable stresses leads to differences in the susceptibility of the XLPE to trap generation during the subsequent endurance test. Both forms of cable stress result in a reduced generation of shallow positive charge traps and an increase of deep positive charge traps compared to the effect of the endurance test upon samples taken from unstressed cable. The reduced endurance life of the samples taken from the thermally stressed cable segment has been associated with a reduction of the amount of shallow trapped charge and an increase in the amount of deep trapped charge that is retained to longer times than in other samples. These changes have been tentatively assigned to the consumption of anti-oxidant and subsequent chemical reaction with chemical species in the polymer.

## ACKNOWLEDGMENT

The authors would also like to thank the members of the ARTEMIS consortium for access to the samples from this project. Also thanked are N. Hampton and U. H. Nilsson for simulating discussions.

## REFERENCES

- [1] B. Garros, "Ageing and reliability testing and monitoring of power cables: diagnosis for insulation systems: the ARTEMIS program", IEEE Electrical Insulation Magazine, Vol. 15, No.4, pp. 10-12, 1999.
- [2] J. C. Fothergill, G. C. Montanari, G. C. Stevens, C. Laurent, G. Teyssedre, L. A. Dissado, U. H. Nilsson, and G. Platbrood, "Electrical, microstructural, physical and chemical characterization of HV XLPE cable peelings for an electrical aging diagnostic data base," IEEE Trans. Dielectr. Electr. Insul., Vol. 10, pp. 514-27, 2003.
- [3] A. Tzimas, S.M. Rowland, L. A. Dissado, Fu Mingli, and U. H. Nilsson, "Effect of long-time electrical and thermal stresses upon the endurance capability of cable insulation material", IEEE Trans. Dielectr. Electr. Insul., Vol. 16, pp. 1436-1443, 2009.
- [4] A. Tzimas, M. Fu, and L. A. Dissado, "Characterization of thermally aged XLPE cable peelings through space charge measurements", IEEE Conf. Electr. Insul. Dielectr. Phenomena (IEEE Cat. No. 05CH37702), Nashville, TN, USA, pp. 30-33, 2005.
- [5] A. Tzimas, M. Fu, L. A. Dissado, U. H. Nilsson, and A. Campus, "Space charge characterization of XLPE peelings with a cable prehistory: Before and after AC endurance tests", IEEE Int'l. Conf. Solid Dielectr. (ICSD), Winchester, UK, pp. 474-477, 2007.
- [6] A. Tzimas, M. Fu, L. A. Dissado, and R. N. Hampton, "Comparison of XLPE cable peelings with different electro-thermal histories using endurance test and residual charge methods", IEEE Int'l. Conf. Properties and Applications of Dielectr. Materials, Bali, Indonesia, pp. 293-296, 2006.
- [7] J. P. Crine, "On the interpretation of some electrical aging and relaxation phenomena in solid dielectrics", IEEE Trans. Dielectr. Electr. Insul., Vol. 12, pp. 1089-1107, 2005.
- [8] 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.
- [9] A. Tzimas, S M Rowland, L. A. Dissado, M. Fu, and U. H. Nilsson, "The effect of dc poling duration on space charge relaxation in virgin XLPE cable peelings", J. Phys. D: Appl. Phys., Vol. 43, 215401(pp. 10), 2010.
- [10] L. A. Dissado, J. C. Fothergill, A. See, G. C. Stevens, L. Markey, C. Laurent, G. Teyssedre, U. H. Nilsson, G. Platbrood, and G. C. Montanari, "Characterizing HV XLPE cables by electrical, chemical and microstructural measurements on cable peeling: effects of surface roughness, thermal treatment and peeling location", IEEE Conf. Electr. Insul. Dielectr. Phenomena (Cat. No.00CH37132), Victoria, BC, Canada, pp. 136-40, 2000.
- [11] K. Fukunaga, "Progress and Prospects in PEA Space Charge Measurement Techniques", IEEE Electr. Insul. Mag., Vol. 24, No. 3, (DOI 10.1109/MEI.2008.4591432), pp. 26-37, 2008.
- [12] P. Morin, J. Lewiner, C. Alquie and T. Ditchi, "Study of space charge dynamics in solid dielectrics by simultaneous measurement of external current and space charge distributions", in Space Charge in Solid Dielectrics, The Dielectrics Soc., U.K., ISBN0 9533538 0 X, pp. 45-57, 1998.
- [13] S. Mitsumoto, M. Fu, L.A. Dissado and J.C. Fothergill, "Short interval decay measurement of space charge in epoxy resin", IEEE Trans. Fundamentals Materials (FM), Vol. 126, pp. 260-261, 2006.
- [14] G. Mazzanti, G.C. Montanari and J.M. Alison, "A space charge based method for the estimation of apparent mobility and trap depth as markers for insulation degradation-theoretical basis and experimental validation", IEEE Trans. Dielectr. Electr. Insul., Vol. 10, pp. 187-197, 2003.
- [15] L.A. Dissado, O. Paris, T. Ditchi, C. Alquie and J. Lewiner, "Space charge Injection and extraction in high divergent fields", IEEE Conf. Electr. Insul. Dielectr. Phenomena (CEIDP), pp. 23-26, 1999.
- [16] K. Wu, X. Chen, X. Liu, X. Wang, Y. Cheng and L.A. Dissado, "Study of the space charge behaviour in polyethylene nanocomposites under temperature gradient", Int'l. Sympos. Electr. Insulating Materials (ISEIM), Kyoto, Japan, paper D4, 2011.
- [17] L.A. Dissado and A. Tzimas, "The effect of Ageing on Charge Traps in XLPE Cable Insulation", Int'l. Sympos. Electr. Insulating Materials (ISEIM), Kyoto, Japan, paper C5, 2011.
- [18] M. Meunier, N. Quirke, and A. Aslanides, "Molecular modeling of electron traps in polymer insulators: Chemical defects and impurities", J. Chem. Phys., Vol. 115, pp. 2876-2881, 2001.
- [19] H. Herman, J. Thomas, and G. Stevens, "Spectroscopic and chemometric analysis of cable condition in the Artemis program", IEEE Int'l. Conf. Solid Dielectr., (ICSD), pp. 623-627, 2004.
- [20] H. Herman, G. Stevens, and J.L. Thomas, "Cable condition through a spectroscopic and chemometric analysis of cable ageing", IEEE Int. Sympos. Electr. Insul., Object Identifier, 10.1109/ELINSUL.2010.5549573, pp. 1-5, 2010.
- [21] Y. Sekii and T. Maeno, "Generation and Dissipation of Negative Heterocharges in XLPE and EPR", IEEE Trans. Dielectr. Electr. Insul., Vol. 16, pp. 668-675, 2009.
- [22] Y. Sekii, "Influence of antioxidants and cross-linking on the crystallinity of XLPE dielectrics", IEEE Conf. Electr. Insul. Dielectr. Phenomena (CEIDP) pp. 719-722, 2007.
- [23] G. Tardieu, G. Teyssedre, and C. Laurent, "Role of additives as recombination centres in polyethylene materials as probed by

luminescence techniques”, J. Phys. D: Appl. Phys., Vol. 35, pp. 40-47, 2002.

- [24] N. Hozumi, G. Teyssède, C. Laurent, and K. Fukunaga, “Behaviour of space charge correlated with electroluminescence in cross-linked polyethylene”, J. Phys. D: Appl. Phys., Vol. 37, pp. 1327-1333, 2004.
- [25] C. Laurent, G. Teyssède, J.L. Auge, G. Mazzanti, G.C. Montanari, L.A. Dissado, J.C. Fothergill and A. See “Space charge and associated electroluminescence processes in XLPE cable peelings”, IEEE Conf. Electr. Insul. Dielectr. Phenomena (Cat. No.00CH37132), Vol. 2, pp. 568-572, 2000.
- [26] Y. F. F. Ho, G. Chen, A.E. Davies, S.G. Swingler, S.J. Sutton, R.N. Hampton and S. Hobdell., “Measurement of space charge in XLPE insulation under 50 Hz ac electric stresses using the LIPP method”, IEEE Trans. Dielectr. Electr. Insul., Vol. 9, pp. 362-370, 2002.



**Antonios Tzimas** (M'08) was born in Volos, Greece in 1979. He received the B.Eng. (2001) and M.Sc. (2003) degrees in electrical and electronic engineering from the University of Leicester where after completing his national service duties in Greece he returned to do research on the ageing properties of cross linked polyethylene. This was the topic of his Ph.D. thesis, he received the degree in 2008. At the end of his Ph.D. he joined the National Grid High Voltage laboratory at the University of Manchester as Post-Doctoral researcher.

Currently he is working on the ageing properties of outdoor composite insulators as well as cable insulation. In 2011 he became the chair of IEEE Dielectrics and Electrical Insulation Society GOLD membership and part of the society's membership committee. He is also a board member of the Annual Conference on Electrical Insulation and Dielectric Phenomena (CEIDP).



**Simon M. Rowland** (SM'07) was born in London, England. He completed the B.Sc. degree in physics at The University of East Anglia and the Ph.D. degree at London University. He was awarded the IEE Duddell Premium in 1994 and became a FIEE in 2000. He has worked for many years on dielectrics and their applications and has also been Operations and Technical Director within multinational manufacturing companies. He joined The School of Electrical and Electronic Engineering in The University of Manchester as a Senior Lecturer in 2003, and was appointed Professor of Electrical Materials in 2009. He is currently President of the IEEE Dielectric and Electrical Insulation Society.



**Leonard A. Dissado** (SM'96-F'06) graduated from University College London with a degree in chemistry in 1963 and obtained the Ph.D. degree in theoretical chemistry in 1966. After rotating between Australia and England twice, he settled in Chelsea College in 1977 to carry out research in dielectrics. Since then he has published many papers and one book, together with John Fothergill, on breakdown and associated topics. In 1995 he moved to the University of Leicester and was promoted to professor in 1998. He has been a visiting professor at the University Pierre and Marie Curie in Paris, Paul Sabatier University in Toulouse, and Nagoya University, and a Senior Visiting Fellow at the Institute of Advanced Studies of the University of Bologna. He also has given numerous invited lectures, including the Whitehead Memorial Lecture in 2002. Currently he is an associate editor of the IEEE Transactions on Dielectrics and Electrical Insulation, and the chair of IEEE DEIS publications Committee. He obtained a D.Sc. degree in 1990 from London University. In 2007 he was honored by the Université Paul Sabatier, Toulouse, with the award of a Doctuer Honoris Causa, and in September 2008 with the award of Honorary Professor of Xi'an Jiaotong University, Xi'an, China.