

## Modification of Space Charge Pattern and Charge Mobility of Low Density Polyethylene (LDPE) in the presence of Crosslinking Byproducts.

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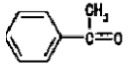
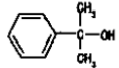
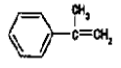
**Abstract:** The formation of space charge in Crosslinked Polyethylene (XLPE) cable has always been related to the byproducts such as acetophenone,  $\alpha$ -methylstyrene and cumyl alcohol. In this paper, the argument is to be justified. Space charge formation in cumyl alcohol soaked Low Density Polyethylene (LDPE) is presented and compared with that of acetophenone and  $\alpha$ -methylstyrene soaked LDPE. Cumyl alcohol causes charge accumulation of both charge polarities with heterocharges appeared when the sample is stressed at low voltage. The total charge accumulated as well as the charge decay in the clean LDPE sample are then compared to that of the chemicals soaked LDPE films. Charge mobility values in all samples were calculated from the decay characteristics and the results reveal that the byproduct chemicals accelerate the decay process in LDPE film, due to their high charge mobility. Since the byproduct chemicals change the space charge decay behavior of LDPE, it is suggested that the chemicals modify the trapping characteristics of LDPE. This is possible either by superseding the deep traps in insulator with shallow traps from the chemicals, or simply adding up the number of shallow traps in the insulator.

**Keywords:** space charge, mobility, LDPE, Dicumyl Peroxide, cumyl alcohol, acetophenone, alpha-methylstyrene, crosslinking byproduct.

### INTRODUCTION

In the crosslinking process of polyethylene (PE) using the crosslinking agent, Dicumyl Peroxide (DCP), three main byproducts are produced which are acetophenone, cumyl alcohol and  $\alpha$ -methylstyrene. Some properties of the byproducts are simplified in Table 1. During recrystallisation process after PE undergoes the crosslinking processes, the byproducts are 'forced' into the new amorphous region and spaces in between the polymer chain in XLPE. It was suspected that the space charge which was found in the XLPE cable is due to the byproducts that reside in the polymer. Space charge formation in the cable has always been related to the deep traps from the byproducts [1]. In the present paper, the real effect of the main crosslinking byproducts on space charge formation and charge mobility are investigated in referring to our previous work [2] that discussed on the effect of acetophenone and  $\alpha$ -methylstyrene on space charge formation.

Table1. Physical properties of the crosslinking byproducts

Byproduct	acetophenone	cumyl alcohol	$\alpha$ -methyl styrene
Chemical structure			
Melting point	19 - 20 °C	32-34 °C	-24°C
Boiling Point	198 - 204 °C	88-90°C	164-168 °C

### EXPERIMENTAL PROCEDURE

The samples with a thickness of 180 $\mu$ m were cut into small circular film with a diameter of 36 mm. In this project, LDPE was chosen over XLPE to eliminate any possibility of having more than one byproduct in the sample. The samples are soaked into  $\alpha$ -methylstyrene and acetophenone at room temperature and into cumyl alcohol at 80°C for 2 hours.

In the space charge measurement, a pulse voltage of 600V with duration of 5 ns is applied to the sample to generate an acoustic signal wave. These samples are stressed at three different positive dc voltages; 5kV, 8kV and 10kV. Readings were taken for every 10 minutes for one hour and after that the charge decay pattern was observed for another one hour. Before the sample was aged at the specified voltage, 2kV dc voltage was applied across the sample for calibration purpose which had been discussed elsewhere [3].

### RESULT AND DISCUSSION

Fig. 1 shows the space charge pattern of cumyl alcohol soaked LDPE. Volts Off measurements are included to eliminate the fast moving charges that exist in the Volts On measurement result. After comparing the space charge patterns in clean LDPE and the soaked samples, all byproducts have different effect on the space charge pattern.

Cumyl alcohol causes charge accumulation of both charge polarities with heterocharges appeared when the sample is stressed at 5kV and 8kV. Both positive and negative charges were introduced into the sample near the cathode and anode respectively. After 10 minutes

charging, the negative charges in samples stressed at 5kV and 8kV, migrated into the bulk, reducing the height of the positive peak. After 20 minutes, the positive peak totally disappears and negative charges are now accumulated in the bulk of sample. Similar observation is seen for the negative peak near the anode. The formation of heterocharge can be seen clearer in the Volts Off measurement result.

At 10kV, heterocharges have not appeared. However, only after 5 minutes charging, negative peak is observed in the sample bulk and the peak height is lowering by charging time. It is not surprising to see that due to the high voltage applied to the sample, the number of homocharges that are injected from the electrodes is big enough to overcome the heterocharges formed due to cumyl alcohol.

Conversely, the space charge result of acetophenone and  $\alpha$ -methylstyrene soaked LDPE show that only homocharges appear in the stressed samples. Acetophenone causes high domination of negative charges in sample bulk. Meanwhile the charge profiles for  $\alpha$ -methylstyrene soaked LDPE and un-soaked clean sample are almost similar except that the positive charges in LDPE increases gradually compared to the easy domination of positive charge in the soaked sample [2].

### Trap control Mobility

The quantities that are extracted from the space charge measurement can be translated into an estimation of the trap-controlled mobility values. As in Fig. 2, the charges in the soaked samples decay very fast. After 20 minutes of decay, almost three quarter of the initial charges had decay. After 20 minutes, the decay

becomes gradually slow. Conversely, after one hour discharging, the clean LDPE still has 75% of the initial charges remaining in the sample. The rate of charge decay is very much dependent on how mobile the charges are in the specific material.

A method to estimate the trap-controlled mobility from the space charge measurement has been proposed in [4]. The simple procedures of this method enable measurement at any charging or poling field and provide the apparent mobility of charges in the insulator which then can be associated with the trap level distribution. In order to analyse the mobility value, the ratio of  $Q(t)$  to the stored charge value at the beginning of depolarisation,  $Q_0$ , is considered. This ratio term will be represented as  $q_0$  and is defined as;

$$q_0 = \left(\frac{1}{L}\right) \int_0^L Q_p(x, t) dx / Q_0 \quad (1)$$

$Q_0$  is obtained immediately after the sample is short circuited and this value is crucial to represent the reference value associated to the applied field and the type of sample. A very simple but highly approximate expression for mobility, which is extensively described in [5], is given below:

$$\mu(t) = 3\epsilon \left| \frac{d}{dt} (-1/q_0(t)) \right| \quad (2)$$

where  $\epsilon$  is the material permittivity. This equation is applied to estimate the value of trap controlled mobility from the evolution of  $-1/q_0$  characteristic as shown in Fig.3. Some assumptions were made in order to impliment this mobility equation such as any charge recombination is neglected, the trapped charge is so much more than the conduction charge and the charges are uniformly distributed in the insulation [4].

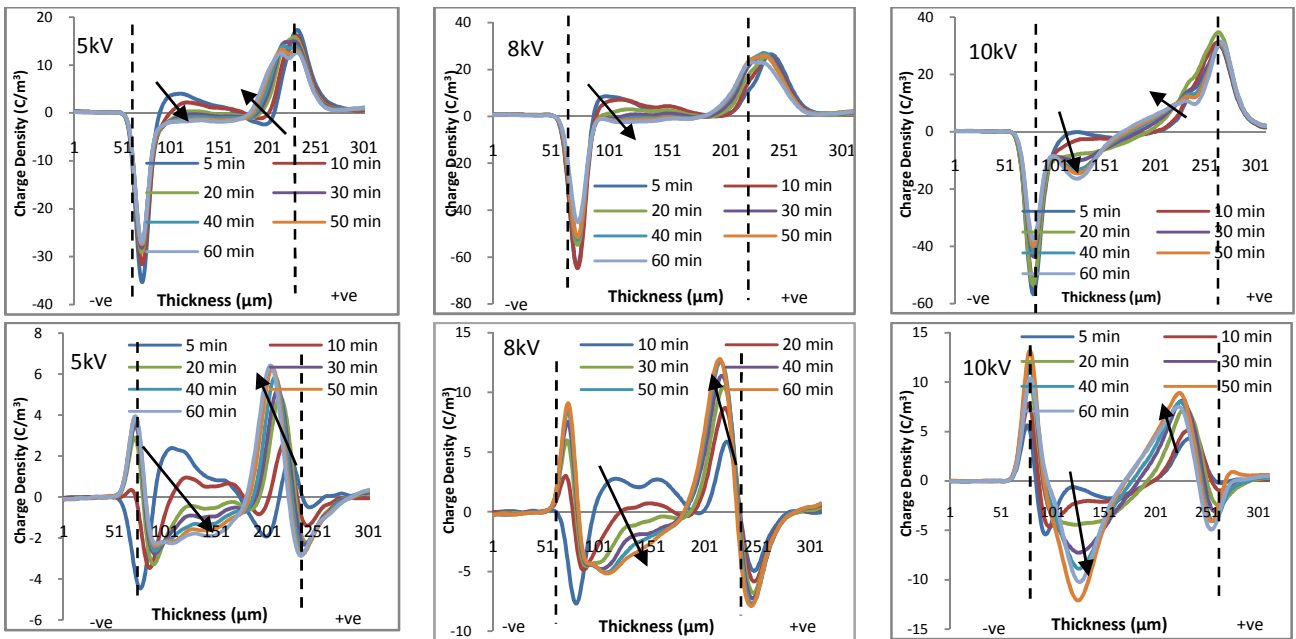


Fig. 1. The space charge density of cumyl alcohol soaked LDPE for Volts On (top) and Volts Off (bottom) measurement.

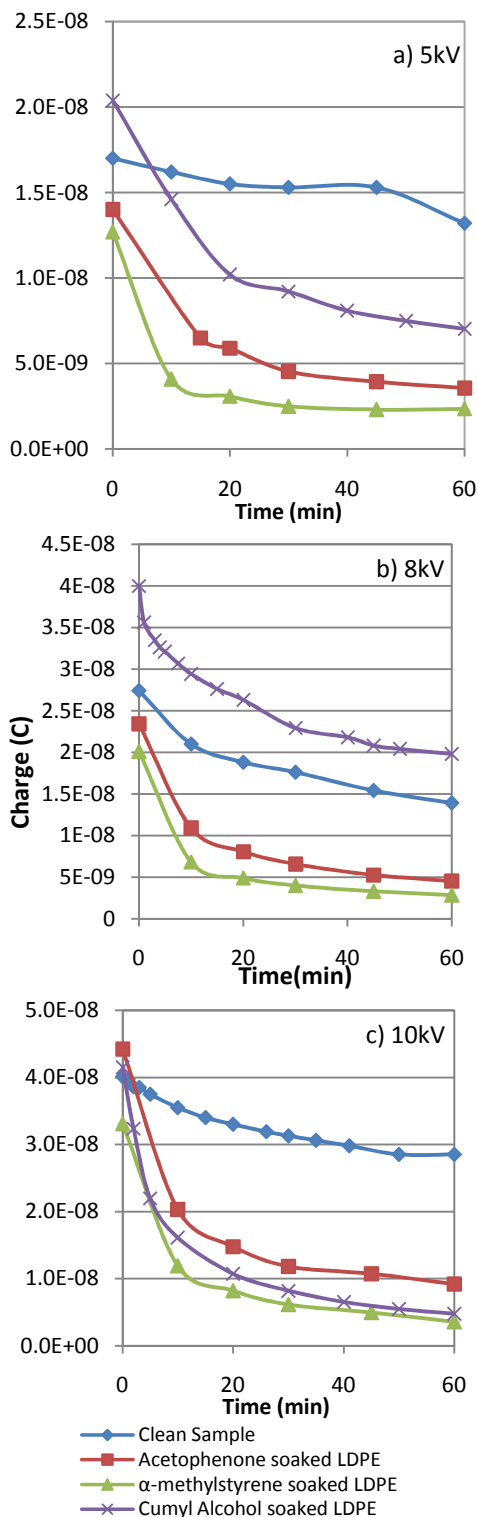


Fig. 2. Total charge decay of soaked and unsoaked samples after stressed for one hour.

As in Fig. 4, the mobility values are time and electric field dependent. The apparent mobility curves shift to lower values as the decay time increases. The mobility values of soaked samples is higher than that of the clean LDPE film.

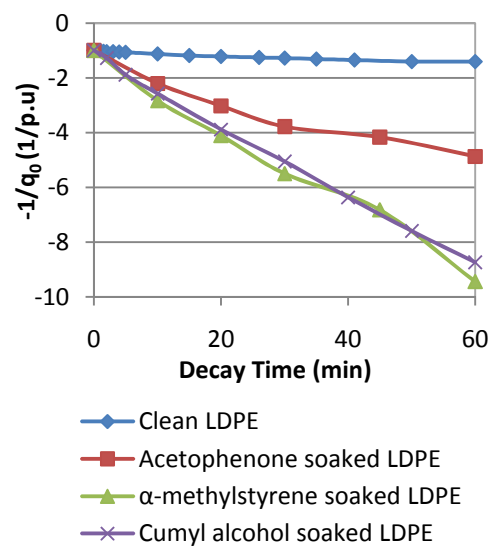


Fig. 3. Time behaviour ( $-1/q_0$ ) characteristics for samples stressed at 10kV.

Among the three byproducts,  $\alpha$ -methylstyrene proved to have the highest apparent mobility value and like clean LDPE film, the mobility value decreases in respect of the applied stressing voltage. On the other hand, mobility value for acetophenone soaked sample increases with the increase of the stressing voltage. Nevertheless, the increment is not very significant.

Last but not least, cumyl alcohol response towards the applied field is inconsistent as the field is increasing. The highest mobility value is observed in the sample that contains  $\alpha$ -methylstyrene. The mobility value decreases dramatically in the sample that was stressed at 5kV. However, at elevated stressing voltage, the curve becomes more linear giving constantly high mobility value during the 60 minutes of decay. In contrast, acetophenone mobility curve becomes more nonlinear as the voltage is increased. No big change is seen for the charge mobility value for cumyl alcohol soaked LDPE.

The mechanism in which that the byproducts mobilise the movement of the charges is still undiscovered. However, it is suspected that some chemical bonds in the byproducts molecules may introduce shallow traps that ease charge transportation especially at the regions that are originally populated by the deep traps.

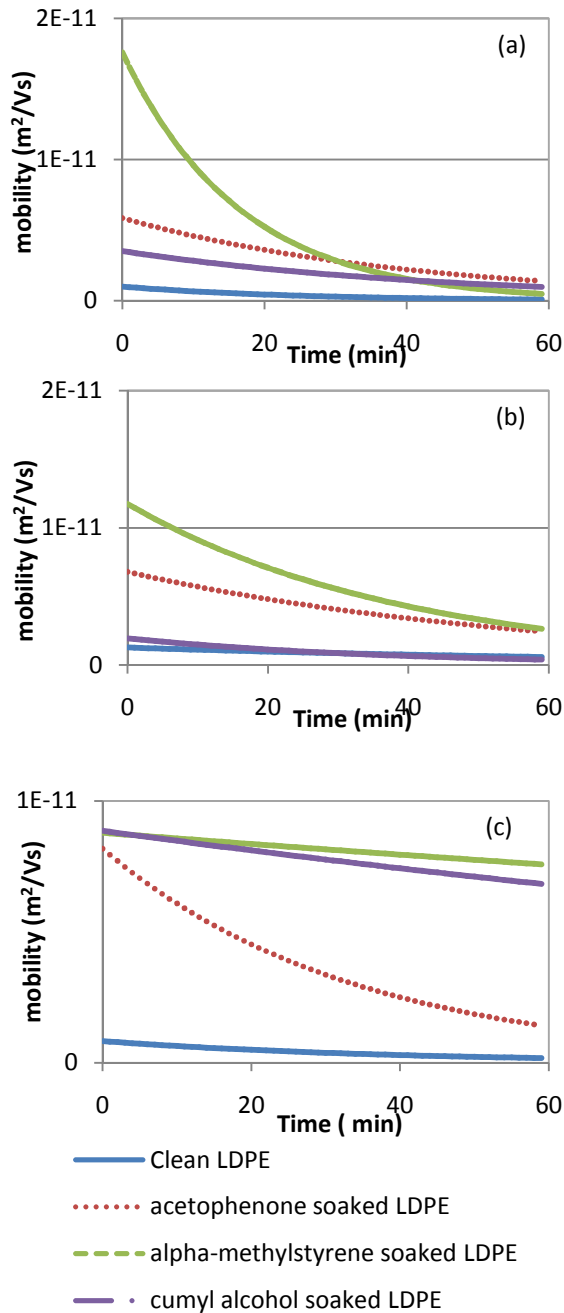


Fig. 4. The mobility of the charges in clean and soaked samples charged at a)5kV, b)8kV and c)10kV.

## CONCLUSION

The effect of crosslinking byproducts on the space charge patterns and charge mobility in soaked LDPE was investigated. Cumyl alcohol causes heterocharge to form in LDPE.

The apparent mobility of charges in the presence of crosslinking byproducts is very high compare to the clean unsoaked sample. The mobility value is time and field dependent. As the mobility value is

being related to the trap depth, the high mobility value is due to the shallow traps that increase with the existence of the crosslinking byproducts. Meanwhile the low mobility value is attributable to the deep traps which present in the polymer structure.

## REFERENCE

1. G. Teyssedre, C. Laurent, A. Aslanides, N. Quirke, L. A. Dissado, G. C. Montanari, A. Campus, L. Martinotto. *Deep trapping centers in crosslinked polyethylene investigated by molecular modeling and luminescence techniques*. Dielectrics and Electrical Insulation, IEEE Transactions on 2001;8(5):744-752.
2. N. Hussin, G. Chen. *The Effect of Acetophenone and alpha-Methylstyrene on the Space Charge Properties of Low Density Polyethylene*. Annual Report Conference on Electrical Insulation and Dielectric Phenomena. CEIDP . , 2008. p 702-705.
3. G. Chen , Y L Chong , M. Fu. *Calibration of Pulse electroacoustic technique in the presence of trapped charge*. Institute of Physicys Meas. Sci. Technol 2006;17:1974-1980
4. G. C. Montanari, G. Mazzanti, F. Palmieri, B. Bertuzzi. *Mobility evaluation from space charge measurements performed by the pulsed electroacoustic technique*. Proceedings of the 6th International Conference on Properties and Applications of Dielectric Materials (Cat. No.00CH36347); 2000; Piscataway, NJ, USA. IEEE. p 38-41. (Proceedings of the 6th International Conference on Properties and Applications of Dielectric Materials (Cat. No.00CH36347)).
5. R. Ciobanu, C. Schreiner, W. Pfeiffer, B. Baraboi. *Space charge evolution in oil-paper insulation for DC cables application*. Proceedings of 2002 IEEE 14th International Conference on Dielectric Liquids. ICDL 2002 (Cat. No.02CH37319); 2002; Piscataway, NJ, USA. IEEE. p 321-4. (Proceedings of 2002 IEEE 14th International Conference on Dielectric Liquids. ICDL 2002 (Cat. No.02CH37319)).