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## Conductivity and Space Charges in PE with Additives

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### Abstract

Temperature dependent conductivity and space charge formation was measured in metallocene catalyst polymerized polyethylene samples of planar geometry with semiconductive electrodes. The temperature dependence of the conductivity was determined based on leakage - current measurements at variable temperatures.

Space charge formation under an applied electrical field of 20kV/mm was investigated by means of the pulse-electro-acoustic method (PEA) at room temperature.

The results were compared to space charge formation and conductivity in common LDPE. The measurements showed considerable differences between materials and only minor influence of crosslinking process and the addition of antioxidant with respect to the electrical properties of the material. Possible correlations between conductivity and space charge formation are discussed in the paper.

The relevance of the findings for application to extruded high voltage cables will be discussed.

### Introduction

Space charge formation in dielectrics might lead to field enhancements resulting in reduced electrical strength of high voltage insulation, as in case of polymer insulation for HVDC-applications. The formation of space charges is, on a macroscopic scale, related to the distribution of permittivity  $\epsilon$  and conductivity  $\sigma$  of the dielectric, given by  $\epsilon/\sigma = f(x) \neq const.$ , where  $x$  is the position in the dielectric in field direction. Under the assumption of an electric current density  $> 0$ , the expression means that an inhomogeneous distribution of the quotient will lead to space charge formation in the dielectric. [1]

This approach, however, does not cover charge carrier injection and other electrode effects affecting the electrical field.

Controlling space charge formation and conductivity by additives has been described earlier [2], but no evidence has yet been given that bulk conductivity control, aiming at a homogeneous material, is sufficient for space charge reduction. In a similar way, controlled change of permittivity distribution can be thought of as leading to a better field distribution, but cannot be considered isolated from conductivity and electrode considerations.

Looking at polymer materials for high voltage insulation under real conditions, additives, necessary for process control and stabilization, are known to have influence on charge distribution and the resulting electrical field in the insulation [3,4]. The question is if an appropriate choice of polymer and additives might lead to proper field control and if the conductivity approach is sufficient.

In the present paper, the effect of crosslinking and addition of antioxidant on space charge distribution and conductivity in a solution polymerized PE will be investigated and compared to common LDPE. Detailed information about the results described in this paper can be found in [5]

### Test Samples and Materials

Conductivity and space charge measurements were performed on flat, circular test samples with two thin electrodes of a conducting polymer bonded to the test material. The electrodes were of an ethylene-ethylacrylate compound with 32w% of carbon black. The thickness of the dielectric layer between the electrodes was 1.6mm. In some cases, the production techniques caused slightly thinner samples, which was compensated for by applied voltage corrections, always giving 20kV/mm as the resulting electric field.

Two base polymers were chosen as test materials:

- Bulk polymerized LDPE without crosslinking and

additives with a density of 920 kg/m<sup>3</sup> was used as reference material for the measurements.

- Solution polymerized polyethylene using a metallocene catalyst commercially available as Exact 0201 with a density of 902 kg/m<sup>3</sup> was investigated pure, crosslinked and crosslinked with antioxidant. In the following, it will be referred to as 0201. Crosslinking of 0201 was done by means of 1.6w% dicumylperoxide. 0.2w% Santonox R was added as antioxidant to some samples of 0201.

## Tests Performed

### Conductivity

Conductivity was measured at varying temperatures and an electrical field at 20kV/mm, since this value can be considered as being close to the maximum field being applied in real high voltage transmission cables. For the same reason a temperature range of 40 to 80 °C was chosen.

Conductivity was measured by applying the temperature profile shown in table 1.

200	+20	+20	+20	+20	+20	+20	+20	+20	hrs
80	70	60	50	40	50	60	70	80	°C

Table 1: Temperature profile applied to most samples

In the case of crosslinked 0201, some temperature levels were omitted in the measurement procedure due to deviations in the temperature control system.

### Space Charges

The space charge distribution was measured by means of the PEA method applied on flat samples at room temperature and at 20kV/mm DC electric field. The system is based on a 2kV square pulse with a pulse width of 20ns. The acoustic wave is detected with a 27µm PTFE-film.

## Results

The results shown here are a selection from several tests showing comparable characteristics.

The scale in x-direction is given in nanoseconds. In order to show the position in the dielectric, two gray markers indicate the electrode peaks with the cathode at the left and the anode at the right. The position is not marked precisely, due to the limited spatial resolution of the measurement system and electrode adjacent space charges affecting the electrode peak. The arrows indicate the direction of the temporal development.

Fig. 1 shows charging of an LDPE-sample, a process which has stabilized after about 96hours at 20kV/mm. Under stable conditions, both electrode areas are characterized by heterocharges, the decay of which is

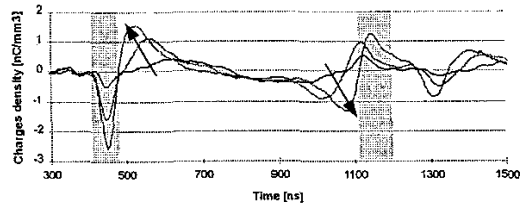


Fig.1: SC-distribution in LDPE, measured without voltage applied after 2, 8, & 96hrs charging at 20kV/mm

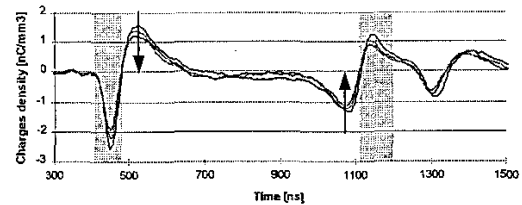


Fig.2: Discharging of LDPE, measured after 0, 32 & 64hours with electrodes shortened.

hardly visible, even after 64 hours (fig.2).

A very different charge distribution can be seen for 0201 in fig.3. Some initial heterocharges at the cathode move towards the anode, finally leaving a minor amount of homocharges, which decay until 96 hours under short circuiting is shown in fig. 4.

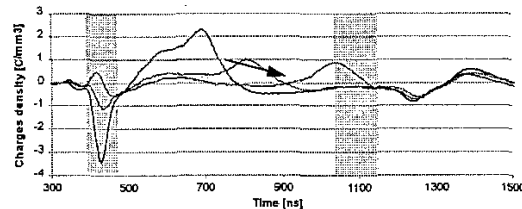


Fig.3: SC-distribution in 0201, measured without voltage applied after 2, 8, & 96hrs charging at 20kV/mm.

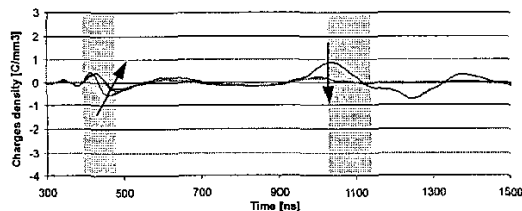


Fig. 4: Discharging of the 0201, measured after 0, 24 & 64h with electrodes shortened.

Quite a similar distribution and dynamic behavior as in pure 0201 could be observed in crosslinked 0201, see fig.5. With respect to discharging under short circuiting, differences were clearly visible between cathode homo-charges decaying pretty fast and more stable negative hetero charges at the anode (fig.6).

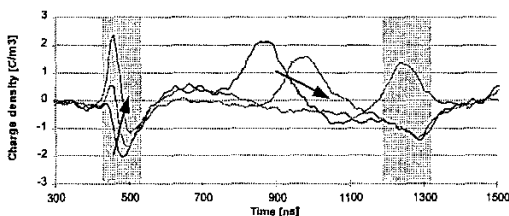


Fig.5: SC-distribution in crosslinked 0201, measured without voltage applied after 2, 4, & 24hrs charging at 20kV/mm.

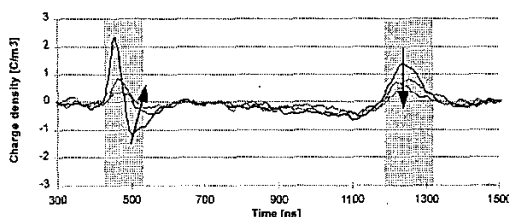


Fig.6: Discharging of crosslinked 0201, measured after 0, 4 & 24h with electrodes shortened.

A more complex situation occurred in case of crosslinked 0201 with antioxidant (fig.7), where only a slight movement of the positive hetero charges can be seen, together with a negative charge build-up at both cathode and anode.

The charge decay (fig.8) was comparable to the one of pure and crosslinked 0201.

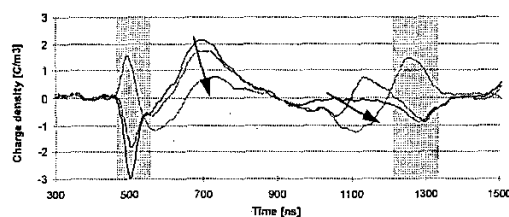


Fig.7: SC-distribution in crosslinked 0201 with antioxidant, measured without voltage applied after 2, 4, & 24hrs charging at 20kV/mm

The temperature dependent conductivity for the materials investigated is shown in fig. 9

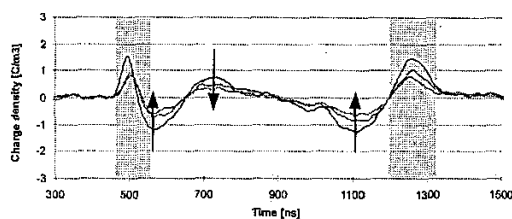


Fig.8: Discharging of crosslinked 0201 with antioxidant, measured after 0, 4 & 24hours with electrodes shortened.

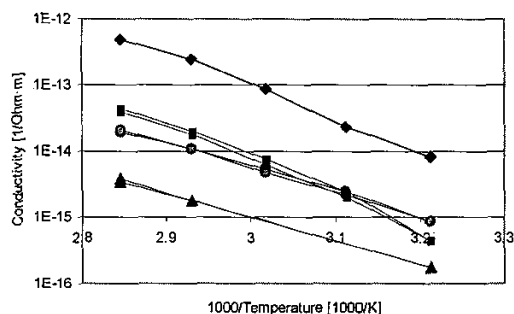


Fig.9: Conductivity in LDPE (♦), 0201 pure (●), 0201 crosslinked (▲) and 0201 crosslinked + antioxidant (■) as a function of temperature.

The double line visible for some material indicates the up and down regulation of the temperature, as specified in table 1. Only very little deviation of the final conductivity from the start value can be seen, even after 360 hours with voltage and temperature (see table 1 for details).

Assuming the conductivity  $\sigma$  given by the simple approach  $\sigma = \sigma_0 \exp\left(-\frac{E_a}{kT}\right)$ , where  $T$  is the temperature,  $k$  is Boltzmanns constant and  $\sigma_0$  is a constant, and applying this expression on the results shown in fig.9, gives the activation energy  $E_a$ :

LDPE	1.1 eV
Exact 0201	0.7 eV
Exact 0201, crosslinked	0.7 eV
Exact 0201, crosslinked with Santonox R.	1 eV

Table 2: Activation energy  $E_a$  [eV] for materials used in the investigations (average values).

It can be seen that the metallocene catalyst polymerized 0201 in its pure condition has a conductivity well below that of LDPE. Crosslinking decreases the conductivity by a factor 5-6. The addition of antioxidant has the effect of increasing conductivity to the level of pure 0201, the activation energy, however, now has increased, as indicated by the slope of the respective curve in fig.9 and calculated in table 2.

## Discussion

In order to discuss the materials' suitability for HVDC applications, the results have to be considered under the aspect of space charge related field enhancements and time constants for charging and discharging. The possible influence of additives has to be known for specific space charge/conductivity control.

The LDPE-samples showed a clear and stable field enhancing build-up of heterocharges at the electrodes.

The results from 0201 show positive bulk charge formations, which, under the influence of the electric field, were able to move through the dielectric and decrease in amplitude. Approaching stable conditions, negative charges were visible at both electrodes, the amount of which had a steadily decreasing tendency.

While crosslinking decreases the conductivity and the antioxidant increases conductivity to the level of pure 0201, no considerable difference between the final charge distributions in 0201 can be observed. The negative electrode charges decrease the field at the cathode and increase the field at the anode and a closer investigation of charge injection/extraction in comparison to the bulk conductivity would be appropriate. Formation of hetero charges in case of the LDPE gives a higher field at both electrodes with a possible higher charge injection, in good correlation with the higher conductivity observed.

With respect to the time constants for charging and discharging, it was surprising that the material with the highest conductivity, LDPE, showed the slowest discharging i.e. the largest time constant as compared to 0201. On the other hand, the additives did not seem to have considerable effect on the charge and discharge dynamics in the dielectric.

Earlier investigations have confirmed the influence of polymer additives and/or crosslinking by-products on space charge formation [2], the influence of which, however, can be minimized by proper conditioning of the sample, as for example heat, vacuum, electric field or a combination of these methods.

This leads to the idea that - under the assumption of inhomogeneous additive distributions in the samples being excluded as main cause for the final space charge formations - electrode effects remain the dominating cause for electrode adjacent space charge formation. In the future, most attention should be paid to this aspect with respect to development of polymer DC-insulation with an optimal field distribution.

A factor contributing to the materials' different electrical behavior might be the lower crystallinity of 0201 (~37%) compared to the crystallinity of LDPE (42%) and the fixed chain length in 0201 as compared to the wider distribution in LDPE. Also conditioning of the material, with respect to reduction of the amount of

by-products, should be taken into account, since this is known to have influence on space charge formation [3].

Optimization of insulation system for HVDC also needs considerations and investigations on the breakdown strength of the material composition. This work is ongoing and results will be published later.

## Conclusion

Conductivity and space charge measurements were performed in metallocene catalyst polymerized polyethylene Exact 0201 and common LDPE.

0201 showed a lower conductivity than LDPE, especially after crosslinking, whereas addition of antioxidant increased the conductivity. While the space charge distribution in LDPE was characterized by stable heterocharges at both electrodes, 0201 showed considerable, but mobile bulk charge formations, decreasing with time.

No direct relationship was found between conductivity and the measured space charge distributions. The results indicate that the simple macroscopic approach on the relationship between conductivity and space charge formation is not sufficient.

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