Bernhard Fruth, Dietmar Meurer

ELECTRICAL PROPERTIES OF HIGHLY STRESSED POLYOLEFINS

1. Introduction

Polyolefins as polyethylene and polypropylene are cheap alternatives to conventional oil-paper dielectrics. However the evaluation of the lifetime of an insulator (e.g. cable) is highly complicated because of a lack of powerful testing procedures.

The electrical strength measured under special laboratory conditions lies in the range of 1MV/mm. But, the operating field strength if e.g. cables is about 10 kV/mm [1] and goes up to 140 kV/mm in impulse capacitors with a fluorcarbon impregnant. This discrepancy is a result of the complex ageing processes in polymers. The maximum operating voltage is limited by the inception of partial discharges at conducting particles or structural inhomogeneities of conductivity such as boundaries between amorphous and crystalline phases or between structural zones resulting from temperature and flow speed gradients in the melt during the manufacturing process [2,3].

Partial discharge inception is closely related to stable and oscillating excess carriers. There is strong evidence that high mobility charge carriers (hot electrons) induce the first breakup of the molecular bond [4].

Conventional dielectrical measurements (dissipation factor, absorption currents, capacitance) on samples stressed by high electrical fields give insight in the conduction and space charge mechanisms and its dependencies on field, temperature, polymer specific phase transitions and the non-electrical properties (molecular weight, crystallinity, structure et.

Dr.Bernhard Fruth,Dr.Dietmar Meurer-Institut für Allgemeine Elektrotechnik und Hochspannungstechnik der RWTH Aachen

They should be added by thermally stimulated polarization (FITSP) and depolarization (TSD) current measurements giving information about contact influence and charge stability [5].

Because of sensitivity problems of conventional current measuring devices, these experiments are usually carried out in homogeneous field arrangements with large measuring areas (sheets or foils contacted by guard electrodes). The maximum testing voltage is limited by partial discharges (pd) in the embedding medium of the contacting electrodes and not by prebreakdown in the polymer under test.

However it is desirable to apply the whole dielectric test spectrum on small volumes up to pd-inception, monitoring the complete dielectric ageing process under thermal and electrical stress. From our experiences with PP and PE this means testing up to at least 800 kV/mm [6].

For this purpose the needle-plane arrangement, originally designed for treeing experiments, is very attractive. Reproducible samples can easily be prepared under normal laboratory conditions.

The first chapter of this contribution gives some examples of the homogeneous field properties of PP. The second part deals with the attempt to apply the needle-plane arrangement to ultra high sensitivity pd-detection and TSD measurements and their correlation to space charge mechanisms.

2. Homogeneous field

a) Experimental

The samples used are PP-sheets of 1,25 mm thickness manufactured either by extrusion or injection moulding with varying process parameters from 87,5%isotactic untreated granulate. The microstructure formed during the cooling process in the tool is revealed microphotography under polarized light (Fig.1). Especially in injection moulded samples we find surface layers with very small and highly distorted spherolites due to cooling gradients up to 1000 K/min.

The samples were contacted by high vacuum evaporated chromium guard electrodes inhibiting surface effects (area 5,3 cm^2). Alternating field properties are measured by a Schering's bridge with automatic guard potential control. Sensitivity is increased using a synchronized lock-in amplifier as zero detector. The HV setup is insulated and thermostated with silicon-oil.

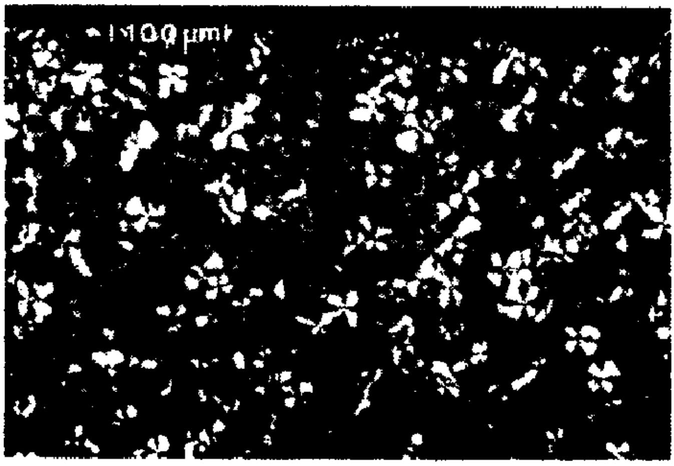

Fig.1. Microstructure of Polypropylene under polarized light

DC-properties and TSD are measured in a coaxial cell. The voltage limits are 40 kV with dry air insulation and 200 kV with siliconliquid.Temperature control and data acquisition is fully computerized with a current sensitivity of 1 fA and a maximum temperature error of 0,05 K.

b) Results

Semicrystalline PP possesses two main phase transitions, glass transition and crystallite melting. The first examples (Fig 2,3,4,5) show the higher sensitivity of dielectric measurements in comparison to the differential scanning calorimetry (DSC). At Glass transition T_G the dielectric loss exhibits a distinct maximum, whereas capacitance, due to thermal expansion and heat flow have a slight change in temperature coefficient.

DSC and TSD deliver formal identical results in detecting the melting process (Fig. 4,5).

It is clearly shown that space charge relaxation is closely related to the melting process.

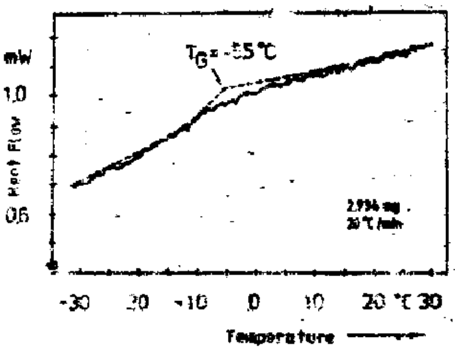

Fig.2. DSC Measurement at glass transition T_g

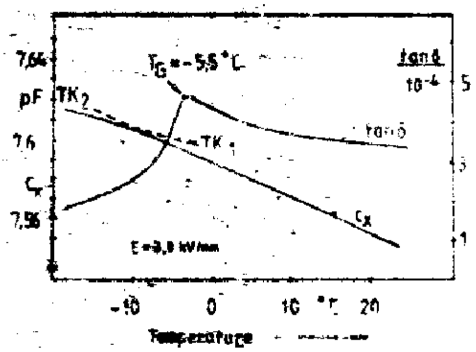

Fig.3. Loss tan δ and capacitance at T_G.

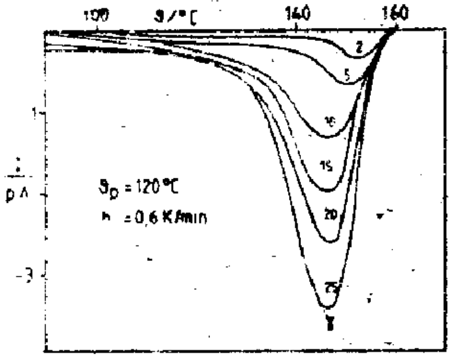

Fig.4. TSD in the melting region

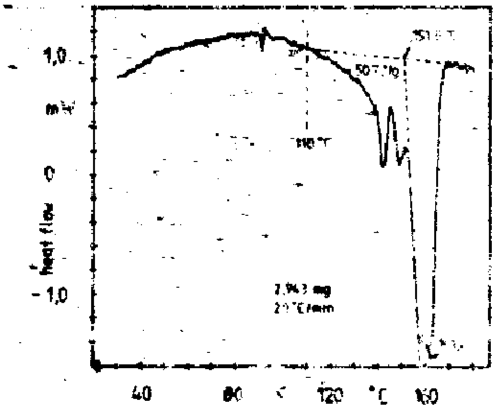

Fig.5. DSC in the melting region

High field conduction mechanisms in PP above glass temperature and below detectable melting processes (110°C) is dominated by injected negative charge carriers [5]. The transport mechanism can be described according to the model of dispersive hopping [5,7]. Experiments in the minute range show an E^n law for the AC and DC-conductivity as a result of the exponential dependency of hopping mobility and contact emission current [5].

At longer times a space charge blocks the injection current and the result is a severe time dependence of conductivity and capacitance. [illegible] shows the typical behaviour of high field dissipation factor vs time. The maximum is produced by counteracting mechanisms: Injection fills the colume with charge carriers on one hand, on the other hand a low carrier mobility produces an electrode blocking space charge.

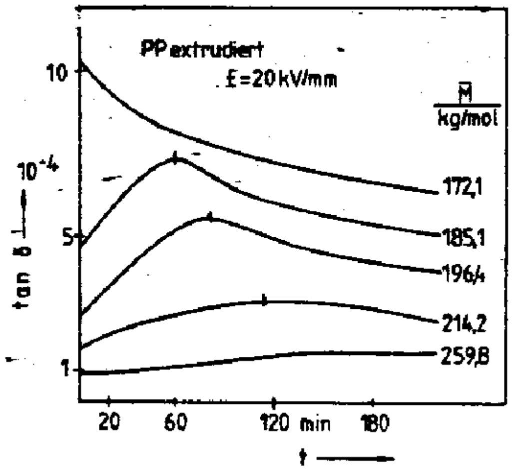

Fig.6. Time dependence of the dielectric loss of PP of different molecular weight .

We see that a decrease in molecular weight is controlled by mechanical damage e.g. by the rotational speed of the extruder leads to an increase of hopping centers and therefore to an increase of mobility.

For a sensitive detection of further dielectrical relaxation mechanisms involved in a specific temperature interval conventional methods have to be completed by TSD- measurements with low neating for optimal resolution. In the case of PP it is possible to detect five different discharging mechanisms ($\rho_1 - \rho_3$, γ and α_c) leading to five TSD-Current peaks reacting specifically on polarization conditions and microstructure of the polymer [5]. The peak ρ_2 characterizes especially the high field carrier transport which affects partial discharge development.

3. Inhomogeneous field

Partial discharges, TSD and FITSP were measured in a needle plane arrangement in LDPE and PP. Needle electrodes of a diameter of 1 mm and tip radii of 5 μm and 20 μ respectively were either melted or pressed into the specimen. The gap distance is 2 mm. The ground electrode does not need to be metallized. The measured sample capacitance os 0,2 pF.

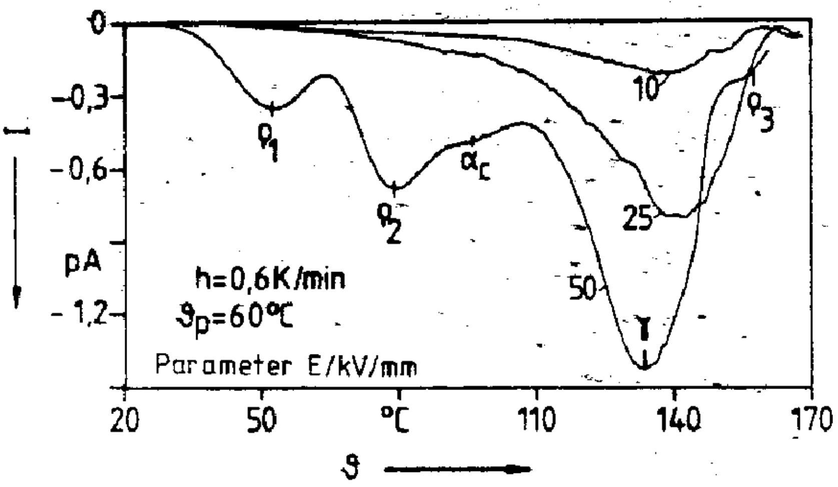

Fig.7. TSD-spectrum of PP (parameter poling field in kV/mm)

Pd are measured at a sensitivity greater than 20 fC in a fully automated setup where a microcomputer controls voltage rise and temperature and stores pd pulse intensity vs. the actual phase angle of the sinusoidal high voltage. (Further details of this measuring system have been published in [8]).

The test setup for measuring TSD currents is also fully automatic.Heat conduction and peak resolution considerations lead to optimal heating rates of 0,5 to 1 K/min. The temperature controller has been designed for minimum linearity error of 0,02 K over the whole range (0°C - 200°C). The current is measured by an electrometer (Keithley 617), the noise amplitude is 2 fA. The test cell provides minimum temperature gradients in the sample; it is designed for DC voltages up to 10 kV (Further details in [9].

a) TSD measurements

The stable part of space charge is detected by the TSD experiment on short circuited samples which have been AC-prestressed without detectable discharges (Fig.8).

In this case two dominant relaxation maxima appear, one in the crystallite melting range and the other around 60°C which is correlated to injected charge carriers. DC prestress results in a formal identical TSD spectrum but with higher current peaks. We therefore can suppose identical discharging mechanisms for both cases.

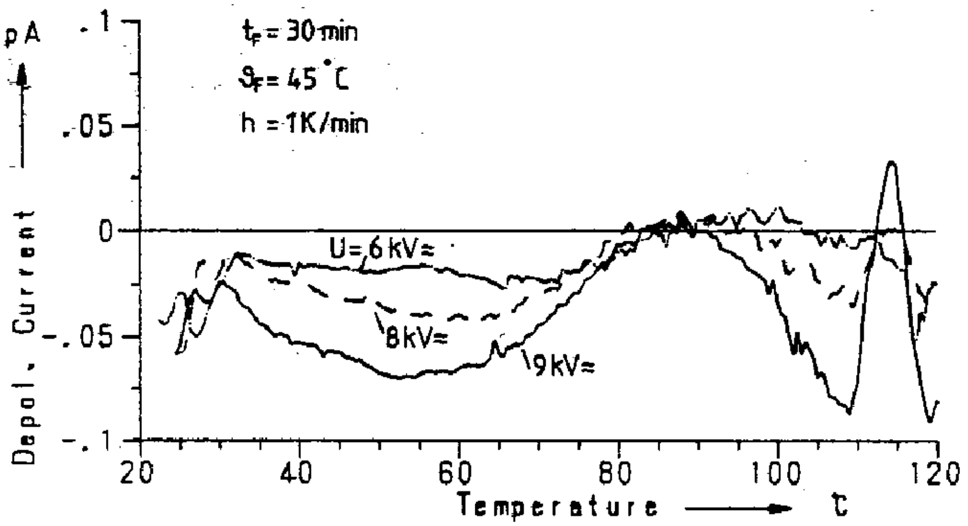

Fig.8. Depolarisation current after AC-stress

b) partial discharge measurements

Fig.9. shows a typical distribution of the first 500 discharge impulses with regard to the actual phase angle of the applied voltage.

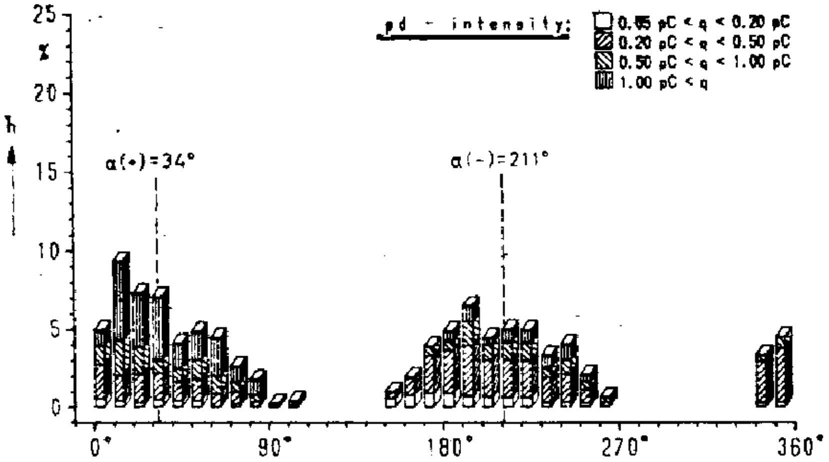

Fig.9. Histogramm of the pd intensity vs. phase angle T = 343 K, tip radius r = 5 µm, dU/dt = 1,25 kV/mm

The average phase angles $\alpha(+)$ and $\alpha(-)$ of the discharge distributions in each half cycle do not coincide with the maximum voltage applied but are shifted to lower values. Assuming that the maximum of the discharge activity happens at maximum of the local field stress, this shift can only be explained by a space charge, distorting the geometric field. Regarding the temperature dependence of the mean phase angle of polypropylene and polyethylene (Fig.10) we see a minimum at that temperature where the space charge due to injected charge carriers relaxes (ρ_2-mechanism for PP, 60°C - peak for PE). Beyond this temperature pd tends to happen in the voltage maximum, coinciding with the ohmic conduction of PE and PP in the melting region. [5].

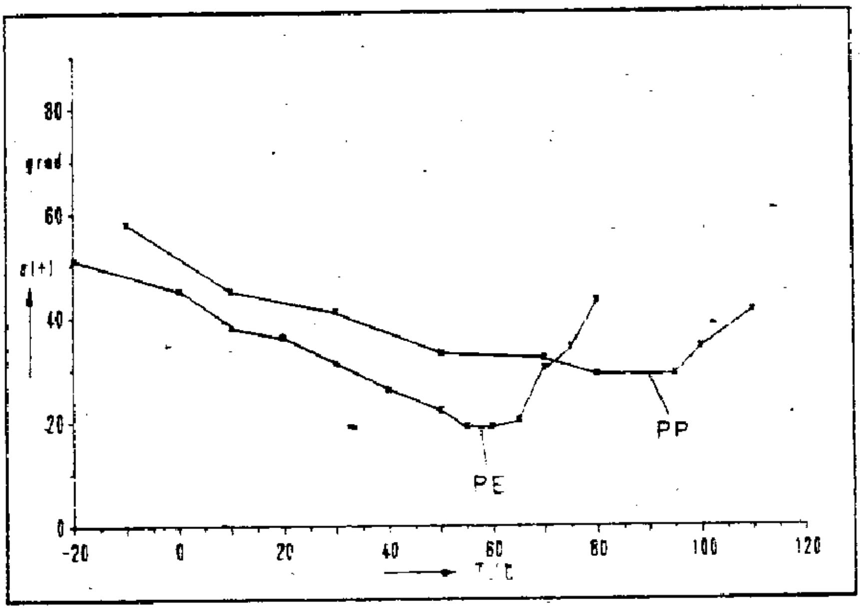

Fig.10. Average phase angle $\alpha(+)$ of the discharge distributions vs. phase angle

It is shown that sensitive pd measurements resolving intensity and phase angle can be attributed to dielectrical properties found in nondestructive independent experiments, which themselves are more closely related to nonelectrical polymer properties. Using the same preparation for the whole experimental spectrum the achieved results will be fully comparable.

As involved time constants of the charge transport and current densities cannot easily be derived from the experiments presented here, two additional setups are under construction:

- direct measurement of the frequency dependence of the injection current in the needle plane arrangement,
- frequency dependent pd measurements.

Acknowledgement

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