

Influence of degree of crystallinity on DC-conduction of polypropylene

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Abstract

Aim of this work was to study how degree of crystallinity influences the DC conduction characteristics of capacitor grade polypropylene (PP). PP pellets were pressed to 0.3 mm plates using two different cooling rates in order to vary the degree of crystallinity. The DC conduction currents were measured at 50 and 70 °C at 8-55 kV/mm. 3 h polarization and 15 h depolarization times were used. It turned out that temperature and the dielectric stress played a more dominant role regarding DC-conductivity than the obtained ~4 wt-% difference in crystallinity. Especially at 39-55 kV/mm, when temperature increased from 50 to 70 °C the DC-conductivity increased 10-15 times. There was however a trend related to the degree of crystallinity: the increase in crystallinity (more slowly sample cooling) led to lower conductivity values.

1. Introduction

The dielectric behavior of a polymer is dependent not only on its chemical composition, chain configuration and additives compounded in it but also how it is processed [1-3]. As a result of orientation of the polymer chains, the degree of crystallinity and density increase leading to improvement in many mechanical and physical properties; modulus, barrier and thermal properties to mention a few [4-6]. Also electrical properties of PP are improved, as was noticed early [7, 8] and all commercial capacitor grade polypropylene films are biaxially oriented. In power capacitors, in which high breakdown strengths, low losses and good ageing characteristics are required, biaxially oriented polypropylene (BO-PP) dielectric is the dominant choice [9-14]. The maximum operating temperature (< 100-105 °C, depending on the wished life time) is unfortunately a limitation with polypropylene dielectrics [9-11, 13, 15]. One way to handle this is to increase the degree of crystallinity and especially the thickness of the crystalline lamellae, by which the start of the melting process can be pushed towards higher temperatures [5].

In order to improve the capability of polypropylene chain to crystallize, the regularity of the chain structure is crucial. How the regularity is influenced in the polymer production step [16], understanding the complex crystal structure of polypropylenes [14, 17] and how the final morphology is created during the film processing [6, 9] are parts in a long chain of efforts to obtain high performance films. The additional step is the understanding of the correlation between different aspects of film, its morphology and electrical performance; also this work has been ongoing for a long time in various contexts [1-3, 5, 7-15]

In the present work the effect of degree of crystallinity on DC-conductivity was studied for two capacitor grade polypropylenes, one normal and one sc. high isotacticity grade. Plates pressed from pellets were studied. Different degrees of crystallinities were obtained by changing the cooling rate during the plate preparation. For the plates prepared from same pellet grade, there was a trend that samples with higher degree of crystallinity (slow cooling rate) showed slightly lower DC-conductivities than the fast cooled samples. The slowly cooled samples showed however breakdowns at lower fields. The DC-conductivities were not constant but field-dependent, especially at 70 °C and > 25 kV/mm (the exact threshold value was not assessed due to limited number measurement fields).

2. Experimental

2.1 Samples

Two grades of commercial, capacitor grade polypropylenes were studied. The samples had as similar additives, additive concentrations, melt flow rates and polydispersities as possible, whereas the isotacticities were different. The isotacticities for the studied grades were following: ~96% for the "Normal" and ~99% for the "High isotactic" grade. This was in accordance to the basic idea; that only the isotacticities of the studied pellets would differ, not other factors that can influence the electrical performance.

2.2 Sample preparation

Materials were received as pellets and pressed to 0.3 mm thick plates using a Servitac Laboratory Press Polystat 300s. Press plates with especially flat surface and a mask plate with a 15 cm hole in the middle were used. Kapton®-films, which were carefully cleaned with ethanol prior to the sample pressing, were used as protective films. For Sample A, a cleaned aluminum foil was used as protective film instead of Kapton®-film.

All samples were pressed twice. Prior to pressing, the pellets were stored under vacuum at 50 °C at least 24 h or longer. During the first pressing cycle the pellets were melted slowly and kept at 200 °C for 30 min under low pressure before cooling rapidly (~ 30 °C /min) to RT. During the cooling, pressure was increased to 200 bars. The obtained plates were cut to smaller pieces which were re-melted according to same procedure as is described above. The plates that were identified as "Fast" cooled were then rapidly cooled to RT under high pressure, in the same way as described above. The plates that were cooled to RT "Slowly" were first cooled from 200 to 125 °C by applying cooling rate of 0.6 °C /min. During this phase the pressure was

stepwise increased to 50 bars. Then the pressure was increased to 200 bars but the cooling rate was kept slow. From 100 °C the samples were cooled more rapidly, by ~ 4 °C/min, down to RT while the pressure was kept high, at 200 bars. Aim of the different cooling procedures was to obtain flat sample plates of good quality, with at least 3-4 wt-% difference in degree of crystallinity. The details regarding the pressure procedures were experiment-based, upper limits for the applied pressure and cooling rate being the max-capacity of the press. The ready plates, examples of which are shown in Figure 1, were carefully wrapped in a capacitor grade aluminum-foil. The samples were then placed in polyethylene bags and stored in dark at RT.

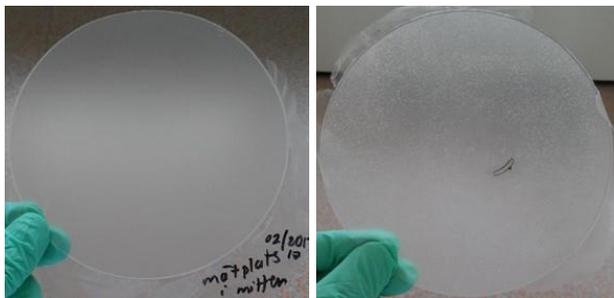


Figure 1. Examples of sample plates. To left, a fast cooled sample with “Normal” isotacticity. To right, same grade but cooled slowly. Dark mark in the sample is a breakdown.

The slowly cooled samples had small cavities, which disappeared if fast cooling rate was applied instead. Longer drying of pellets and reduced pressing temperature did partly, but not completely remove the cavities. It turned out that the amount and size of the cavities depended on the cooling rate. As the slow cooling rates led to higher degrees of crystallinities and therefore higher density, main reason for the observed cavities was believed to be local shortage of polymer

The sample thickness was assessed after the electrical measurements, under the location of the measurement electrode, by using a Mitutoyo micrometer.

3. Methods

3.1 Degree of crystallinity

Degrees of crystallinities were assessed by using Mettler-Toledo DSC1 differential scanning calorimeter with a scan rate of 10 °C/min. Heats of fusions were analysed and degree of crystallinity was calculated by using 207 J/g as ΔH° , heat of fusion for ideal, 100% crystalline polypropylene. Three samples per each plate were analysed and average of these was given as the sample crystallinity. The $6.5 \text{ mg} \pm 0.5 \text{ mg}$ samples were taken from the locations where the electrical measurement had been performed.

3.2 Optical Microscopy

Even if the assessment of spherulitic structure or other more detailed morphological analysis was outside the scope of this work, two samples were analysed by optical microscopy. Thin slices out of two plates were cut out at RT with a very sharp knife. One of the plates was “Normal” isotacticity, the other of “High” isotacticity. The “Normal” plates were cooled fast whereas the “High” isotacticity plate was cooled in stepwise

manner and very slowly (this sample was excluded from the electrical measurements due to cavities).

3.3 DC conductivity

Current-voltage (I/U) measurements in the time domain were performed to determine the DC conductivity at different electric fields. The measurement cell was placed in a climate chamber (CTS C-20/350) in which the temperature and humidity could be regulated. Both the measurement electrode (diameter of 30 mm) as well as the guard were made of brass. The HV electrode was made of stainless steel. Samples were measured as such. In other work regarding assessment of dielectric loss, both sputtering and silver paint were tested in order to improve contacting. As it turned out that both the coatings influenced the PP-sample behavior, it was therefore decided that no additional contacting material should be used.



Figure 2. A typical sample on the high voltage electrode and current measurement- and guard electrode mounted on the sample. The area of the measurement electrode is 7.1 cm^2 .

The voltage steps (U_a) were applied across the sample using a Heinziger PNC5 20000 voltage generator (max voltage 20 kV). An active HV filter was used between the HV generator and the sample to improve the stability and decrease ripple of the voltage source. On the low-voltage side of the sample the guarded electrode was connected to a Keithley 6512 Electrometer to measure the current. The guard electrode ensures a homogenous electric field and an exact effective measuring area not influenced by leakage currents. The current was measured continuously every second. Before analyzing data, signal to noise ratio was improved by averaging the data so that each measurement point was average of 50 previous and 50 next data points. An example of averaged current values, for measurements performed at 9 kV/mm and 70 °C, is shown in Figure 3 below.

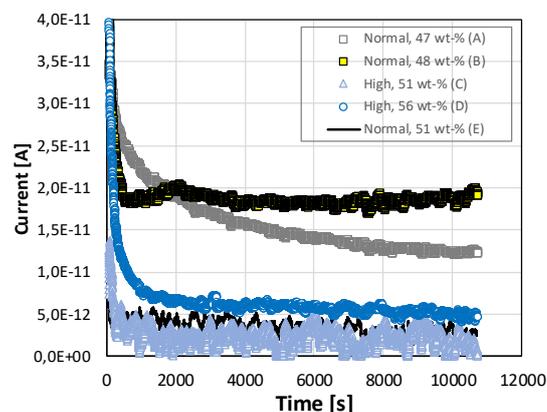


Figure 3. Averaged currents at 70 °C and 9 kV/mm for the studied samples.

The influence of different electrode positions on the sample plates was tested and some scatter was observed. Measurements on different plates prepared from the same pellets and using the same preparation procedure also showed some scatter. The detailed reason for this is not known. However, the process of pressing the PP-plates was not easy, and small, local variations in the obtained plates could be the reason for the observed scatter.

During the application of a constant voltage, we measured the lapse of the total current (i_{pol}), as a function of time (t), including both DC conduction and polarization. By applying the voltage for a very long time the material will, ideally, become fully polarized at the specific voltage application and the measured current will, ideally, be constant. In reality, the measured current contains contribution from both conduction and displacements from polarization processes. The contribution from displacements may be reduced for the estimation of DC conductivity by using both the polarization and depolarization currents. The theory for estimation of DC conductivity by use of both polarization and depolarization currents is described in more detail in [18,19].

The estimation of DC conductivity was calculated based on the polarization (i_{pol}) and depolarization (i_{depol}) currents,

$$\sigma = \frac{d}{A} \left(\frac{i_{pol} + i_{depol} - 2 \cdot i_{offs}}{U_a} \right) \approx \frac{d}{A} \left(\frac{i_{DC}}{U_a} \right) \quad (1)$$

where A is area of the measurement electrode, d is thickness of the sample, U_a is applied voltage, i_{offs} is offset current measured before voltage-on and i_{DC} is DC current. The offset current was so low that it did not influence the results and was neglected. Polarization time was 3 h. Some long measurements with 72 h charging times were also made, but no significant difference to 3h-values were found. For practical purpose the 3-hour values were therefore concluded to be good enough. DC-conductivity was based on an average of current values (i_{pol} and i_{depol}) during the last 600 s from each measurement.

Between each voltage step, the voltage was turned off and the HV electrode was grounded. The samples were allowed to relax under this condition for 15 h (including measured depolarization for 3 h). This was done to ensure that the sample has achieved a more virgin like condition before a new measurement is started. When the oven temperature was changed, additional 4 h conditioning time was added before a new measurement was started.

The polarization and depolarization currents were measured at 2-3 temperatures (T) 30, 50 and 70 °C and 2-5 different voltages (U) between 3 and 18 kV resulting in electric fields (E) of 8-55 kV/mm. Some different humidity levels were tested at 50 and 70 °C. As no significant difference was found at 50 and 70 °C, most of the measurements were performed at 5% RH, which was as dry conditions as possible in the climate chamber.

4. Results and Discussion

DC conductivity was studied for PP plates prepared from pellets with two different isotacticities and applying two cooling rates, see Table 1. The resulting crystallinity is also shown in Table 1.

Table 1. Sample description.

Sample	Pellets	Isotacticity	Second cooling	Crystallinity (wt-%)
A	P1	Normal	Fast	47
B	P1	Normal	Fast	48
C	P2	High	Fast	51
D	P2	High	Slow	56
E	P1	Normal	Slow	51

When the samples with different isotacticities but same cooling rate were compared, the sample with higher isotacticity showed a higher degree of crystallinity, as expected. The fast-cooled Samples A and B from the same pellets showed practically the same crystallinity, 47-48 wt-%. The third sample which was cooled fast, Sample C, had a high isotacticity and showed a higher degree of crystallinity, 51 wt-%. Sample D which was a slowly cooled high isotacticity sample, had an average degree of crystallinity of 56 wt-%, whereas the slow cooling of Sample E with normal isotacticity grade, gave 51 wt-%, Table 1.

The resulting DC conductivities for Samples A, B, C, D and E are seen in Figure 4 to Figure 7 with semi-logarithmic scales. Due to breakdowns, Samples E and D show fewer measurement points in the higher electric field region than Samples A, B, and C. It can be seen that temperature and the dielectric stress, played a more dominant role for the conductivity than the obtained differences in crystallinity. The strong temperature and stress dependence are especially clearly seen in Figure 4.

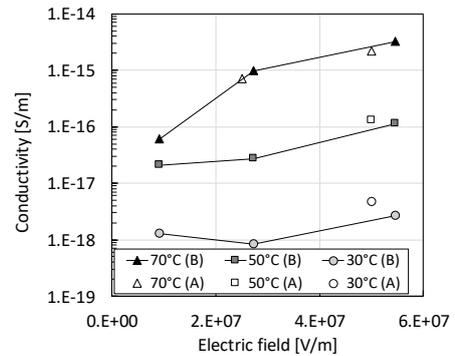


Figure 4. DC conductivity vs. electric field at 30 to 70 °C for Samples A and B: *Normal isotactic*, Fast cooling, 48 wt-%.

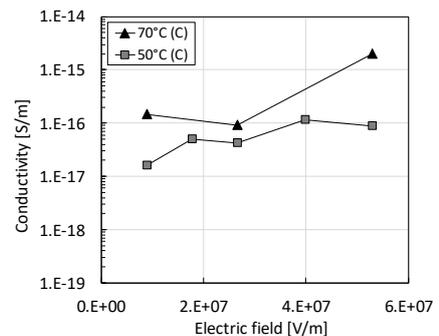


Figure 5. DC conductivity vs. electric field at 50 and 70 °C for Sample C: *High isotactic*, Fast cooling, 51 wt-%. At 50 °C, a different measurement position on the sample plate was used for 18 and 40 kV/mm compared to for 9, 26 and 53 kV/mm.

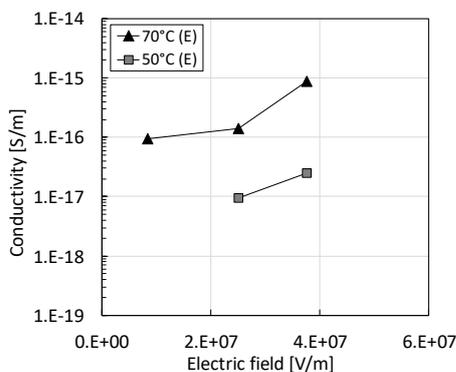


Figure 6. DC conductivity vs. electric field at 50 and 70 °C for Sample E: *Normal isotactic*, Slow cooling, 51 wt-%.

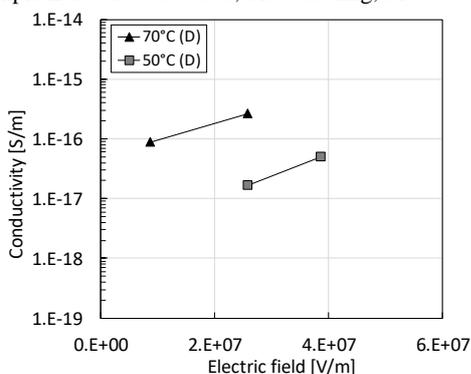


Figure 7. DC conductivity vs. electric field at 50 and 70 °C for Sample D: *High isotactic*, Slow cooling, 56 wt-%.

At 30 °C the DC-conductivity values for Sample A and B were very low, around $1 \cdot 10^{-18}$ S/m. Due to very low currents, conductivity was not assessed for the other samples at 30 °C.

At 50 °C the DC-conductivity values for Samples A, B, C, D and E were higher, although still low, between $1 \cdot 10^{-17}$ and $1 \cdot 10^{-16}$ S/m, Figure 4 to Figure 7. Conductivity increased by roughly $+2 \cdot 10^{17}$ S/m per every 10 kV increase in voltage. No major differences between the conductivity values for the different samples could be identified. The obtained conductivity-values can be compared with a conductivity level of about $2 \cdot 10^{-16}$ S/m, obtained at 50 °C between 30 and 60 kV/mm for a commercially available 7 μ m thick capacitor-grade BOPP film [12]. Above 80 kV/mm a conductivity increase for this film was seen, conductivity being about $1 \cdot 10^{-14}$ S/m at 400 kV/mm [12].

When the temperature increased from 50 °C to 70 °C the DC-conductivities at stresses of 39-55 kV/mm increased ~ 10 -15 times for Samples A, B and C, Figure 4 - Figure 5. Also at 25 kV/mm the increase was almost at the same magnitude, except for sample C. When the electric field at 70 °C was increased from 9 kV/mm to ~ 26 kV/mm, the conductivity values for the 48-wt% crystallinity (sample B) increased from $6 \cdot 10^{-17}$ to $1 \cdot 10^{-15}$ S/m indicating a threshold behavior in conductivity, Figure 4. Also the other samples showed at 70 °C similar type of behavior, Figure 8, but at higher fields.

Classically, the conduction for many polymers at low electric fields is described to be Ohmic and dominated by thermally activated hopping of charge carriers between “traps” [11, 12]. When energy gained from the electric field becomes

comparable with the thermal energy (for many polymers typically >10 kV/mm), it is expected to contribute to detrapping process [11]. This results in a significant increase in conductivity with the electric field [11].

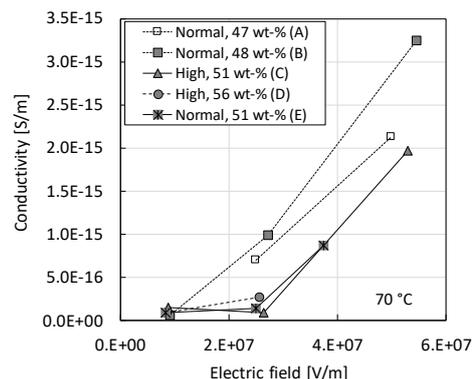


Figure 8. DC conductivity vs. electric field 70 °C for Samples A, B, C, D and E. Please note linear x and y-axis.

In work of Ho and Jow [12] measured transition from Ohmic to nonlinear conduction for the BOPP film took place at fields in the range of 100 kV/mm. In the present study the conductivity values were field-dependent above ~ 25 kV/mm, Figure 8, or earlier depending on the sample type and temperature (~ 10 kV/mm or below for sample B), Figure 4 to Figure 8. It should be clarified that the exact threshold values were not assessed due to limited number of measurement fields.

Ho and Jow [12] use readily prepared BOPP films. In present work the polymers were of capacitor grades as well, but the samples were uniaxially pressed and cooled in the press, not extruded and then biaxially oriented. Morphology in the studied plates consisted of spherulitic structures, Figure 9, not the fibrillar structures typically observed biaxial stretched PP-films [7, 9].

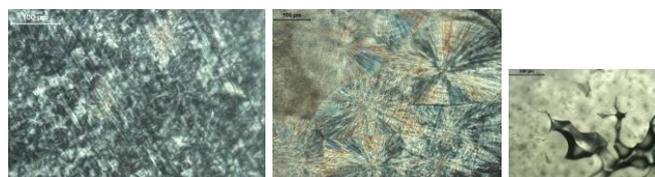


Figure 9. LOM pictures of spherulitic structures in Fast cooled *Normal* sample (left), very slowly cooled *High Isotactic* sample with crystallinity of 58 wt-% (middle) and cavities inside the latter (right). Straight lines on the sample surfaces originate from cutting of the sample. Scale bars from left to right are 100 μ m, 100 μ m and 500 μ m.

It was noticed that the slowly cooled samples, Table 1, suffered of early breakdowns during the electrical measurements. For these samples no data could be obtained >40 kV/mm, Figure 6 and Figure 7, whereas the fast cooled samples were measured up to fields of 55 kV/mm, Figure 4 and Figure 5. For these samples the limitation was merely the max output of the used measurement set-up (20 kV), not the sample itself.

For the electrical strength of the polyolefine samples that are not stretched, the spherulite borders and size are important, and the breakdown strengths decrease with increasing spherulite size [1-2, 9-10, 20]. Spherulite-like structures in the fast cooled “Normal” isotacticity sample are much smaller than for the very

slowly cooled “High” isotacticity sample, Figure 9, which could be the reason for the breakdowns. On the other hand, the cavities observed inside of the slowly cooled samples (more clearly in the High than in the Normal isotactic ones) could have been the dominant reason for the early breakdowns.

What comes to the degree of crystallinity; despite of the modest influence of changes in crystallinity on the conductivity, there was a trend related to the degree of crystallinity: When Samples A, B and E prepared out of *same, normal isotacticity pellets*, see Figure 4 and Figure 6, *but with different cooling rates* were compared, the higher level of crystallinity in Sample E (more slow cooling) tended to keep the conductivity on a lower level at most of the studied stresses and temperatures.

When the samples with *same degree of crystallinity (51 wt-%)*, *but different isotacticities* were compared (Samples C and E, Figure 5 and Figure 6, respectively), it was clear that the high isotacticity Sample C did not show lower conductivity.

In the recent work of Moyassari et al. [21] a realistic semi-crystalline structure of polyethylene (PE) was created with MD (molecular dynamics)-simulations. DFT (Density Functional Theory) was applied to the modelled amorphous, crystalline and semicrystalline structures of PE in order to obtain information about electronic structure. Their results indicate that interfacial regions at crystal/amorphous interface and especially increased free volume are interesting for conduction processes [21]. Updhyay and Reddy [22] combine experimental and theoretical work and apply them to a semicrystalline structure of PE. They conclude as well that boundary between the crystalline and amorphous regions plays a “crucial role” in determining the overall conductivity for a semicrystalline polymer [22]. These findings indicate the importance of interfaces for conductivity of polyolefines.

In the present study the slow cooling rate widened the melting range of the PP-samples slightly, both towards thinner lamellae melting at lower and thicker lamellae melting at higher temperatures. At the same time the total degree of crystallinity increased clearly. There was simply more time for the PP chains to undergo crystallization process and organize themselves better. The obtained differences influenced both the DC-conductivity and breakdown performance, in opposite directions. (Possible degradation was neglected due to well stabilized systems and for relatively short times in warm press).

For sample B, the conductivity was studied for three temperatures, see Figure 4. The activation energy of conduction was calculated by use of the Arrhenius relation for this sample, see Table 2. The activation energies were between 0.9 and 1.6 eV. This can be compared to an activation energy of about 0.8 eV determined in the electric field region 57 to 290 kV/mm between 30 and 100 °C for the 7 µm thick capacitor-grade BOPP films [12]. Generally, the conductivity of an unfilled solid dielectric often follows an Arrhenius relationship with an activation energy in the range of 0.5-1.5 eV [11].

Table 2. Activation energy of DC conduction for sample B at different electric fields based on measurements at 30, 50 and 70 °C.

Applied voltage (kV)	Electric field (10 ⁶ V/m)	Goodness of model fit R2	Data points No.	Eaσ (eV)
3	9	0.95	3	0.9
9	27	1.00	3	1.6
18	55	1.00	3	1.6

5. Conclusions

For the studied plates out of capacitor grade PP, the temperature and also the dielectric stress played a more dominant role for the DC-conductivity (based on 3h-data) than the obtained differences in crystallinity. At 50 °C the increase in the dielectric stress did lead to roughly +2·10¹⁷ S/m increase in conductivity per every 10 kV increase in voltage. At 70 °C the conductivity changes caused by increasing dielectric stress could be more drastic, indicating a threshold behavior in conductivity. When the temperature increased from 50 to 70 °C the DC-conductivity increased 10-15 times at stresses of 39-55 kV/mm.

Despite of the modest influence of changes in crystallinity on the conductivity, there was however a trend related to the degree of crystallinity. For the sample of same isotacticity the increase in crystallinity (more slow cooling) tended to keep the conductivity on lower level at most of the studied stresses and temperatures.

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