

Characterization of Isotactic-polypropylene-based Compounds for HVDC Cable Insulation

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Abstract— This paper aims at a preliminary characterization of isotactic-polypropylene-based blends as a candidate to be used for High-Voltage Direct Current HVDC cable insulation in the framework of the European project NEWGEN (New GENERation of HVDC insulating materials, cables and systems). Isotactic-polypropylene (i-PP) is blended with various propylene-based elastomers (PBEs) and heterophasic PP random copolymers (HECO) with different percentages. Results show that, compared to XLPE, i-PP blends have a comparable breakdown strength, but higher DC conductivity. Noticeable space charge accumulation is observed in the i-PP blends. TSDC measurements show that i-PP blends are characterized by two great peaks (that attribute shallow and deep traps) compared to only one deep-trap peak in the XLPE.

Keywords—HVDC transmission, cable insulation, polymer, thermoplastic.

I. INTRODUCTION

High-Voltage Direct Current HVDC cable projects have been progressively increasing during recent decades as a result of the integration of renewables in the electrical grid as well as the transmission of electricity over long distances. Cross-linked Polyethylene (XLPE) has been the most widely used polymer in extruded insulation for power cables due to its good electrical and mechanical properties. However, XLPE, as a thermosetting insulation, has many limitations e.g., the complex and long production processes (including crosslinking and degassing), the formation of crosslinking byproducts as well as the recycling difficulties [1]. The limitations of XLPE push towards developing innovative, reliable, and environmentally friendly insulating materials that are also able to withstand higher operating temperatures and electric fields [2]. This paper presents the initial characterization of isotactic-polypropylene-based (i-PP) blends to be used as HVDC cable insulation. Other blends based on polypropylene random copolymers are subjected to the same initial characterization in Niittymäki et. al [3]. Four measurements have been chosen to characterize the dielectric properties of the specimens during the initial screening phase i.e., space charge measurement on flat specimens (using Pulsed Electro-Acoustic (PEA) method), Breakdown Strength (BD), (transient) DC Conductivity, and Thermally Stimulated Depolarization Current (TSDC).

II. COMPOUNDING AND MATERIAL CHARACTERIZATION

Polypropylene homopolymer has excellent electrical and thermal properties, however, its mechanical properties have limited its application in high voltage cables [4]. For this reason, isotactic polypropylene (i-PP) is often blended with elastomeric components to enhance its mechanical properties. Table I presents the composition of each i-PP based blend considered in this study. The used i-PP is a medium-flow homopolymer with a melt flow rate of 3.0 g/10 min (230 °C/2.16 kg). Two different types of propylene-based elastomers, namely PBE1 and PBE2 (both being essentially propylene-ethylene co-polymers), are used to modify the i-PP. The ethylene contents of PBE1 and PBE2 are 9 wt-% and 16 wt-%, respectively. In addition, a heterophasic PP random copolymer (HECO), comprising essentially of a PP homopolymer matrix with a dispersed ethylene-propylene rubber (EPR) phase, is also studied as a PP modifier. An antioxidant (AO) comprising of a synergetic 1:1 blend of a hindered phenolic antioxidant (Irganox® 1010) and a phosphite processing antioxidant (Irgafos® 168) was added to the blends to prevent thermo-oxidative degradation. The materials were melt-blended by using a mini-scale twin-screw extruder (temperature 230 °C, screw speed 100 rpm, 4 min mixing time) after which the molten compound was mini-injection moulded into 25×25×3 mm³ specimens. Finally, the mini-injection moulded specimens were hot-pressed to obtain flat specimens. For PEA, the thickness is approximately 0.25 mm, while it is approximately 0.1 mm for the other measurements. For the sake of comparison, state-of-the-art XLPE flat samples (fully degassed) undergo the same characterization process, as a reference material.

TABLE I. i-PP BLEND COMPOSITIONS.

Code	Polymer component weight ratio				iPP blend	
	i-PP	PBE1	HECO	PBE2	Polymer [wt-%]	AO [wt-%]
i-PP/PBE1	0.5	0.5	-	-	99.7	0.3
i-PP/HECO	0.5	-	0.5	-	99.7	0.3
i-PP/PBE2	0.5	-	-	0.5	99.7	0.3
i-PP/HECO/PBE2	0.5	-	0.25	0.25	99.7	0.3

Differential scanning calorimetry tests were made on the insulation compound samples using a TA Instruments MDSC2920 (temperature range -50...+230 °C, dynamic heating rate of 10 °C/min, constant nitrogen gas purge of 33 ml/min, samples encapsulated in aluminum pans). Tensile and

flexural properties of the PP blend were tested by Instron 4505 universal testing machine in accordance with ISO 527 and ISO 178 standards, respectively. Cross-sectional sample morphologies were imaged using JEOL JSM 6360-LV scanning electron microscope (SEM). The polymer samples were fractured in liquid nitrogen and sputtered with Au before imaging.

III. DIELECTRIC CHARACTERIZATION OF SPECIMENS

The initial dielectric characterization of the i-PP blends is carried out using four measurements, i.e., space charge, breakdown strength, DC conductivity, and TSDC. The initial characterization is planned to be carried out (as applicable) at a high temperature $T=90^\circ\text{C}$, and an average electric field of 30 kV/mm representing the target of the future HVDC cables. It aims at a fast screening of possible variations of i-PP based blends before proceeding with more comprehensive characterization of the selected blends in the future.

A. Space Charge (using PEA Method)

Space charge measurement aims at finding the charge carriers behavior, dynamics, and stability inside the i-PP blend flat specimens. The PEA method measures space charge density distribution inside the thickness of the specimen over time. Three hours of polarization (volt-on) and one hour of depolarization (volt-off) periods are chosen as a compromise between time restriction of each measurement and the time enough to obtain a quasi-stable space charge pattern. Specimens with 0.25 mm thickness are tested in a climatic chamber at 70°C as the maximum design temperature of the PEA cell. Then, the raw acquired data is post-processed to obtain the deconvoluted pattern. The net charges and the maximum Field Enhancement Factor (FEF) are calculated as follows to compare the PEA results of i-PP based blends [5]:

$$E_{x_i} = \frac{1}{\epsilon_0 \epsilon_r} \int_{x=0}^{x_i} \rho_x dx \quad (1)$$

$$E_{max} = \max(E_{x_i}(x)) \quad (2)$$

$$FEF = \frac{E_{max} - E_{mean}}{E_{mean}} * 100 \% \quad (3)$$

where E_{x_i} is the calculated local electric field at the point x_i inside the specimen (m), and ρ_x is the charge density at the point x inside the specimen (C/m^3), E_{max} is the maximum electric field in the thickness of the specimen (kV/mm), FEF is the percent maximum field enhancement (%).

B. Breakdown Strength

Breakdown strength is an intrinsic property of the insulating material that can be used to determine its applicability in HV power cable insulation. The flat sample is placed between two electrodes (diameter of 25 mm, edge rounding of 3 mm) immersed in mineral oil (Shell Diala S4 ZX-1) and designed according to IEC60243 [6]. The testing cell is kept at the room temperature ($T \approx 20^\circ\text{C}$) during the test. The applied ramp voltage has two slopes; during the first slope, the ramp rate is 1 kV/s until approximately 70% of the expected breakdown voltage, while during the second slope the ramp rate becomes 0.25 kV/s until the breakdown. Eight samples of each i-PP based blend are tested. After the breakdown, the thickness of a sample is measured near the breakdown hole. Then, the results are subjected to statistical processing and fit to two-parameter Weibull distribution.

C. DC Conductivity Measurement

The DC conductivity is a key characteristic to determine the suitability of the insulating materials in HVDC cable systems. Specimens with a thickness of 0.15 mm are subjected to 24-hour polarization at each of the following electric fields: 20, 30, 40, 50 and 60 kV/mm. This protocol was repeated twice at 30 and 50°C . The leakage current is measured using a femto-/pico-ammeter during the polarization. Then the DC conductivity is calculated and plotted for the considered specimens. In a subsequent stage of the characterization process, more of the selected PP blends will be subjected to the long DC conductivity measurements.

D. Thermally Stimulated Depolarization Current TSDC

For thermally stimulated depolarization current (TSDC) measurements, circular gold electrodes with diameter of 22 mm and thickness of 100 nm were deposited on both sides of samples using a custom-built e-beam evaporator (Instrumentti Mattila, pressure $<1 \times 10^{-6}$ mbar, deposition rate of 0.18 nm/s) inside an ISO 6 class clean room facility. The evaporated samples were short-circuited and stored in a vacuum oven at $+50^\circ\text{C}$ for at least 24 hours prior to electrical measurements. TSDC is used to identify the charge relaxation processes as a function of temperature. For TSDC measurements, a DC source (Keithley 2290E-5) and an electrometer (Keithley 6517B) were utilized. Shielded sample cell equipped with a PT100 temperature sensor was used for all the measurements. Temperature control (accuracy of $\pm 0.1^\circ\text{C}$) was realized using the Novocool system. The TSDC measurement procedure was: (i) polarization at DC field $E_p=15$ kV/mm for 40 min under isothermal conditions at $T_p=90^\circ\text{C}$, (ii) rapid cooling to $T_0=-50^\circ\text{C}$; hold isothermally for 5 min, (iii) removal of the poling voltage and short-circuiting of the sample through the electrometer; hold isothermally for 3 min, (iv) linear heating at $\beta=3.0^\circ\text{C}/\text{min}$ up to $T_{max}=140^\circ\text{C}$ while recording the depolarization current. Electrical polarization/conduction data from TSDC experiments (during poling period at 90°C) were also used in the analysis.

IV. RESULTS AND DISCUSSION

A. Morphology and mechanical properties

Table II summarizes the tensile and flexural properties of the studied PP blends including tensile modulus (E_{tens}), stress at break (σ_{break}), elongation at break (ϵ_{break}) and flexural modulus (E_{flex}). Overall, the tensile properties of the i-PP blends are in line with the minimum requirements presented in Cigré TB 852 [7], i.e. minimum tensile stress and elongation at break of 8.5 MPa and 350%, respectively. Moreover, it is seen that the i-PP/PBE1, i-PP/PBE2 and i-

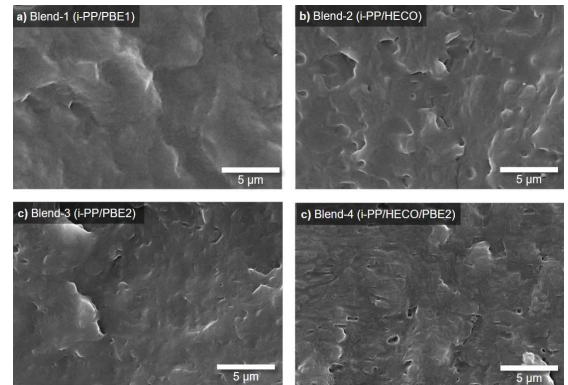


Fig. 1 SEM cross-sectional morphology of a) i-PP/PBE1, b) i-PP/HECO, c) i-PP/PBE2 and d) i-PP/HECO/PBE2.

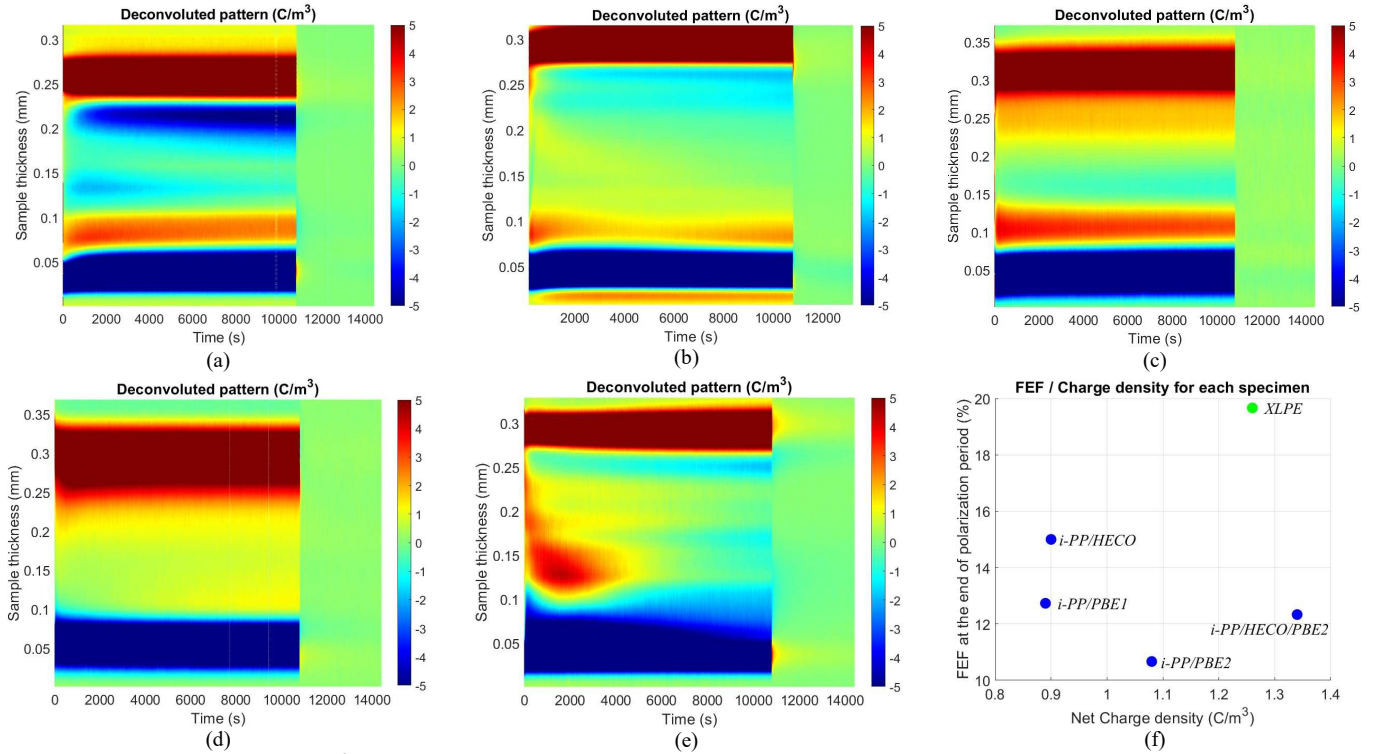


Fig. 2 Space charge (color bar in C/m^3) evolution inside the insulation thickness (y-axis) over time (x-axis) (Figs 1. a – e), and FEF/net charge density (Fig. 1 f), at electric field $E=30kV/mm$, and a temperature $T=70^\circ C$, for: a) i-PP/PBE1, b) i-PP/HECO, c) i-PP/PBE2, d) i-PP/HECO/PBE2, e) Reference XLPE.

PP/HECO/PBE2 systems exhibit significantly reduced flexural moduli (in the range of 285–390 MPa) in comparison to that of plain i-PP homopolymer (1450 MPa), hence fulfilling the project internal target of $E_{flex}=300\pm 100$ MPa. However, i-PP/HECO blend still exhibits E_{flex} of 530 MPa which could be considered too high for cable application. DSC measurements of the i-PP blends (see Table II for a summary of the data from the second heating cycle) showed a main crystalline melting peak at $\sim 159\text{--}161^\circ C$ corresponding to α -form PP crystallites. i-PP/HECO showed the highest melting enthalpy (*c.f.* degree of crystallinity) whereas the PBE-containing blends showed lower melting enthalpies, likely associated with the higher total ethylene content of the blends and thus reduced crystallizability.

TABLE II. SUMMARY OF TENSILE, FLEXURAL AND DSC RESULTS.

Code	Tensile			Flexural	DSC	
	E_{tens} [MPa]	σ_{break} [MPa]	ϵ_{break} [%]	E_{flex} [MPa]	T_m [°C]	ΔH_f [J/g]
i-PP/PBE1	516	26.8	482	375	161.3	53.9
i-PP/HECO	412	13.8	451	530	159.6	60.2
i-PP/PBE2	301	21.5	497	285	161.9	47.9
i-PP/HECO/PBE2	527	24.7	472	390	161.0	54.1

Finally, SEM cross-sectional morphology of the studied i-PP blends are presented in Fig. 1. i-PP/PBE1 is seen to exhibit relatively smooth blend morphology similar to plain PP homopolymer. On the other hand, varying degrees of phase separation is evident for the other i-PP blends incorporating PBE2 and/or HECO.

B. Space Charge Measurement

Fig. 2(a–e) present the space charge pattern inside the specimen during 3 hours of polarization followed by 1 hour of depolarization for all blends shown in Table I, respectively. While Fig. 2(f) is a scatter plot that shows the FEF with respect to the net charge density calculated according to (1) – (3) for

all considered blends. Space charge pattern in i-PP/PBE1 is illustrated in Fig. 2(a). The heterocharges dominate the vicinity of both anode and cathode. In Fig. 2(b), i-PP/HECO blend shows less amount of charge accumulation with the same pattern of heterocharges near the opposite electrodes. i-PP/PBE2 shows an accumulation of positive charges near both electrodes with negative charges in the bulk, as illustrated in Fig. 2(c). While i-PP/HECO/PBE2 shows a strong accumulation of positive charges near the anode with a dominance of the positive charges also in the bulk. XLPE, in Fig. 2(e), shows a more complex pattern of both positive and negative charge dynamics and accumulation. In Fig. 2(f), XLPE has the greatest FEF (20%) over all tested i-PP based blends. Overall, all tested i-PP/PEC blends still have a great FEF of 10 – 15 % which can affect the reliability of the HVDC cable system [2].

C. Breakdown Strength Measurement

Table III shows the parameters of the Weibull distribution that fit the breakdown strength results for the i-PP blends in addition to the reference XLPE. The scale parameters of the breakdown strength of i-PP/PBE1 and i-PP/HECO/PBE2 are greater than that of the XLPE. While the breakdown strengths of i-PP/HECO and i-PP/PBE2 are lower than that of the XLPE

TABLE III. WEIBULL DISTRIBUTION OF THE BREAKDOWN STRENGTH.

Code	$E_{50\%}$ [kV/mm]	$E_{63.2\%}$ [kV/mm]	β
i-PP/PBE1	520.3	542.2	12.9
i-PP/HECO	325.0	339.6	12.2
i-PP/PBE2	410.0	433.8	9.5
i-PP/HECO/PBE2	486.3	510.0	10.3
XLPE	451.3	471.0	11.5

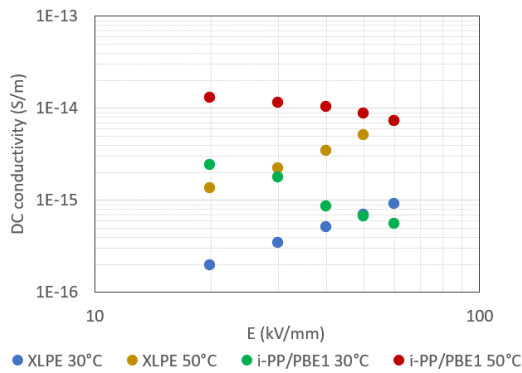


Fig. 3 DC conductivity calculated at the end of 24-hour polarization for XLPE and i-PP/PBE1 at $T=30^\circ\text{C}$ and $T=50^\circ\text{C}$.

D. DC-conductivity Measurement

Fig. 3 shows the DC conductivity calculated from the polarization current measurements taken at the end of each 24 h voltage application. The DC conductivity trends for XLPE and i-PP/PBE1 exhibit opposing behaviors, indicating differences in the space charge dynamics and conduction mechanisms between the two materials. DC conductivity in i-PP/PBE1 is relatively higher than that of XLPE up to a specific electric field threshold, beyond which the relationship reverses. The value of this electric field threshold increases the higher the temperature.

E. Thermally Stimulated Depolarization Current TSDC

Fig. 4 presents the TSDC current in picoampere with respect to the heating temperature in the range ($0 - 140$) $^\circ\text{C}$. The reference XLPE is characterized by the presence of only one weak peak in the temperature range ($70 - 110$) $^\circ\text{C}$. Dissimilar to all tested i-PP based blends which all have two distinguishable peaks, the deep-trap peak falls in the temperature ≈ 100 $^\circ\text{C}$, while the shallow-trap peak falls in a various range of lower temperatures. i-PP/PBE1 shows the greatest peaks and followed by the other blends (i-PP/HECO, i-PP/PBE2, and i-PP/HECO/PBE2) that have comparable peaks. Fig. 5 shows the (transient) DC-conductivity calculated from the polarization current measured during the TSDC measurement at $E=15$ kV/mm and $T=90^\circ\text{C}$. XLPE (black curve) shows the lowest value of DC-conductivity compared to all i-PP based blends under study. i-PP/PBE1 and i-PP/HECO have greater DC-conductivity values with 2 and 1 orders of magnitude greater than that of the XLPE, respectively. While both i-PP/PBE2 and i-PP/HECO/PBE2 have a comparable DC-conductivity that is still greater than that of XLPE.

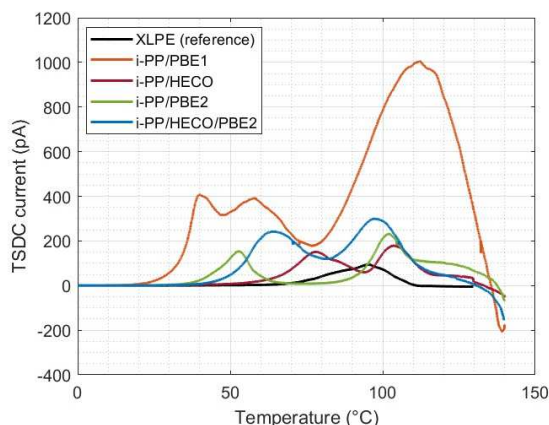


Fig. 4 TSDC current in (pA) with polarization at $E=15$ kV/mm and $T=90^\circ\text{C}$ for all blends, and the reference XLPE.

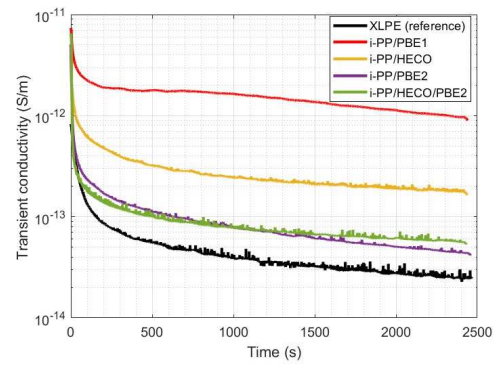


Fig. 5 Transient DC conductivity calculated during the polarization time of TSDC at $E=15$ kV/mm and $T=90^\circ\text{C}$ for all blends, and the reference XLPE.

V. CONCLUSION

In this paper, i-PP based blends are subjected to initial characterization for the application as HVDC cables insulation. Compared to XLPE, some i-PP based blends show greater breakdown strength, while other blends show lower values. The DC conductivity of all the tested i-PP based blends are greater than that of the XLPE. i-PP blends show noticeable heterocharge accumulation near the electrodes. i-PP blends are characterized by the presence of two distinguishable great TSDC peaks, compared to one high temperature peak in the case of the XLPE. Further investigations are currently carried out to enhance the characteristics of the i-PP blends by either varying the blends composition or by introducing grafted additives in the insulation matrix for reliable HVDC cable systems.

ACKNOWLEDGMENT



Funded by the European Union

Funded by the European Union Grant Agreement No 101075592. Views and opinions expressed are however those of the author(s) only and do not necessarily reflect those of the European Union or CINEA.C. Neither the European Union nor the granting authority can be held responsible for them.

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