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Effect of Reverse Polarity on Space Charge Evolution in Polypropylene with Different Concentration of Natural and Synthetic Nano Clay

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Abstract- The space charge evolution in the Polypropylene (PP) loaded with two types of nanoparticles, synthetic and natural clay with weekly reversing polarity was studied. The space charge in these two polymer nanocomposites (PNCs) were measured with the PEA technique after the each period of poling. The charge distribution in the host materials was substantially smaller after reverse polarity compared to dc negative polarity. This could be related to the mitigation of charge bay the compatibilizers added to the host materials. The charge accumulation measured after the negative periods was lower than the positive periods. The PNC with 2-wt% of synthetic clay stocks lots of charges after the end of the positive periods. Then, it will not be optimal concentration as it was for dc poling. It is found out that the two optimal concentrations for reverse polarity were the 4-wt% (PP1-S4%) for the synthetic PNC and the 2-wt% (PP1-N2%) for the natural PNC. With reverse polarity and before the 12th week of poling, all filled and unfilled materials stock charges of the same order. The unfilled materials show increasing of charges that crosses the values of filled materials, except the PP1-S2%, after the 12 weeks of poling with reverse polarity. More poling time is necessary to investigate the effect of long term poling on the charge evolution and investigate the continuous increase of charge in the unfilled materials.

II. INTRODUCTION

Polypropylene (PP) is extensively used as electrical insulation in power capacitors, cable wraps, phase separators for rotating electrical equipment, etc. Loading this material with low weight percentages of nanoparticles such as natural or synthetic nanoclay generates Polymer Nanocomposites (PNC) with superior electrical, mechanical or thermal properties compared to the unfilled material [1, 2]. These composites could have better resistance to partial discharges, higher working temperatures, less build-up of space charges, etc.

It is generally accepted that the interface between nanoparticles and surrounding media plays a major role in controlling the carrier transport through PNCs [3]. The electric transport properties are closely related to the charge injection, retention and bulk dynamics [4]. Nanoparticles can change the density and depth and density of traps, which in turn change the density and

mobility of space charges in the PNCs. In other words, small quantities of nanoparticles could mitigate charge injection (or trapped charges) and slow down the degradation of these materials, although, excessive quantities of fillers could lead to lose this advantage. Therefore, the percentage ratio of fillers to PP must be limited to the so-called percolation threshold [5].

In our recent work, the effect of long term poling with dc negative polarity, on the evolution of space charges and dc conductivity for two types of nanoclay, synthetic and natural organoclay was studied [6]. But, the majority of equipments used for high voltage dc (HVDC) in power systems will be frequently subject to polarity reversal. Designing a PNC for dc applications must take into account the effect of reverse polarity, especially on the evolution of space charge.

After determining the optimal concentration for dc poling in our recent works [6, 7], in this work, the optimal concentration on nanoparticles that can mitigate the maximum of space charge for each PNC material will be investigated. The evolution of space charge density during a weekly reversed polarity of a 25 kV/mm dc field will be explored. At the end of each week, the space charge distribution was measured using the Pulse Electro-Acoustic technique, (PEA).

II. EXPERIMENTAL

The tested specimens were composed of two sets of PNCs. The first set was composed of an isotactic PP (Profax HL-451H from Basell) filled with different wt% of Topy synthetic tetrasilisic mica from Topy Co., Ltd. of Japan. The second set was composed of the same PP material but filled with a different quantity of Cloisite®20A natural montmorillonite clay from Wyoming. The main purpose of the project was to develop better insulation materials for power capacitors.

The two unfilled PP materials are called as PP1-0% and PP2-0%. PP1-0% is used as the host material for the PP-synthetic clay composite and the PP2-0% is the host material for the PP natural nano clay composite. The only deference between these two materials is the percentage of compatibilizers (17.4% and 12.8% respectively). The filled specimens are referred to in this paper as "PP1+Sx%" if they contain synthetic clay and "PP2+Ny%" if they contain natural clay. x% and y% indicate the percentage by weight content of

Topy and Cloisite®20A (or the synthetic and natural) organoclay, respectively.

The specimens of each composite poled for 12 W (week)-one W= 168 h- with -25 kV/mm dc field and for 12 W with +25 kV/mm dc field, in weekly steps of polarity reversals. Before starting the poling process and to set the reference distribution of space charge the PEA was performed at the unpoled (0 W) condition. The space charges were performed with the Five Labs PEA measurement system. This system have a resolution of ~15 μm measured with Tektronix 7404, 4 GHz, 20 GS/s digital oscilloscope. The first week of polarity was negative and then immediately after removing specimens from the poling setup, the PEA was carried out before poling with the positive polarity. Figure 1 illustrates the weekly reversing polarity of the poling process and the time at which the PEA measurements were performed. For convenience the week is used as a time unit instead of hours in this work.
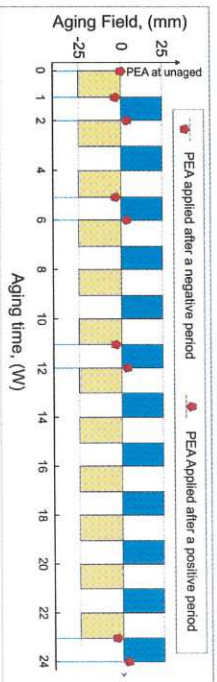

Fig. 1. Description of periodically reversing polarity and the time at which the PEA was applied 1 period is 1 week or 168 h of poling.

During the PEA tests, a thin layer of silicon oil was applied on each side of the sample to ensure a good contact between the specimen and the electrodes. A 0.2 mm thick, 30 mm diameter semicon disk was inserted between the specimen and the upper brass electrode during dc poling.

Two specimens of each material were tested and the average value of the charge distribution will be used for this paper.

III. RESULTS AND DISCUSSION

III-1. Space charge distribution

Figures 2 and 3 depict the space charge distribution in the two host materials, PP1-0% and PP2-0%, poled for 12 W of weekly reversed polarity. To clearly see the impact of reversing polarity on the charge density, the dc poling for 12 W was drawn at the same figure. No charge accumulated in the third of the right for polarity reversal in spite of having a large negative homocharge for the dc poling. The dashed line in Fig. 2 shows a positive peak near the ground electrode after the 6th week of negative polarity. This homocharge peak is followed by a heterocharge peak generated from the past opposite polarity periods. But, after the next period or the positive polarity, the point-dash line shows a large homocharge peak. At the right-hand electrode, all charges were mitigated after both periods of polarity. The dc single negative poling of the PP1-0% shows lots of charge injected from both electrodes and more charges

were injected from the right-side electrode compared to the ground side one; see the continuous line of Fig. 2.

Fig. 3 depicts the charge distribution in the PP2-0% with dc and reverse polarity after 12 weeks of aging. It is well observed the injection of charges from both electrodes after the negative period (dashed line) and the positive period (point-dash line) for the reverse polarity poling. These peaks of charge are very low compared to the large quantity of charge injected by the dc poling (continues line). It was reported in [6] that both host materials PP1-0% and PP2-0% showed a large injected charges under a dc negative polarity with significant difference between each other. The peaks of charge for the PP1-0%, were anti-symmetric and higher compared to the PP2-0%, see the continuous lines in Fig. 2 and Fig. 3.
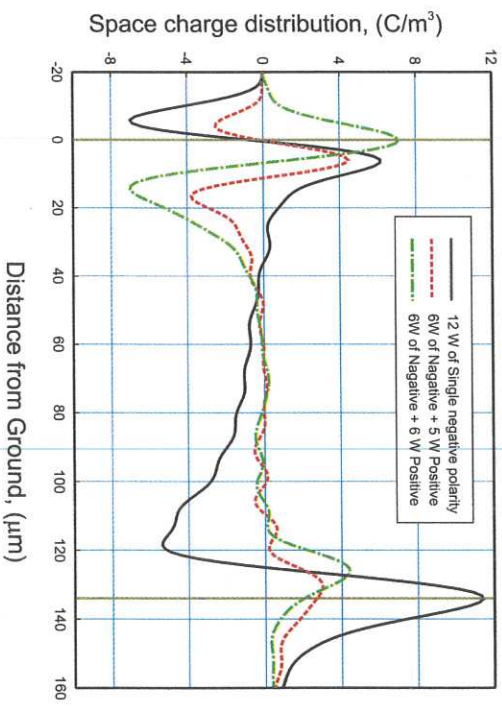

Fig. 2 Space charge distribution after 12 W of single negative polarity and 6 weeks negative + 6 weeks positive polarity for (a) PP1+0%.
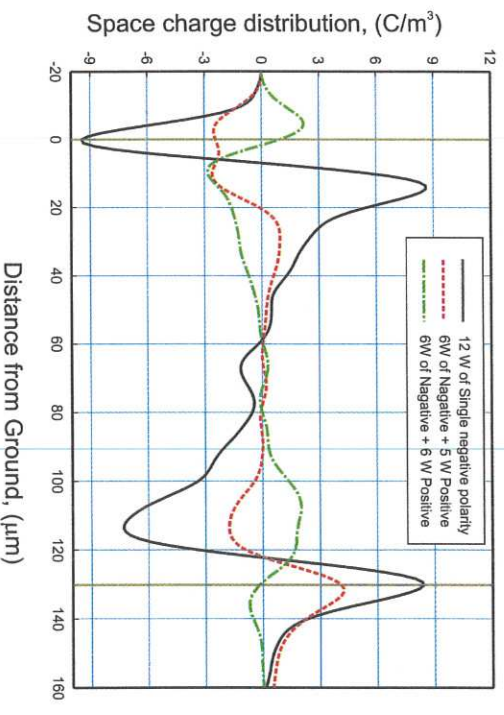

Fig. 3 Space charge distribution after 12 W of single negative polarity and 6 weeks negative + 6 weeks positive polarity for PP2-0%.

The homocharge in these materials is due electron injection or extraction (hole injection), which is likely generated by the compatibilizers. The difference between the charge quantity and distribution is only due to the different percentage of compatibilizers, which was added to make the interaction between the PP and the nanoparticles more favorable [8-10]. More work is required to study the effect of compatibilizers on the dielectric properties of the PNCs in general and space charge evolution in particular.

The charge distribution with reverse polarity in the PP filled with different wt% of synthetic clay, PP1-Sx%, is depicted in Fig. 4 and Fig. 5. The first figure shows the charge distribution after the 12th **negative period** (23 W of total poling time) and the second after 12th **positive period** (24 W of total poling time). The peak of heterocharge near the ground electrode in the PP1-0% is decreased with the loading 2-wt% of the synthetic fillers, and reversing polarity (becomes homocharge) for the 4-wt% and higher, see Figure 4. All the filled PNCs show low value of charge in the central zone.
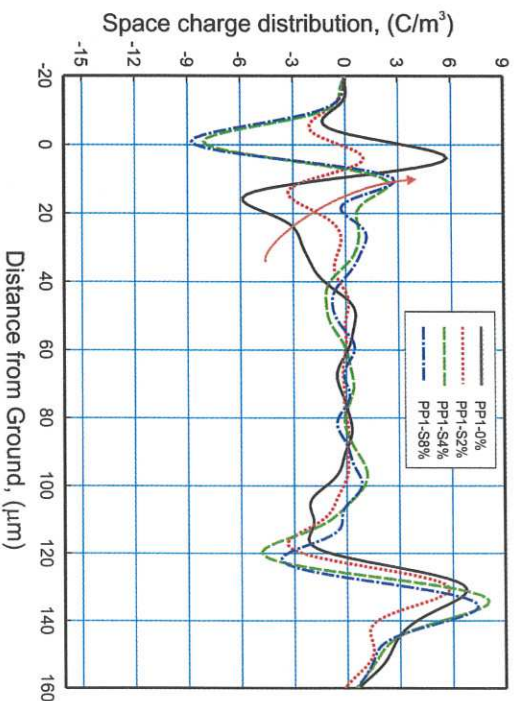
composite is the 4-wt% (PP1-S4%) and this is valid for the dc and the weekly reversed polarity poling.

Space charge distribution, (C/m³)
(y-axis values: 9, 6, 3, 0, -3, -6, -9, -12, -15)
Distance from Ground, (µm) (x-axis values: -20, 0, 20, 40, 60, 80, 100, 120, 140, 160)
Legend: PP1-0%, PP1-S2%, PP1-S4%, PP1-S8%

Fig. 4. PP with synthetic clay. Poled for 12 W negative + 11 W positive. Space charge distribution measured after the 12th W of **negative polarity**.

Close to the ground electrode and after the 12th period of **positive polarity**, Fig. 5 shows a similar evolution of decreasing charges with the concentration, see the arrow. A significant quantity of injected charges is observed for the PP1-S2% compared to all the other concentration including the PP-0%. Many large peaks with opposite polarity are seen in the central zone as illustrated in the eclipsed area. This could be related to the earlier periods of opposite polarity. Then, the PP1-S2% stocks a large quantity of charges, especially at the vicinity of the ground electrode and in the central zone. Fig. 4 and Fig. 5 demonstrate that the optimal value of space charge for the PNC with synthetic composites is either the PP1-S4% or the PP1-S8%. But the later concentration has shown a very high conductivity and more than one sample failed under dc aging [6, 7]. Then, the optimal concentration of the synthetic

Space charge distribution, (C/m³)
(y-axis values: 9, 6, 3, 0, -3, -6, -9, -12, -15)
Distance from Ground, (µm) (x-axis values: -20, 0, 20, 40, 60, 80, 100, 120, 140, 160)
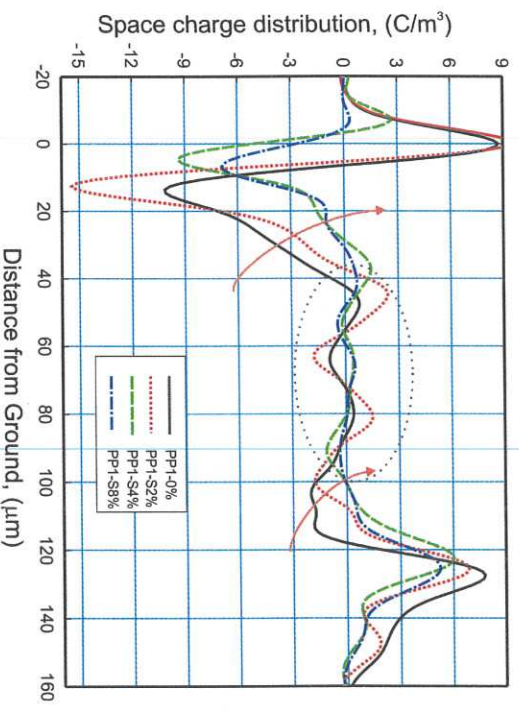Legend: PP1-0%, PP1-S2%, PP1-S4%, PP1-S8%

Fig. 5. PP with synthetic clay. Poled for 12 W negative + 12 W positive. Space charge distribution measured after the 12th W of **positive polarity**.

Figure 6 shows that all concentrations including the unfilled material accumulate a very low quantity of charge.

In Fig. 7 the homocharge peaks from both electrodes decrease before being inversed and become heterocharges and increased for the PP2-N6%. Heterocharges increase the electric field at both electrodes; sharply increase conductivity and leads the specimens to failure. The 2-wt% concentration of natural clay stocks the lowest quantity of charge in the central zone compared to the PP2-0% and the PP2-6% after both the negative period and the positive periods. Then, the optimal concentration of the PP filled with natural composites is the 2-wt%.

Space charge distribution, (C/m³)
(y-axis values: 9, 6, 3, 0, -3, -6, -9, -12, -15)
Distance from Ground, (µm) (x-axis values: -20, 0, 20, 40, 60, 80, 100, 120, 140, 160)
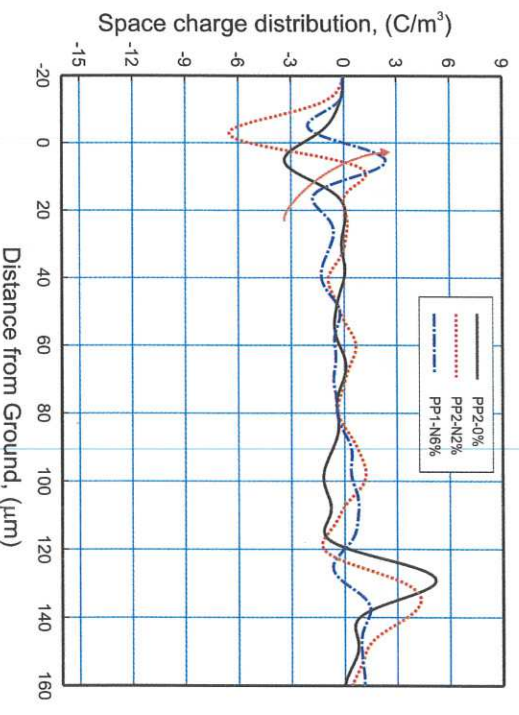Legend: PP2-0%, PP2-N2%, PP1-N6%

Fig. 6. PP with natural clay. Poled for 12 W negative + 11 W positive. Space charge distribution measured after the 12th W of **negative polarity**.
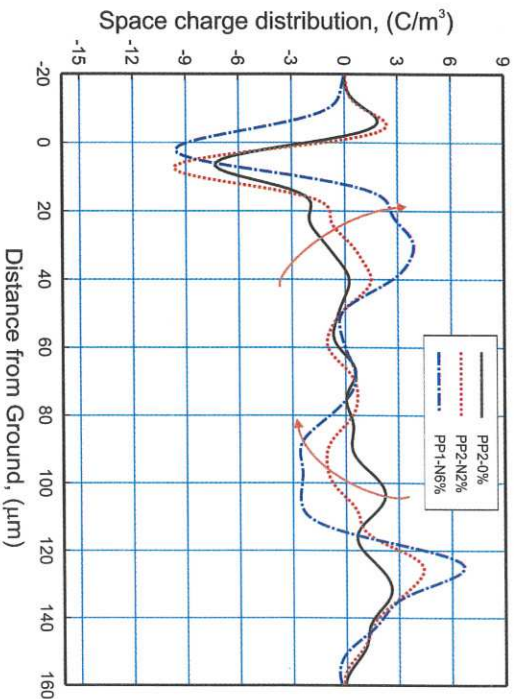
Fig. 7. PP with natural clay. Poled for 12 W negative + 12 W positive. Space charge distribution measured after the 12th W of positive polarity.

Space charge distribution, (C/m³)

Distance from Ground, (μm)

Legend: PP2-0%, PP2-N2%, PP1-N6%

Drawing the two optimal composites PP-S4% and PP-N2% in Fig. 8 after the negative and positive periods shows lots of general similarity. Many positive and negative small peaks of charges with opposite polarity are observed in this figure. This indicates the existence of charge packets induced by the alternative polarity reversal, which push some quantity of charges deeper into the specimen's volume. These charge peeks were created by the precedent poling periods.
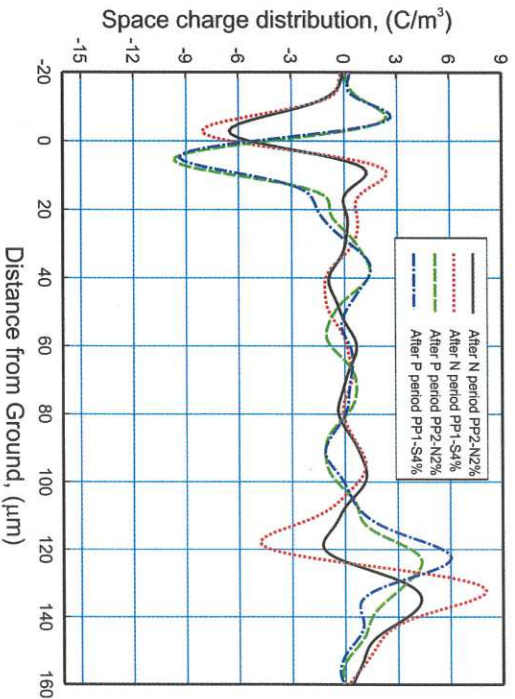
Fig. 8. Space charge distribution in the PP2-S4% and PP1-N2% specimens after the 23rd week (negative polarity) and the 24th week (positive polarity).

Space charge distribution, (C/m³)

Distance from Ground, (μm)

Legend: After N period PP2-N2%, After N period PP1-S4%, After P period PP2-N2%, After P period PP1-S4%

III-2. The integral of the charge density

The absolute value of the integral of charge between both electrodes, Q, for the PNC with synthetic clay for all the measured periods is depicted in Fig. 9. The left side of this figure shows Q measured after the negative periods and the right side after the positive periods. For comparison purpose the Q calculated from the dc poling is inserted on the top of the

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Fig. 9. From this figure the following observations can be noticed: 1) the highest Q is observed in the unfilled material PP1-0%. 2) the PP1-S2% has shown the high value of charge for reverse polarity. 3) the general tendency of Q for all the filled material, except the PP1-S2%, is not dramatically changed after 24 weeks of reverse polarity poling, 4) the value of Q for the dc and reverse polarity poling of the PP1-0% shows a clear tendency of increasing value with aging time. 5) This confirms that the PP1-S2% is not convenient for the reverse polarity applications where it stocks lot of charge, especially after the positive period of poling. 6) The value of Q was lower than the filled materials before the 2nd week of dc poling and 12 W for the reverse polarity. For reverse polarity, the charges could continue increasing with poling time and could become much larger than the filled materials after long term poling.

Absolete value of Space charge, (mC/m²)

Aging time, (W)

Legends: Weekly Reversed Polarity Tested with PEA After Negative Periods; Single Negative Polarity; Weekly Reversed Polarity Tested with PEA After Positive Periods
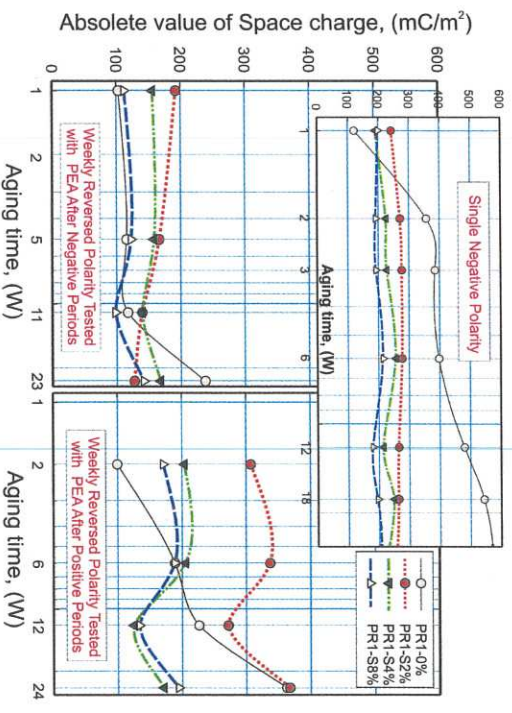
PR1-0%, PR1-S2%, PR1-S4%, PR1-S8%

Fig. 9. The integral of charge Space charge distribution in the PP with synthetic nano particles composites for all the poling time.

The absolute value of the integral of charge, Q, for the PNC with natural clay is depicted in Fig. 10. The parts of this figure are similar to Fig. 9. However, the very clear advantage of filling with natural clay for the dc poling, no real effect on charge accumulation is seen for the reverse polarity after 24 W of poling with reverse polarity for the PNC with natural clay. The interface between nano-particles and neighbouring materials controls carrier and electric transport properties, which are closely related to the charge injection, retention, and bulk dynamics [10, 11]. Nanoparticles can change the depth and density of traps, which in turn change the density and mobility of space charge. From another point of view, it is probable that the interface could not be simple charge sources but the seats of progressive chemical, mechanical, and electrical degradation. Small quantities of nanoparticles could mitigate the charge injection (or charge trapping) and slow down material degradation [12]. However, an excessive quantity of nanofillers makes the distance between the adjacent platelets short enough to trigger local percolation processes. This can increase the

conductivity of the highly doped materials like the PP1-S8%, which could lead the specimen to failure. Lower quantity of charge for reverse polarity proves that these materials can withstand the reverse polarity as well as the dc polarity. Therefore, the optimal concentration of both composites can be used successfully as insulators for HVDC applications that will be frequently subject to reverse polarity.
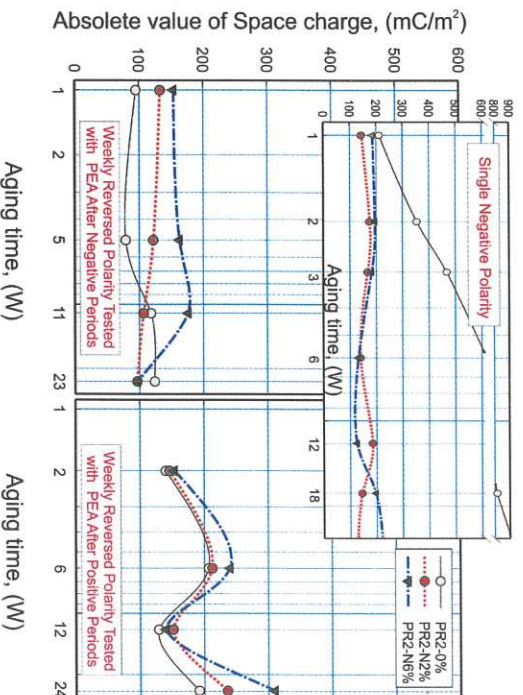

Fig. 10. The integral of charge Space charge distribution in the PP with natural nano particles composites for all the poling time.

Finally it is necessary to note that the 24 W of reverse polarity was not enough to fully see the advantage of reverse polarity for the PNCs. More work is in progress and the results will be the subject of future report

IV. CONCLUSIONS

The evolution of a space charge of two types of PP nano composites (PP based synthetic and natural clay) with weekly reversed polarity was investigated.

The quantity of accumulated charges was much lower in the case of the two base materials of PP with reverse polarity compared to dc single polarity.

The PP-S2%, which accumulated low quantity of charges at dc polarity and was accepted as an optimal concentration, will not for reverse polarity, because it stocks the highest value of charge.

The optimal concentrations of both composites were 4-wt% for the PP-synthetic clay and 2-wt% for the PP-natural clay. These concentrations were valid for both dc single negative polarity and weekly reversed polarity.

There is a tendency of continues increase of the total charge for the PP1-0% after 12 W, but, more work is necessary to confirm this tendency for long term poling with reverse polarity. These results can help in finding PP-nanocomposites can be efficiently used as power insulation for the dc and reverse polarity applications.

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