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Space Charge Evolution in Polypropylene loaded with Synthetic and Natural Nanoclay Aged at 50 °C Temperature

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Abstract- The evolution of space charges in Polypropylene (PP) filled with synthetic and natural nanoclay after aging with -25 kV/mm DC field under 50 °C were investigated. Following certain periods of aging, the space charge was measured by the PEA technique. The PP materials, bases for the two types of Polymer nanocomposites, i.e. PP were loaded with synthetic and natural nanoclay. The unfilled PP contains grafted maleic anhydride as compatibilizers. It was observed that the percentage amount of compatibilizers affects the charge distribution in the PP materials. The results indicated that aging with 50 °C temperature of the PNC with 2 and 4-wt% synthetic clay stocks high quantity of charges compared to the other filled materials. These two concentrations could not be optimal for aging under 50 °C. Insignificant accumulation of space charge and independent from aging time was observed for the PP loaded with 2-wt% of natural clay. This could be due to the smaller aspect ratio of natural nanoclay platelets compared to the synthetic nanoclay platelets. The clay platelets limit the molecular motion in the amorphous phase due to the interaction between the platelets and PP matrix. But, more work is required to investigate the effect of the percentage of compatibilizers on the charge accumulation property. Highly doped PNCs like the PP1-S8% and PP2-N6% can enhance overlapping of the diffuse double layer charge clouds around nanoparticles and promotes movement of space charges, thereby leading to a sharp increase of the conductivity of PNCs.

I. INTRODUCTION

Developing new insulation materials with superior quality, particularly for DC high voltage applications, has drawn great attention during the last decade. It was recently reported that polymer materials such as polypropylene (PP) with low weight percentages of nanoclay generates Polymer Nanocomposites (PNCs) with better mechanical, thermal tensile and fire retardant properties compared to unfilled materials [1-2]. It was also reported that these composites can show higher resistance to partial discharges and less build-up of space charges [3-4].

It is generally accepted that the interface between nanoparticles and base polymer plays a major role in controlling the carrier transport through PNCs. The electric transport properties are closely related to the charge injection, retention and bulk dynamics [5]. Nanoparticles can change

the depth and density of traps, which in turn changes the density and mobility of space charges in the PNCs. This could be related to the aspect ratio of the nano particles. The lower the aspect ratio, the higher the mitigation of charge is [4]. A small quantity of nanoparticles could mitigate charge injection and slow down the degradation of the PNCs. But, excessive percentage of fillers can enhance the overlapping processes of the diffuse double layer charge clouds around nanoparticles and promotes movement of space charges, thereby leading to a sharp increase in the conductivity of PNCs. Accordingly, it was reported that the percentage ratio of fillers to the base materials must be limited to the percolation threshold [6].

In our previous work the space charge and conductivity evolution in the PP filled with different concentrations of synthetic and natural clay were reported. This work illustrated the optimal concentrations of nanoparticles that can mitigate the space charge accumulation with dc single and periodically-reversing polarity aging [7] in room temperature (RT).

Since the majority of power equipment works at temperatures between 50 and 70 °C and even more, it will be interesting to find the optimal composites that can mitigate charges under these temperatures.

In this work and as a part of a continuing project, the evolution of space charge with aging time under 50 °C for 1008 h will be studied. More work is required to investigate the impact of using different percentage of compatibilizers on the property of space charge accumulation. In this work, space charge measurements were carried out using the Pulses Electro-Acoustic (PEA) technique.

II. MATERIALS AND METHODS

The tested PP-based nanocomposites in this study, which were prepared by the master batch method of melt compounding, can be categorized into two groups of PNC. The first group is composed of PP with 0, 2, 4 and 8 weight percent (wt%) of synthetic organoclay and the second one includes composites of PP with 0, 2 and 6 wt% of natural organically Cloisite 20A (montmorillonite) or tetrasilic mica. Both groups were developed in obtaining improved

power capacitor insulating materials. The master batch was prepared by melt compounding of isotactic polypropylene with maleic anhydride (1:1 mixture of maleic anhydride from Eastman (Epolene- 3015) and from Crompton (Polybond 3150), antioxidant and either natural or synthetic organoclay. More details about the process of preparation of the master batch can be found in **Error! Reference source not found.** The nanoclays were pre-intercalated through replacement of the Na^+ of the clay by an organic cation, thereby facilitating an acceptable degree of exfoliation for the resultant composite [9].

Table 1 summarizes the content of tested composites and their corresponding name codes. It is to be noted that the only difference between the unfilled PP, the PP1-0%, and PP2-0% is the wt% of compatibilizers; where PP1-0% contains 17.4-wt% and PP2-0% contains only 12.8-wt%.

Sample aging was conducted under 25 kV/mm DC negative field and 50 °C temperature for 1008 hours, and space charge measurement was conducted using the Five Lab[®] pulsed electroacoustic (PEA) technique under 4 kV/mm DC positive field. The resolution of the PEA system was 15 μm and PEA signals were captured by a Tektronix 7404 oscilloscope with 4 GHz bandwidth and sampling rate of 20 GS/s. In this work, the anode is the ground electrode, and the cathode is the HV electrode during the aging time. The measurements were conducted for unaged (0 h) as well as 168 h, 504 h, and 1008 h of aging. A silicone oil layer was added to both sides of the tested specimens prior to measurement in order to ensure a good contact between the electrodes and specimens.

Table 1. Description of the tested composites

Sample code	Base Polymer	Anti-Oxidants (wt%)	Compatibilizers		Filler	
			Level	(wt%)	Type	(wt%)
PP1-0%	PP	0.2	PP grafted with Maleic Anhydride	17.4	Synthetic clay	0
PP1-S2%				17.1		2
PP1-S4%				16.7		4
PP1-S8%				16		8
PP2-0%			Natural clay	12.8	Natural clay	0
PP2-N4%				12.5		2
PP2-N6%				12		6

III. RESULTS AND DISCUSSION

The evolution of space charges in the unfilled materials is depicted in Figures 1-2. The arrows inserted in Figures 1-2 at the cathode side illustrate the movement of homocharge charge towards the bulk of the specimens with decreasing magnitude as a function of aging time. The peaks of charge in the PP1-0% unfilled material are opposite to these in the PP2-0% material. The first material, PP1-0% shows a larger quantity of heterocharge peaks relative to the PP2-0% composites, with an increase in magnitude and advancement towards the specimen center with aging time. In contrary, PP2-0%, which contains a lower quantity of compatibilizers, showed homocharge peaks in the vicinity of the anode. In Fig. 2 the positive peaks of homocharges was depleted with

aging time. Note that the positive peak in the vicinity of the anode decreased and became negative after 1008 h of aging, see inserted arrows in Figs. 2. This means that aging with this temperature creates heterocharges at the vicinity of the anode. Since compatibilizers are added to PNCs to facilitate interaction between nanoparticles and host material, it is expected to have an effect on the space charge accumulation.

Loading the PP1-0% with 2-wt% or 4-wt% of synthetic nanoclay and aging with -25 kV/mm DC field in 50 °C temperature, changes dramatically the charge distribution. Figures 3-4 show low quantity of charge at the cathode side, but large homocharge peaks with increasing magnitude close to the anode side. Therefore, the PP1-S2% and PP1-S4% could not be optimal PNCs. It is necessary to note that both concentrations were considered optimal for RT single polarity aging and only the PP1-S4% was found optimal for weekly reversed polarity aging [7].

Figure 1. Space charge measurement of PP1-0% samples aged with -25 kV/mm DC field at 50 °C.

Figure 2. Space charge measurement of PP2-0% samples aged with -25 kV/mm DC field at 50 °C.

The specimens of PP1-S8% and PP2-N6%, which contain high concentrations of nanoclays, showed lower charge quantity than the PP1-S2% and PP1-S4%, Figures 5-6. But, the fact that the PP1-S8% and the PP2-N6% composites possessed relatively high conductivity and many

failures, as reported in [4], didn't encourage considering any of these composites as optimal compositions. This could be related to the excessive nanoparticles concentrations, which may increase the overlapping of the diffused double layer charge clouds. Therefore, the concentration of nanoparticles should be lower than the percolation threshold [6].

Figure 3. Space charge measurement of PP1-S2% samples aged with -25 kV/mm DC field at 50 °C.

Figure 4. Space charge measurement of PP1-S4% samples aged with -25 kV/mm DC field at 50 °C.

Figure 7 presents the evolution of charge in the PP2-N2% after aging with -25 kV/mm DC field under 50 °C for 1008 h. It is clearly observed that the quantity of charge in this type is small and independent of the aging time. This concentration is the most effective in suppressing charges under 50 °C and can be considered optimal for this condition of aging. It is useful to note that the 2-wt% of natural clay was also found optimal for aging with single polarity and RT degree and also under weekly reversing polarity.

Figure 8, shows the total charge or the integral of the absolute value of charge, Q . It is clearly observed that the highest Q was obtained for the PP1-0% composites. However, the PP2-0% presented high quantity of charges up to 168 h of aging. After this aging time, the quantity of total charge started to decrease, reaching a relatively low value after 1008 h.

Figure 5. Space charge measurement of PP1-S8% samples aged with -25 kV/mm DC field at 50 °C.

Figure 6. Space charge measurement of PP2-N6% samples aged with -25 kV/mm DC field at 50 °C.

Figure 7. Space charge measurement of PP2-N2% samples aged with -25 kV/mm DC field at 50 °C.

For the filled specimens, the following points were observed: (i) the highest Q , which increased with aging time, was obtained for the PP1-S2% (ii) The lowest quantity of charge was observed for the PP2-N2%, this type of PNC also presented independence of aging time.

The lower charges found for the PNC loaded with natural clay, PP2-N2%, as compared to the PNC with synthetic clay, PP1-S2% and PP1-S4%, could be related to the ratio of the individual dimensions of the clay nanoplatelets. The aspect ratio of the individual platelets is ≈ 6000 for synthetic nanoclay; whereas, the aspect ratio was ≈ 286 for the natural nanoclay [11]. The smaller the nanoplatelets the larger the interfacial interaction zone between nanoparticles and the host material is [12]. Therefore, PP2-N2% would block charge injection more efficiently than the synthetic clay. This is in agreement with the findings of other authors [12, 13].

Figure 4. The total space charge, Q for all concentrations and both types of PNCs.

In addition, PP material can be affected by the manufacturing process where the percentage of compatibilizers can lead to different space charge evolution.

This is a continuing project aimed to find the best insulation materials for power capacitors. The next step is the investigation of the effect of different temperature and longer aging time on the evolution of space charge.

IV. CONCLUSIONS

The evolution of space charge distribution in the PP polymer filled with different concentrations of synthetic and natural organoclays aged with -25 kV/mm DC field under 50 °C was investigated. It was observed that the quantity of compatibilizers affect space charge distribution and quantity.

The PP1-S2% and PP1-S4%, which were accepted as optimal concentrations for single polarity with RT, stocked large amounts of homocharges close to the anode side when aged with 50 °C. Though, none of the synthetic PNCs could be considered optimal for this condition of aging at least until 1008 h.

The lowest quantity of charge after aging with 50 °C was observed in the PNC with 2-wt% of natural clay, PP2-N2%. It was reported that this type of PNC was optimal for aging with single polarity as well as for reverse polarity at RT. This may be due to smaller aspect ratio of the individual platelets of natural nanoclay compared to the synthetic nanoclay.

These results could be a big step towards searching for new insulation composites with superior insulation properties for power equipment in general and power capacitors in particular. More work is required to investigate the space charge evolution with different percentage of compatibilizers, longer aging time aging and higher temperature, which will be the subject of our next reports.

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