Electrical Treeing Behavior in Polyethylene Filled with Aluminum Oxide Nanoparticles

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Abstract

Electrical treeing behavior in low density polyethylene and its nanocomposite filled with 1.0 wt% and 3.0 wt% aluminum oxide nanoparticles was investigated by means of a real-time microscope recording system and a synchronized partial discharge measurement. For preparing the nanocomposite, uncoated aluminum oxide nanoparticles with 99.5% purity (Nanodur, CAS 1344-28-1) were used. These were of almost spherical shape with an average diameter of 45 nm. The test objects were prepared in form of the wire-plane electrode geometry [1]. The electrical treeing tests were carried out by applying 50 Hz AC voltage at a ramping speed of 22 V/s. The obtained experimental results indicate that the addition of the nanoparticles has a positive impact on the resistance to electrical treeing. Both the scale and the shape of the distribution of the treeing initiation voltage improved. The magnitude of partial discharge activity associated with the treeing in polyethylene nanocomposites was also lower than in the reference material.

1. Introduction

The historical development of electrical insulation has mainly been based on introduction of polymeric materials, which often contain various additives. Nanocomposites are in this respect a new class, containing filler particles of a few tens of nanometers and forming a well dispersed homogeneous blends [2]. The process of obtaining well-dispersed polymeric nanocomposites based on pure or silanized aluminum oxide nanoparticles in polyethylene has been developed [3]. However, for proving the applicability of these nanocomposites in electrical insulation applications, especially for insulation of cables of highest voltage levels, electrical testing is required to evaluate their behavior.

Electrical treeing is one of the main mechanisms of the degradation of polymeric insulation. There are three stages describing the formation of the trees: initiation, propagation and finally breakdown [4, 5], detection of which is usually based on optical observations. Since nanocomposites are often non-transparent, the optical observations of electrical tree inception have to be accompanied by electrical detection based on partial discharge (PD) measurements. The change in the number of detected PD events as well as the PD amplitudes provides information about the ongoing degradation process [6]. The work presented in this paper uses both the methods for investigating the electrical treeing behavior in low density polyethylene (LDPE) and its nanocomposites filled with aluminum oxide filler of different concentrations. For comparison purposes, reference experiments were also performed on samples made of pure LDPE. The influence of a prolonged thermal and electro-thermal treatment was also studied.

2. Experimental procedure

2.1. Nanocomposite preparation

LDPE pellets for manufacturing test samples were supplied by Borealis AB, Sweden. Aluminum oxide (Al_2O_3) nanoparticles with 99.5% purity (Nanodur, CAS: 1344–28–1), were supplied by Nanophase Inc. USA. Polyethylene nanocomposites containing 1.0 wt% and 3.0 wt% Al_2O_3 nanoparticles were prepared at School of Chemical Science and Engineering, Fibre and Polymer Technology, Royal Institute of Technology, Stockholm, Sweden [7].

2.2. Manufacture of test object

A schematic diagram of the test object used in this work is shown in Figure 1. It consists of a tungsten wire embedded between two sheets of the polymeric material, connected directly to an externally attached aluminum tape. Here the aluminum tape is used as the voltage connection. The wire- plane distance is 3-4 mm. The diameter of tungsten wire (provided by Luma Metal, Sweden) is 10 μm. The manufacturing method of this type of treeing test object was earlier developed at Chalmers University of Technology [1].

Premade sheets of polyethylene are joined together while the wire electrode is placed in between them. The whole assembly is melted in a special mold placed in a hot press at 130 °C and 2 kN for 3 minutes, thereafter a press-force of 200kN is applied. After 6 minutes, the temperature is ramped down to the ambient at a rate of 7.33 °C/min. Thereafter, the manufactured objects are degassed at 60 °C in vacuum for 7 days. A typical problem occasionally appearing during the preparation is formation of kinks at the wire electrode, but no particular precautions are needed as the trees growing at them can be censored [8, 9].

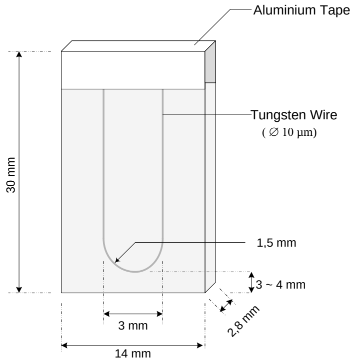

Fig. 1 – Treeing test object

2.2. Thermal and electro-thermal ageing

In order to study the effects of thermal and DC electric stresses on the tested materials, some samples were aged in air at 80 °C for 400h. It has been earlier shown that the influence of the voltage polarity was not significant [9]. In this work -10 kV voltage sources was used for the electro-thermal ageing. During the ageing, both the thermally and the electro-thermally aged samples were kept separately in the oven in special containers.

3. Measurements setup

3.1. Optical observation

The treeing tests were performed in a special cell with a transparent bottom wall for facilitating the microscopic observations. The whole test system, shown in Figure 2, consisted of a microscope, a CCD camera and a cold light source to provide transmitted illumination. A personal computer was used for real-time microscope digital recordings of the first four tree development. During the experiments, the samples were immersed in a transformer oil.

3.2. Electrical test arrangement

The electrical treeing tests were performed by applying a 50 Hz AC voltage with a ramp rate of 22 V/s. The test setup consisted of a variable AC source and a step-up transformer, connecting the test voltage to the sample through a current limiting resistor. A coupling capacitor in series with an active coupling quadripole AKV 568 was connected in parallel to the test object for PD measurements. The used coupling quadripole is supplied by +15 V and has two outputs; one is for PD signal detection and the other one is for synchronize signals, which also functions as trigger signal. A data acquisition (DAQ) module NI-5133/USB and a personal computer (PC) were used to registering the PD events. The electrical test setup together with the system for microscopic observations is shown in Figure 2. The whole arrangement allows realizing real-time video recording of the treeing process and the synchronized with it PD measurement. Due to the limitations of PCs data processing and internal memory, PD record length was set to 20000 points per sample and with 5 cycles per one record time (first cycle will be neglected).

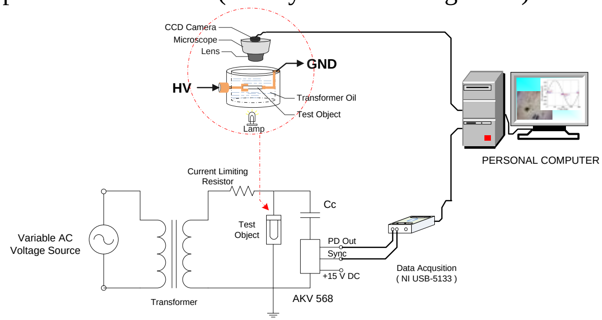

Fig. 2 – Combined test setup for optical observation and PD detection of electrical treeing

4. Result and discussion

Analysis of the recorded tree inception voltages were carried out by means of extreme value Weibull statistics, according to the IEEE 930 guide [10, 11]. The Weibull distribution plots used in this paper are shown with confidence intervals of 95% (95 % CI). This confidence interval refers to the scale parameter (α) defining the characteristic measured variable, i.e. the tree inception voltage and the shape parameter (β).

4.1. Electrical treeing

The tree inception voltages of the first four trees were analyzed, but the trees appearing on the kink were neglected. Weibull distributions shown in Figure 3 indicate that the samples containing nanoparticles have higher scale parameters comparing to the pure LDPE samples, indicating this way an increased resistance to electrical treeing. The sample with higher filler content (3.0 wt%) of aluminum oxide has inception voltage of 15.88 kV, whereas the tree inception voltages of the lower content (1.0 wt%) aluminum oxide nanocomposite and the pure LDPE are respectively 14.43 kV and 14.28 kV.

After the ageing at 80 °C for 400 h there is no influence on the inception voltage for the pure LDPE. However, nanoparticle filled materials yields an increase of the inception voltage. The inception voltages for 3.0 wt% and 1.0 wt% aluminum oxide nanocomposites respectively became 16.58 kV and 17.38 kV. The results obtained for the electro-thermally aged samples also show increases of the tree inception voltage for the pure LDPE and the nanocomposites.

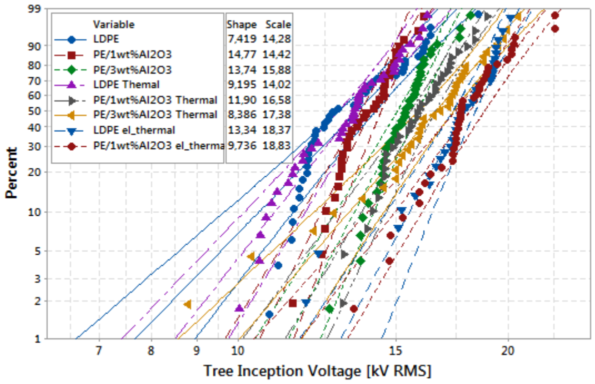

Fig. 3 – Two-parameter Weibull distribution of tree inception voltage in the studied materials (with 95% CI).

4.2. Partial discharge measurements

PD measurement data are presented as interval plot of maximum PD magnitudes (pC) and PD numbers (PDs/cycle) in Figures 4 and 5. The indicated individual standard deviations are used to present the intervals, while the mean values are determined with 95% of confidence. Comparing to the pure LDPE, the polyethylene filled with nanoparticles shows lower PD numbers and maximum PD magnitudes. For the pure for LDPE the PD number at 18 kV was 29 PDs/cycle, while it became only 5 PDs/cycle for the 1.0 wt% filled LDPE, and 22 PDs/cycle for the 3.0 wt% filled one. The respective mean values of maximum PD magnitudes were at the same time 316 pC, 293 pC, and 263 pC.

PD characteristics show after the thermal ageing a similar trend. For the material filled with 1.0 wt% aluminum oxide, the mean value of maximum PD magnitude slightly decreased, from 293 pC to 289 pC. The average PD numbers decreased further from 22 to 15 PDs/cycle for the 3.0 wt% filled LDPE, whereas the average PD magnitude decreased from 263 to 242 pC.

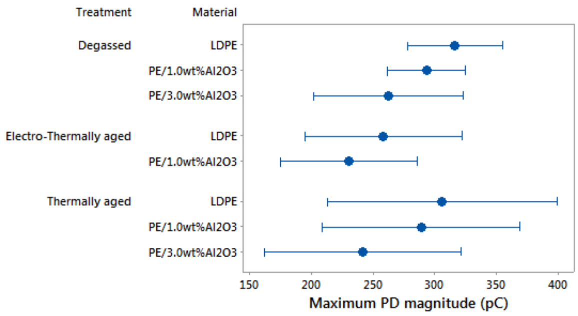

Fig. 4 – Interval plot of maximum PD magnitudes

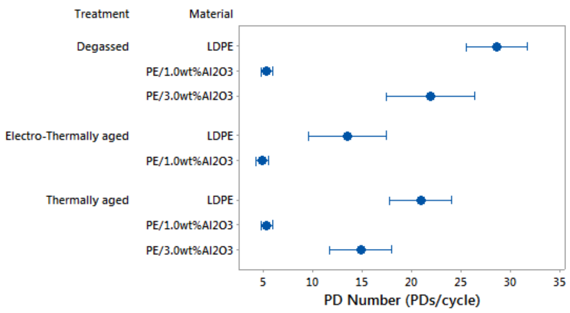

Fig. 5 – Interval plot of PD number (PDs/cycle) at voltage level of 18 kV

4.2. Correlation between electrical tree observations and PD activity

A strong correlation was found between the microscopic recordings and the PD measurements, similarly to those reported in [6]. In Figure 6 the number of PD events and their magnitudes are illustrated with respect to the phase of the applied voltage as well as related to it images. A small differences appearing in the detection of the first trees and the respective PD initiation voltages can result for three reasons; (1) PD magnitude threshold is set due to background noise to 100 mV (equal to 20 pC), (2) kink-trees are neglected in the optical observations but not in the PD measurements, and (3) the test objects are too dark for proper microscopic observations.

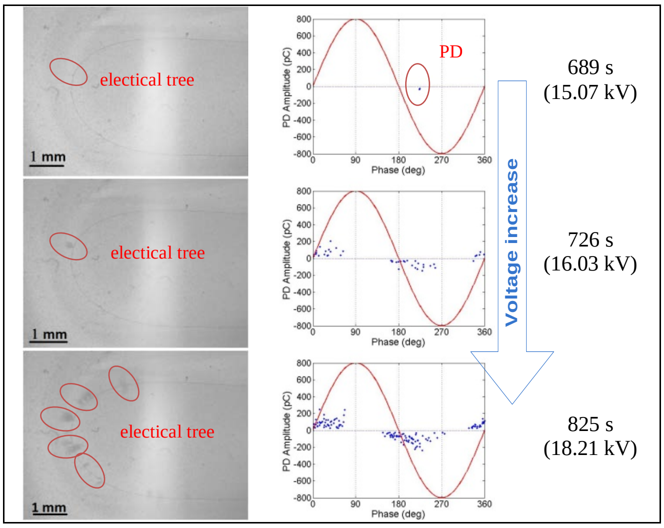

Fig. 6 – Electrical tree images and respective PD events detected during measurement with 22 V/s ramping rate in LDPE filled with 3 wt% of Al_2O_3.

4.3. Material analyses

To distinguish the ageing effects on the tested samples, differential scanning calorimetry (DSC) was used. The obtained melting curves are characterized by presence of two different peaks. The first peak, often called the 'annealing peak' [12], appears around 63-64°C for untreated samples, whereas for the aged samples it shifts to 93-96 °C. On the other hand the melting peak remains stable at between 109-111 °C.

Table 1 – Degree of crytallinity and melting behavior of tested samples

Material	Degree of Crystality (%)	Melting Temperature	
		First peak (°C)	Second Peak (°C)
LDPE	40.66	63.33	110.71
PE/1wtAl_2O_3	44.96	64.66	110.67
PE/3wtAl_2O_3	45.03	63.83	109.80
LDPE-$_{Thermal}$	53.03	93.98	110.09
PE/1wtAl_2O_3 $_{-Thermal}$	47.96	96.63	110.98
PE/3wt$Al_2O_3$$_{-Thermal}$	47.17	93.49	111.16
LDPE $_{Electro-Thermal}$	46.87	93.17	109.98
PE/1wtAl_2O_3 $_{Electro-Thermal}$	47.29	93.49	110.65

DSC in the scan mode is frequently used to determine the heat of fusion and the degree of crystallinity. In order to obtain these parameters, the melting peak has to be integrated and the obtained heat of fusion has to be compared with the heat of fusion of a perfect crystal (100% crystallinity) [12]. With the specific enthalpy of LDPE being 293 J/g [13], the estimated degree of crystallinity and the melting behavior of the test objects are listed in Table 1. An interesting behavior of the correlation between tree inception voltage and degree of crystallinity has been observed, as shown in Figure 7. This effect however still needs further elucidation as the calculated crystallinity ranges are within overlapping confidence intervals, indicating rather than the positive effect in nano-filled materials has another origin.

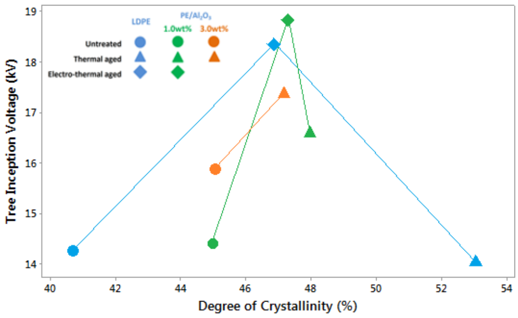

Fig. 7 – Tree inception voltage versus degree of crystallinity in the tested samples of pure and nano-filled LDPE.

5. Conclusions

The obtained experimental results indicate that the addition of Al_2O_3 nanoparticles to LDPE has a positive impact on the resistance to electrical treeing (the scale parameter of Weibull distribution is improved). In addition, increase of the filler content strengthens this effect. At the same time, the magnitude of PD activity associated with the treeing becomes weaker as compared with the discharges in pure LDPE.
The preformed thermal ageing had not affected the electrical tree inception voltage in the pure LDPE samples, while it resulted in a further increase of the inception voltage in the nano-filled samples. As a matter of fact, the electro-thermally aged samples exhibited higher tree inception voltage of both nano-filled and pure LDPE samples as compared to the non-aged ones.

6. Acknowledgments

The Swedish Foundation for Strategic Research (project EM110022) and Chalmers Area of Advance in Energy are acknowledged for providing the financial support that allowed conducting this work. Borealis AB is thanked for providing the base polyethylene material.

7. References

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