

# The Impact of the Inclusion of MgO Nano-Fillers in a Polyethylene Matrix on Dielectric Strength and Resistance to Partial Discharges

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## Abstract

One commonly used strategy to enhance polymers specific properties such as the resistance to partial discharges erosion is the incorporation into the polymeric matrix of inorganic micro or nanoparticles. This study focused on the dielectric properties of Low-Density Polyethylene (LDPE) filled with nano-sized Magnesium Oxide (MgO) particles compounded by thermo-mechanical process and one of the purposes was to establish appropriate processing parameters in order to reach the desired dielectric properties. LDPE was used as a matrix and was reinforced by MgO particles having a nominal average size of 30 nm. The MgO nanoparticles were treated with a silane coupling agent (3-Glycidyloxypropyl Trimethoxysilane). The samples were initially prepared in a melt-mixing chamber with a MgO content of 1% wt. These pre-mixed samples were further treated by the means of thermo-mechanical mixing in a conical co-rotating twin-screw extruder in order to improve the dispersion and distribution of the MgO particles. In this report, both lifetime under a PD activity and AC dielectric strength of pure and nano-filled LDPE samples have been measured and compared. Nano-filled LDPE samples were found to exhibit an improve lifetime, without any detrimental impact on their short-term dielectric strength. This suggests that nano-filled LDPE may be for electric applications for which the dielectric materials may be exposed to partial discharge activities. This is significant result for the use of MgO-reinforced PE as an insulating material for HV cables since the resistance to PD is closely related to treeing resistance which is the main electrical degradation mechanism that leads to failure for shielded extruded power cables.

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## Keywords

Low Density Polyethylene, MgO Nanofillers, Dielectric Strength, Partial Discharges Resistance

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## 1. Introduction

Physical properties of polymer mixed with micro- and nano-fillers strongly depend on several parameters such as the chemical nature of both phases and their interaction, as well as the concentration, shape, distribution and dispersion of particles used as fillers, with usually a better dispersion leading to enhanced properties. Numerous works have reported this rule for dielectric properties [1] [2] [3] [4] as well as for mechanical and rheological properties [5] [6] [7] [8]. More specifically, well-dispersed MgO nano-sized particles when incorporated into a polyethylene matrix were found to prevent space charges injection [9], to increase the electrical resistivity [10], and to some extent to improve DC breakdown strength [11] without any unacceptable increase of dielectric losses [12]. Accordingly, MgO-based polymeric nanocomposites are currently promising candidates to replace XLPE for extruded power cable applications, particularly for HVDC cables. However, another important aspect that has not yet been addressed in the literature is the resistance to partial discharge erosion of these materials, which is very relevant for HV applications. A significant improvement of this property could indeed translate in orders of magnitude improvement in the inception time of a tree-like degradation process (known as electrical treeing) and this is highly relevant since electrical treeing is usually the main failure mechanism in HV applications. In this work, the impact of the inclusion of 1 wt% of MgO particles into a Low-density Polyethylene (LDPE) matrix on electrical properties is reported. It was found that the presence of nano-sized MgO particles led to a significant increase of the material lifetime when exposed to partial discharges without any detrimental impact on the short-term AC breakdown strength.

## 2. Experimental

### 2.1. Materials

All nano-composites were prepared with unpigmented LDPE powder (additive-free - density: 0.922 g/cm<sup>3</sup>) and MgO nano-particles (1% weight-nominal mean diameter: 30 nm). The nanoparticles were first dried in vacuum during 24 h at 130°C, which is not expected to change the surface chemistry as this temperature is much lower than the thermal decomposition temperature of surface hydroxide groups [10]. They were then ultrasonically dispersed in ethanol until a homogenous suspension was formed and subsequently a solution of (3-Glycidyoxypropyl) trimethoxysilane was slowly added to the suspension with the mixture stirred at 50°C for 48 h in vacuum. The resulting slurry was washed

with fresh ethanol several times and dried in a vacuum oven at 80°C for 24 h. This type of surface treatment is expected to improve the compatibility between the organic matrix and the inorganic particles and has been reported to improve the dielectric properties for several metallic oxides polyolefins composites [13] [14].

Following the nanoparticles treatment, LDPE/MgO masterbatches were produced using a conventional hot mixer. The mixing was conducted at a temperature of 140°C, a rotation speed of 60 rpm, and for 30 min. This was followed by micro-extrusion conducted by the means of a mini-extruder.

Pellets obtained from the extrusion were finally pressed at 140°C and under a pressure of 10 MPa to obtain films with thicknesses ranging from 175 to 200 µm. The after-fabrication concentration was assessed by Thermo Gravimetric Analysis (TGA) using the following procedure: the temperature was risen from 50°C to 600°C at the rate of 20°C/min under nitrogen and then maintained at 600°C for 30 min under air. The residual weights averaged on several samples are listed in **Table 1**.

A SEM Hitachi SU3500 was used for the microstructural characterization of the samples. A reasonable dispersion and distribution of the MgO particles was observed with the mixing procedure previously described, as shown in **Figure 1**.

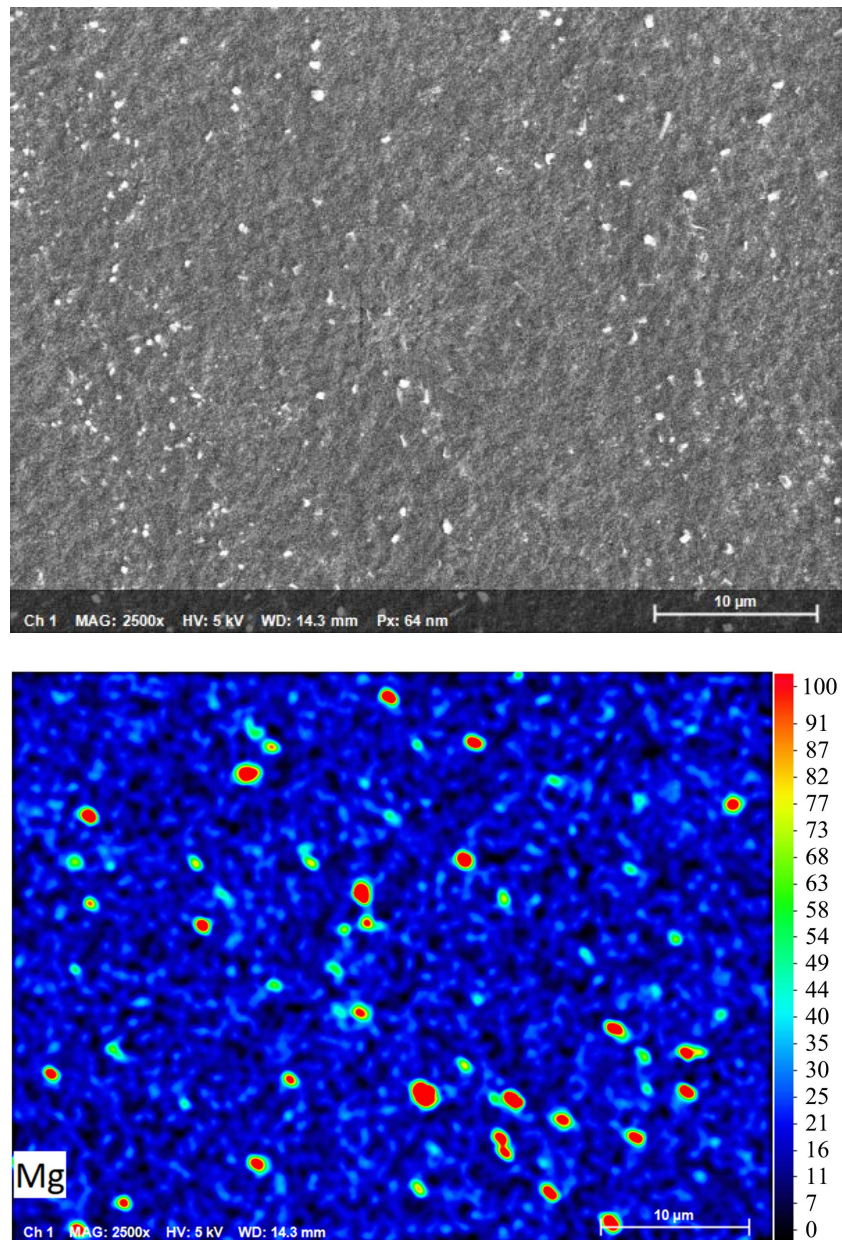
## 2.2. Experiments

The short-term AC dielectric breakdown strength was measured by using a classical experimental set-up composed of a 50 Hz high voltage transformer fed by a motorized autotransformer. The voltage rising rate was fixed at 1 kV/s. The experimental set-up is fully automatized, *i.e.*: the autotransformer is stopped when the current flowing through the sample reaches 10 mA and the corresponding breakdown voltage is then recorded. The measuring cell made of plexiglass (**Figure 2**) was equipped with two stainless steel electrodes: one plane electrode (diameter: 25 mm) and one semi-spherical (diameter: 1 mm). Samples were sandwiched in between the two electrodes and the measuring cell was filled with an insulating surrounding medium (dielectric liquid: GALDEN® HT55) to avoid any flashover or surface dielectric breakdown.

The thickness of each specimen was estimated after each experiment in the breakdown area, *i.e.*: four measurements around the breakdown point channel in order to calculate an average thickness. The variability in samples thickness was less than 50 µm. The Weibull's statistical analysis was used to treat the recorded data according to the IEEE std-930 [15]. The two-parameter Weibull's distribution cumulative probability of failure is given by:

**Table 1.** TGA measurements on the LDPE/MgO samples. (UT) stands for without the silane treatment and (T) stands for with the silane treatment as described above.

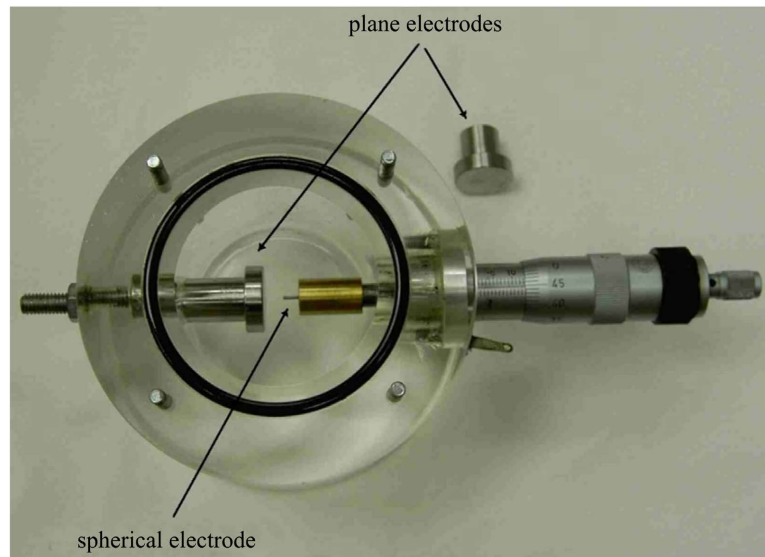
	T-95% (°C)	Residual Weight (%)
Pure LDPE	462	0.00
LDPE + 1% MgO (UT)	457	1.01
LDPE + 1% MgO (T)	463	0.72



**Figure 1.** SEM image of a 1 wt% PE/MgO sample (above) and Energy dispersive X-ray spectroscopy (EDX) map based on the Mg-K band (below).

$$P(x) = 1 - \exp\left[-\left(\frac{x}{\alpha}\right)^\beta\right] \quad (1)$$

where  $\alpha$  and  $\beta$  are respectively the scale and shape parameters and  $x$  is the breakdown field.  $\alpha$  and  $\beta$  were obtained by using the weighted least-squares regression technique. Since the estimated values obtained from a single experiment involving a finite number of specimens are unlikely to equal the true values of  $\alpha$  and  $\beta$ , the range of conceivable values for  $\alpha$  and  $\beta$ , given the experimental data, was obtained by computing the 90% confidence bounds using the procedure described in [15].



**Figure 2.** Experimental setup used for the dielectric breakdown measurements.

Lifetime measurements have been performed by applying square pulse voltages (duty cycle: 50%). The frequency has been adjusted from 1 to 20 kHz, the voltage at  $\pm 1$  kV. All measurements were performed at room temperature (*i.e.*:  $20^{\circ}\text{C}$ ; RH = 55%). The accuracy of the thermal chamber was about  $\pm 0.5^{\circ}\text{C}$ . The rise and fall times of the square voltage have been estimated to be about 30 and 50  $\text{kV}/\mu\text{s}$  respectively. During these tests, the level of applied voltages was always higher than the partial discharge inception voltage (PDIV) which has been estimated to be  $\pm 800$  V at  $20^{\circ}\text{C}$ . The experimental lifetime bench is shown in **Figure 3**.

Samples have been fixed on grounded stainless-steel plates. The square wave high voltage has been applied on stainless steel spheres (diameter: 4 mm) deposited on the sample surface (**Figure 4**). An automatic system detects any sample short-circuit, extracts the broken sample for the test bench and record its lifetime. Partial discharges take place between the spherical electrode and the sample surface. Submitted to ions bombardment, heat and oxidation mainly due to ozone formation, the sample surface erodes progressively until the sample final dielectric breakdown. As partial discharges appear during the rising and falling fronts of the square voltage, it is expected that the higher is the frequency, the lower is the lifetime.

Since partial discharges generate heat around the discharge area, all the samples have been put into a climatic chamber (at the left of **Figure 3**), rather than in a simple oven, in order to maintain the medium temperature at  $20^{\circ}\text{C}$ .

### 3. Results and Discussion

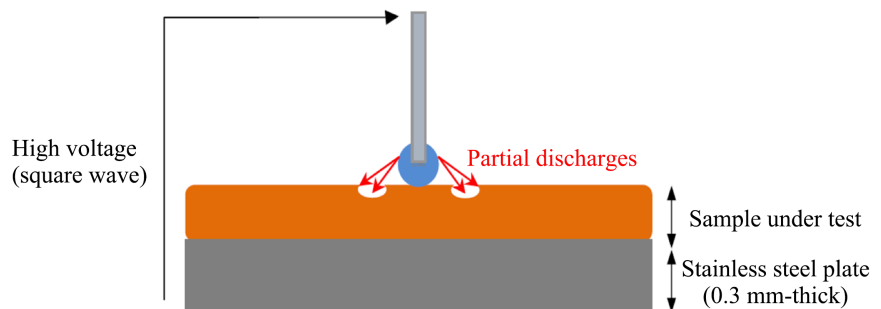
#### 3.1. Dielectric Strength

The dielectric strength results of both pure and nano-filled LDPE samples are reported in **Table 2**. The electrical field at breakdown is simply calculated by





**Figure 3.** Experimental test-bench used to measure the lifetime under a PD.



**Figure 4.** Cross-section of the samples for lifetime measurement under partial discharges.

**Table 2.** AC dielectric breakdown results.

	$\alpha$ (kV/mm)	$\beta$
<b>Pure LDPE, Mean thickness: 182 <math>\mu\text{m}</math></b>	101.8	10.5
<b>LDPE + 1% MgO, Mean thickness: 190 <math>\mu\text{m}</math>-thick</b>	108.4	9.0

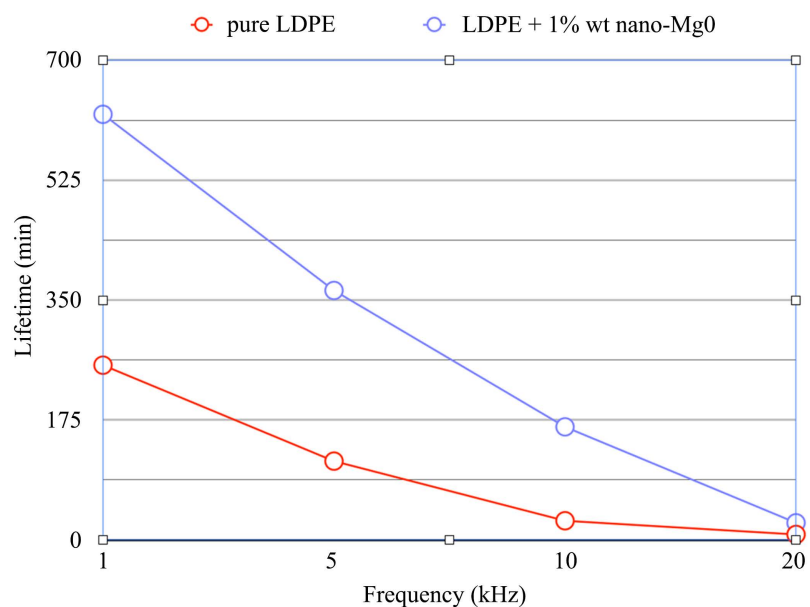
dividing the breakdown voltage by the sample thickness. This is an average breakdown field across the sample since the electrical field is not uniform due to the electrode geometry. For each breakdown test, the sample was examined in order to assess that the breakdown occurred through the sample and was not due to surface breakdown or flashover. Values reported in **Table 2** correspond to the Weibull's shape and scale parameters, obtained from measurements on 8 samples for each material. The slight improvement observed is in agreement with reports found in the literature where similar tests were reported [16] [17].

### 3.2. Resistance to Partial Discharges

The lifetime results of both pure and nano-filled LDPE samples are reported in **Figure 5**. Each dot in this figure corresponds to the Weibull's scale parameter obtained from 8 measured lifetime.

### 3.3. Discussion

The AC dielectric strength of pure and nano-filled LDPE measurements showed data very similar, *i.e.* a marginal difference of about 6% with an overlap of the confidence bounds. As expected, the lifetime data reported in **Figure 5** clearly shows that the inclusion of the nano-fillers improves the resistance to partial discharges. The lower the frequency (*i.e.*: the lower the number of PD per second), the higher was this enhancement of PD resistance. These results show that PE/MgO composites are potentially interesting material to be used for the insulation of high voltage AC cable with an improved resistance to local PD initiated in defects (*i.e.*: voids) that may appear during the LDPE insulator ageing. This is expected to lead to improved resistance to electrical treeing. Since PD is the main mechanism leading to the propagation of electrical trees in polymeric dielectrics, as well as much longer inception times. This behavior is also in good agreement with other reports in the literature on the dielectric properties of MgO-based nanocomposites. As an example, it was found that the lapse time to tracking failure increases with the content of nano-MgO in the 0 to 5 wt% range in the case of epoxy [18]. More complete reviews can be found in the literature on the improvement of erosion resistance in polymeric nanocomposites (see [1] [19] for example) that also are in good agreement with the observed trend in this report.



**Figure 5.** Lifetime (in minutes) of pure and nano-filled LDPE samples submitted to partial discharges under a square voltage versus frequency. Voltage:  $\pm 1$  kV,  $T = 20^\circ\text{C}$ .

## 4. Conclusion

The inclusion of a low content of nano-MgO into a LDPE matrix has been shown to improve the material dielectric properties, particularly the resistance to PD degradation without comprising other properties such as dielectric breakdown strength. This promising result will be supplemented by a more exhaustive investigation in order to optimize the dielectric performance of LDPE/MgO composites. The effect of filler content, the effect of particle size and of the processing parameters will be investigated and presented in a future report.

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## Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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