# From Laboratory to Industrial Scale: Comparison of Short- and Long-Term Dielectric Performance of Silica-Polypropylene Capacitor Films

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***Abstract*- In this paper a route from laboratory scale samples to full industrial scale production of biaxially oriented silica-PP nanocomposite capacitor films is for the first time shown and verified. Morphological and dielectric characteristics of laboratory scale produced and by full industrial tenter -process produced nanocomposite and neat-PP reference films are presented and compared. In addition to the short-term characteristics also ageing performance of the industrial films are studied. The results show closely comparative dielectric behavior of the films and thus verifies the successful up-scaling of the used nanocomposite recipe. Although certain amount of impurities and voids were observed in both nanocomposite films the low probability dielectric breakdown strength characteristics were considered promising. Modified trap distribution, decreased conductivity and improved dielectric loss behavior has been verified for the nanocomposite film while areas of further development are also recognized.**

## I. INTRODUCTION

Current trends in development of power grids towards future smart grids include a growing demand for sustainable production in distant regions and utilization of modern power electronics-based technologies for cost and landscape friendly power transmission and power quality. In practice this means often e.g. DC power transmission at different transmission voltage levels and voltage source converter (VSC) based technology for voltage conversion. Metallized film capacitors are one of the key components in this technology for which extreme reliability requirements are imposed. As DC capacitor banks form a large part of a converter stations both in volume and mass, development of new high energy density and high reliability capacitor dielectrics is highly desired.

Next generation capacitor and cable dielectrics are currently developed for HVDC use in European project GRIDABLE by nanostructuration of polypropylene base material. This paper focuses on development of capacitor dielectrics. For the first time the route from laboratory scale optimized biaxially oriented polypropylene (BOPP) sample films to industrial thin film nanocomposite prototype is shown. Project's and world first silica-BOPP prototype film is produced in full industrial production line. The main purpose of this paper is not to focus on the dielectric performance of the films where areas of further development are still recognized but to show the up-scalability of nanocomposite technology in the challenging production of capacitor thin film BOPP, which is one of the main objectives of the project.

## II. EXPERIMENTAL

### *A. Materials and film processing*

One silica-polypropylene (PP) nanocomposite BOPP film material and a neat PP reference material were produced, –both using a laboratory scale processing equipment as well as using an industrial BOPP film production line.

For nanocompounds a 600 kg masterbatch of capacitor grade isotactic polypropylene (PP) and 4.5 wt-% of hydrophobic fumed silica nanoparticles was at first manufactured using KraussMaffei Berstorff ZE 25 × 49D UTX compounder (screw speed 275 rpm, output 15 kg/h; nitrogen gas & a screen pack). The compounded strands were cooled in filtered water bath, pelletized and dried. For industrial film production the masterbatch compound was diluted by mixing with virgin PP to reach ~1 wt-% silica content, followed by cast film production and biaxial orientation in an industrial tenter line to 5.5 µm film thickness. For laboratory scale samples the masterbatch was diluted during extrusion of a ~350 µm cast film to 1 wt-% silica content. In the last phase 10 cm × 10 cm cast film specimens were biaxially stretched into ~7–8 µm thin films inside a semi-clean room using a Brückner KARO IV laboratory stretcher (temperature ~157 °C, stretch ratio 5.6 × 5.6). For neat PP reference films as identical processing as possible was utilized. Virgin PP 'masterbatch' was made similarly with the same compounder, the batch was mixed with virgin PP and finally cast film and orientation was carried out at the industrial BOPP film line to produce a 5.5 µm industrial reference film. Also laboratory scale reference film samples were produced, similarly as described above in case of silica-BOPP samples.

### *B. Dielectric breakdown strength characterization*

Breakdown strength characterization of the films were carried out using a method [1] based on large-area film samples and self-healing breakdown events. Test samples with an active area of 81 $cm^2$ were constructed by sandwiching commercial metallized BOPP electrode films on both sides of the sample film, the metallization facing towards sample film. DC test voltage was ramped up in atmospheric

air conditions utilizing the slow-rate-of-rise method of IEC 60243-1. Totally ~490 $cm^2$ of film was measured for each breakdown distribution. Analysis of the multi-breakdown data sets were done in MATLAB as detailed in [1].

### *C. Dielectric spectroscopy, thermally stimulated depolarization current and absorption current measurements*

For these measurements electrodes consisting of 10 nm Ni and 100 nm Au layers were deposited on both sides of the thin film samples using a e-beam evaporator (Instrumentti Mattila) where high vacuum ($<1\times10^{-6}$ mbar) and low deposition rate (0.05–0.20 nm/s) were utilized to minimize thermal and radiative stress on film sample. After evaporation the samples were short-circuited and stored in desiccator in vacuum several days prior to the measurements.

Dielectric spectroscopy was carried out using Novocontrol Alpha-A dielectric analyzer equipped with Novocool temperature control system. Thermally stimulated depolarization current (TSDC) measurement comprised of a Keihley 2290E-5 DC source, Keithley 6517B electrometer and the Novocool temperature control system. For absorption current measurements the same DC source and electrometer were utilized. Shielded sample cell (Novocontrol BDS 1200) was used in all measurements. Temperature control system (accuracy ±0.1 °C) was based on Novocool system or PID controlled resistive heating element (for absorption current measurements).

TSDC measurement procedure was: (i) polarization (40 min @ 100 $V_{DC}$/µm, 80 °C), (ii) rapid cooling to −50 °C (hold isothermally for 5 min), (iii) removal of the poling voltage and short-circuiting of the sample (hold isothermally for 3 min), (iv) linear heating at 3.0 °C/min up to 130 °C while recording the depolarization current. Absorption current measurements were conducted under 100 $V_{DC}$/µm field while holding the sample isothermally at +100 °C. The currents were recorded for 24 h, followed by 6 h measurement of depolarization current.

### *D. Ageing tests*

For long-term performance evaluation of the industrial films a 1000 h ageing experiment was conducted. Ageing was conducted at stable 200 $V_{DC}$/µm, +65 °C conditions under inert $N_2$ ambient, utilizing self-healing electrodes and a total tested film area of ~3200 $cm^2$ per film type. The test arrangements are detailed in [2]. Effects of electro-thermal ageing were analyzed by the characterizations described in sections B and C, made for the films before and after ageing. In addition to the electro-thermally aged samples a set of samples were aged only isothermally under +65 °C stress.

## III. RESULTS

### *A. Film structure and morphology*

Basic properties of the studied laboratory- and industrial-scale produced BOPP films are presented in Table I. Based on DSC experiments, the BOPP films exhibited typical α-form PP crystallinity, showing α-form peak melting temperature of ~169 °C and degree of crystallinity in the range of 47–54 %. Morphology of the BOPP films was studied by both scanning electron microscopy (SEM) and optical microscopy (OM). The industrially produced unfilled reference film (PP-ref) exhibited structural characteristics similar to those of typical commercial capacitor-grade BOPP films, with shallow "crater-like" surface texture being apparent by optical microscope. No signs of inhomogeneities were observed in the unfilled films. Similar film structure was also observed for the industrially produced nanocomposite film (PP-$SiO_2$), however, a small amount of voids and agglomerates were observed (Fig. 1). Very similar agglomerates were found also in the laboratory scale nanocomposite film. Real filler content of the PP-$SiO_2$ film was measured by 'ash method' to be 0.72 wt-%, being thus slightly below the target value (1 wt-%). Realized silica dispersion and particle size distribution were determined by analyzing over 80 SEM pictures of the masterbatch compound, the mean size being ~104 nm and the mode ~57 nm. Silica was reasonably well dispersed and distributed, forming mostly small aggregates but scarcely also larger agglomerates were observed.

TABLE I
Tested BOPP films

| Code | Description | Thickness | Crystallinity |
|---|---|---|---|
| PP-$SiO_2$ | Industrial nanocomposite film | 5.5 µm | 49.0 % |
| PP-ref | Industrial reference film | 5.5 µm | 54.2 % |
| PP-$SiO_2$-lab | Laboratory nanocomposite film | 7-8 µm | 48.9 % |
| PP-ref-lab | Laboratory reference film | 7-8 µm | 47.1 % |

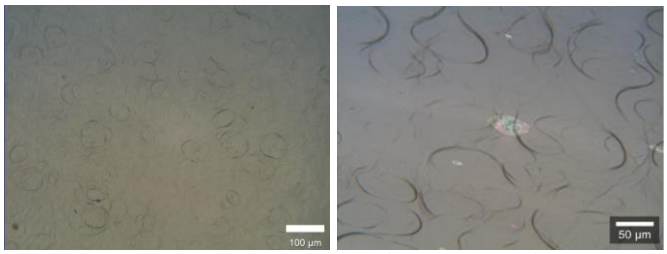

Fig. 1. Optical microscopy (OM) pictures of PP-$SiO_2$-lab (left) and PP-$SiO_2$ films. Cluster of impurities + void visible in the industrially produced film

### *B. Dielectric breakdown strength*

Short-term large-area dielectric breakdown strength (DBS) characteristics of the studied films are given in Fig. 2 together with the main Weibull distribution parameters. For the industrially produced films the high temperature performance at +80 °C is also shown. In addition, breakdown distributions of two commercial BOPP film samples (thickness 5.0 µm and 5.5 µm) based on the same base PP grade are also given to show typical performance range of comparable commercial products.

The overall DBS performance objectives of the project were not yet fully achieved in this first industrial scale production trial. However, promising signs towards the main objective, to achieve a clear improvement especially in the low probability breakdown performance region, may be observed. Improved scatter (Wbl β) is seen for both nanocomposite films and no weak points were observed especially in the industrially produced nanofilm. This is a very positive result especially considering the notable amount of impurities and voids in both nanocomposite films. The

addition of $SiO_2$ nanoparticles in the PP matrix thus seemed to be able to improve the breakdown strength of the film points with non-optimal morphology. From engineering point of view this is more valuable than improvements in the high probability region, even in the self-healing capacitor designs. At +80 °C both films exhibited a slightly higher decrease in DBS than what was expected (~25–30 % at 5–10 % probability regime) although low scatter and practically non-existing weak-point population can also be observed.

The performance of the laboratory scale and industrial scale films were closely similar indicating both successful upscaling and high-quality laboratory production of films. As expected, the industrial scale film production yielded to slightly improved film performance compared to the laboratory scale films.

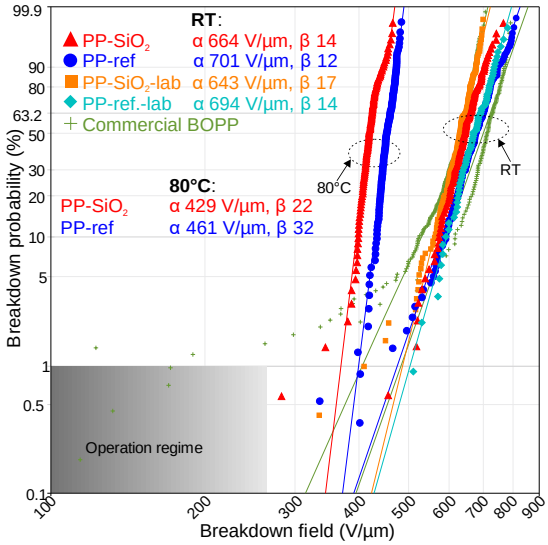


Fig. 2. Dielectric breakdown strength distributions of the experimental films at room temperature (RT) and for the industrially produced films also at +80 °C. Confidence bounds omitted for clarity reasons.

### *C. Conductivity, dielectric loss and charge trapping properties*

Thermally stimulated depolarization current measurement technique was used to study the charge trapping properties of the films (Fig. 3). For all the films TSDC peaks were observed around/above the glass transition temperature $T_g$ (Fig. 3, inset) and above ~90 °C, indicative of the presence of shallow and deep traps, respectively. In case of both nanocomposite films the signal above the $T_g$ is more pronounced compared to the non-filled reference films, indicating slightly higher presence of shallow traps (trap depths ~0.75–0.9 eV range, based on the MTSDC analysis according to [3]). In addition, the deep trap related TSDC peak is greatly reduced in nanocomposite films indicating reduced density and/or more uniform distribution of deep traps (~1.08–1.1 eV). Both changes are typically observed for nanocomposites as well as the shift to deeper trap levels (higher temperature) observable for PP-$SiO_2$-lab film [4].

Absorption current characteristics of the films during isothermal charging at +100 °C for 24 h together with the

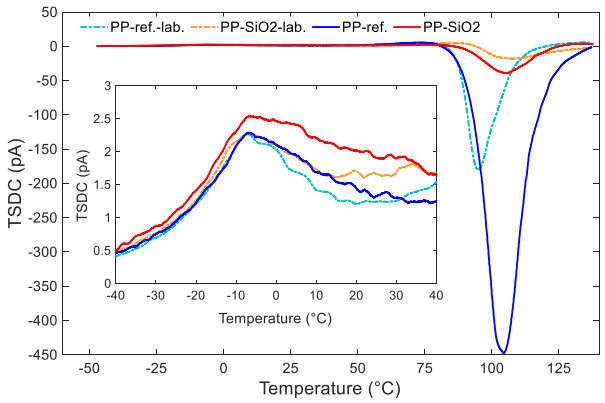


Fig. 3. Thermally stimulated depolarization currents of the studied laboratory and industrial scale produced films. Enlargement of the shallow trap region is given in the inset.

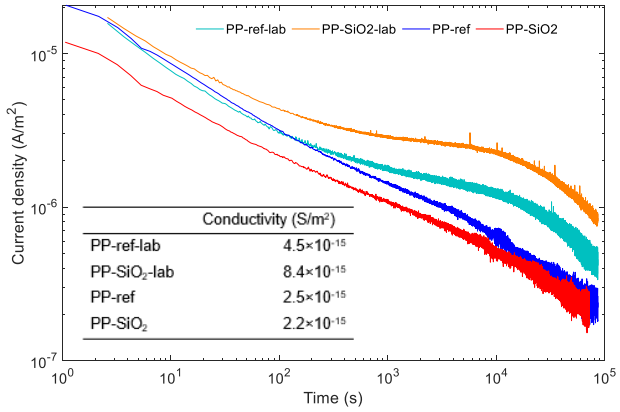


Fig. 4. Current density of the films during 24 h isothermal charging at +100 °C. Conductivity estimates are based on the measured current densities at the end of the measurement period.

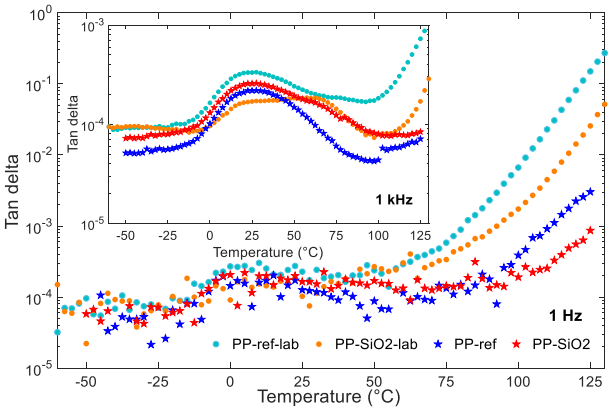


Fig. 5. Dielectric loss (tan δ) behavior of the studied laboratory and industrial scale produced films as a function of temperature at 1 Hz and at 1000 Hz (inset).

conductivity estimates based on the current densities at the end of measuring period are given in Fig. 4. Despite the slightly increased density of shallow traps the industrially produced PP-$SiO_2$ film shows lower conductivity and charging compared to the non-filled reference (under studied stress level), a behavior typically measured for nanocomposite films [4]. Correspondingly, the 1 Hz dielectric losses are lower at high temperatures in the PP-$SiO_2$ films compared to their references, also indicative of the ability of

nanofilled BOPP to suppress charge build up. For the laboratory scale films a non-typical performance was measured. A very slow charging phenomenon can be observed in both films and a slightly higher conductivity was estimated for the nanofilled film. The reason for this will be studied further later.

*D. Ageing performance*

Ageing performance of the industrially produced films were studied by the 1000 h electro-thermal ageing experiment, utilizing combined 200 $V_{DC}$/µm and +65 °C stress level. A notable decrease in the DBS performance was observed (Fig. 6) both in $SiO_2$-PP and in the neat-PP reference film. Although the decrease is slightly more pronounced in the $SiO_2$-PP film, the change is astonishingly similar in the reference film. Separate film samples were also aged only isothermally. DBS distribution measured for the thermally aged $SiO_2$-PP was closely identical to that of the electro-thermally aged sample over the whole distribution. For thermally aged reference film DBS decrease was observed only below ~10 % region being there closely identical to the performance of electro-thermally aged film. Thermal ageing seemed to be the predominant ageing mechanism, -for reference film in localized spots while for $SiO_2$-PP film more comprehensively.

Ageing induced changes were measured also in the other measured dielectric parameters. Dielectric losses increased in both films (Fig. 7, left), most notably above 25 °C (1 Hz) being maximally at the level of ~$10^{-3}$ over typical operating temperatures. The increased loss levels most probably originate from the changes in trap states, especially from the increases in the shallow trap density of states (Fig. 7, right). DC conductivity of the aged films (@ 100V/µm, +100 °C) did not increase over the ageing at the measured stress level.

Electro-thermal ageing of the films was studied also by testing elements (96 µF) wound of the same films (225 $V_{DC}$/µm, 65 °C, 1000 h). Results were promising as no measurable capacitance decreases were noticed over the test.

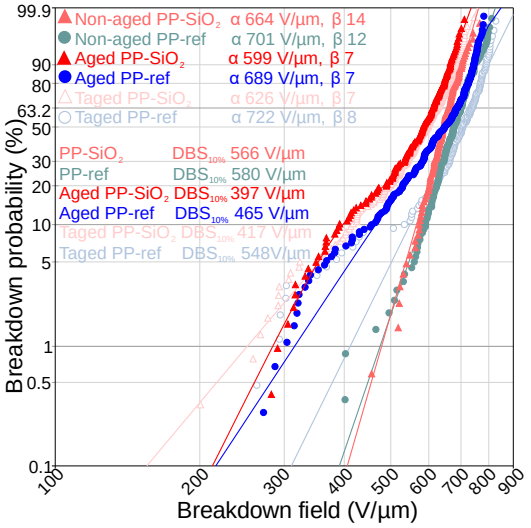


Fig. 6. Dielectric breakdown strength distributions of the electro-thermally and only thermally aged ("Taged") industrially produced $SiO_2$-BOPP and neat-BOPP films before and after the 1000 h ageing.

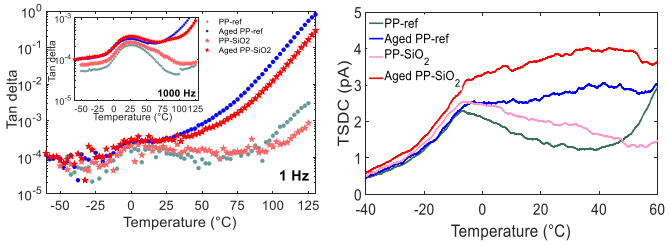


Fig. 7. Dielectric loss (left) and TSDC current at shallow trap region for the industrial films before and after electro-thermal ageing.

## IV. CONCLUSIONS

A route from laboratory samples to full industrial silica-BOPP nanocomposite film prototype for capacitor insulation has for the first time been verified. Dielectric and morphological characteristics of the comparable laboratory and industrial silica-BOPP films were close to each other, the mass production ensuring typically a slightly improved performance. Successful production of several tons batch of only 5.5 µm thick silica-BOPP film and further winding it to capacitor elements verifies good processability of the used silica-PP recipe. However, certain level impurities and voids, originating from laboratory scale compounding process, were observed in both the laboratory and industrial nanocomposite films.

Dielectric breakdown strength characteristics of the $SiO_2$-BOPP film were still on the range of comparable commercial products. However, signs of targeted improvements at the low probability DBS region were observable at room temperature conditions despite the impurity and void problem and lower than planned silica content. Modified structure of trap states and decreased dielectric losses, charging current and conductivity were measured for the nanocomposite. In addition to impurities and voids, challenges were recognized also in the long-term performance while in general this first up-scaling trial may be considered as a successful step towards further developments in the project.

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