

# Polymers in Film Capacitors - The Next Generation Material is available!

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## Abstract

Polypropylene is the polymer of choice for most film capacitors, but there is an inherent high temperature limit for its usage. New polymer materials are therefore required to overcome these temperature limitations. Accordingly, a new class of engineering materials, EPN (Ethylene-Propylene-Norbornene), has been developed for capacitor films, combining the advantages of polypropylene and cyclic olefin copolymers. This new material class can represent a breakthrough on the design of film capacitors for high temperatures.

## 1 Introduction

Over the history of film capacitors, from a material perspective, the major breakthrough started with the move from paper to polymers, and especially to polypropylene, which finally became the dominant dielectric in film capacitors today. While polypropylene was not the first polymer used for film capacitors, it is now the material of choice for most applications. Here, we summarize the advantages of polypropylene vs. other polymers [1, 2].

### 1.1 Polypropylene in Film Capacitors

The reason why film capacitors can be used in applications with high electrical fields is that they exhibit a “graceful failure”, provided by the self-healing mechanism [1].

The self-healing capability depends on the composition of the polymer, in particular the ratio of carbon to other elements such as oxygen, hydrogen, etc. A low carbon ratio is associated with excellent self-healing performance because the graphite deposition is minimized [2]. This is the case for polypropylene, which features the best self-healing ability of non-cellulosic polymers [1].

Other well-known advantages of polypropylene are its low dissipation factor, low leakage current and high breakdown strength.

In addition to these technical features, the key contributors to the unrivalled success of polypropylene films so far are:

- the availability of film processing technology, which allows its production on an industrial scale.
- the ability to be processed to very thin films (downgauging) in order to achieve a high volume efficiency in the capacitor, while keeping adequate tensile strength. Polypropylene films down to about 1.9  $\mu\text{m}$  are commercially available.

Over time, a tailor made technology evolved for the production of capacitor-grade polypropylene resins. These are now produced in special polymerization plants where catalyst residues from the polymerization process are deactivated and removed from the polymer [3].

Additionally, in contrast to other polypropylene grades for film applications (e.g. packaging films), the manufacturers of capacitor grade polypropylenes optimise the additive content as these have a detrimental impact on the electrical performance. This leads to ultra-high purity polypropylene resins, characterized by an extremely low ash content after polymer combustion [3].

During the early 2000's, an improved polypropylene polymer was obtained by increasing the orientation of the methyl groups (isotacticity), plus a tailored polymer weight distribution. This allowed the production of thinner films in parallel with higher temperature performance; the temperature rating for capacitors could be increased from 85°C to 105°C which had major implications for the design of capacitors [4]. Despite the advantageous properties described above, the thermal-electrical behaviour of polypropylene deteriorates drastically above 105°C, defining an inherent high temperature limit for its usage in film capacitors [5]. To overcome this temperature limitation, new polymer materials are required.

### 1.2 Cycloolefin Copolymers for High Temperature Film Capacitors

In the 1990's, an alternative class of polymers, cyclic olefins copolymer (COC), was evaluated as new promising dielectric materials [6].

Chemically, COCs are polyolefins, as they are solely composed of carbon and hydrogen. COCs are derived from the monomers ethylene and 2-norbornene. 2-norbornene is produced from ethylene and dicyclopentadiene, both of which are common raw materials in the chemical industry.

The polymerization of the monomers in a solution process allows the separation of residual catalyst via a filtration step. Thus COCs comply with the high purity prerequisite for usage in film capacitors - just like polypropylene.

In contrast to other polyolefins such as polyethylene and polypropylene, which are semicrystalline polymers characterized by a melting point, COCs are amorphous materials that are defined thermally through a glass transition temperature ( $T_g$ ). COCs will only start softening when the temperature approaches  $T_g$ . Since COCs with  $T_g$  up to to 200°C can be designed by the appropriate fraction of 2-norbornene in the polymer, they are a good high-temperature polymer candidate.

As COCs are non polar, the dielectric loss factor is very low and remains constant to a temperature just below  $T_g$ ; similarly, the dielectric constant is very stable up to  $T_g$  [6].

However, COCs are quite brittle materials and therefore hard to process on existing BOPP-film lines. Therefore, in spite of these promising properties, the challenges to process these materials to thin films on an industrial scale have prevented commercialization [7].

### 1.3 From COC/PP blends to EPN: A New Class of Materials

It has been shown that COCs are able to form heterogeneous blends with PP without the need of a compatibilizer, improving the thermal and mechanical properties compared with pure PP [8]. COC/PP blends showed processability into thin capacitor films under state-of-the-art process technology used for polypropylene, both in sequential and in simultaneous film lines [9]. Results from several performance evaluations for PP/COC blends confirmed better thermal performance than PP [9, 10, 11].

Based on that experience, Borealis and TOPAS Advanced Polymers have developed Stelora™ EPN (Ethylene-Propylene-Norbornene) polymers, for industrial-scale production of dielectric films. EPNs combine the advantages of linear and cyclic olefins in a single resin, representing a new class of engineering materials for capacitor films. In line with growing sustainability requirements, Stelora™ EPN for capacitor films is mostly based on renewable feedstock.

Herein we present some basic polymer features of the EPN system and results from testing at film and capacitor level.

## 2 Samples

An EPN resin manufactured by Borealis has been transformed into cast or biaxially oriented (BO) films:

- 200 µm EPN cast films have been produced through a SML Cast Coex line at Borealis.
- 6 µm BOEPN films have been produced in a sequential tenter line at Tervakoski Films Group in Slovakia. Then these films have been metallised at Tervakoski and at TDK facilities, and finally converted into capacitors in TDK (capacitor characteristics are described in Table 1).

State-of-the-art machinery has been used all along these processes.

For comparison, a commercially available 6 µm BOPP film based on a highly isotactic PP grade manufactured by Borealis have also been tested.

**Table 1** Capacitor characteristics

Film metallisation	Al and/or Zn film met
Winding element	1 & 27 µF, flat pressed
Terminals	Tinned Cu, radial
Casing	PBT box + epoxy resin

### 3 Experimental

At Borealis laboratories:

Atomic Force Microscopy (AFM) images were collected with an Oxford Instrument Cypher microscope, equipped with Olympus AC160TS cantilevers and operated in non-contact mode.

Differential Scanning Calorimetry (DSC) was performed with a TA Instruments Q2000 calibrated with indium and operated under nitrogen atmosphere with a flow of 50 mL/min. The first heating and cooling steps were performed as in a standard DSC with maximum temperature of 225°C, 10 minutes dwell time, and heating and cooling rates of 10°C/min.

High temperature optical microscopy was performed with an Olympus BX50 microscope equipped with a LINKAM THMS 600 (Hot stage) operated under nitrogen flow.

At TDK laboratories:

Film shrinkage was measured on square samples with 120 mm side, after 15 min. exposure to temperature.

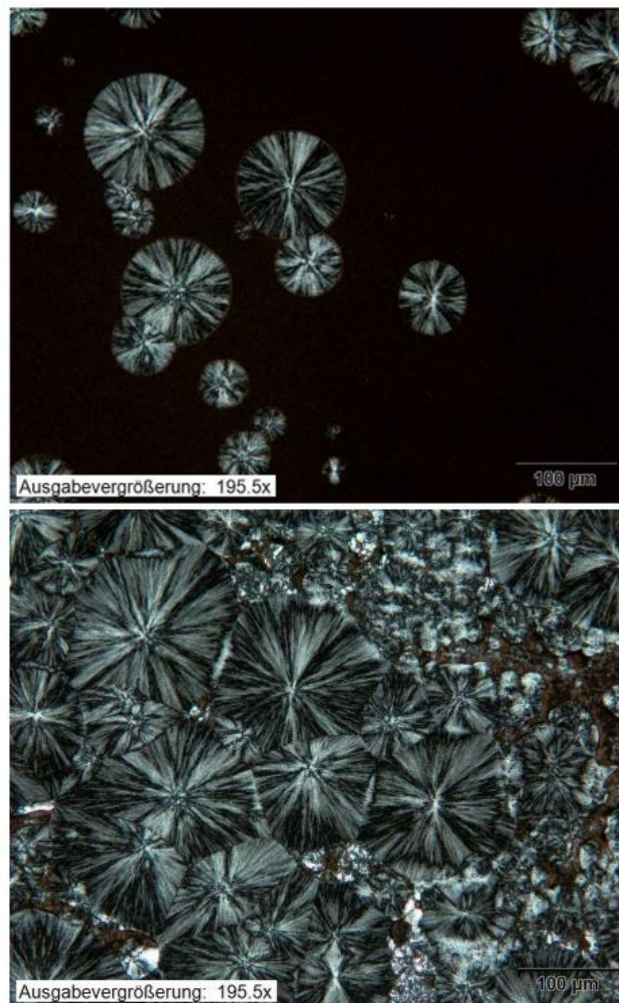
Electrical Conductivity (EC) under DC field and DC Breakdown Strength (BS) were measured in air, inside an oven, using a Chroma 19057 Hipot Analyzer. In BS, the DC voltage was raised at a rate of 100 V/s until breakdown; in EC, the voltage was kept for 2 min. The film samples were placed between an aluminium foil and a plane electrode with rounded edges, 25 mm in diameter. For BS, one electrode was used at a time, in EC 15 electrodes were connected in parallel.

Long-endurance tests (LETs) were carried out on six to ten capacitors per group, in ovens applying ambient temperatures of 125°C and above, and under constant DC voltages applied by FUG HCK power supplies. The typical duration for the LETs is a few thousand hours, with intermediate measurements of the electrical parameters (capacitance, loss factor  $\tan\delta$ , and insulation resistance Riso) every 250 hours.

### 4 Polymer Properties

Recrystallization from the melt of EPN pellets under a heating stage optical microscope was performed. EPN pellets were melted at 225°C and cooled at -3°C/min until fully recrystallized.

Figure 1a shows an intermediate stage, i.e. at 121°C, where spherulites have been nucleated and are growing. Fig. 1b features the final stage of



**Fig. 1** Cross section of an EPN pellet under a microscope cooled from 225° to 117°C at -3°C/min. a) intermediate b) final.

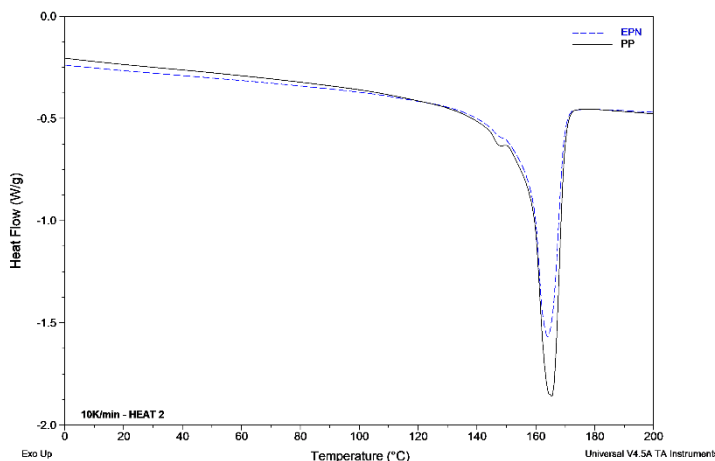
the crystallization, where spherulites are fully grown.

Rapid growth of  $\alpha$  crystallites followed by crystallisation in  $\beta$  crystals at lower temperature is observed. Interesting to note is that the space is filled by spherulites in the typical  $\alpha$  phase crystalline form of iPP, but also by areas with high birefringence typical of the  $\beta$  phase form.

This crystallisation behaviour of EPNs is comparable to the crystallisation behaviour of iPP with highest purity.

The fundamental crystallization behaviour of the multicomponent EPN system is not different to polypropylene. The DSC measurements of PP compared to a typical EPN material reveals that the melting temperature is not affected due to composition change from iPP to EPN. Only the heat of fusion to melt the EPN material as compared to the iPP reference is somewhat reduced (Fig. 2).

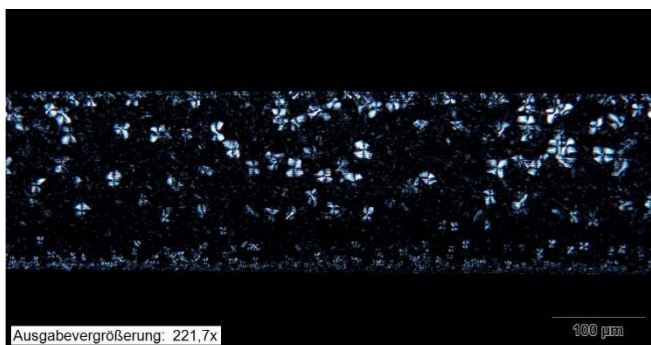




**Fig. 2** DSC of iPP (Borealis Borclean HC300BF) vs. a typical EPN.

In order to understand the steps involved during the film making of a BOEPN film, an interim model cast film was produced and evaluated with a microscope (Fig. 3)

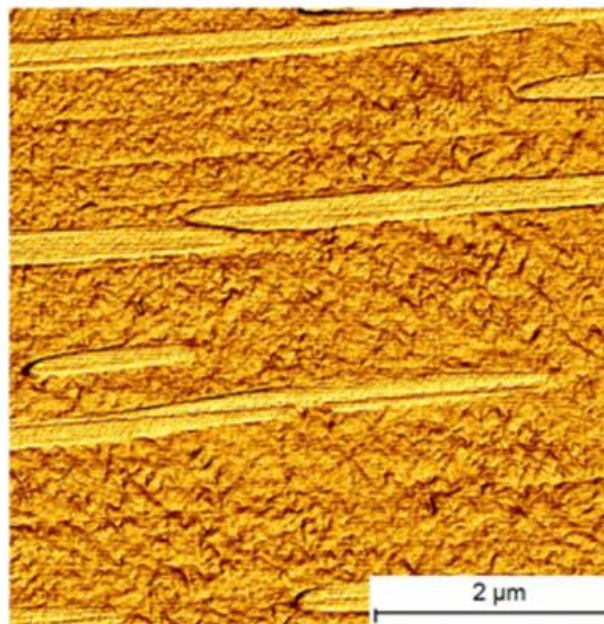
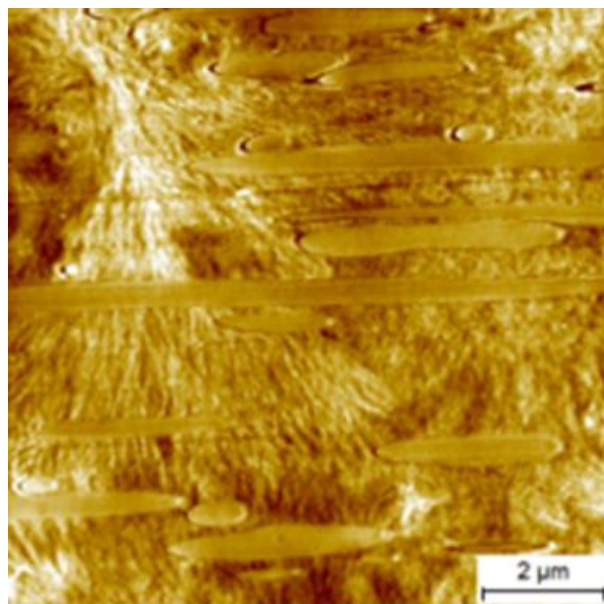
The cast film features prominent  $\beta$ -crystals of PP, visible as white regions in Fig. 3. The formation of  $\beta$ -crystals is a well-known characteristic of the manufacturing of BOPP films for capacitors and is controlled by film producers via temperature setting of the chill roll of the BOPP line [12].



**Fig. 3** EPN derived cast film: microscopy of the cross section; under the conditions  $\beta$ -crystals are visible as bright areas.

This  $\beta$ -phase formation is pre-requisite to obtain a surface of desired roughness in the stretching step. Roughness is fundamental to help processability of the film in subsequent processes such as metallisation, slitting and winding [12]. Furthermore, a certain surface roughness is required so that self-healing develops safely.

Additionally, this intrinsic path of formation of surface roughness during the film processing prevents the usage of additives in the resin, which would have a detrimental impact on the electrical behaviour.



**Fig. 4** AFM images of an EPN-based cast film section along machine direction a) AFM surface scan b) Imaged AFM.

We used an AFM to determine the topography of the section on a cast film cut by a microtome. The scanning of the section reveals details on a much smaller scale than the inspection with an optical microscope. Here, discrete domains dispersed as small layers in a matrix can be observed. The topography picture even shows a crystallite growing around several heterophasic domains (Fig. 4a).

Via advanced AFM methods, the formation of discrete heterophasic domains in the cast film can be better visualized. Layered domains of only a few microns in length can be detected in the matrix (Fig. 4b). It is probable that this layer structure is

maintained after stretching and still present in the final biaxial stretched film.

### 5 Results on Films

The film shrinkage of 6  $\mu\text{m}$  BOEPN and BOPP films at 120°C and 140°C is compared in Figure 5. BOEPN films feature lower shrinkages at high temperatures, and particularly it is nearly zero in the Transverse direction (the most critical direction in capacitor film processing), as an excess shrinkage may deteriorate the critical joint between the end sprayed layer and the film metallisation layer [13].

In Figure 6, the electrical conductivity (EC) of BOPP and BOEPN films are compared. The BOEPN film shows a conductivity nearly one order of magnitude lower than the BOPP film.

The results of the breakdown strength (BS) are shown in Fig. 7. At 125°C, the BOEPN film shows a 70  $\text{V}/\mu\text{m}$  higher characteristic breakdown strength than the BOPP film.

The relationship between these two advantageous properties (low EC and high BS) at such high temperatures is as follows [14]: a breakdown under DC is normally a thermal runaway process caused by the positive feedback resulting from the combination of temperature-dependent conductivity and field-dependent conductivity. Above a certain voltage, the rate of heat generation in the dielectric is higher than the rate of heat dissipation to the surrounding environment, which results in thermal runaway.

### 6 Results on Capacitors

Figure 8 shows the results of a LET at 125°C on capacitors based on BOEPN and BOPP films. A field of 125  $\text{V}/\mu\text{m}$  has been applied. The reference PP-based capacitors show considerable ageing by a high degree of self-healing activity, manifested as loss of capacitance, a drastic increase of  $\tan\delta$  and collapsing Riso (i.e. surge of leakage current, thus risking a thermal runaway of the dielectric). In contrast, the EPN-based capacitors only show a gradual increase of  $\tan\delta$  that can be attributed to electrode oxidation.

Figure 9 shows a LET at 135°C on capacitors based on EPN films. A field of 105  $\text{V}/\mu\text{m}$  has been applied. The evolutions remain approximately flat, indicating minimal ageing.

The much lower extent of dielectric ageing of EPNs at high temperature is related to the low leakage current, already demonstrated in blend-based capacitors [11] and now similarly present in

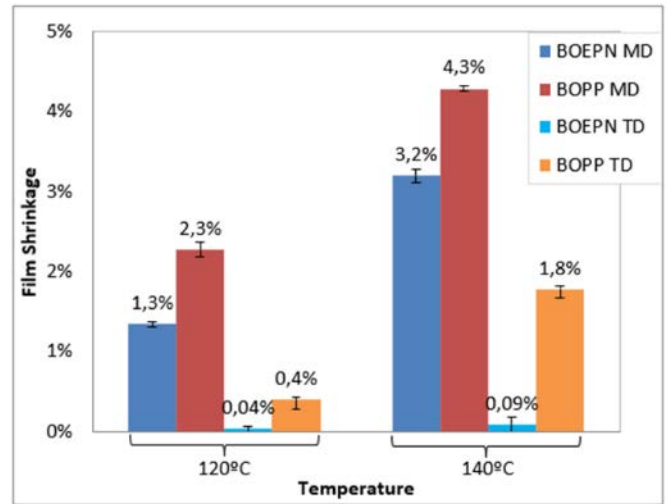


Fig. 5 Shrinkage of BOEPN and BOPP films in Machine (MD) and Transverse (TD) directions after 15' at 120°C and 140°C.

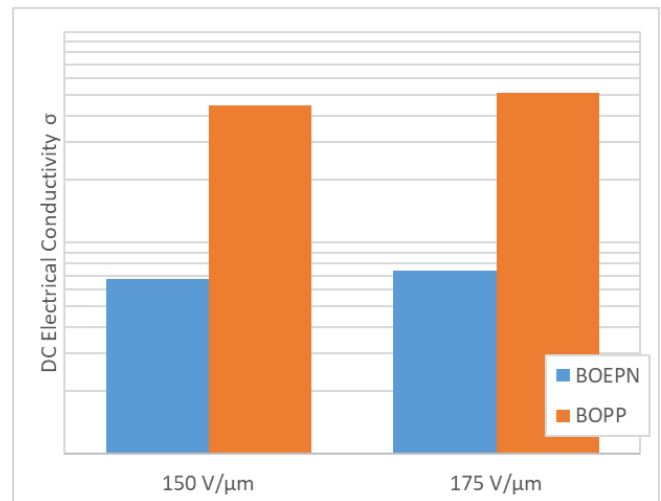


Fig. 6 Electrical conductivity of films in DC at 125°C, under fields of 150 and 175  $\text{V}/\mu\text{m}$ .

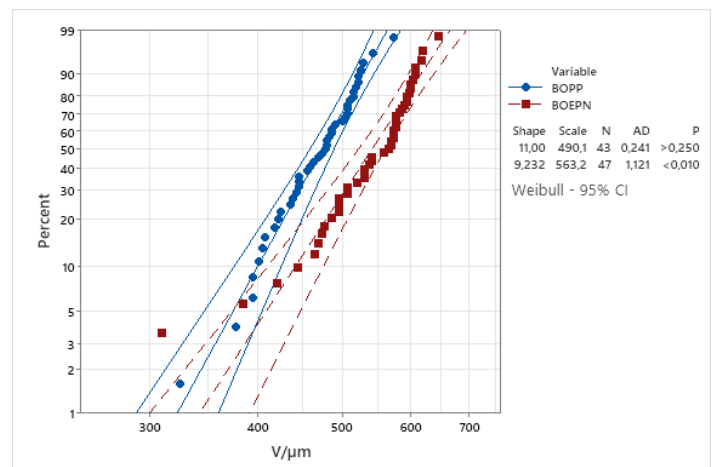
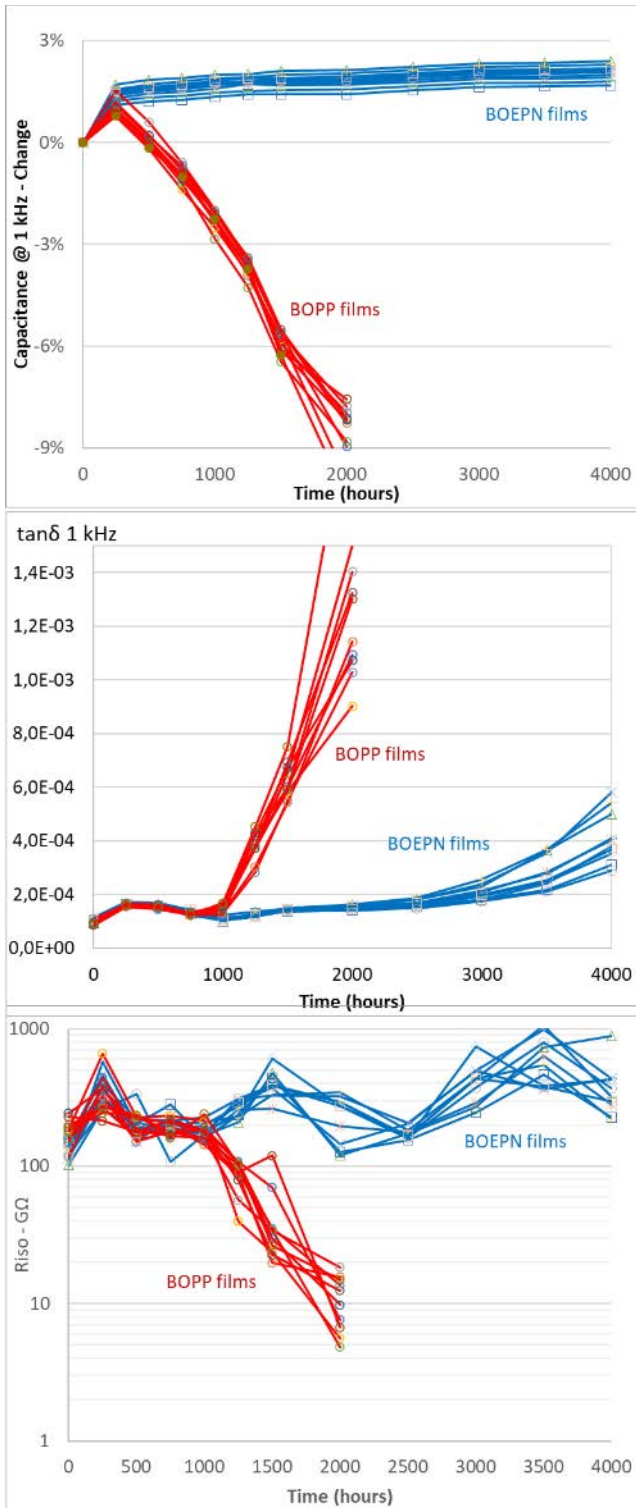
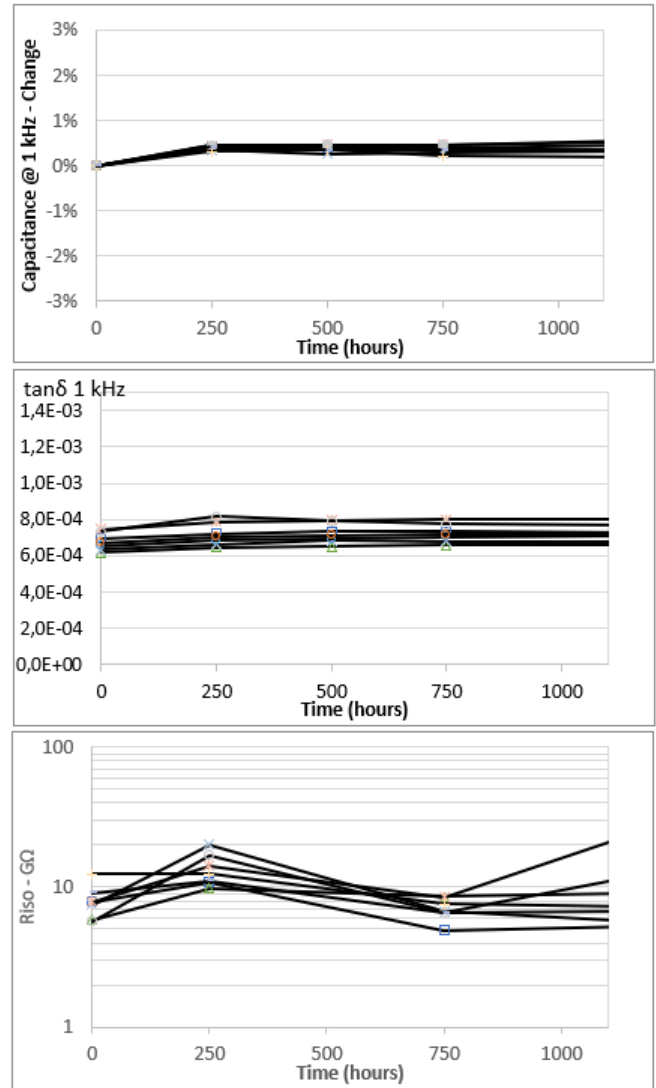


Fig. 7 Weibull probability plot of breakdown strength of BOEPN and BOPP films at 125°C.



**Fig. 8** Evolution of electrical characteristics of BOEPN-based and BOPP-based capacitors (1  $\mu$ F, 10 pcs./group) during a LET at 125°C: a) change of capacitance at 1kHz b)  $\tan\delta$  at 1 kHz c) insulation resistance after 10" at 500V.

EPN-based ones, as can be deduced from the lower film DC conductivity.

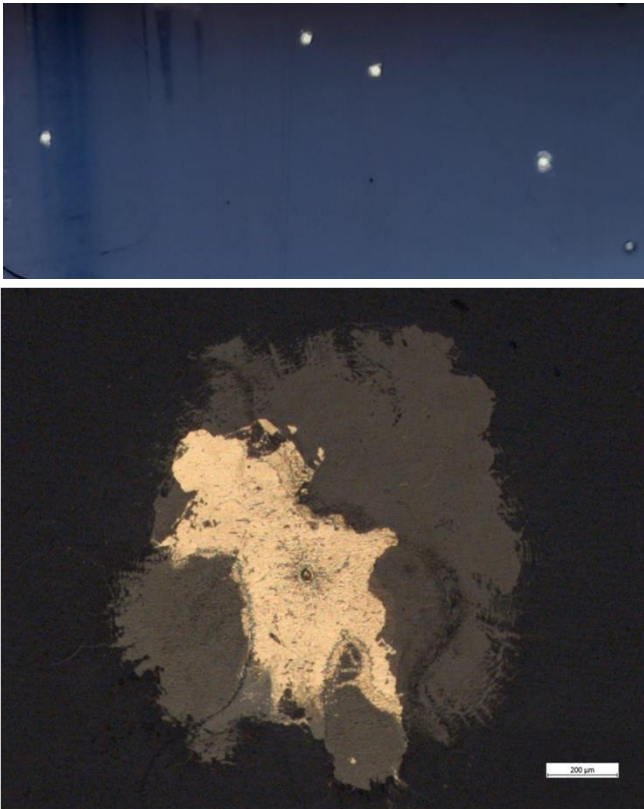


**Fig. 9** Evolution of electrical characteristics of BOEPN-based capacitors (27  $\mu$ F, 7 pcs.) during a LET at 135°C: a) change of capacitance at 1kHz b)  $\tan\delta$  at 1 kHz c) insulation resistance after 15" at 500V.

The analysis of films extracted from capacitors after LET shows that the self-healing ability is kept even under high temperature operation, as seen on Fig. 10. This confirms that EPN-based capacitors present good self-healing characteristics, as was expected from two inherent properties: a) composition (the carbon ratio of EPN is only slightly higher than that of PP), and b) adequate surface characteristics (provided by the  $\beta$ -phase of PP). Therefore, the self-healing mechanisms described in [2] are also applicable to this class of polymer.

Based on a substantial number of DC LETs, such as those shown in Figs. 8-9, a curve for the de-rating of DC field vs. temperature has been defined for capacitors based on the EPN dielectric.





**Fig. 10** Views of two overlapping BOEPN films extracted from a capacitor after a 2.500-hour LET at 130°C. a) General view showing self-healings (white fuzzy areas). b) Detail view of a self-healing.

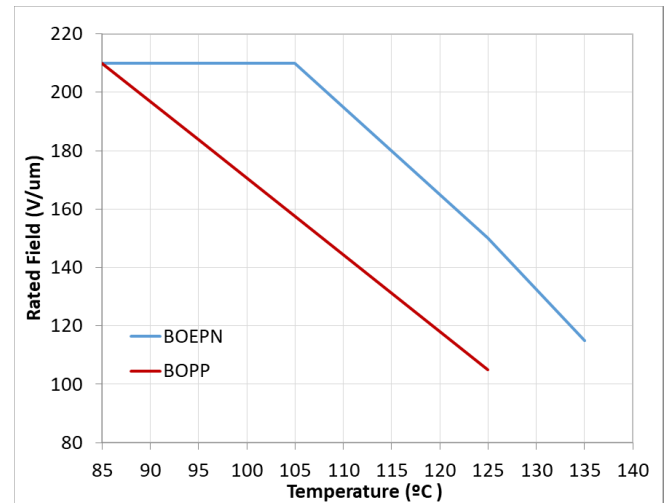
The curve is shown in Fig. 11 in comparison with a typical curve for BOPP-based capacitors. The curves demonstrate that above 85°C, EPNs allow a considerable increase in the operating fields vs. PP-based capacitors, and can operate up to 135°C. Conversely, for a given operating field and temperature, thinner BOEPN films can be used and thus the capacitor size can be reduced.

## 7 Conclusions and Outlook

We have demonstrated that as a polymer, EPNs present the similar fundamental crystallization behaviour of PP, featuring the  $\beta$ -phase of PP. As a result, EPNs can be processed in state-of-the-art stretching lines for PP capacitor films, and develops operation-related characteristics such as surface roughness which plays a role in self-healing ability.

Compared to BOPP, BOEPN films present lower shrinkage, lower DC conductivity and higher breakdown strength.

EPN-based capacitors present a low degree of dielectric ageing under temperatures as high as



**Fig. 11** Curves of DC field derating vs. temperature for the BOEPN capacitors vs. BOPP capacitors.

135°C. Operation under higher temperatures will be checked in future investigations.

EPN-based capacitors present good self-healing abilities even under high temperature operation.

Overall, this new material class improves the current temperature ratings compared to PP in capacitors by at least 20°C.

EPNs have been designed to allow repetitive production in industrial scale. The availability of the polymer resin in industrial volumes, the ability to produce thin films commercially with existing technology, as well as the fit to typical film capacitor designs, make EPNs a breakthrough to allow film capacitors to achieve higher operating temperatures in a straightforward manner.

Based on these characteristics, the new material class is expected to have a major impact on the design and usage of film capacitors, facilitating the transition to wide band gap technologies based on SiC or GaN.

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