



# Global Journal of Engineering Science and Research Management

## DRASTIC CHANGE IN DIELECTRIC BREAKDOWN STRENGTH OF OIL SATURATED POLYMER FILM WITH TEMPERATURE

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DOI: 10.5281/zenodo.3766188

**KEYWORDS:** Capacitor; Metallized polypropylene foil; Breakdown strength; Oil impregnation.

### ABSTRACT

The ever-growing need for high-energy density and high operation temperature capacitive energy storage for next-generation applications has necessitated research and development on new dielectric materials for film capacitors. Consequently, various new approaches offering unique ways to tailor dielectric properties of polymers have recently emerged, and new materials such as dielectric polymer nanocomposites (PNC) are envisioned as potential next generation dielectrics. Establishment of optimized formulation and processing conventions is however necessary in order to achieve improvement in dielectric breakdown properties. Importantly however, such material development puts dielectric breakdown strength assessment of polymer films in a central role in guiding material development process towards highly optimized functional materials. This is not a trivial task though, as the current state-of-the-art breakdown strength measurement techniques rarely provide statistically sufficient amounts of breakdown data from the application point-of-view, thus leading to impaired evaluation of the practical breakdown performance in film capacitors. So many polymer films are used in capacitors since last many year. Their specific properties are high breakdown strength, low dielectric losses, and availability make this film well suitable for use as capacitor dielectric. At the moment, the typical energy density achieved with PP film at room temperature is about 1.2 J/cm<sup>3</sup>. Present work on this film focus on improving the permittivity, breakdown strength, thermal withstand ability, and thermal conductivity, by producing copolymers, blending with other polymers, or adding inorganic particles. The influence of impregnating rapeseed oil on the dielectric breakdown behavior of thin metallized polypropylene (PP) foils used for high power capacitors was investigated. With oil impregnation the dielectric breakdown strength of the PP foil increases strongly. The breakdown behavior of rapeseed oil impregnated PP foils was compared to silicon oil and pentaerythrit-tetrapelargonate oil impregnated PP foils. Based on these experiments a guideline for the selection of impregnating oil for high power capacitors can be given. The breakdown strength of PP capacitor foils decreases with increasing temperature. In addition to this behavior, breakdown strength measurements performed as a function of temperature showed discontinuities for dry and saturated PP foils at defined temperatures. Investigations are performed to study the effects of variation of temperature.

### INTRODUCTION

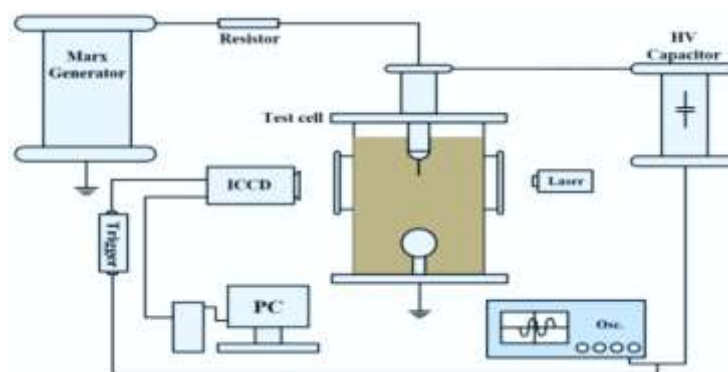
Capacitors, which allow energy to be stored in an electric field within a polarized dielectric medium and then released over short periods of time under controlled conditions, are vastly utilized in virtually every field of electrical engineering for storage and management of electrical energy. Typical targets of application include low and high frequency filtering duties in both AC and DC systems, pulsed-power applications, power conditioning and power factor correction as well as various coupling, tuning and bypass operations. Currently, the major commercially available capacitor technologies include film, ceramic, electrolytic and electric double layer capacitors, each of which are based on a different dielectric medium and have their own application-specific advantages and disadvantages. As the focus of this thesis is on the dielectric breakdown properties of film capacitor insulation, the other classes of capacitor technology are not discussed further here. Essentially, film capacitor elements comprise of two electrodes separated by a thin laminar dielectric layer in a wound configuration. Thin ( $\mu\text{m}$ -range) dielectric polymer films of e.g. polypropylene (PP) are almost exclusively used in film capacitors today as they offer excellent dielectric breakdown strength, low dielectric losses, easy film process ability, good mechanical properties and low cost. Film capacitors are further categorized into film-foil and metallized film capacitors. Oil-impregnated film-foil capacitors comprising of separate sheets of thin metal foil electrodes and dielectric film are generally directed towards very high current applications in which low inductance and low dielectric losses are required, a typical example being high-voltage AC capacitors for power factor correction in power systems. On the other hand, metallized film capacitors, which comprise of polymer



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films with extremely thin metal electrodes evaporated directly on the film surface, offer several advantages over film-foil capacitors with the single most significant one being the self-healing breakdown capability. Metallized film technology enables gracefully-aging dry capacitor designs with operation voltages substantially close to the large-area breakdown strength of the dielectric film, thus leading to much higher energy densities than those achievable in film-foil capacitors. Such features are crucial for many pulsed power and power electronics applications which require high volumetric capacitive energy density without compromising operation reliability. Capacitors are important passive components for electrical networks and apparatus. They can either improve the efficiency of HVAC power networks, or act as energy storage and stabilize the voltage level in HVDC systems. Dielectric materials in HV capacitors play a key role for charge control and energy storage. Before the seventies impregnated Kraft paper was the main capacitor dielectric and it was used in combination with mineral oil, polychlorinated diphenol (PCB), as impregnated liquid [1]. However, due to the low dissipation factor, high insulation resistance, good stability, and high availability, the polymer films gradually replaced the Kraft paper in capacitors. A switch from paper to polymer film also shortened the production process for capacitors, by reducing the drying time needed before impregnation. In general, the energy density of a dielectric material can be expressed as:

$$U_e = 1/2 \epsilon_r \epsilon_0 E^2,$$



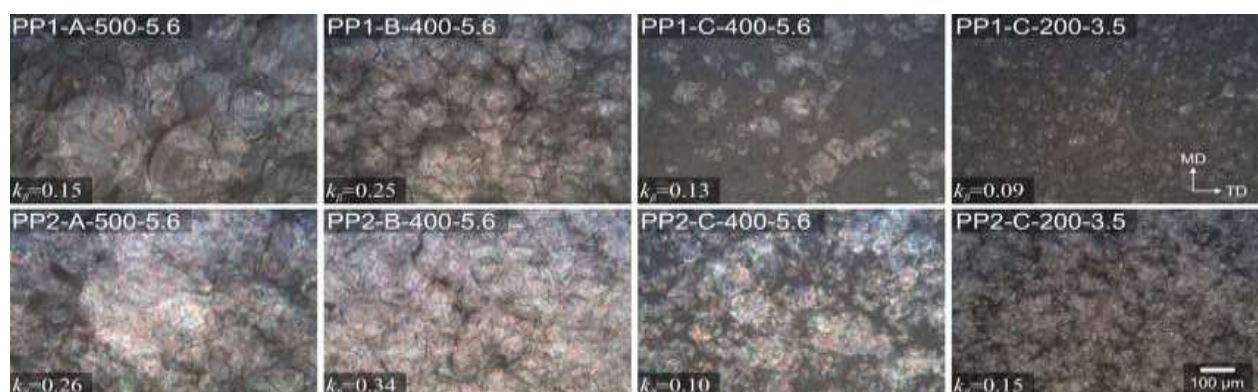
**Fig. 1. Experimental set-up for breakdown strength measurements**

Where  $\epsilon_0$  is the vacuum permittivity ( $=8.85 \times 10^{-12}$  F/m),  $\epsilon_r$  is a relative dielectric permittivity of linear dielectric materials and  $E$  is applied electric field. Although polymers exhibit high breakdown strength, their low dielectric permittivity (most,  $<10$ ) still limits the energy density [2]. Fig1 is used for breakdown measurement. As a result, an improvement of the intrinsic electrical properties of the most commonly used polymers is highly requested. It will have a high impact of the size of power capacitors. The main characteristics of these polymer films will be reviewed in the following sections of the Webster's Dictionary defines a capacitor as: "A device giving capacitance, usually consisting of conducting plates or foils separated by thin layers of dielectric with the plates on opposite sides of the dielectric layers oppositely charged by a source of voltage and the electrical energy of the charged system stored in the polarized dielectric." Capacitors have existed for nearly 250 years; until the invention of the battery, the capacitor was the only mobil means of electrical storage. Today, the capacitor is one of the most important elements in electronics and electrical engineering. During the last 50 years, great progress has been achieved in capacitor technology. The energy density of medium voltage capacitors has increased by a factor of more than 30 [1]. The improvement in capacitor technologies in the last few years was possible by introducing new dielectric materials like polymeric films and by changing from metal-foil electrodes to vapor deposited metallic layers. Furthermore impregnants avoiding air enclosures in the capacitor winding gaps were also optimized. Due to increasing energy and also power density, breakdown protection concepts had to be improved [2]. Materials used for the dielectric isolation are chosen with regard to the application of the capacitor. Each dielectric shows a characteristic behaviour as a function of frequency, temperature and in the case of electric shocks. Most capacitors presently installed onelectrical locomotives as intermediate DC-link capacitorsand series



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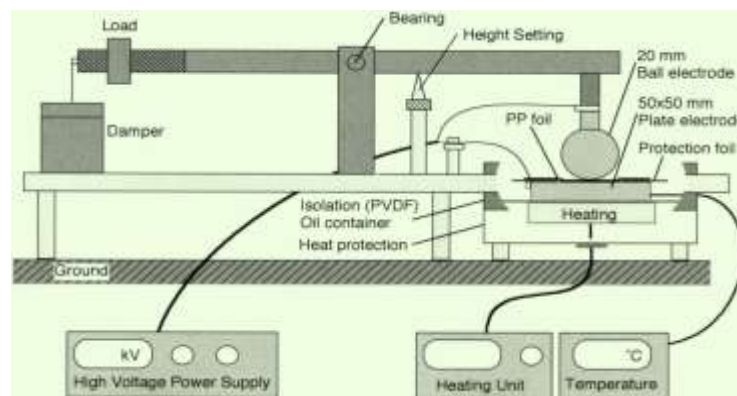
resonant circuit capacitors use a thin polypropylene (PP) foil in the range of 5–20 mm thickness as dielectric. Latest capacitor technologies use patterned electrodes instead of all-over metallized films. This technology allows control of the breakdowns initiated by unavoidable defects in the dielectric film, unavoidable defects in the dielectric film, even at high electric fields and high temperatures. Actually, electric DC-fields of 200 V mm<sup>-1</sup> and more are used for Montena capacitors (Condis product) resulting in energy densities of about 200 J dm<sup>-3</sup> for a required lifetime of more than 20 years. The lifetime depends on temperature, the applied field and the tolerated capacitor loss, which depends on the application. For traction applications, the currently accepted loss is 2%. There is incessant demand for drives of higher power and therefore the demand for higher energy density capacitors continues. The capacitors have to be reduced in weight and size. Moreover a reduction of capacitor size results in an important cost reduction. To reduce the volume and weight, the energy density of the power capacitors has to be increased. The energy density depends quadratically on the electric field in the dielectric. The aim for optimization of capacitors consists of determining the maximum electric field at which the lifetime of the capacitor can still be guaranteed. The applied dielectric field on the metallized polymer foil of a power capacitor in use is about three times lower than its breakdown strength. For a specific dielectric and a given voltage, an enhancement of the energy density is only possible by increasing the electric field strength. Fundamentals of capacitor technology evolution and electronics of dielectric capacitors are discussed in [3,4]. As mentioned above, the lifetime of a capacitor depends on its operational temperature. For traction applications, the capacitor temperature reaches values up to 90°C, depending on the type of engine. For electric fields greater than 200 V mm<sup>-1</sup> and temperatures higher than 50°C, a strong decrease in capacitance was measured as a function of time [9]. Thus, the lifetime of the capacitor is strongly reduced at higher temperatures. To study this behavior in detail, we investigated the breakdown strength of impregnated PP capacitor foils as a function of temperature.



**Fig 2. Cross-sectional OM images of cast film specimens under cross-polarized reflected light, growth. Chill roll (CR) side down. Relative  $\beta$ -phase content ( $k\beta$ ) is given for each cast film for reference. showing mixed type spherulites and trans- crystalline**

**EXPERIMENTAL**

The investigations were made on a metallized isotactic polypropylene (PP) capacitor foil with a thickness of 15.290.1 mm. The degree of crystallinity of the PP foil (c-PP) is 60%. One side of the film is covered with an evaporated 15 nm thin metallic Al:Zn layer to form the electrode. The metallization consists of individual segments interconnected by measurements were performed with an experimental set-up described in [5] according to DIN 0303. The breakdown strength of extremely thin polymeric materials like PP capacitor foils is usually measured with a ball-plate electrode configuration. Fig. narrow current gates. In case of breakdown in one of the segments, the gates serve as fuses and they isolate the segment and therefore the breakdown channel from the rest of the electrode. So the damage of the capacitor is localized capacitance only decreases slightly and therefore the capacitor is protected against complete destruction. This behavior is called self-healing. To avoid air enclosures in the capacitor rolls Montena uses dehydrated rapeseed oil as impregnation medium. The breakdown strength shows the experimental set-up for the breakdown strength measurements. In our case the upper electrode consists of a 20-mm steel ball and the opposite electrode is a 50\_50 mm<sup>2</sup> steel plate polished to optical quality. The contact velocity of the ball on the polymer foil is controlled with a damper and a soft contact is guaranteed. The force between the electrodes is load adjusted. With a heating supply below the plate electrode the polymer foil can be heated up to 100°C. This allows us to measure the breakdown strength of thin dielectrics as a function of temperature. The applied voltage for the breakdown strength measurements is furnished by a power supply with a variable DC voltage between 0 and 15 kV. The slope of the voltage ramp

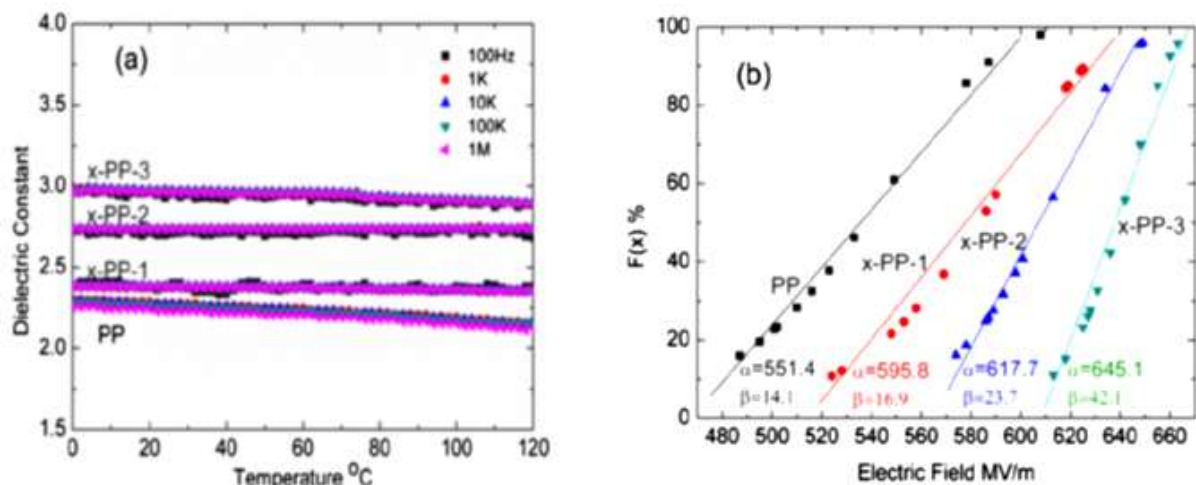


**Fig. 3. Experimental set-up for breakdown strength measurements (DIN 0303)**

was 500 V s<sup>-1</sup>. For a series of tests, metallized PP foils were stored in rapeseed oil at temperatures of 70 and 90°C for 20 days. Furthermore, PP foils were stored for 50 days at 80°C in silicon oil and 20 days at 90°C in pentaerythritetrapelargonate oil. Fig. 3. Shows the Experimental set-up for breakdown strength measurements the impregnations were done in closed glass containers in a circulation air oven. For the breakdown strength measurements the impregnated metallized PP foils were cleaned without using solvents, placed for 30 s on the preheated plate electrode and then the breakdown strength measurements were performed while maintaining the same temperature. For that purpose thermo elements are placed on the plate electrode as well as on the PP foil. All breakdown strength measurements were repeated 20 times for better statistics. The mean breakdown strength value was corrected with respect to the oil film between the PP film and the grounded plate electrode, which increased the breakdown strength slightly. The micro calorimetrical measurements (DSC, Differential Scanning Calorimetry) have been performed with a Perkin-Elmer DSC-7 instrument. A heating rate of 10K: min was used from -70°C up to 200°C. The measurements on samples of 4–7 mg were performed under nitrogen in Al-crucibles. For the mechanical relaxation measurements, metallized PP foils ‘as received’ and in use in a capacitor on a TGV composition of the French railways SNCF for over 1 year were investigated. All foil samples, quadratic and 10\_10 cm in size were stored at 100°C in a circulation air oven.

**RESULTS AND DISCUSSION**

3.1. *Breakdown strength* Breakdown strength measurements of metallized PP capacitor foils before and after rapeseed oil impregnation at different storage temperatures are shown in Fig 3. With increasing temperature, the breakdown strength decreases for the dry PP foil as well as for the impregnated PP foils. This is confirmed by the work of Ieda [6], where commercial isotactic PP shows a slight decrease of the dielectric breakdown strength over the temperature range from  $-50$  up to  $100^{\circ}\text{C}$ . It is shown in Fig. 4 (a) Dielectric constants (vs frequency and temperature) and (b) Weibull distribution for breakdown strength of PP, x-PP-1, x-PP-2, and x-PP-3 thin film dielectrics, which respectively have 0, 0.64%, 2.97%, 3.65% BSt units. From this, two phenomena can be detected. First, comparing dry and impregnated PP foils, the impregnant is increasing the breakdown strength of the foils. Second, a strong discontinuity of the breakdown strength occurs in temperature regions around  $50^{\circ}\text{C}$  and additionally there is a considerable fall-off above  $80^{\circ}\text{C}$  for impregnated foils.



**Fig. 4 (a) Dielectric constants (vs frequency and temperature) and (b) Weibull distribution for breakdown strength of PP, x-PP-1, x-PP-2, and x-PP-3 thin film dielectrics, which respectively have 0, 0.64%, 2.97%, 3.65% BSt units. [6].**

3.2. *Oil impregnation of PP capacitor foils*

Fig. 2 shows that with increasing impregnation temperature the breakdown strength, measured at room temperature (RT), increases from  $640 \text{ V mm}^{-1}$  for dry PP foils up to  $810 \text{ V mm}^{-1}$  for foils impregnated at  $90^{\circ}\text{C}$  for 20 days with rapeseed oil. The PP foils impregnated at  $70^{\circ}\text{C}$  for 20 days exhibit a breakdown strength of  $800 \text{ V mm}^{-1}$  at RT. From XRD (X-ray diffraction) and TEM (transmission electron microscopy) analysis, we know that the amorphous regions of the PP foil intersperse the whole foil thickness and they are coupled together. All impurities, in particular oils, penetrate into the amorphous regions and continuous diffusion channels result perpendicular to the foil. With diffusion of the impregnating into the dielectric, the PP foil swells insignificantly. The mass increase of the PP foil due to rapeseed oil uptake in PP after impregnation at  $80^{\circ}\text{C}$  for 25 days was 10%. This behaviour shows that the increasing breakdown strength with oil impregnation is attributed to the diffusion of the rapeseed oil into the for 20 days PP foil. In an amorphous polymer, the molecules are not packed regularly with the highest possible density. Between the macromolecules empty spaces exist which form the total free volume of the polymer. Artbauer to impregnant explain the breakdown phenomena of polymers [7]. Fig. 5 shows Temperature dependence of the dielectric permittivity of PVDF/PP blend proposed a volume breakdown theory composite in a 1/1 volume fraction at selected frequencies as indicated. The free volume in the dielectric decisively determines the breakdown behaviour of the PP foil. With oil diffusion, the PP foil structure becomes more regular as the free volume of the dielectric is filled with the impregnant PP foil. In an amorphous polymer, the molecules are not packed regularly with the highest possible density. Between the macromolecules empty spaces exist which form the total free volume of the polymer. Artbauer proposed a free volume breakdown theory to explain the breakdown phenomena of polymers [7]. The free volume in the dielectric decisively determines the breakdown behaviour determines the breakdown behaviour of the PP foil.



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With oil diffusion, the PP foil structure becomes more regular as the free volume of the dielectric is filled with the impregnant. . Therefore the electric field distribution in the case of applied voltage becomes more homogeneous and thus the breakdown strength of the PP foils increases. Further investigations of the interaction between polymer foils and insulating oils are discussed in [8].As argued above, the increasing breakdown strength for impregnated PP foils is attributed to the diffusion of the rapeseed oil into the PP foil. To investigate the breakdown behaviour of PP foils treated with other insulating oils, we measured the dielectric breakdown strength of metallized PP capacitor foils impregnated with silicon oil and pentaerythrit-tetrapelargonate oil. Silicon oil is one of the most widely used impregnation media for capacitor applications, with a lower viscosity n than rapeseed oilPentaerythrit-tetrapelargonate oil has a viscosity comparable to that of rapeseed oil.

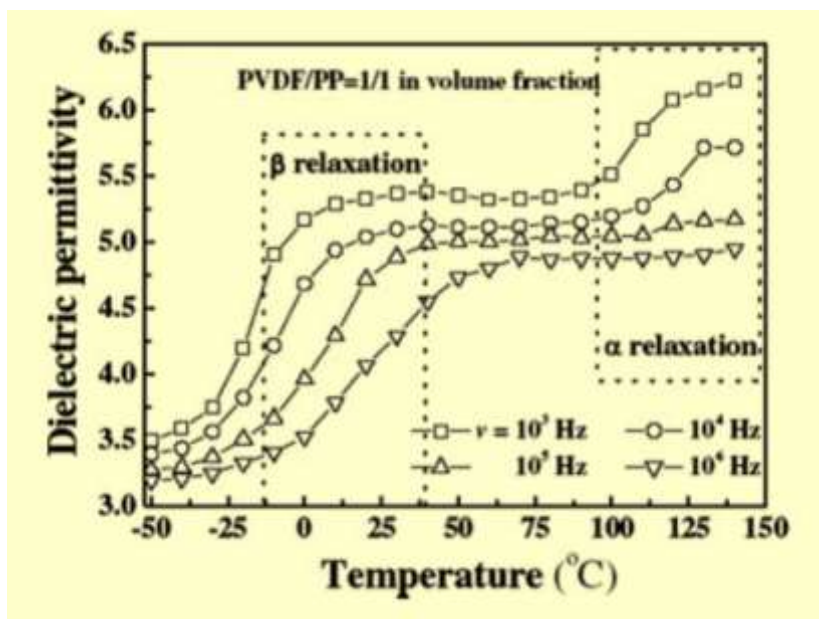


Fig. 5. Temperature dependence of the dielectric permittivity of PVDF/PP blend proposed a volume determines the breakdown behaviour of the PP foil.breakdown theory composite in a 1/1 volume fraction at selected frequencies as indicated

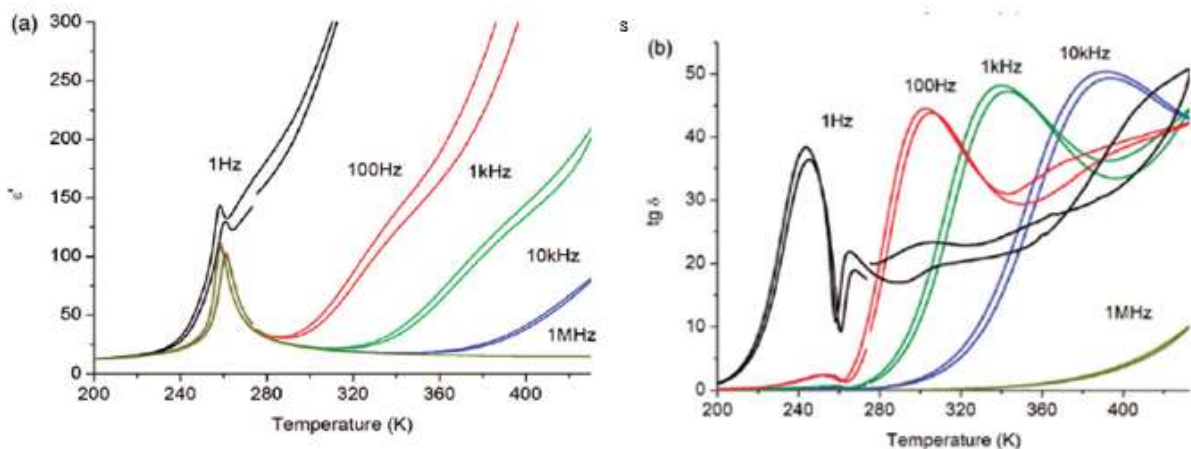
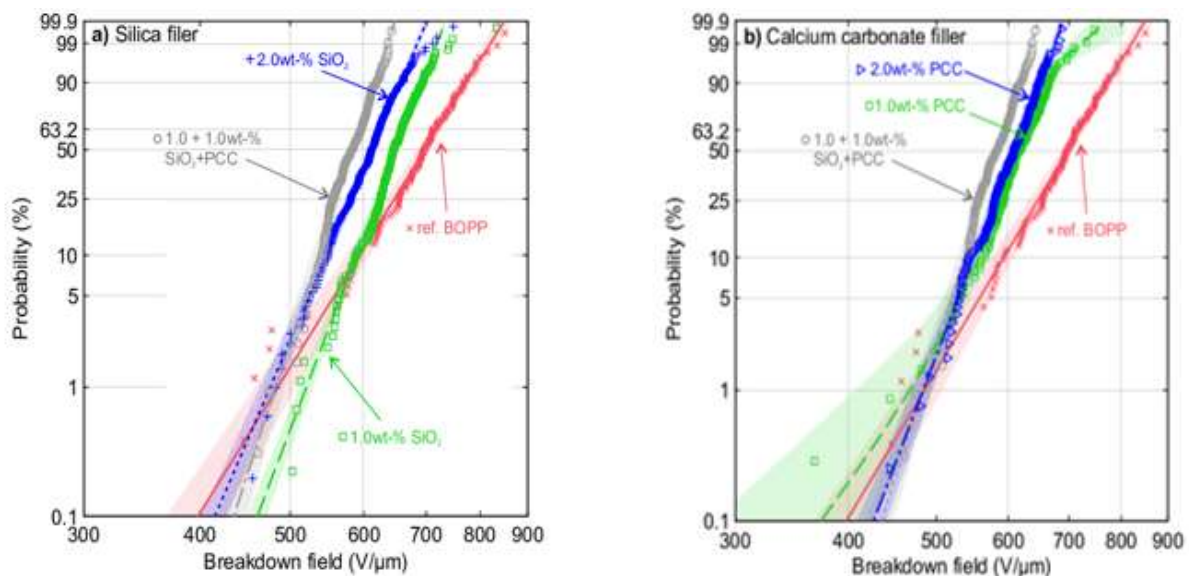


Fig. 6. Temperature dependence of the dielectric permittivity of PVDF/PP blend composite in a 1/1 volume fraction at selected frequencies as indicated



Silicon oil is one of the most widely used impregnation media for capacitor applications, with a lower viscosity than rapeseed oil. Pentaerythrit-tetrapelargonate oil has a viscosity comparable to that of rapeseed oil. The molecular structure of rapeseed oil contains several C-C bonds. Fig. 6. Represents Temperature dependence of the dielectric permittivity of PVDF/PP blend composite in a 1/1 volume fraction at selected frequencies as indicated. In case of a discharge in the winding gaps, C-C bonds in the molecular structure of the impregnating oil represent a risk for carbonization and better conductivity of the breakdown channels.

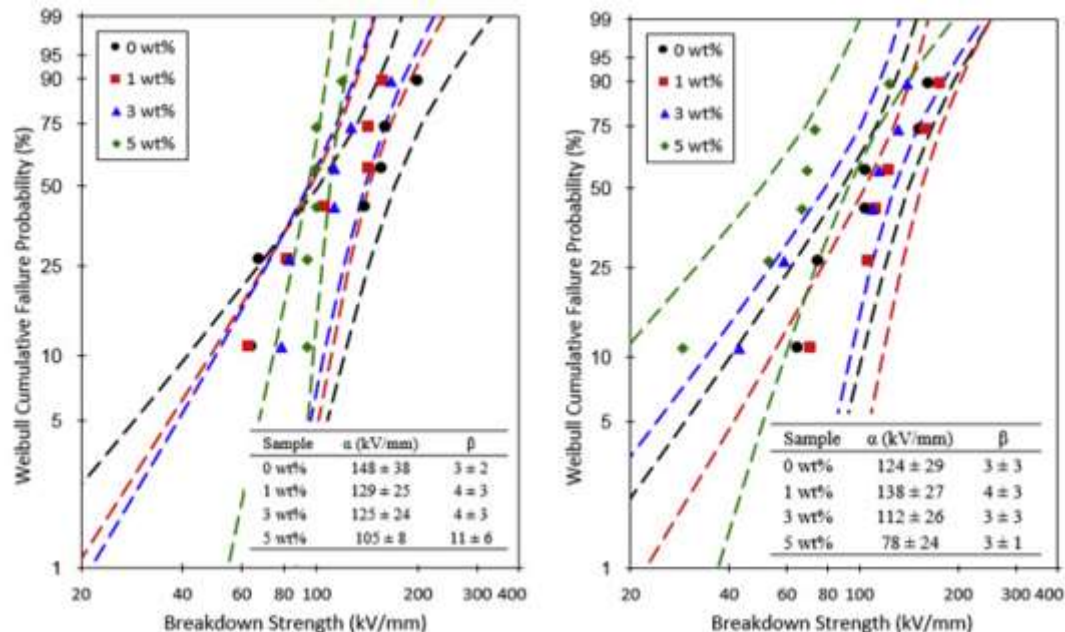


**Fig. 7. Breakdown strength of metallized PP capacitor foils before and rapeseed oil impregnation at temperatures of 70 and 90°C Effect of (a)Silica and (b)calcium carbonate fillers on large-area multi breakdown performance of BOPP. The Shaded areas represent the 90% confidence bounds**

Thereby the self-healing behavior of the capacitor can be affected, resulting in a large number of decoupled segments in the case of a breakdown. Therefore a reduction of the capacitor lifetime results. Pentaerythrit-tetrapelargonate oil is an impregnation medium without C-C bonds. The breakdown strength of PP foils stored in tetrapelargonate oil for 20 days at 90°C was measured as a function of temperature and compared to rapeseed oil impregnated and silicon oil impregnated foils (Fig. 3). While the tetrapelargonate oil impregnation shows nearly the same diffusion behaviour as the rapeseed oil and reaches a breakdown strength of 810 V mm<sup>-1</sup> after 20 days for an impregnation at 90°C, the silicon oil impregnated PP foil stored for 50 days at 80°C shows only a slight increase in breakdown strength from 640 V mm<sup>-1</sup> up to 680 V mm<sup>-1</sup> at RT. Even after longer impregnation times, the silicon oil impregnated PP foils reveal no significantly higher breakdown strengths. Fig. 7. Breakdown strength of metallized PP capacitor foils before and rapeseed oil impregnation at temperatures of 70 and 90°C Effect of (a)Silica and (b)calcium carbonate fillers on large-area multi breakdown performance of BOPP. The Shaded areas represent the 90% confidence bounds. We conclude that the impregnation with silicon oil leads only to a partial impregnation of the dielectric and therefore the breakdown strength is only increased slightly compared to the rapeseed oil impregnation. This effect may tentatively be ascribed to a much lower solubility (lower uptake) of the silicon oil compared to tetrapelargonate or rapeseed oil in PP. The measurements showed that the capacitor foil should be impregnated as completely as possible to obtain a high dielectric strength. Poor treatment during production of the capacitor or an unsuitable impregnation medium may cause discharges or an unfavorable breakdown behaviour. This effect may tentatively be ascribed to a much lower solubility (lower uptake) of the silicon oil compared to tetrapelargonate or rapeseed oil in PP. The measurements showed that the capacitor foil should be impregnated as completely as possible to obtain a high dielectric strength. Fig. 8. Breakdown strength of metallized PP foils impregnated with rapeseed oil at 70°C and 90°C for 20 days, PP foils impregnated with pentaerythrit-tetrapelargonate oil at 90°C for 20 days and metallized PP foils after impregnation with silicon



oil at 80°C for 50 days. Poor treatment during production of the capacitor or an unsuitable impregnation medium may cause discharges or an unfavorable breakdown behaviour.



**Fig. 8. Breakdown strength of metallized PP foils impregnated with rapeseed oil at 70°C and 90°C for 20 days, PP foils impregnated with pentapentaerythrit-tetrapelargonate oil at 90°C for 20 days and metallized PP foils after impregnation with silicon oil at 80°C for 50 days**

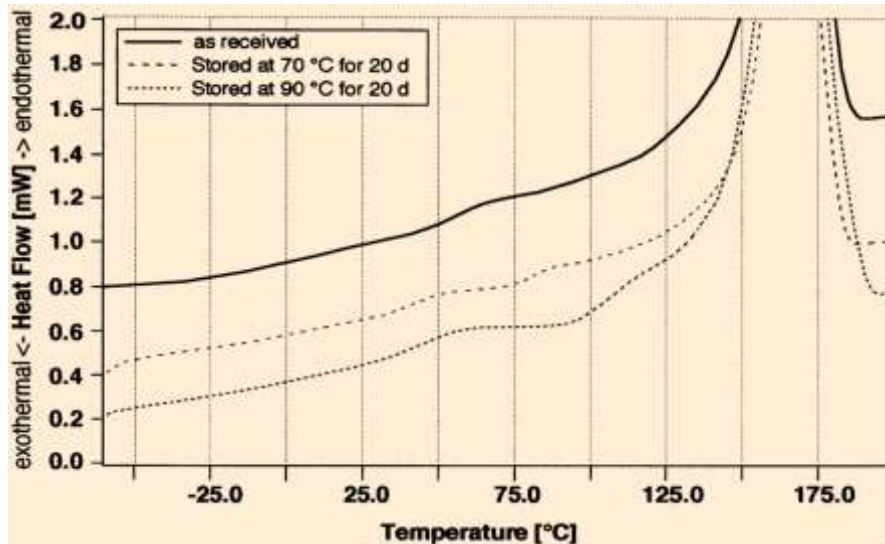
### 3.3. Discontinuity of the breakdown strength

All investigated foils, dry or impregnated, show a stepwise drop in the breakdown strength at around 50°C (Figs. 2 and 3). The impregnated PP foils show an additional decrease in breakdown strength at temperatures over 80°C. Due to this additional drop observed for oil impregnated foils the breakdown strength for temperatures >90°C is lower compared to the breakdown strength of the 'as received' foils. Thermal analysis yields information about structural properties, glass transition temperature, specific heat, melting and crystallization properties of polymers. To investigate the structural behaviour of metallized PP capacitor foils, DSC measurements of PP foils stored at different temperatures have been performed. The results are presented in Fig. 4. The main peak at temperatures around 170°C is due to the melting of the crystalline phase of PP. The untreated PP foil shows a shoulder in the DSC plot around 50°C. In DSC measurements such a shoulder usually indicates a structural phase transition in the examined polymer. For capacitor foils stored at 70°C the shoulder, already observed for dry PP foils and discussed above, is moving to higher temperatures and is registered at 75°C in the DSC plot. Furthermore, a second shoulder is located around 50°C in the plot. PP foils treated at 90°C also show two shoulders similar to PP foils impregnated at 70°C. One shoulder is beginning at 50°C and the second shoulder occurs around 95°C in the DSC plot. From the DSC results we conclude that PP foils stored at higher temperatures cause a splitting of the phase transition peak. One part of the shoulder is moving with increasing storage temperature to higher temperatures in the DSC plots and is beginning at the storage temperature while the other part is always located around 50°C. As DSC measurements of impregnated PP foils reveal shoulders at the same temperatures as those of dry PP foils stored at corresponding temperatures, the structural changes in the PP cannot be attributed to the influence of the oil impregnation. The DSC measurements reveal structural phase transitions in the same temperature regions as the PP foils show discontinuities in the breakdown strength. Comparing these results we conclude that a strong correlation exists between structural changes and the dielectric breakdown behavior of the PP foils. Due to the structural changes occurring at defined temperatures electrons in the polymer may be liberated





[7]. The free electrons in the polymer are accelerated in the high electric field applied to the dielectric. These phenomena favour the breakdown of the PP foil and thus decisively lower its dielectric strength.



*Fig. 9. DSC measurements of metallized PP foils as received and stored at temperatures of some specific temperatures*

#### (a) PP foil

The structural phase transition observed for impregnated PP foils at temperatures over 80°C results in a considerable fall-off of the breakdown strength. As the untreated PP foil reveals no structural changes in this high temperature region and thus no drop in the breakdown strength occurs, the impregnated foils reveal lower breakdown strength values than the 'as received' foil at temperatures over 90°C (Fig. 9). For traction applications, the capacitor temperature reaches values from 50°C up to 90°C, depending on the type of engine. Our investigations showed structural changes in the PP foil exactly in this temperature range as well as stepwise drops in the breakdown strength. These results point to critical temperature regions for PP capacitors, confirmed by accelerated lifetime measurements [9]. They showed that for operational temperatures higher than 50°C a strong decrease in the lifetime of the capacitors results.

#### 3.4. Mechanical relaxations

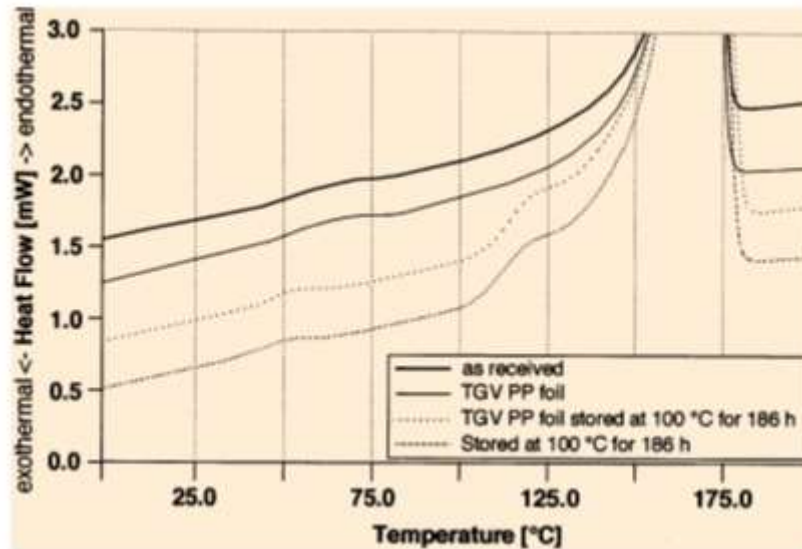
To control if the origin of the structural phase transitions observed in the DSC measurements can be attributed to pure mechanical relaxations we performed DSC measurements before and after heat treatment of the samples at 100°C. After a treatment time of 64 h and 186 h, the geometrical changes of the samples were measured. The results 'as received' PP foil with an original size of 10\_10 cm shows different side lengths after 64-h heat treatment. Sides A and C decreased in length while sides B and D increased by 10%. These geometrical changes are still unchanged even after 186 h of heat treatment. PP foils after 1 year in use on a TGV and therefore fully impregnated with rapeseed oil show decreased side lengths in all directions after a storage time of 64 h at 100°C. Sides A and C reveal the same results as the unimpregnated PP foil. Sides B and D are decrease around 20%. After heat treatment time of 186 h it was not possible to measure the geometrical values of the sample. The foil had a wavy profile and began to disintegrate. The mechanical relaxations (anisotropic length changes) we observed in the case of heat treatment are due to frozen tensions in the PP foil and can be attributed to the foil production process (extrusion and calender process, stretching in rolling direction). The shrinkage in both foil directions we observed for the PP foil in use on a TGV as well as the complete disintegration due to the 100°C storage test suggest severe damage in the polymer. For capacitors in use on a TGV composition the peak ambient temperature reaches 55°C and short term hot spot temperatures up to 70°C are measured. As shown in the section above, PP foils stored at 70°C cause a splitting of the phase transition peak and two shoulders are present in the DSC curve (Fig. 4). In Fig.5, the curve of the TGV PP foil before heat treatment shows the same feature as the untreated foil



with a corresponding shoulder around 50°C. Therefore the short term hot spot temperatures up to 70°C measured for the TGV capacitor foil in use have no influence on the structural changes of the foil. Based on these results we conclude that for foils stored at temperatures up to 55°C as the TGV PP foil, no splitting of the transition peak occurs in the DSC curves. In contrast to this, PP foils stored or impregnated at 70°C or higher show two critical temperature regimes in the DSC as well as in the breakdown strength measurements. This confirms the crucial role of temperature in the region between 50 and 70°C with regard to the breakdown behaviour as the structural behaviour of the PP foil. The DSC results of the examined PP foils before and after heat treatment at 100°C are presented in Fig. 5. The untreated metallized PP foil shows the well-known curve feature already seen in Fig. 4. After storage at 100°C for 186 h the PP foil shows a first shoulder in the temperature range around 50°C and a main transition peak beginning at 105°C. The TGV PP foil stored at 100°C for 186 h shows two transition peaks in the DSC curve (Fig. 9). A first shoulder in the temperature range around 50°C and a main transition peak beginning at 105°C. Compared to the 'as received' PP foil heat treated at 100°C the TGV PP foil shows exactly the same curve feature. After the heat treatment the structural phase transition peaks are still visible and the DSC measurements reveal the same results as the DSC curves of metallized PP foils presented in Fig. 4 and discussed in the earlier section. Therefore the origin of the structural phase transitions observed in the DSC analysis and correlated to the discontinuities in the breakdown strength measurements cannot be attributed to pure mechanical relaxations in the polymer. It is known that a polymer usually contains a small amount of impurities such as residual catalysts, additives and process residues. It has been reported that impurities can affect the dielectric properties of PP [10]. To determine if the origin of the structural phase transitions observed in the DSC measurements can be attributed to the impurities in the examined PP foil, further investigations are necessary. For that purpose the structural properties and the breakdown behaviour of PP capacitor foils after removing the impurities have to be investigated.

#### 4. Numerical Analysis of Sample Thickness-Dependent DC Electrical Breakdown

At a high electric field, charges in an electrode can be injected into a dielectric material via Schottky thermionic emission or quantum mechanical tunneling. The accumulation of space charges under high voltage causes electric field distortion inside the dielectric material [14–16]. The maximum local electric field  $F_{\max}$  would be much higher than the applied electric field. Electrical breakdown would occur when  $F_{\max}$  exceeds a critical value  $F_c$ . This is named as the space charge modulated electrical breakdown model (SCEB) or bipolar charge transport model [14–16]. In addition, a charge transport and molecular displacement modulated electrical breakdown model (CTMD) is utilized to investigate the sample thickness dependent DC electrical breakdown of polymer [24]. Since free volume exists in polymers, it may be enlarged by the displacement of molecular chains with trapped charges by the Coulomb force under the high electric field [24,25]. Electrons are accelerated in the free volume under the effect of the electric field to obtain energy. The energy  $w$  of electrons gained from the electric field depends on the local electric field  $F$  and the length of the free volume  $\lambda$ , namely  $w = eF\lambda$ . The energy of electrons increases with an increase in the local electric field and the enlargement of free volume caused by the displacement of molecular chains. When the maximum energy of electrons  $w_{\max}$  is higher than the deep traps energy  $ET$ , electrons have enough energy to jump over the potential barrier, which will result in electrical breakdown.



*Fig. 10. DSC measurements of metallized PP foils and PP foils after 1 year in use on a TGV composition before and after heat treatment at 100°C for 186 h. 188*

## CONCLUSION

In conclusion, the large-area breakdown response of laboratory- and PP films was found to be strongly dependent on the composition and film processing. While the optimum silica content presumably resides at the low fill-fraction range (~1 wt-%), it is not the only determining factor, as compounds with equal silica content but differences in e.g. compounder screw speed were found to exhibit large differences in the breakdown response. Indications of possible silica-antioxidant interaction were also observed. The SEM-imaging analysis of the films showed that nano-structural features cannot solely explain the observed large-area breakdown behavior –this aspect points towards large-area approach being necessary for the imaging techniques as well in order to reliably establish a link between structural properties and engineering breakdown strength. Overall, the results suggest that up-scaling of polymer production is sensible with the traditional melt-blending technology, although further development and optimization of nanocomposite formulations and processing is necessary. The dielectric breakdown strength of thin metallized PP foils, measured at RT increases from 640 V mm<sup>-1</sup> in suitable impregnation medium may cause discharges or an unfavorable up to 810 V mm<sup>-1</sup> for foils impregnated in rapeseed oil at 90°C for 20 days. We showed that the increasing breakdown strength observed for oil impregnated PP foils is attributed to the diffusion of the impregnant into the dielectric. With oil diffusion, the PP foil structure becomes more regular as the free volume of the dielectric is filled with the impregnant. Therefore the electric field distribution in the case of applied voltage also becomes more homogeneous and thus the breakdown strength of the PP foils increases significantly. The mass increase of the PP foil after impregnation in rapeseed oil was determined to be 10%. The breakdown behaviour of PP foils treated with different insulating oils was compared to rapeseed oil impregnated foils. The tetrapelargonate oil impregnation reveals nearly the same diffusion behaviour as the rapeseed oil and therefore also reaches breakdown strengths of 810 V mm<sup>-1</sup>. The silicon oil impregnated PP foil shows only a slight increase in breakdown strength from 640 V mm<sup>-1</sup> up to 680 V mm<sup>-1</sup> at RT [ Fig. 10]. The impregnation with silicon oil leads only to a partial impregnation. The measurements showed that the capacitor foil should be impregnated as completely as possible to obtain a high dielectric strength. Poor treatment during production of the capacitor or an unsuitable impregnation medium may cause discharges or an unfavorable breakdown behaviour. With increasing temperature, the breakdown strength decreases for dry PP foil as well as for impregnated PP foils. Furthermore the breakdown strength measurements show a strong discontinuity in breakdown behavior in temperature regions at 50°C and over 80°C for impregnated foils. DSC measurements reveal phase transition peaks in the same temperature regions as the breakdown strength discontinuities occur. The transition peaks can be attributed to structural changes in the PP. Due to these structural changes occurring at defined temperatures, electrons in the polymer may be liberated which are accelerated in the



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high electric field applied to the dielectric, favouring the breakdown the PP foil and thus decisively lowering its dielectric strength. We conclude that the discontinuities in the dielectric breakdown strength of the PP foils are due to the structural changes in the dielectric observed in DSC measurements at defined temperatures. To control if the origin of the structural phase transitions can be attributed to pure mechanical relaxations we performed DSC measurements before and after heat treatment of the samples at 100°C. The examined PP foils reveal mechanical relaxations in both foil directions (anisotropic length changes) due to frozen tensions in the PP foil which can be attributed to the foil production processes. The DSC measurements have shown that the origin of the operation have no influence on the structural behavior of the PP foil. The results confirm the crucial role of the structural phase transitions observed in the DSC analysis and correlated to the discontinuities in the breakdown strength measurements cannot be attributed to pure mechanical relaxations in the polymer. Further measurements of the structural properties and the breakdown behaviour of PP capacitor foils after removing the impurities are currently being investigated to determine the parameters affecting the structure changes in the foil. PP foils in use on a TGV composition for over 1 year at a peak ambient temperature of 55°C were investigated. We could show that short term hot spot temperatures up to 70°C occurring in the capacitor during temperature between 50 and 70°C with regard to the breakdown behaviour as well as the structural behaviour of the PP foil.

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