TREEING GROWTH AND LIFETIME OF EPOXY FILLED WITH 26% OF SILICA NANOPARTICLES AT 50 HZ AC-VOLTAGE

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This paper presents first results of treeing growth analysis and lifetime tests of epoxy resin with 26 weight percent of nano-scale SiO_2-filler compared to samples with established neat epoxy resin. The test specimens made of hot curing BADGE epoxy consist of a moulded-in tungsten needle electrode with a tip radius of about 7 µm and a distance of 3,5 mm to the ground plane electrode. At nano particle loaded epoxy the inner wall surface of the treeing channel is covered with spherical nano particles that are half dug in the epoxy wall matrix. These nano particles act as an inorganic shield and avoid direct erosion of polymeric epoxy material by partial discharge attack. Therefore the growth and expansion of slim channels to broad ones is hindered by these nano particle layers like experienced at surface corona withstand tests. With the defined needle-plane configuration and specimen preparation it can be stated that the silica nanocomposite insulation prolongs treeing lifetime by a factor of two to nine depending on field strength and compared to pure epoxy. Further research work is needed to understand the electrical treeing process at the interface between inorganic nano particle and polymer resin matrix. Does the otherwise published 3-layer multi-core model fit?

1 INTRODUCTION

The world wide market of electrical energy became highly competitive after government directions of de-regulation and liberalization in most countries. Under this economic pressure the utilities had to reduce their plant operation costs and had to increase the reliability of their power plant assets. This forces the manufacturer to develop new generator designs with higher efficiency, longer maintenance intervals and much lower life cycle costs. As part of this development a fundamental research program has been started to improve the electrical properties of high voltage (hv) winding insulation by using nano particle.

Due to Nanotechnology it has become possible to produce silica (SiO_2) filled epoxies with particle size in the nm-range. With a suitable preparation technique the spherical nano-sized filler particle can be dispersed homogenously into the polymer matrix and without agglomerates. A new group of special additive treated SiO_2 material with a high amount of inner boundaries has been designed. The effect of this large inter-phase to volume ratio on the performance profile of the nanocomposite has to be analyzed by stressing the insulation system under high electrical fields which a needle-plane electrode configuration generates.

This paper presents the first results obtained on treeing growth behaviour and electrical lifetime tests of epoxy nanocomposites to be used for impregnation of mica-paper taped high voltage stator winding insulation. Comparison between established neat impregnation epoxy resin and new epoxy-nanocomposite is performed in electrical tests and discussed.

2 SAMPLE PREPARATION

For all samples a hot curing epoxy resin system is used. To avoid any possible influence of sample preparation effects all needle-plane electrode specimen had been manufactured by applying the same vacuum moulding process and a well defined curing program with stepped temperatures and smooth cooling down.

2.1 Insulation Materials

Neat Epoxy: The two-component epoxy resin consists of bisphenol-A diglycidylether (BADGE) resin and an anhydride hardener mixed by ratio of 100:75 parts by weight. A special curing catalyst is used. The curing process takes 5 h at 80 °C and 10 h at 140 °C. The cured specimen are cooled down smooth and slow for more than 24h, to minimize dilatation stresses at the insulation material.

Epoxy with nanoscale SiO_2-fillers: To the same type of BADGE resin a different amount of nano scale silica filler was homogeneously dispersed and mixed with the anhydride hardener. The content of dissolved nano particle varied between 5 wt% to 26 wt% for different batches that were produced for electrical treeing tests.

Figure 1 shows a transmission electron microscopy photo (TEM) of two nanocomposite probes of about 80 nm thin layer which were taken from test samples by a microtome cut. This type of spherical nano particles got an average diameter size of d_{50} = 23 nm. Taking into consideration some electron trace image scattering of the TEM the pictures in Figure 1 could verify the given nano particle size quite good. Due to the well approved dispersion technique the nm-particles are homogenously distributed in the resin matrix without forming any agglomerations. This uniform dispersion could be achieved independent of the weight amount of the nano particle up to a filler content of 26 wt% as the TEM pictures in Figure 1 demonstrate.

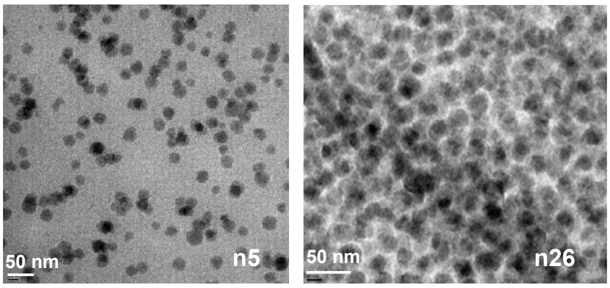


Figure 1: TEM-Photos of epoxy specimen filled with different amount of spherical nano-scale SiO_2-particles; 5 wt% (n5 - left) and 26 wt% (n26 - right)

2.2 Needle-Plate Sample Geometry

To generate treeing-channels in solid insulation material a high electric field is needed. A typical needle-plane test configurations which can be used to obtain an extreme electrical high field enhancement and to start the treeing process is given in Figure 2.

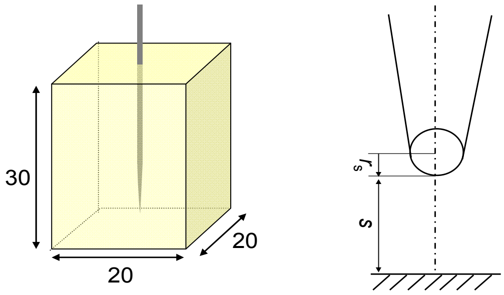


Figure 2: Geometry of the test sample and the needle-plate-electrode system

The thin needle electrode of tungsten carbide has a diameter of 0.5 mm and a length of 32 mm. The tip radius of r_s = 7,25 µm ± 0,25 µm is well defined and small enough to generate a high electrical field. The electrode configuration is designed to a nominated insulation distance of s_0 = 3,5 mm between the needle electrode (high voltage) and the plane electrode (ground).

To ensure a minimum influence of manufacturing all ten needle configurations of a test set are prepared in one casting mould. After the curing process the individual needle samples are separated and polished on each side to get better visibility of the growing treeing-channels. The bottom side of the sample is coated with a silver paint to achieve a void-less contact to the metal ground electrode.

The local field enhancement at the needle tip has been calculated with the geometric model of a hyperboloid electrode contour opposite to a large ground plate as given in [1]. The results for different test voltages which would be applied to the needle electrode are presented in Table 1.

Table 1: Electrical field enhancement E_{max} at needle tip for different test voltages with an electrode distance of 3.5 mm.

Test Voltage	16 kV	18 kV	20 kV
E_{max} [kV/mm]	575	650	715

2.3 Permittivity and Dissipation Factor

The dielectric permittivity ε_r and dissipation factor tanδ may have an influence on the local field distribution and on treeing propagation mechanism at nanocomposites when exposed to high local fields. Therefore these dielectric values ε_r and tanδ were measured at epoxy resin specimen loaded with a different quantity φ of nano-scale SiO_2-particle at room temperature, line frequency of 50 Hz and an electric field of 1 kV/mm (Figure 3).

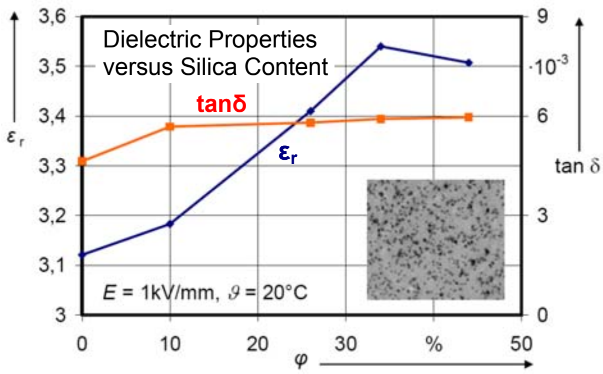


Figure 3: Dielectric permittivity ε_r and dissipation factor tanδ of epoxy resin specimen loaded with a different quantity φ of nano-scale SiO_2-particle

The values of ε_r and tanδ of the two materials used for the treeing samples, which are neat epoxy resin (oEP) and 26 wt% SiO_2-particle filled epoxy (n26), are summarized in Table 2.

Table 2: Permittivity ε_r and dissipation factor tanδ of oEP and n26 at room temperature and 50 Hz

Material	oEP	n26
ε_r	3.1	3.5
tanδ [10^{-3}]	4.6	5.8

Mixing pure epoxy resin with silica nano particle results in significant higher permittivity ε_r and dissipation factor tanδ than neat epoxy. With increasing content of nano particles the values of ε_r and tanδ raise in the same way as measured for epoxy to which about 60 wt% micro-scale silica was added [2]. In the literature there can be found two different explanations for an increase in permittivity and dissipation factor:

(1) Blocking and trapping of free charge carriers and polymeric dipole by nano particle interface (outer layer) resulting in higher polarization losses

(tanδ) and larger restrained dipoles which would raise the dielectric permittivity ε_r [3].

(2) Epoxy nanocomposites are highly sensitive to water absorption from ambient humidity. A larger mass content of nano-sized silica in the epoxy matrix will take up a higher amount of water, which is bound to the individual nano particle (water shell model) [4]. The extreme dipole characteristic of the water molecules with a permittivity of ε_r = 81 increases the dielectric permittivity ε_r in correlation to the filler content as shown in Figure 3.

On the other hand, depending on filler type, particle treatment and wt%-loading the interfacial polarizations at epoxy-to-particle interlayer and the relaxation mechanism may also result in a decrease of permittivity and dissipation factor [5]. The influence of nano filler concentration and particle agglomeration on decrease of permittivity due to hindered dipole mobility in [6].

Based on the above given information it becomes very clear how difficult it will be to understand the tree growth mechanism at nanocomposites because of the many possible and hard to define influence factors to local interface behaviour.

2.4 Frozen mechanical stresses

A further important influence factor on inception condition of the very first treeing channel at the needle tip is provided by residual mechanical stresses (frozen strain), which are unavoidable after hot curing and cooling down of the resin samples. The thermal extension coefficient of the tungsten needle is about 4 ppm/K while the epoxy material has an extension of about 60 ppm/K, which is 15 times higher. The huge difference results in frozen mechanical stresses around the needle electrode during polymerisation and shrinking of the epoxy resin. The frozen mechanical strain can be seen and analysed by taking polarized light pictures as demonstrated in Figure 4.

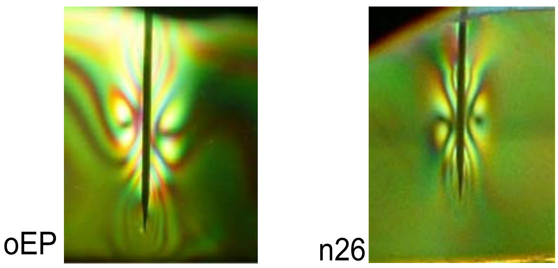


Figure 4: Polarized light pictures of equal-stress lines of frozen shrinkage strain at oEP and n26

Beneath the different transparency properties of neat epoxy (oEP) and the 26%wt nano particle loaded epoxy (n26) both samples present the same distribution of stress lines. Thus the influence of the frozen strain on the treeing growth is the same for both materials.

In Figure 5 polarized stress pictures of two samples of 26 %wt nano particle loaded epoxy (n26) at the needle tip area are shown. Despite the fact that all samples were smooth cured with an optimized temperature regime and very slowly cooled down high mechanical shrinkage stress still exist at the needle tip and could produce micro cracks at the material interface tungsten-epoxy.

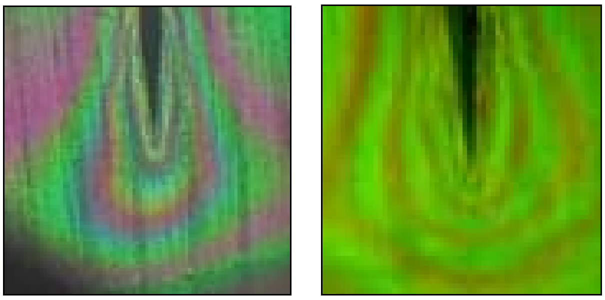


Figure 5: Polarized light pictures of equal-stress lines in nano-epoxy n26 at needle tip area

3 EXPERIMENTAL TEST SETUP

For treeing behaviour analysis of nano-composites voltage endurance tests at two different setups with 50 Hz AC are performed. At the first test bench 8 needle samples are stressed continuously in parallel without any interruption until breakdown. This test will work out the difference in electrical life time and treeing channel structure between neat epoxy and SiO_2 nanocomposite on a statistically ensured basis.

The second test setup is used to study the treeing growth in detail at one special prepared sample only, but with continuous observation of tree development by a stereo microscope with digital camera. The partial discharge (pd) activity is measured in parallel to treeing growth.

3.1 Parallel test bench

The voltage endurance parallel test set of eight samples is energized by a 20-kV-transformer. The AC high voltage is measured by a capacitive divider. Each of the eight samples is connected to the high voltage with a mechanical fuse, which consists of a conductive foil that burns away within a response time of 150 ms after breakdown of the individual sample. The other treeing samples running in parallel are not affected by an individual breakdown.

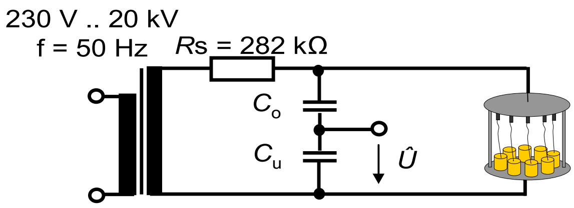


Figure 6: HV setup of the parallel test bench

3.2 Microscopy test bench

The electrical hv circuit of second test setup, which was build to study the treeing growth, is given in Figure 7. The setup consists of a capacitive divider (C_o-C_u) to control the high voltage and a hv coupling capacitance C_K with a coupling device (AKV) to measure the partial discharge (pd) activity during treeing expansion process. The samples are stressed in an oil-filled cubic glass vessel to avoid parasitized discharge activity. A cold-light source creates the luminous exposure of the treeing structure for recording with a stereo microscope and digital camera. The external pd noise level at the unscreened hv test room of the laboratory is about 2 pC.

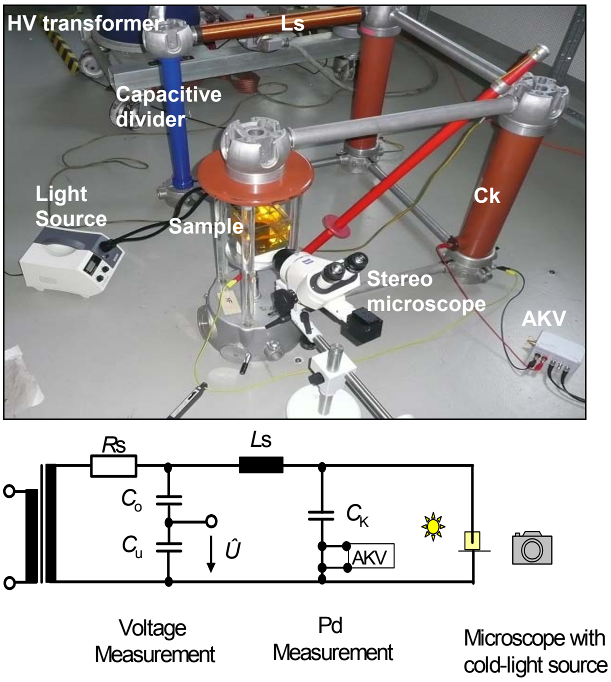


Figure 7: HV test setup of one sample for studying the treeing growth by using a stereo microscope with camera and simultaneous pd measurement

4 EXPERIMENTAL RESULTS

The tree propagation at the prepared needle specimen was observed at three different voltage level of 16 kV, 18 kV and 20 kV AC 50 Hz. To get mean values for breakdown time from the individual life time data of the sample collective the 2-parameter Weibull probability statistics are used. With these mean values electrical life time curves for neat epoxy and nanocomposites can be drawn. Microscopic pictures of typical tree propagation and tree channel shape could be captured at successive growing stages.

4.1 Treeing growth of neat epoxy resin

The results of voltage endurance up to electrical tree breakdown for neat epoxy (oEP) performed in the parallel test bench are plotted in the Weibull probability grid in Figure 8. Table 3 summarizes the characteristic parameter like 63% mean value of lifetime $t_{63\%}$ [min], shape (scatter) b and number of specimen n of the test collective.

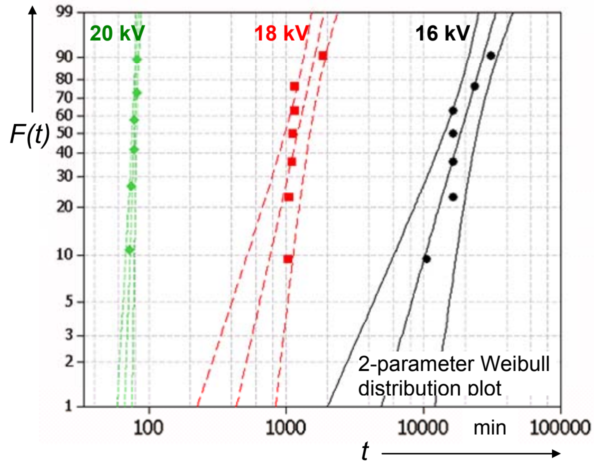


Figure 8: Weibull probability plot of electrical treeing lifetime of neat epoxy **oEP** at 16 kV, 18 kV and 20 kV, 50 Hz at room temperature

Table 3: Weibull distribution parameter of neat epoxy (oEP): 63% mean value of lifetime $t_{63\%}$ [min], shape (scatter) b and number of specimen n

Test voltage	20 kV	18 kV	16 kV
$t_{63\%}$ [min]	79,50	1318	20873
b	26,16	4,14	3,18
n	6	7	7

Because of the extreme inhomogeneous field distribution of the needle-plane electrode configuration a small increase of test voltage of about 10% results in more than one decade shorter electrical breakdown time of the specimens.

The treeing structure of a pure epoxy specimen after 60 min at 16 kV is characterised by a few clear and broad channels growing directly forward to the ground electrode as the microscopic picture in Figure 9 demonstrates. The leading channel is already well defined and will be the breakdown path later on.

Figure 9: Treeing structures of neat epoxy (oEP) after exposure to 18 kV for 60 min

4.2 Treeing growth of nano particle filled epoxy resin

The same voltage endurance tests which were performed for the specimens of neat epoxy had to be repeated with samples of 26 wt% silica nano particle loaded epoxy resin (n26) for comparison. The results of treeing lifetime are given in the Weibull plot of Figure 10 together with summarized data in Table 4.

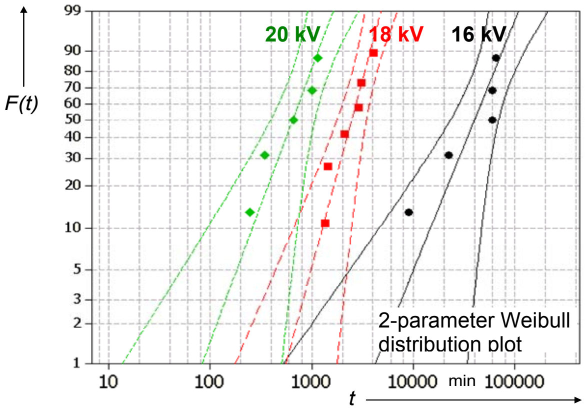


Figure 10: Treeing lifetime of nano particle loaded epoxy **n26** at 16 kV, 18 kV and 20 kV, 50 Hz, RT

Table 4: Weibull distribution parameter of nano particle loaded epoxy **n26** (see Figure 10)

Test voltage	20 kV	18 kV	16 kV
$t_{63\%}$ [min]	776	2 829	49 168
b	2,06	2,84	1,88
n	5	6	5

The microscopic photo series in Figure 11 shows the tree growth at 18 kV in a 26 wt% nano filled epoxy specimen after different time intervals. Due to the embedded spherical SiO_2 nano particle the tree path is retarded and has to go zigzag between the nano particle interface layers [7] and cannot generate a more or less direct path to the ground plane. Therefore the treeing is indicated by many fine and short branches giving the tree a more bush like structure [8].

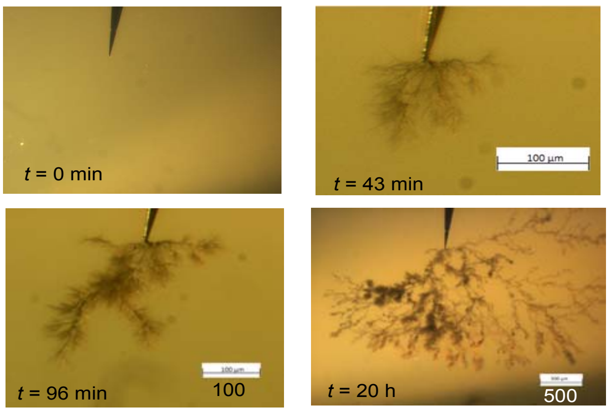
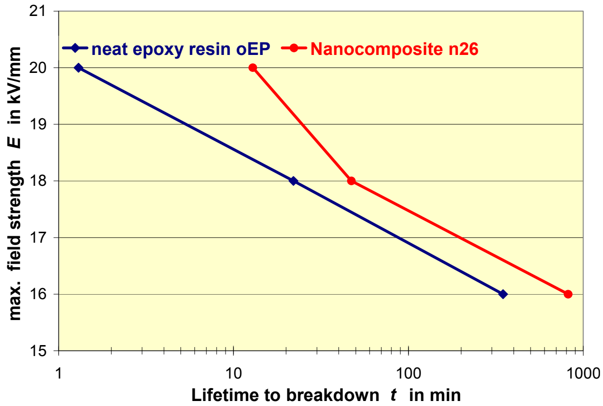


Figure 11: Development of treeing structure of nano particle loaded epoxy **n26** at voltage of 18 kV

After 20 h several parts of the branches tend to broaden up because of pd erosion activity but no leading channel to the ground electrode can be identified. The nano particle slow down treeing growth significantly.

With the lifetime results summarized in table 3 for neat epoxy and in table 4 for nanocomposite a lifetime curve versus maximum electrical field strength at needle tip can be drawn as given in Figure 12. Based on this type of needle-plane configuration and specimen preparation it can be stated that the nano-composite insulation instead of pure epoxy can extend the lifetime by 2,2 up to 9,9 times. Treeing tests in [8] with epoxy/alumina nanocomposites resulted in a factor of 2 – 3.



E_{max} [kV/mm]	575	650	715
oEP $t_{63\%}$ [h]	347,9	22,0	1,3
n26 $t_{63\%}$ [h]	819,5	47,2	12,9
Relation n26/oEP	2,4	2,2	9,9

Figure 12: Comparison of electrical lifetime of neat epoxy oEP and nanocomposite n26

5 ANALYSIS OF TREEING CHANNEL WALL

Samples at 16 kV endurance test with treeing channels of about 2 to 3 mm length were taken out of the hv test setup to analysis the inner treeing channel structure. The channel wall of specimens of pure epoxy and specimens of 26 wt% SiO_2 nano filled epoxy had been examined by using a scanning electron microscopy (SEM).

5.1 Neat epoxy resin

Figure 13 shows a cut through the main treeing channel near the needle tip of a neat epoxy sample (oEP). The distance from the needle tip is about 100 µm. The channel has a diameter of 6 µm and is opened by the cut over a length of 25 µm. The channel has a cylindrical shape without any macroscopic wall structures. In deeper sections the wall surface is unaffected by the preparation process and presents a rough and fleecy wall structure like ash deposits in a chimney.

One hypothesis may be that the fleecy rough coating at the channel wall is a deposit of carbon flakes which were produced by partial discharge activity when expanding the tree channel by erosion of epoxy resin. This carbon coating is the explanation for the black coloration of the broad main treeing channel. Beneath this fleecy coating the treeing wall has a smooth surface.

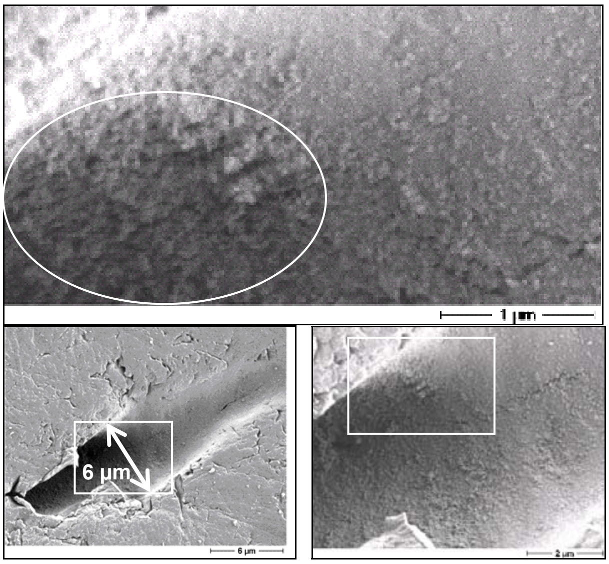


Figure 13: SEM photo of main treeing channel of neat epoxy, about 100 µm distance to needle tip

5.2 SiO_2 nano particle filled epoxy

There are many brittle and broken particles around the treeing channels. The SEM photo in Figure 14 shows an opened tree channel about 2000 µm away from needle tip. The channel wall is covered with nano particles that are half dug in the wall surface. The SiO2 particles act as an inorganic shield like mica does and avoid direct erosion of polymeric epoxy by pd attack. Furthermore there are µm-size structures in the wall, formed after long pd activity.

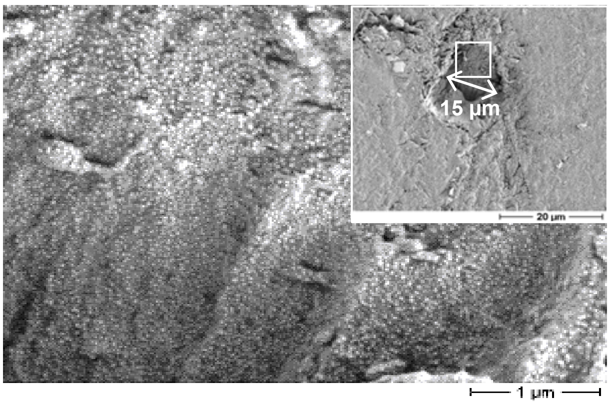


Figure 14: SEM photo of main treeing channel of n26, about 2 mm distance to needle tip

6 CONCLUSION

Summarising following conclusions can be given:

(1) For electrical treeing analysis of SiO_2 nano particle filled epoxy samples a needle-plate electrode configuration with a tip radius of 7,25 µm had been used. At test voltages of 16 - 20 kV a max. electrical field at the needle tip of 575 - 715 kV/mm is achieved.

(2) With the defined needle-plane configuration and specimen preparation it can be stated that the silica nanocomposite insulation prolongs treeing lifetime by a factor of two to nine, depending on field strength, compared to pure epoxy.

(3) At nano particle loaded epoxy specimen the surface of the treeing channel wall is covered with spherical nano particles that are half dug in the wall matrix material. These nano particles act as an inorganic shield and avoid direct erosion of polymeric epoxy material by partial discharge attack. The growth and expansion of slim channels to broad ones is hindered by nano particle layers like experienced at surface corona withstand tests.

(4) The treeing shape of nanocomposite specimen differ from neat epoxy samples because of the influence of silica nano particle. Due to the barrier effect of nano particle the tree channels develop slowly and highly distributed in small branches without a leading channel to ground electrode.

(5) Further research work is needed to understand the electrical treeing process at the interface between nano particle and resin matrix.

7 ACKNOWLEDGMENT

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