Study of Electrical Treeing in Dielectrics with Inorganic Fillers

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List of Abbreviations

3D: Three Dimension
AC: Alternate Current
APTES: Aminopropyl Triethoxysilane
BCC: Body-Centred Cubic
BNNS: Boron Nitride Nanosheet
BSE: Backscattered Electron
CCD: Charge-coupled Device
CTM: Computerized Tomography Method
DC: Direct Current
EDX: Energy Dispersive X-ray
EM: Electron Microscopy
FEGSEM: Field Emission Gun Scanning Electron Microscopy
FTIR: Fourier-Transform Infrared Spectroscopy
HV: High Voltage
LDPE: Low-Density Polyethylene
LSCM: Laser Scanning Confocal Microscopy
MXIF: Manchester X-ray Imaging Facility
PA: Polyamide
PD: Partial Discharge
PE: Polyethylene
**PRPD**: Phase Related Partial Discharge

**PSA**: Phase Sequence Analysis

**PSI**: Paul Scherrer Institute

**PTFE**: Polytetrafluoroethylene

**RMS**: Root Mean Square

**SBFSEM**: Series Blocked Face Scanning Electron Microscopy

**SE**: Secondary Electron

**SEM**: Scanning Electron Microscopy

**SSM**: serial sectional Method

**TEM**: Transmission Electron Microscopy

**UCL**: University College London

**UV**: Ultraviolet

**XLPE**: Cross-Linked Polyethylene

**XCT**: X-ray Computed Tomography

**XUM**: X-ray Ultra-Microscopy
Abstract

Electrical trees are degraded paths in polymeric insulation and are one mechanism of electrical failure of high voltage insulation systems. High-quality insulation system is needed to meet the increasing demand for electricity transmission and also to reduce energy loss and maintenance costs. Traditional polymer dielectrics filled with inorganic fillers are commonly regarded as the ideal design due to excellent physical and mechanical properties, but how tree propagation is influenced in these composites is still debatable even though there have been a lot of studies carried out in past years.

The main objective of this project is to investigate how and why these filler particles influence the tree propagation. Electrical trees were grown in the HV laboratory using the needle-to-plane geometry and there were four sample groups made for comparison: unfilled, micro-filled, nano-filled (untreated) and nano-filled (surface treated). Previous studies have confirmed the application of X-ray Computed Tomography (XCT) imaging of electrical trees using phase contrast enhancement. In this thesis, XCT imaging was further developed for both unfilled and filled epoxy. A multi-stage treeing and imaging experiment was carried out in an unfilled epoxy sample to analyse the development of tree branches in three consecutive treeing steps. The impact of x-ray dose on the epoxy resin studied was also evaluated by using tensile tests and Fourier-transform infrared spectroscopy (FTIR). Synchrotron XCT was applied for the imaging work in both micro- and nano-filled materials and based on the 3D replicas, several quantified parameters were used to describe the electrical tree structures in different dielectric systems. Partial discharge (PD) measurement was also introduced into the treeing studies so as to discuss treeing characteristics.

The achievements of this project include: the correlation between the tree volume and corresponding PD signals has been characterized and compared by the multi-stage test on an early tree structure. Electrical trees in microsilica-filled systems were successfully imaged and reconstructed in epoxy system with different load levels and their geometries were quantitatively compared. Interactions between tree channels and micro-sized fillers were also visualized in 3D replicas and have been used to illustrate the characteristics of
the tree propagation in micro filled systems. A ‘fine tree’ growth model was developed based experimental results and also the transition from non-conducting to conducting tree are discussed, evidenced by PD measurement. The model is used to explain the impact of filler particles on tree growth, which is also introduced into the characterization of the tree growth in both micro- and nano-composites.
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1 Introduction

1.1 Background

In electrical engineering, the transfer and distribution of power are dependent upon materials used for electrical insulation, so the reliability of the insulation system is an important factor for network performance. As equipment operates beyond its design life, reduced performance is inevitable. For operators, unreliable insulation will influence the operation of electrical equipment and it may increase the effort required to maintain networks. To reduce energy loss and maintenance costs, increasing demands of insulation reliability need to be met.

Insulation ageing is a complex part of dielectric science. Multiple aspects result in insulation breakdown such as thermal issues, chemical problems and mechanical damage [31]. Electrical trees are degraded as paths in polymeric insulation. In solid dielectrics, this kind of track has a characteristic topology and resembles botanical tree structures, as shown in Figure 1-1. From last century [32], research work on electrical trees has been carried out. In the past decades, further research has revealed a lot of features and developed many good methods for analysis of electrical trees. Electrical trees are regarded as a main cause of electrical breakdown of polymers, but unfortunately, the basic mechanism of tree generation is not well understood so tree initiation is still hard to eliminate. However it is clear that the tree growth is accompanied by partial discharge activity which may be used to monitor tree development.

Figure 1-1: Electrical tree in a polyethylene cable [33].
Discovery of novel materials has never stopped. From 1920s, synthetic polymer composites were introduced into electrical engineering and new kind of materials were adopted [6]. With development of applications in electrical insulation, micro-filled composites such as epoxy resin filled with quartz flour started to replace traditional insulation materials and where used for dry distribution transformers because of its excellent electrical, thermal and mechanical properties. In the 1990s, nanocomposites were commercially developed and applied in applications as the second generation of filled composite [34]. Approaching the 21st century, some research on the electrical properties was carried out and after a series experiments to explore the mechanism of how nanoparticles influenced the dielectric properties [34, 35], intensive interest started to focus on these novel materials. With the development of nanoscale technology in recent decades, nanocomposites have become one of the most popular topics in dielectric research. A 250 kV DC-XLPE nanosilica-filled cable in Japan has passed the post-installation test in accordance with CIGRE TB219 and then a 45 km length was put into operation in 2012 [36] as the world’s highest voltage extruded DC cable in service at that time.

A lot of research has been done on both thermoset epoxies and thermoplastic polymers such as polyethylene, Polyimide and Polypropylene. It has been proposed that the improved insulating properties of filled polymers may be caused by the inhibited treeing. For example, inorganic fillers were commonly regarded as physical barriers to block electrical tree growth. However, so far only few studies have been carried out because filled composites are generally opaque and most standard techniques cannot reveal the internal features. Therefore a better understanding of the treeing mechanism in filled dielectrics is important for a reliable asset management.

1.2 Aim and objectives

The overall aim of this research is to investigate treeing characteristics of different dielectrics and determine why materials filled with micro- and nano-silica appear to have improved treeing resistance. As a powerful technique to study the tree structure, X-Ray Computed Tomography (XCT) is further developed for the imaging of electrical trees in filled polymers. The specific objectives are:

- To develop 3D imaging techniques to monitor tiny structural changes at consecutive treeing stages.
To visualize electrical trees in both micro- and nano-silica filled epoxy resin by using a synchrotron X-ray source.

To characterize the mechanism of tree growth in epoxy resin by combining quantified imaging results and collected PD data.

To determine how inorganic fillers impact electrical tree growth in the epoxy resin system under AC field and consider the mechanisms of treeing suppression.

To compare and explain how different particle sizes and load concentrations influence geometry and growth features of trees in filled systems.

To study if functionalized nano particles suppress the treeing and associated PD activities in epoxy system.

1.3 Methodology

Adopting the most common method to investigate electrical trees, a needle-to-plane geometry was used for experimental growth of electrical trees. Epoxy resin was selected as the base insulating materials and silica (SiO$_2$) with different particle sizes was blended into the dielectric for comparison. University College London provided silane treatment of filled particles for comparison.

Electrical trees were grown in the high-voltage (HV) laboratory under alternating current (AC) voltage with 50 Hz frequency. Optical imaging and partial discharge (PD) measurement were synchronised with the progress of tree growth. In some cases, in order to determine the 3D tree structure, the sample was imaged by XCT. Micro-XCT imaging was applied with different sources: a laboratory source in the Manchester X-ray Imaging Facility (MXIF) for unfilled samples and a synchrotron source provided by the Diamond Light Source for the imaging task in filled epoxy samples.

1.4 Main contributions

The main achievement of this research is the structure-based analysis of electrical treeing phenomenon in both unfilled and filled epoxy based dielectrics. 3D images of electrical trees in an unfilled sample have been revealed at three separate times during its growth and structural changes were extensively linked with PD data to achieve further understanding of the tree growth mechanism.

The summarized contributions in this thesis are:
An early tree growth was captured in unfilled epoxy in three consecutive stages where the whole tree structure grew only few micro-metres in length between every two images. The change of physical and chemical properties of epoxy resin materials before and after X-ray irradiation was also estimated in consideration of three imaging stages.

Electrical trees in the microsilica-filled epoxy system were imaged and reconstructed from 3D data using a synchrotron source. Geometries of electrical trees in materials with different load levels (0, 5, 15 and 30 wt %) were quantified and compared. Interactions between tree channels and micro-sized fillers were visualized in three dimensions and have been used to understand tree propagation in filled systems.

PD data were correlated with the quantified 3D structural increment by comparing the dissipated discharge energy and the evaporation energy required to form a given tree volume. The role of PD was further explained during the tree growth and it helped to understand the result collected from PD measurement.

The “fine-tree“ growth model was developed according to the specific features of the different tree types and both ‘fine trees’ and ‘dark trees’ were defined in detail. Moreover, the transition between these two tree types was also characterized by analysing the corresponding PD activities.

Both micro- and nano-filled epoxy systems were characterized in terms of treeing resistance. As an important factor for filled dielectrics, the particle dispersion was quantified and compared in different systems. The treeing mechanism in different filled systems was discussed in the context of the “fine-tree” growth model. It was proposed that the role of nanosilica could be essentially different with the microsilica in terms of the impact on the propagation of electrical trees.

The improved dielectric properties of functionalized nanosilica particles were demonstrated by the treeing experiment. Quantified dispersion suggested the optimised particle separation and its improvement were also evaluated according to its PD resistance.

1.5 Thesis outline

This thesis is organised into 8 chapters as follows:

Chapter 1: Introduction
Chapter 1: Introduction

This chapter introduces the overall background of dielectric materials used in HV applications and presents some potential challenges and opportunities. Aims and objectives are also stated.

Chapter 2: Literature review

This chapter gives a brief introduction to electrical trees in insulation materials and some published treeing models to explain the treeing mechanism. Imaging techniques are described from the literature and some recent 3D imaging results are introduced and discussed. The different dielectric properties in filled systems are summarized based on theoretical models and some challenges and inconsistencies are revealed.

Chapter 3: Experimental description

This chapter firstly describes the general approaches applied to achieve objectives and aims in this thesis. Techniques to generate electrical trees in laboratory, how to reveal tree structures in different systems and experiments designed to compare different treeing features are summarized. Then the experimental setup and facilities used for treeing tests are given in details. Both the lab-source and synchrotron-source XCT are introduced and imaging settings are presented with the sample information. Material preparation is provided with the whole procedure of manufacturing treeing samples. The silane treatment is introduced and composites mixed by different methods are compared in this chapter.

Chapter 4: Processing of SEM and XCT images

In this chapter, the reconstruction approach is firstly discussed. The reconstructed 3D tree model from micro-XCT is then measured and quantified with several structural parameters. The particle dispersion in filled materials is estimated by introducing quantitative parameters on SEM images.

Chapter 5: Correlation between PD and tree growth

This chapter introduces the 3D imaging work on one small electrical tree at three consecutive stages of growth. The influence of the exposed epoxy to the required high doses of X-ray irradiation from imaging is considered in both physical and chemical aspects. The early-stage electrical tree is characterized by both 3D structural changes and the corresponding PD data.
Chapter 6: The impact of inorganic fillers on electrical tree growth

The results of treeing breakdown tests in both unfilled and filled epoxy resin samples are summarized in this chapter. The “fine tree” growth process is identified from the optical images and characterized by the corresponding PD activities in unfilled epoxy. For filled systems, the interactions between electrical trees and embedded particles are visualized and analysed based on 3D reconstructions. Different treeing mechanisms are differentiated and compared in both micro- and nano-filled systems. The influence of the functionalized particle surface is also discussed.

Chapter 7: Discussion

This chapter discusses on the test results shown previously and proposes some treeing models with the consideration of ‘fine tree’, ‘dark tree’ and filler particles.

Chapter 8: Conclusions and further work

This chapter summarizes the main findings and conclusions of the research work presented in this thesis. Some potential opportunities and further work are identified.
2 Literature review

2.1 Introduction

This chapter introduces some related knowledge about electrical treeing phenomena from previous research. This review covers both experimental and theoretical research about how electrical trees are generated and propagated in polymeric materials. Structural characteristics of electrical trees are summarized as well as the techniques used for tree imaging. The study of electrical trees in filled systems is also introduced.

2.2 Electrical treeing

Electrical trees are regarded as a deterioration phenomenon in polymeric insulation exposed to a high level electrical stress [31]. These paths of damaged material are easily initiated at sites where defects cause excessive electrical field stress [37]. It is an important factor which seriously affects the practical lifetime of high voltage equipment. Several parameters such as voltage duration, temperature and interface quality are known to impact the electrical tree growth [7, 9, 53].

The study of electrical discharges that damage dielectrics can be tracked back to 1912, when Rayner firstly reported the activities of discharges which can result in breakdown [32]. In 1972, the announcement by Vahlstrom [38] about treeing phenomena caused a great concern. After that a wide range of studies on electrical treeing phenomenon were carried out.

2.2.1 Electrical tree types

Electrical trees can be categorized accorded to their structural characteristics. Under AC voltage at 50 Hz, three typical types of tree structure were classified in most unfilled polymers by Dissado et al [11]: branch tree, bush tree and bush-branch tree. Generally a branch tree has multiple main branches with several small branch tips, while tubule distributions are more densely packed together in the bush type. The bush-branch tree is identified by a bush tree structure but with one or more projecting branches. However according to the treeing test in XLPE by Chen et al [1], it was also found that when the
bush tree approached the earth electrode, some branch types could be also found, which met the description of the bush-branch tree. Based on that, the bush-branch tree might be also considered as a later stage of the bush tree development. Figure 2-1 shows the basic structural difference between branch trees and bush trees. Moreover, other tree shapes have been also described in the literature such as the pine tree in silicon rubber [39], stagnated tree in XLPE [1] and ‘fine tree’ in epoxy resin [24, 25].

Many researchers have pointed out that the magnitude of AC voltage applied was closely linked with the formation of specific tree structures [1, 40, 41]. The effect of voltage magnitude was demonstrated by Brown et al [42] and it was found that the branch tree was usually formed at lower voltages while a bush tree was generated at higher voltages [31]. It was also found that sometimes these two tree types could be changed from one to the other by varying applied voltages [40, 41]. On the other hand, other factors such voltage waveform, voltage frequency and temperature also influenced tree shapes [41, 43-45]. Based on the tree morphology, the lifetime of dielectrics has been further investigated. Guastavino et al [46] has showed that a branch tree type could lead to electrical breakdown more quickly than a bush structure. With the same topology and field distribution, it has been suggested that a field threshold determines the lifetime and average growth speed of electrical trees [1]: When the field is below a critical value, the propagation speed might decrease with the increasing field level because the structural features played a more important role in field determination than applied voltage.
The different treeing characteristics of branch and bush type trees might be explained by a change of conductivity. Jiang et al. [40] explained that discharges in most tree channels in branch trees were generated at tree tips which prevented the formation of a dense structure. Just like a short circuit, these conducting paths prevented PD activities along the branch channels. Bush trees channels, however were full of discharges and significantly distorted the local electric field, which lead to slower tree growth. Vaughan et al. [47] suggested an optical method to determine the conductivity of branches: generally conducting channels contained graphitic carbon so they were observed as dark under reflected light and non-conducting branches appeared as white due to the existing of fluorescent decomposition residues [7]. An example is shown in Figure 2-2. As shown in Figure 2-3, the light emission from partial discharges collected by a sensitive charge-coupled device (CCD) camera also supported the conductivity concept in different tree structures [17]. With the help of a partial discharge model, it was found that
the PD amplitude in a conducting tree was much smaller than that in non-conducting one [48]. It was also suggested that the glass transition temperature T_g was a key factor for both types of trees: Conducting structures were found to be generated at room temperatures (20 °C) in the glassy epoxy (T_g = 55 °C), while non-conducting trees were seen in the flexible epoxy resin (T_g = -8 °C) [17].

### 2.2.2 Treeing mechanism

#### 2.2.2.1 Treeing stages

In order to understand how electrical treeing happens in insulation materials, tree growth models have been proposed and developed for decades. According to specific growth rates, three basic stages were distinguished by Dissado et al [11] as shown in Figure 2-4.

![Figure 2-4: Typical electrical tree growth stages according to growth rate [11].](image)

The inception stage is the time taken from energizing to the formation of the early tree (normally defined by a tree length of ~10 µm [11]). After initiation, the electrical tree will continue to propagate further towards the earth electrode with different shapes. When the tree channel is about to reach the ground, the runaway stage takes place. Wider branches will generate and finally lead to the dielectric failure. Iddrissu et al [25] divided the whole treeing process into 5 stages with the consideration of tree structures and PD patterns: tree initiation, forward tree growth, fine tree growth, fine tree darkening and
reverse tree growth. Figure 2-5 shows the corresponding PD features in each stage. It was found that the classification of PD activities was also consistent with the three-stage model if the stage 3 and stage 4 in Figure 2-5 were merged.

![Figure 2-5: Five stages of tree growth characterized by partial discharge activities in epoxy resin](image)

**2.2.2.2 Tree initiation**

The inception of an electrical tree is mainly considered as the consequence of high electric field stresses from impurities, voids or artificial defects in a polymer [49]. For most treeing tests in laboratories, electron injection from the metallic electrode was thought to be the pre-initiation process, and once enough energy was injected then bond scission and molecular excitation could happen in the surrounding dielectrics, which resulted in chemical degradation [50, 51]. The cause of the first tree appearance has been suggested to be due to various mechanisms. Shimizu et al [49] suggested that the void formation cause by degradation processes could finally result in tree initiation under an AC source and this was generally a long-term initiation (>> 1s). While under higher impulse and DC voltages, the initiation time was shorter (typically < 1s) and the mechanism should be attributed to local avalanches in the solid (such as PE and XLPE). Wu et al [52] further explained the initiation mechanism by the generation of new charge traps enabling great transfer of energy and then resulting in the ignition of electron
avalanches especially in epoxy resins. Tanaka [53] also summarized the possible initiation mechanism as the combination of electrical, mechanical and thermal causes.

From experimental results, the tree initiation time was found to be influenced by several factors. Shimizu et al [54] compared PE samples with different degassing conditions and found the time to tree initiation was clearly improved by degassing, which was explained by the removal of absorbed oxygen in the polymers. Auckland et al [55] found that the geometry of the electrode (radii of the needle tip) could influence the inception time in polyester and epoxy resins. From comparison it was found that there was no huge difference at 10 kV, 50 Hz when the tip radii increased from 1 µm to 3 µm while 5 µm and 10 µm could result in much longer inception times. This phenomenon could be explained by the shielding effect caused by space charges, which electrostatically shield the sharp needle tip [56]. The external factors such as temperature also changed the initiation characteristic. Du et al [57] carried out treeing tests with an ambient temperature from 30 °C to 90 °C to investigate the effect of temperature on initiation time in silicon rubbers. He revealed two important observations: The tree inception time increased with the rising temperature and the probability of tree initiation was higher at lower temperatures.

2.2.2.3 Tree growth

After initiation, tree propagation is commonly thought to be essentially linked with PD activity [16]. The characteristics of PDs during the tree inception and tree propagation stage have been studied by observing luminous phenomenon. It was found that the light emission recorded at these two stages is different: in the early initiation stage only low-level emission is detected at the needle tip without an obvious cavity [58] but more intense light was found with the appearance of a cavity [59]. The initial luminescence phenomenon was considered to result from charge injection [60] and the latter one was mainly from partial discharges in microtubules [58]. Propagation of PDs and their nature was associated with the tree shape as mentioned before: in branch trees partial discharges mainly occurred at tree tips while PDs could be observed near the pin electrode and propagating along branches when a bush tree formed [61]. Figure 2-6 shows the PD characteristics summarized by Laurent et al [16].

The growth of branch trees was characterized by the decreased PD number and magnitude after initiation. By combining with the luminous test as shown in Figure 2-7,
the same author proposed that most of charges could be trapped on the channel wall and generated the opposite local electric field against the original field, which finally lead to the intensive PD only at the tree tips [16]. The PD extinction phenomenon during the tree growth might be caused by conducting products newly generated in the gaseous form [62]. The similar PD extinction feature during the tree growth was also found by Bahder [63] but he attribute this phenomenon to the result from the increased pressure after material decomposition. As for a bush tree, discharges were generated along the whole channel and there was no obvious PD extinction when tree propagated. Instead, most the PD characteristics were roughly linear with the increment of the tree length. This is consistent with later literature [3, 64].

![Graph 1](image1)

![Graph 2](image2)

**Figure 2-6:** Discharge characteristics for (top) a branch-type tree at 10 kV and (bottom) a bush-type tree in polyethylene at 20 kV [16].

There are some other tools to define the tree growth characteristics based on experimental results. The PD pattern extracted from Phase-resolved Partial Discharge (PRPD) is also widely studied to characterize tree propagation [3, 65-67]. Lv et al [3] found there were two typical PD patterns during the early treeing stage in epoxy resin:
‘wing-like’ PD and ‘turtle-like’ PD (as shown in Figure 2-8). It was considered that the ‘wing-like’ PDs with different magnitudes corresponded to the different tree branch lengths and these PDs were mostly directly ejected from the needle tip. The larger PDs in the ‘wing-like’ pattern could directly reach the tree tip and lead to further tree growth. ‘Turtle-like’ PDs generally contained PDs with similar magnitudes independent of the voltage phase. This PD pattern might be generated from junctions where the new tree channel was short. It was considered that the main trunk of an electrical tree is conducting so that most PDs only occurred near the tree tip instead of the main branch.

In addition to the PRPD pattern, Pulse Sequence Analysis (PSA) was also applied to characterise PD data [4, 19, 68, 69]. PSA is an analytical method firstly proposed by Patsch et al [19] and it was used to evaluate the voltage difference between consecutive PD signals. Patsch et al [19] firstly proposed simulated sequential PDs for three types of defects as shown in Figure 2-9 and the specific PD characteristics in different defects were identified by a $dV$-$dV$ pattern which reflects the change of the voltage between every two consecutive discharges.

Figure 2-7: Luminous activities in the (top) branch tree structure and (bottom) bush tree structure [16].
Lv et al [4] further linked the PSA with the laboratory tests and found there were two pairs of voltage differences: $dV_1$ with smaller absolute values occurring in the same half cycle and $dV_2$ with higher value corresponded to the voltage difference between consecutive events in two half cycles. These two characteristic voltage differences increased with applied voltage and decreased with time. Figure 2-10 shows some examples of the $dV$-$dV$ patterns of PDs as well as the corresponding PRPD patterns.
It can be found that when the applied voltage is too low, there is no treeing phenomenon observed so there are only few PDs in each voltage cycle, which leads to a straight line pattern. Once the applied voltage is increased to 10 kV, the PRPD pattern shows a typical ‘wing-like’ shape which also means the electrical tree has started to grow [3]. The $dV$-$dV$ pattern is a combination of a rhombus route and a hexagon at the beginning of the energization. After treeing for 480 s, both PRPD pattern and the $dV$-$dV$ pattern change their shapes. The previous ‘wing-like’ PD turns into ‘turtle-like’ pattern and a hook-like hexagon appears on the $dV$-$dV$ plot containing six points.

As shown in Figure 2-11, the author [4] then proposed three typical PD combinations during tree growth referring to experiment results. It can be found that the number of PDs in each cycle is regular (2, 4 and 6) and essentially determined by the applied voltage. On the other hand, two more apparent parameters were given by:

$$V_j = \frac{(dV_1 + dV_2)}{2}$$

(1)
where $V_I$ was the PD inception voltage and $V_X$ was the extinction voltage. From the experimental results it was also found $V_I$ could decrease in time while $V_X$ almost kept constant, which means the inception threshold of PD event during the tree growth can become lower [4].

As with the initiation time, the tree growth time (time to breakdown) is affected by different factors. The applied voltage mattered apparently due to the changed electric field at the needle tip as it controlled PD activity (PD magnitude and PD number [70]) so several treeing experiments [31, 41, 71, 72] with different epoxy materials showed similar results: the breakdown time monotonically decreased when the voltage was increased. However Fothergill et al [72] suggested that once the voltage was above a certain stress level the tree growth time could be abruptly increased due to the change of tree structure from branch to bush. Chen’s [1] treeing data were also consistent with this finding but he also found that the breakdown time of bush tree can be then significantly decreased with the increasing voltage level as shown in Figure 2-12.

The ambient temperature did not only affect tree initiation but was also an important factor for the tree growth. It was found that the time to breakdown in epoxy resin at 20 °C could be more than 4 times longer than 80 °C however the structural change of the electrical tree also happened with the varying temperature [21]. Figure 2-13 reveals a correlation between the tree growth rate and the temperature and three characteristic
zones could be distinguished. The growth rate in the glass state from 20 °C to 40 °C shows little dependence on the temperature while in the flexible state (70 °C to 85 °C) it becomes more sensitive to the temperature change. This phenomenon may be explained by the molecular reconfiguration of the polymeric matrix in the flexible state.

![Figure 2-12: Electrical tree growth in XLPE with different voltages: (a) 13 kV; (b) 15 kV; (c) 18 kV; (d) 23 kV; (e) 27 kV. Arranged by breakdown time: c>b>a>d>e. f, g, h, i, j show the respective fractal dimension of the optically observed tree structure in (a), (b), (c), (d) and (e) [1].](image)

![Figure 2-13: Average tree growth rate as a function of the temperature in CY1301 epoxy resin at 13.5 kV, 50 Hz [21].](image)

### 2.2.2.4 Pre-breakdown

When an electrical tree channel approaches the counter electrode, dielectric breakdown may then occur. Partial discharge sometimes does not immediately cause insulation breakdown in polymers but it generates erosion and an electrical tree eventually does cause electrical failure [11]. Breakdown is most likely characterised by the appearance of huge and conducting breakdown channels but the time required for the formation of these channels is not found to be consistent [16, 73, 74]. Zheng [24] and Iddrissu [25] revealed
a newly characterised tree type and they called it ‘fine tree’ in the epoxy system. The ‘fine tree’ could be either initiated from the needle tip directly or from the end of the traditional tree structures. When these thin and slight tree structures grew to reach the earth electrode, the electrical breakdown would not immediately happen. Budenstein [75] suggested that there were some return streamers causing the final dielectric failure by enlarging the tree channel and also increasing the conductivity. Just recently, some literature has reported the appearance of ‘reverse tree’ in the final breakdown stage of epoxy resin under AC voltages [24, 25, 76]. As shown in Figure 2-14, the ‘reverse tree’ was identified by the reverse propagation of the tree structure from the plane electrode towards the needle electrode and the ‘reverse tree’ could be classified as a non-conducting tree according to its PD features [24]. However the mechanisms for the formation of a ‘reverse tree’ and a ‘fine tree’ are still unknown.

Figure 2-14: A reverse tree observed in the epoxy resin at 15 kV, 50 Hz for (left) 71 minutes and (right) 77 minutes [24].

2.3 Electrical tree imaging

2.3.1 Optical imaging

Most characteristics of the tree growth come from 2D images. Generally, the structure of electrical trees can be directly identified by normal imaging technology if the polymer material is translucent such as unfilled epoxy resin, polyethylene or silicone rubber [2, 7, 77, 78]. The most frequent way of imaging, the optical method, is non-destructive and flexible due to its availability and convenience. The requirement of optical imaging is easy to achieve and samples can be designed in various geometries, for example thin sheets and small cubes. In most cases, only a smooth sample surface is required to ensure low scattering of light. In an experimental setup, optical imaging systems typically
Chapter 2: Literature review

involve a charge coupled device (CCD), magnification lens and background light source. An example is shown in Figure 2-15. DC illumination is often used instead of AC power to avoid the periodic change of light intensity [2].

![Diagram](image)

Figure 2-15: Real-time observation system for the AC and DC treeing test of transparent epoxy samples [2].

Many results for characterisation of electrical trees have been based on 2D optical images because it is a most effective technique to record the nature of the tree in real time using parameters such as tree length, channel radius and growth rate [16, 39, 79-82]. In order to accurately describe the tree structure, fractal dimension was introduced into the analytical work. Fractal dimension was firstly introduced by Mandelbrot [83] and recent researchers developed and used it in the dielectric area [46, 65], by which the shape of electrical trees could be described quantitatively. There are several methods to calculate fractal dimension $D_f$ such as the box-counting method, the power spectrum and the fractal measure relations [84]. The box-counting dimension, also known as Minkowski dimension, is commonly used to determine the fractal dimension value by dividing the object into small spaces like cubes or circles. The relationship can be defined in the equation:

$$N(r) \propto r^{-D_f}$$  \hspace{1cm} (3)

Where $r$ is the box size and $N(r)$ stands for the number of boxes required to cover the objects. By rearranging the equation, the value of $D_f$ can be calculated by:

$$D_f = - \frac{\log N(r)}{\log r}$$  \hspace{1cm} (4)
Electrical trees are regarded as fractal structures [85] and some similarities in terms of fractal geometry have been found [84]. Three tree shapes have been found in XLPE as shown in Figure 2-16 and each of them gave a corresponding fractal dimension by using ‘box-counting’ method. It was suggested that for 2D fractal dimension measurement, the fractal dimension parameter $D_f$ of electrical tree structure always varied between 1 and 2 [86]. Bush-type structures have a relatively high $D_f$ which is normally between 1.7 and 2 [1]. However, it was also revealed that sometimes the difference between branch-type tree and bush-types tree was indistinct [87] and the observed structure could be influenced by the observation angle of projections, which lead to different values of fractal dimension [29]. Hence the fractal analysis of electrical trees measured from 2D projected images is still limited because the tree extension is actually in three dimensions.

Electrical trees in filled compounds are a challenge to observe especially for microcomposites because of the opaque nature of the material. Kurnianto et al [27, 88] developed thin ‘leaf’ samples with 200 µm thickness to overcome the challenge. As shown in Figure 2-17, it proved the possibility of using optical microscopy to image the propagation of electrical trees in a micro-filled epoxy system (50 µm particle size and 30 wt%). However the evidence provided by 2D images about the tree propagation might contain overlapping structures concerning the interaction between tree channels and fillers so it was still not clear whether the growing tree channel was actually touching the filler particle’s surfaces or just going around them.

Figure 2-16: Three kinds of tree structures varying from (a) a branch tree to (c) a bush tree with specific fractal dimensions [3].
Most tree structures reported in the literature were observed and defined by optical techniques whose resolution is normally about few micrometres. However due to the limitation of the resolution and the transparency of the objects, the actual resolution could be poorer so some features in sub-micro size or nano size will be lost in the optical microscopy.

2.3.2 Electron Microscopy (EM)

Electron microscopy (EM) uses the electron beam as the illumination source, which provides much stronger power of revealing micro objects than light microscopy. There are two commonly used imaging techniques: Scanning Electron Microscopy (SEM) and Transmission Electron microscopy (TEM). TEM uses transmitted electrons (the sample thickness is usually tens of nanometres) to generate an images so it is widely used in material science to reveal the inner features [89]. SEM is achieved by collecting reflected electrons from the sample surface so it provides more information on the surface so in order to maintain the completeness of the tree structure, SEM is often used for the study of electrical treeing. Chen et al [7] showed a comparison between SEM micrographs of a conducting channel and a non-conducting channel in XLPE. As shown in Figure 2-18, the conducting channel, or carbonized channel, has some clear deposits (regarded as graphitic carbon) inside of the channel but the non-conducting one gave a relatively smooth channel. These carbon residues were considered as part of a conducting channel which stimulated the discharge phenomenon to occur at the end of channels instead of within the main trunk [17].
SEM also provided a good way to analyse the chemical characterization of electrical trees by using energy dispersive X-ray (EDX) analysis which allowed the identification of elements and their relative proportions. The detector in the EDX setup can measure the x-ray energy from the interaction between the electron beam and the sample. Due to the fundamental principle that each element has unique set of peaks on its X-ray spectrum, the abundance of specific elements can be determined. Some elemental analysis has been carried out into the study of electrical damage in polymers [9, 90, 91]. Du et al [9] used EDX to analyse the elemental difference of polypropylene samples under different ambient temperature. As shown in Figure 2-19, both carbon and gold (came from the coating on the sample surface) are detected in all samples. It was found that the oxygen atom is additionally found in the breakdown channels generated from 70 °C to 130 °C.

The conventional SEM has been also widely used for the treeing studies in filled systems to investigate tree propagation and particle dispersion [30, 92-96]. Iizuka et al [23] used SEM to study internal features of electrical trees of epoxy/silica nanocomposites. Figure 2-20 shows the scanned image along the direction of the tree growth. Two distinct regions were observed: one is the large region consisting voids and nano particles, another one is the thin tree channel [23]. The author postulated that an initial tree was firstly formed between neighbouring nano particles. SEM imaging can also be used for cross-sections of the tree channels where treeing holes with different diameters are able to be found in both epoxy and PE samples [30, 97].
The resolution of SEM imaging could be less than 1 nm in the case of imaging conducting materials while for polymeric dielectrics the resolution might be decreased to tens of nanometres [89]. In contrast to SEM, electrons in TEM are transmitted through the specimen [89]. As a result, the sample must be prepared as a very thin slice (normally ~100 nm) to ensure electrons can pass through the slice. This technique is mainly used for imaging below nanometres size [98] so some micro structures are easily found by using this technique, for example the very early degradation tubes just after tree initiation [99]. However, due to the limitation of the sample thickness, only few tree channels might appear in the sample so this technique is mainly used for the observation of nano-sized fillers [100-102].

Figure 2-19: EDX analysis of polypropylene samples: (a) a raw surface without degradation; (b) A breakdown channel formed at 17 kV, 70 °C; (c) a breakdown channel formed at 17 kV, 130 °C [9, 10].
2.3.3 3D imaging

2.3.3.1 Optical techniques

In the past, 3D imaging on electrical trees was achieved by using the Serial Section Method (SSM) and the Computerized Tomography Method (CTM) [29, 103] based on optical microscopy. The tomography was achieved by taking images every 2° from 0° to 180° and then reconstructed based on the 90 projections. SSM is a destructive method and the sample is sliced in 10 µm thickness sections, until the whole treeing area was cut. A 3D tree model was then established by stacking up these cross sections. By using these two imaging systems shown in Figure 2-21 (a) and (b), Uehara et al [29] successfully reconstructed the 3D electrical tree structure in XLPE. It was found that SSM could provide better image quality even on the denser tree structure. Based on the 3D reconstruction, different tree patterns could be clearly characterized by fractal dimension and it was found a fractal dimension value of 2 was a threshold value for different tree types as well as the characteristics of tree propagation [29]. However according to the limited resolution of the optical camera, most of micro details were still lost and it failed to observe the inner structural features of electrical trees.

For a better definition and identification of electrical trees, 3D fractal dimension measurement has been carried out. Figure 2-22 showed the correlation of between the 3D fractal dimension and tree structures obtained from the computerized tomography method (CTM) and serial sectioning method (SSM) [104]. It suggested that in the three-dimensional world, bush-type trees displayed a 3D fractal dimension exceeding 2. The most recent 3D tree models from XCT imaging also supported this hypothesis [105].
2.3.3.2 X-ray computed tomography (XCT): laboratory imaging

X-ray tomography is a non-destructive technique for characterizing specific areas of a scanned object. The method has been widely used in the biological and medical fields [106] and it is also of great help in material science. The tomography technique allows observing the object in a 3D view and developing numerical models based on 3D

Figure 2-21: 3D optical techniques achieved by (a) the CTM system; (b) The SSM system. Image (c) and (d) show cross sections of the same bush-type tree in XLPE at 16 kV by using CTM and SSM, the thickness of each slice was 10 µm [29].

Figure 2-22: Theoretical fit to voltage dependent tree propagation data in polyethylene at f = 50 Hz. The assumed tree fractal dimensions necessary to produce the fitted line are given by the circular symbols [84].
geometries. Just as a 2D image is composed of pixels, a 3D tomographic image is assembled from voxels. When X-rays pass through each voxel, the radiography is able to measure the transmission features corresponding to the X-ray attenuation. The 3D volume is based on a series of 2D projections that are taken when the sample is rotated in front of the X-ray source (as shown in Figure 2-23). The basic imaging system always includes an X-ray source, a turntable for supporting and rotating the sample and a detector to measure the intensity. After enough time periods for exposure, a large number of radiographs are collected and computed to create the 3D replica by using specific algorithms. With the continuous development of XCT sources and facilities, the cone beam imaging system now can realize the very accurate resolution even down to sub-micro size [107, 108].

![Figure 2-23: Principle schematic of the cone-beam X-ray tomography [109].](image)

Sometimes when imaging objects with little attenuation in low contrast materials such as polymers, phase contrast can be applied to greatly enhance edge differences [110, 111]. In phase contrast imaging, the varying intensity is determined by the phase shift and its sensitivity can be three orders larger than the traditional absorption contrast imaging system [112]. An example of the improvement on image quality using phase contrast is shown in Figure 2-24.
From 2011 to 2015, Schurch et al [5, 105] started to develop the 3D XCT technique for the study of electrical trees. Some well-established imaging systems were proved to successfully image electrical trees in the University of Manchester. Table 2-1 summarizes most of experiment details about the imaged samples.

Table 2-1: Description of imaged electrical trees in unfilled epoxy resin: U1 was energized at 7.5 – 9 kV for 6h; U2 and U3 were energized at 10 kV for few seconds; U4 was energized at 8.5 kV for 3 h; U5 was energized at 10 -12 kV for 40 minutes; U6 was energized at 10 kV for 1. 8h; U7 was energized at 10 kV for 5.5h. All XCT and Series Blocked Face Scanning Electron Microscopy (SBFSEM) techniques were provided by University of Manchester [14].

<table>
<thead>
<tr>
<th>Sample</th>
<th>Description</th>
<th>XCT</th>
</tr>
</thead>
<tbody>
<tr>
<td>U1</td>
<td>Bush tree in transparent polymer</td>
<td>Micro-XCT &amp; SBFSEM</td>
</tr>
<tr>
<td>U2</td>
<td>Branch tree in glassy epoxy</td>
<td>Nano-XCT &amp; SBFSEM</td>
</tr>
<tr>
<td>U3</td>
<td>Small branch tree in glassy epoxy</td>
<td>Micro- &amp; Nano-XCT &amp; SBFSEM</td>
</tr>
<tr>
<td>U4</td>
<td>Low density bush tree in transparent polymer</td>
<td>Micro-XCT &amp; Synchrotron</td>
</tr>
<tr>
<td>U5</td>
<td>Small dense bush tree in flexible epoxy</td>
<td>Micro-XCT &amp; SBFSEM</td>
</tr>
<tr>
<td>U6</td>
<td>Big branch tree in glassy epoxy</td>
<td>Synchrotron XCT</td>
</tr>
<tr>
<td>U7</td>
<td>Two stages bush tree in flexible epoxy</td>
<td>Micro-XCT</td>
</tr>
</tbody>
</table>

In the table above, ‘glassy epoxy’ is the mixture of Araldite LY5052 and Ardur HY5052 whose glass transition temperature is 120-130 °C; The ‘flexible epoxy’ has a 25-29 °C glass transition temperature and it is made up by mixing Araldite CY221 and Aradur HY2966. As shown in Figure 2-25, all samples were processed to a 2-3 mm diameter cylinder containing treeing regions to meet the requirement of imaging techniques.
The Zeiss Xradia Versa 520 system was used for the micro-XCT in Manchester X-ray Imaging Facility (MXIF) and as a sub-micro resolution instrument, it provided up to 20X magnification lens with a resolution from 5 to 100 μm [113]. The whole scanning period took about 22 hours and 1501 radiographs were captured in total. As for Nano-XCT, an Ultra 810 system was used. There were 721 projections collected over 24 hours and the corresponding voxel size was 64 nm.

After reconstruction, the comparison between Micro-XCT and Nano-XCT was carried out for the sample U3 and U5. As shown in Figure 2-26, the small tree created in the glassy epoxy sample could be regarded as a traditional ‘branch tree’. It was found that the replica generated from nano-XCT had a similar tree shape with the one from micro-XCT but the diameter of the tree channel was at least 3 times thinner. It was explained by
the unsatisfactory sample geometry which resulted in a poor signal-to-noise ratio and so influenced the image quality negatively in this case [5].

The two-stage tree imaging was achieved in sample U7 as shown in Figure 2-27. Both stages were energized and imaged at the exactly same settings except the treeing times of 5 minutes for the first stage and a further 30 minutes for the second stage. The micro-XCT took 21 hours in each stage for 10001 radiographs respectively with a pixel size of 0.45 µm. The tree length in the 2nd stage was 605 µm and it was 7.6 times longer than that in the 1st stage. It was found that both of 3D tree renderings showed traditional bush tree features and the 3D fractal dimension remained similar (2.12 for the 1st stage and 2.18 for the 2nd stage) even through the number of nodes was 22 times at the 2nd stage, which suggests the relatively same fractal nature during the bush tree growth.

2.3.3.3 X-ray computed tomography (XCT): synchrotron imaging

The Diamond Light Source synchrotron is a large machine with half a kilometre in circumference and it is mainly designed to produce intense beams of X-rays, ultraviolet light and infrared [13]. Because of the high flux and coherence, X-rays from large synchrotron facilities can be applied for rapid imaging. The simplified scheme of a synchrotron in Diamond Light Source (UK national synchrotron science facility) is presented in Figure 2-28. As shown, electrons are generated in an electron gun and then accelerated by the linear accelerator (Linac). After further acceleration by the booster synchrotron, electrons are injected into the storage ring where these electrons travel.
around at almost the speed of light. With the direction of a magnetic field, the high energy produced from electrons at relativistic speeds can be converted into photons (X-rays). Radiation X-rays are emitted from the channelled electrons and distributed into beamlines where people can carry out experiments inside enclosed space.

![Typical synchrotron scheme in Diamond Light Source](image1)

Figure 2-28: Typical synchrotron scheme in Diamond Light Source [13].

![Schematic of the parallel beam geometry of synchrotron XCT imaging system](image2)

Figure 2-29: Schematic of the parallel beam geometry of synchrotron XCT imaging system [5].

The synchrotron beamline has a parallel beam (as shown in Figure 2-29) of high brilliance and the high flux x-ray beams can ensure a long-distance scanning without loss of intensity. This results in better contrast of objects and fewer artefacts [114]. A long distance between sample and detector is set in synchrotron XCT system so as to involve a big field of view from the sample even for a sub-micron resolution, which is not available in laboratory experiments.

Schurch et al [5] applied Synchrotron facilities in the Diamond Light Source to image electrical trees. As shown in Table 2-1, U4 and U6 were imaged by using the X-ray source in I13 Manchester beamline. For Synchrotron imaging, the whole scanning time was reduced to 2 hours. There were 1891 projections scanned with a voxel size of 0.45 µm. As shown in Figure 2-30 and Figure 2-31, reconstructed 3D trees of U4 from the lab
source and synchrotron source were compared. By comparing the number of tree channels in each slice, it was found that even with the larger voxel size, the 3D reconstruction of the tree structure from the synchrotron source could still collect more detail than that with laboratory source. More disconnected branches were also observed in the synchrotron image but the reason for these was still not clear. The possible cause might be that these disconnected channels had different properties and so showed with worse contrast.

![XCT (Laboratory) vs. XCT (Synchrotron)](image)

Figure 2-30: Low density bush tree U4 reconstructed from laboratory source (left) and synchrotron source (right). U4 was energized at 8.5 kV for 3 h [53].

![Number of tree channel counted](image)

Figure 2-31: Number of tree channel counted from laboratory XCT (black) and synchrotron XCT (red) in each slice as the distance increased from the needle tip [53].

The synchrotron XCT was also applied to filled epoxy systems [115]. However the results suggested that even with a synchrotron source, it was still challenging to obtain the complete tree structures in micro-silica filled epoxy. Roger’s results showed that only part of the tree structure (~100 μm from the needle tip) could be revealed by synchrotron micro-XCT imaging (11-13 keV energy and 0.45 μm pixel size) but as the concentration increased from 5 wt% to 20 wt%, most of details were lost. The core problem was that
the contrast between the tree channel and the epoxy was too weak to identify and so significantly increased the difficulty of the later reconstruction.

Synchrotron imaging was also done by Pallon et al [116] in Paul Scherrer Institute (PSI), Switzerland. In this case the electrical tree was grown at -45 kV DC and 40 kV lightning impulses in Low-density polyethylene. The synchrotron source in PSI provided a resolution of 2 nm ptychographic tomography which was the most accurate 3D imaging technique so far in terms of treeing study. The whole tree structure could be well reconstructed while some isolated bits were still found, which may suggest the existence of the disconnections in the tree structure. The authors proposed these isolated features as pre-treeing bits of degradation and would be connected to the main trunk as the treeing proceeded. However this theory is still tentative and more work is required to clearly interpret such special structures.

2.3.3.4 Other 3D techniques

Recent development of SEM imaging has led to the 3D EM technique achieving the automatic process for large voxel volumes. Series Blocked Face Scanning Electron Microscopy (SBFSEM) is introduced as an automated block-face imaging combing with Backscattered Electron (BSE) so that high-resolution 3D reconstruction can be realized by the stack of EM images. This 3D imaging approach now can be achieved using the commercial Gatan 3View system in an FEI Quanta 250 Field Emission Gun Scanning Electron Microscopy (FEGSEM) in which phase contrast imaging can be used to generate high-resolution images [20]. The mechanism of the Gatan X-ray Ultra Microscope (XuM) [20, 115], which is operating in the University of Manchester, is shown in Figure 2-32. FEGSEM with variable pressure mode is also available in the Gatan system so the specimen is not only limited to conducting materials and also no conducting coating is needed on the sample surface. The resolution of SBFSEM can be below 50 nm.

SBFSEM is a destructive technology because all the data are acquired by serial sectioning by ultra microtomy (usually at 0.1 mm/slice) inside a low-vacuum chamber. The schematic procedure is shown in Figure 2-33. Damage can be generated during the ultra-microtomy such as knife marks during the cutting (named as ‘chatter’) and tracks formed from plucking out hard particles from a soft matrix [20]. Factors such as cutting
angle and cutting speed should be well controlled according the different catalogues of the material.

![Diagram](image)

Figure 2-32: Schematic diagram of Gatan XuM system located within the FEI Quanta250 SEM chamber, X-ray target arm, specimen stage and camera [20].

![Diagram](image)

Figure 2-33: A schematic of a sliced sample prepared for SBFSEM [14].

Just as shown in Table 2-1, U1, U2, U3 and U5 used SBFSEM after X-ray irradiation [14]. Here sample U3 is selected to compare the reconstructive ability of Micro-XCT and SBFSEM because of its excellent image quality. After the laboratory source XCT imaging, U3 was placed into SBFSEM chamber (FEI Quanta 250 in the University of Manchester). For 3D SEM technology, a smaller cube whose edges should be less than 1mm was needed according to the microtomy window size and the metal needle was removed from the sample to avoid damaging the slicing knife. The FEI Quanta FEG 250 Instrument offered more accurate pixel size which could reach to about 5 nm at 1.5 kV accelerating voltage [37]. For detailed sample scanning, the scanning cycle lasted more
than 20 hours and finally there were 1000+ scanning slices. As shown in Figure 2-34, by comparing with micro-XCT, the tree model scanned by SBFSEM retains a similar tree shape but many finer branches were revealed appearing on the main trunk than seen in the structure obtained from XCT.

As a direct stack of SEM images, SBFSEM gave an excellent resolution and revealed more morphological features for electrical tree imaging. While, due to its destructive mechanism, it might be not a good choice for a highly degraded region such as dense bush trees and heavily carbonized trees since slicing process could destroy fragile matrices. In addition, filled samples were shown to be not suitable for SBFSEM imaging either because the filler particles (silica) are harder than the epoxy resin, so that the microtome knife could push the filler and leave artefacts (as marked in Figure 2-35) which may confuse the reconstruction of the real tree structure [115].

Figure 2-34: Small branch tree in epoxy resin (U3: 10 kV energized for few seconds) reconstructed by Micro-XCT (left) and SBFSEM (right) [5].

Figure 2-35: Slice of sample using SBFSEM. Red circled: the artefact due to the slicing generated around the filler [101].
Another technique is Laser Scanning Confocal Microscopy (LSCM) which can provide non-contact and high-accuracy surface measurements with excellent resolution. The main advantage of this technique is that it can be used for measurement on rough surfaces. With a small-size laser beam, the imaging system can collect more accurate details of the surface in comparison with a conventional microscope and it also provides the ability to reconstruct objects with different depths by collecting fluorescence from the source and then to create 3D replica models. Moreu et al [117] applied LSCM to image water trees and the result showed great resolution to image small objects about 0.22 µm. Erozo et al [15] used Laser Scanning Microscopy to characterise the degradation channel at the interface between epoxy and silicon rubbers. The topography of treeing region is shown in Figure 2-36. However, the object can be only scanned over a few micrometre thicknesses so some deeper objects may be hidden.

![Figure 2-36: 3D characterization of the epoxy surface roughness by using LSCM on the silicon rubber/epoxy interface which was energized at 32 kV for 96 hours from a wire electrode [15].](image)

### 2.4 Electrical treeing in filled systems

#### 2.4.1 Electrical treeing in micro-filled system

The addition of fillers has been shown to improve some properties including cost and the treeing resistance over the neat polymers [27, 88]. As the first generation of filled dielectrics, microcomposites have been applied in industries for several decades. Factors such as particle size and loading rates can influence the physical properties of the compounds. Researchers have revealed that the fracture toughness of silica filled epoxy
composites would be significantly affected by the particle size [118, 119]. On the other hand, in an electrical treeing study, Fujita et al [120] found that the use of smaller micro-size fillers (5 µm in his experiments) resulted in higher breakdown voltage than bigger fillers (500 µm) and ~30 wt% micro-composite had the maximum electrical strength.

Kurnianto et al [27, 88, 121] did a series of tests and found a higher filler concentration tended to have better treeing resistance at the early treeing stage as shown in Figure 2-37. The fractal dimension of the tree determined by 2D optical images was also observed to rise when more fillers were embedded.

![Figure 2-37: Electrical tree growth in microcomposites with the filler (50 µm silica) concentration from 5 phr to 20 phr (phr: parts of silica per hundred parts of resin) [27].](image)

In order to understand how the filler changed the tree growth, the interaction between the electrical tree and filled particles has been considered. The most popular theory is that the silica filler works as an obstruction to the propagation of electrical trees [122]. As shown in Figure 2-38, Kurnianto et al [27] took an optical image in a 200 µm leaf sample and found that the propagation direction of branches was changed when encountering filler particles and also the number of branches increased with filler concentration. Kurnianto explained this phenomenon by the increased interfaces of filler-resin which branches tended to grow along locally. Moreover, the electric field distribution simulated by Chen [30] also supported this treeing model. Because of the different electrical permittivity between silica particles and the bulk epoxy resin, the local field would be significantly distorted around the filler particle as shown in Figure 2-39, which may guide the tree propagation toward the particle. High electrical field becomes easier to gather around
particles so finally the tree channels tend to extend along the surface of particles. It was proposed that the effect of interfaces was the most important factor in the micro-filled epoxy resin but the experimental result shown in [123] did not show significant difference in the 1.6 µm microsilica filled epoxy composites even with silane treatment. It should be noted that images taken by optical method such as Figure 2-38 is actually projections which may not show the real interaction between tree channel and fillers. At the same time the propagation of the tree might not only a field driven procedure but may be combined with other inducements such as mechanical forces [124, 125], so only considering the local electrical field may be not enough to explain the treeing mechanism.

Figure 2-38: Image of trees in filled sample (left) and illustration of tree propagation (right) [27].

Figure 2-39: Field distribution for micro-silica filled samples: the tip radius of the needle is 5 µm and the micro silica filler with the diameter of 1 µm [30].
2.4.2 Electrical treeing in nano-filled system

With the fast development of nanotechnology, materials with nano fillers are gradually considered as a new innovation of insulation systems. Even though nanodielectrics in power engineering have been rarely applied into industry so far, much more research work has been published in nanocomposites rather than microcomposites.

Several kinds of nano particles have been tested in dielectrics such as ZnO, TiO$_2$, SiO$_2$ and Al$_2$O$_3$ [126-130] and their potential applications are considered to be varied [131-133]. An essential difference between the nanocomposite and the microcomposite is the interface volume as shown in Figure 2-40. With a similar content (wt%) in the composite, nano fillers have much larger surface interacting with polymer matrices and generate more nano-sized structuration. As a result, it is very important to understand the mechanism of how the nano particle works in dielectrics. The most commonly agreed concept in nanocomposites is the multi-core model established by Tanaka in 2005 [28]. As shown in Figure 2-41, a multi-layer structure of the interaction zones contains: a bonded layer, a bound layer and a loose layer. The first layer is tightly bonded to the particle surface if a coupling agent is applied. The second layer normally has the thickness of 2 to 9 nm and it contains polymer chains which strongly interact with the particle. The loose layer is a weakly coupled region and it is characterized by different chain conformation, mobility or even free volume, which may be ascribed to the less stoichiometrically crosslinked layer. A Gouy-Chapman diffuse layer may additionally surround on these three layers. This model mainly describes the physico-chemical and electrical structure of the interface regions in nanocomposites [28].
The improvement from pure material or microcomposite to nanocomposite could be comprehensive in all properties such as mechanical (e.g. tensile strength, bending strength, fatigue), thermal (e.g. heat decomposition temperature, weight deflection temperature) and electrical properties (dielectric loss $\tan \delta$, permittivity, breakdown strength) [134, 135]. Here the PD resistance is introduced because it is relevant to the treeing phenomenon. In most of the literature, PD resistance is estimated by the PD
erosion test under a rod-to-plane geometry and measurement of the surface roughness or degradation depth caused by PD. According to Kozako’s results [26] as shown in Figure 2-42, PD resistance was found to be enhanced by adding 2, 4 and 5 wt% nano-sized layered silicates into polyamide (PA). Preetha et al [136] also tested the epoxy composites with nano Al₂O₃ particles from 0.1 wt% to 15 wt% and his result suggested even with a very low content (i.e. 0.1 wt%) the PD resistance could be considerably improved by nano particles. The mechanism of the PD resistance was explained by Tanaka [28] with his multi-core model: With the different size and specific surface area, the polymer binding of nanoparticles is able to change the structure of the polymer [137].

According to the concept of the multi-core model, the first layer was generated if the coupling agent was applied. It is assumed to strengthen the binding with the surrounding polymer matrix so it could help to resist PD damage. The second layer is generated under the influence of the first layer. It is well ordered by surrounding crystalline, or spherulite region, which also enhances PD resistance.

![Graph](image)

**Figure 2-42:** The change of surface roughness with the time for PD erosion test. PA: polyamide-6 without fillers. NC-2, NC-4, NC-5: polyamide-6 with 2 wt%, 4 wt% and 5 wt% [26].

The treeing phenomenon has been investigated mainly in epoxy or XLPE nanocomposites with a needle-to-plane design. Several researchers found significantly improved treeing resistance in nanocomposites. For example Imai et al [138] found the silica filled epoxy nanocomposite had a more than 2 times longer treeing breakdown time at room temperature at 10 kV and 1 kHz. Tian et al [139] also measured the electrical treeing life under different contents from 0.1 wt% to 10 wt% ZnO in LDPE at 10 kV and
the results suggested the time to breakdown was significantly increased. However, Tanaka [8] found there was a crossover phenomenon occurred under high AC voltages (as shown in Figure 2-43) and a similar phenomenon was also found in DC breakdown tests [140]. The performance of nano particles under higher voltages seemed to be worse than the pure dielectric, which implied that the performance of nanocomposites should be further evaluated. The mechanism for this crossover is still unknown but Calebrese et al [100] mentioned the importance of the particle dispersion in nanocomposites, which could definitely influence the consistency of the experimental results from different research groups.

![Graph showing tree channel propagation](image)

Figure 2-43: Tree length at 4 minutes after energizing with different voltage levels in pure epoxy (red) and 10 wt% nanocomposite (black). Two lines crossover over at about 17 kVrms [8].

The interaction between the nano particle and the treeing path has been further studied to explain the enhanced treeing resistance. Similar to the case of microcomposites, nano-sized fillers were also considered as barriers for the tree propagation [22]. There were three possible situations proposed by Danikas [22] as shown in Figure 2-44:

(a) The tree channel propagates through the polymer without touching particles;
(b) The tree channel propagates through the polymer contacting particles;
(c) The tree channel propagates through the polymer circumventing particles;
The situation of (b) and (c) were more consistent with the tree growth model in nanocomposites stated by other literatures [30, 141, 142]. Except for prolonged treeing paths guided by the local electric field and the relatively weak interfaces, Ding et al [142] also ascribed the better treeing resistance to more sub-micro voids in the nano-filled system instead of micro-size voids generated by micro fillers. As a result, the treeing resistance was enhanced by expanded size of the damage zone around the tree tip. However, there is no further work to support this opinion. However, there is a situation people usually ignored in nanocomposites: the diameter of the tree branch (several microns) is actually much larger than the nano-size particle (less than 100 nm). As a result the treeing models shown in Figure 2-44 and the other literature do not actually represent the real tree propagation in nanocomposites.

To assess the electric field distortions, space charge assessment was implemented by Nelson et al [129]. In contrast to the micro-size fillers, epoxy resin with nano-size fillers displays a different performance concerning charge accumulation. It was proposed that with the nano-size particles, the internal field enhancement will be mitigated. The particles inside of the tree can generate charge traps which are distributed on the wall of the tree channel [129, 143]. Deposited charges from avalanches creates further avalanches so as to reduce the surface charge at the end of the channel, which interferes the tree propagation [144]. Figure 2-45 shows the inner surface of the tree channel in which some silica products were observed [18]. It was suggested by the author that these deposited fillers could prevent the widening of tree channels so as to help increase the breakdown time.

Figure 2-44: Possible paths of the tree growth in nanocomposites [22].

(a)                                            (b)                                   (c)
2.5 Summary

A lot of work has been done on the study of electrical treeing phenomenon. Although there are still many questions about the tree initiation and propagation, characterisation of the treeing phenomenon has been widely developed such as the identification of the treeing stages, some important structural information as the tree grows and the conductivity of tree channels. Moreover, several treeing models have been proposed which do contribute to an understanding of the treeing phenomenon.

Because of the limitation of 2D imaging methods, 3D imaging techniques are necessary for the further understanding of electrical trees. Imaging results from researchers have proved the capability of using XCT and SBFSEM technique to realize 3D tree imaging. As a non-destructive method, XCT imaging is suitable for most insulating materials such as epoxy resin and PE. The resolution of micro-XCT with lab source is in sub-micron and it can be used to find out general tree features. Synchrotron XCT has more intense X-ray energy and can provide more accurate details. SBFSEM can provide finer details than Micro-XCT but it is a destructive technique and sometimes it is limited by the structure of electrical tree, for example the densely packed or severely degraded tree structure. 3D imaging in micro-filled epoxy system remains a challenge, but by using synchrotron it is possible to image internal tree structures. Laboratory X-ray source and SBFSEM may be capable to do it but higher X-ray energy would be required. In order to make the 3D imaging tool more useful in treeing studies, a combination between these imaging techniques and PD measurement is now suggested and more dynamic treeing and imaging experiments are described in this thesis.

Figure 2-45: SEM image of electrical tree cross-section in a 5 wt% epoxy nanocomposites. The tree structure was generated under 10 kV for 240 minutes [18].
Chapter 2: Literature review

The propagation of electrical trees in both micro- and nano-filled composites may be different from that in pure dielectric and more electrical and physico-chemical aspects should be considered. So far the understanding of the interaction between tree channel and nanoparticles is still based on theoretical models and the photographic evidence is not strong enough to support the entire hypothesis. In particular more work is still required to verify the real propagating direction when the tree tip is approaching the micro- and nano-sized barriers.

Considering the literature review presented and objectives proposed in Section 1.2, the following areas are identified to be targeted in this thesis:

- High-quality 3D imaging of electrical trees in filled epoxy resin systems.
- The application of 3D tree models to understand treeing mechanisms.
- Development of an experimentally verified model of the relationship between tree channel propagation and the micro- or nano-size particles.
3 Experimental description

3.1 Methodology

3.1.1 Introduction
The key aim of this research is to study the treeing phenomenon in epoxy resin and compare the difference between unfilled and filled systems. Hence, the commonly proved methodologies for treeing experiments and data collection were applied. 3D imaging has proved to be a powerful tool to reveal electrical trees in polymeric materials so most analysis about the structural information of electrical trees were based on XCT systems. The following strategies were taken for the study of the treeing phenomenon.

3.1.2 Tree creation in laboratory
Epoxy resin is selected as the main dielectric because of its great transparency and its relatively low viscosity which help the mixing process. On the other hand it is the commonly used material in the University of Manchester previously so the experimental result can be readily compared with the literature. All samples were designed by using the conventional needle-to-plane geometry and energized at the same conditions (13 kV$_{\text{rms}}$, 50 Hz and room temperature). The methodology used for sample preparation and experimental setup has been developed in the University of Manchester’s HV laboratory over many years. The whole treeing progress was monitored in the real time and some necessary data were collected such as PD signals and optical images (if the material is transparent enough). Details about the treeing experiment and sample preparation are included in Chapter 3.

3.1.3 XCT imaging
It has been known that 3D imaging of electrical trees is achievable by lab-source XCT in unfilled epoxy system while for micro-filled materials it is still challenging because of the very low contrast among the tree channel, the host epoxy and additional interference from the filler particles. SBFSEM has better resolution and contrast but its reconstruction can be affected by the artificial defects caused by microtomy. Therefore in this thesis, the
same micro-XCT system in the University of Manchester was still used to reveal the tree structure in unfilled sample, but the filled composites were imaged by an advanced synchrotron source in the Diamond Light Source facility.

Image acquisition is important in all 3D analysis followed by the image processing and reconstruction. In order to retain the real tree structure as much as possible, methods used for structure visualisation and characterisation are developed in this study. The 3D tree structure was fully quantified by professional software (Avizo and ImageJ) and algorithm developed in Matlab.

In addition to the normal tree imaging, a multiple-stage treeing and imaging experiment was designed based on the previous attempt [14] but in this case more treeing stages were captured and each stage was controlled to only create a smaller physical change (tens of µm). The effect of the X-ray irradiation was an important consideration for this experiment so an additional evaluation of the material properties was undertaken after substantial doses of X-rays to ensure the imaging process was not itself damaging or weakening the polymer.

Details about the XCT imaging and 3D visualization are introduced in Chapter 3, Chapter 4 and chapter 5.

3.1.4 Characterization of treeing phenomena in different epoxy systems

The investigation of the effect of inorganic fillers on tree propagation in epoxy resin was achieved by treeing breakdown tests on different sets of samples. As suggested in Literature chapter, Fujita et al [120] suggested that a smaller particle size (5 µm used in his experiments) could contribute to the better breakdown strength rather than the filler with the particle size of hundreds of micrometres, so in this thesis a microsilica filler with the 1-10 µm particle size was selected. As for nanocomposites, ~20 nm was usually chosen because it is a sufficient size that SEM facilities can easily observe. Two commonly used mixing methods (high-speed shearing and sonication) were compared before the electrical test and the better mixing strategy was used for all the other samples.

By doing breakdown tests a straight-forward understanding of the treeing resistance was achieved and compared in unfilled, micro-filled and nano-filled systems. Some filled
samples with different concentrations and particle sizes were also prepared for the synchrotron XCT. The new ‘pink-beam’ system was applied to dramatically increase the contrast of the tree channel. As the most important aim of using the XCT imaging technique on filled samples, the potential interactions between tree channels and embedded particles were further analysed based on 3D reconstructions.

For filled materials, the particle dispersion is always of concern because of its great impact on the properties and reliability of the system. SEM is commonly used to study the particle dispersion but most of times only objective judgement were provided in the literature. So in this project, some quantification methods are developed to describe the degree of the particle dispersion and separation. The interfacial effect in nano-filled systems has also proved to be important for the improvement of dielectric properties so materials were prepared with surface silanisation of the filler designed to compare its effect in terms of the treeing features. This silanisation was prepared by colleagues at UCL.

In order to find out how filler impacts the tree propagation, the treeing mechanism in the both unfilled and filled epoxy system was characteristics by imaging results and PD data. As a reference group, treeing and breakdown progress in unfilled epoxy system was firstly analysed by combining experimental results such as optical images and PD measurements. The treeing model proposed in the unfilled material was then set as the theoretical basis for the further characterization in filled batches so as to obtain a more coherent conclusion about the treeing mechanism in filled materials.

Details about treeing analysis and sample characterization are introduced in Chapter 3, Chapter 4 and Chapter 6.

3.2 Sample preparation

3.2.1 Introduction

Experimental techniques adopted for all the experiments are introduced in this section. Selected materials for sample fabrication are described. Factors such as mixing methods and curing procedure are described in detail. Different mixing methods can affect material characteristics [100], so samples made in different ways are compared firstly to optimise the process. All fillers selected in this research are silicon dioxides but with
different particle sizes. The silane surface functionization was provided by the Department of Material Discovery, University College London (UCL) and the surface designed is expected to give better compatibility with epoxy matrix.

Tests introduced in this part include: electrical treeing experiments, 3D XCT imaging, Scanning Electron Microscopy (SEM), tensile tests and Fourier-transform infrared spectroscopy. Among them, electrical treeing experiments and 3D XCT imaging are the core experiments, so both of them are most carefully described in this chapter.

### 3.2.2 Geometry of epoxy samples

Generally in HVAC cables, the transmitted electrical power is usually at above 50 kV but even so the electrical treeing phenomenon can take tens of years. In order to accelerate the whole degradation cycle, a needle-to-plane system was designed to enable high electric fields at the needle tip with relatively low voltages (5 kV - 30 kV). Figure 3-1 shows an illustration of the geometry of treeing test samples.

![Figure 3-1: A schematic of a needle-to-plane geometry for the treeing test.](image)

The maximum field stress is generated at the tip of the needle point and the theoretical value can be calculated by the Mason equation [56] in the absence of space charge:

\[
E_{\text{max}} = \frac{2V}{r \times \log (1 + \frac{4d}{r})}
\]  

where \( V \) is the applied voltage, \( r \) is the radius of the needle tip and \( d \) is the distance between the needle tip and the ground. In this research, all steel needles are provided by
Chapter 3: Experimental description

Ogura® with a tip radius of 3 µm and 30° angle, and the distance between the needle tip and the earth is 2 mm. If the rms voltage applied is 13 kV, the theoretical electrical field (rms) at the needle tip is 2.53 MV/mm. However in practice it is expected that space charge accumulated around the electrode can substantially reduce the field.

3.2.3 Materials for unfilled samples

Epoxy resin was chosen as the main dielectric material in this research according to its great transparency and hardness. Epoxy resin is a commonly used thermoset polymer made from the base epoxy resin and a hardener. The hardener accelerates the curing cycle and also develops the completely solid state during the cross-linking process. Two epoxy resin systems were selected: Araldite® LY5052/Aradur® HY5052 and Araldite® CY225/Aradur® HY925, both from Huntsman Advanced Materials. Both of these epoxy systems were used to make the unfilled samples but with different purposes.

The room-temperature-curing epoxy LY5052 is a translucent liquid at room temperature and the hardener HY5052 is a mixture of polyamines. This combination is selected as the dielectric system for samples preparing for multi-stage imaging as it has been used in previous laboratory tests [2, 3, 24] and so the tree structure in this composite is well known. The epoxy resin was mixed with the hardener at the ratio of 100:38 by weight and then stirred for 5 minutes using magnetic bars. After mixing, the epoxy resin mixture was degassed in a vacuum oven for 30 minutes to remove air bubbles. Once there were no more bubbles appearing, the degassed composite was ready to cast into cylindrical moulds as shown in Figure 3-2. A mould with a diameter of 20 mm and 30 mm in height was made from polytetrafluoroethylene (PTFE) which is generally non-sticky and nonreactive to most materials. The Ogura needles were used as the electrode for all samples and before the casting, the needle was firstly cleaned with acetone solution and placed under a microscope to ensure it was not damaged or polluted. Then it was installed in a metal cap fixer and a fixed distance was set between the needle tip and the bottom surface by using a 2 mm rubber slab as a spacer. After pouring the epoxy into the PTFE mould and setting the needle, the manufacturer specification [145] required curing for at least 24 hours at room temperature in a dry environment, followed by a post-curing cycle at 100°C for 4 hours.
The hot curing epoxy resin CY225 is a colourless bisphenol-A epoxy resin while the hardener HY925 is an anhydride curing agent with an amber colour. The whole mixing and degassing procedure was exactly the same as for LY5052/HY5052 but the curing condition was different: 6h at 80 °C and 10h at 130 °C. After fully curing and post-curing, all unfilled samples were machined and polished to thin sheet with about 5 mm thickness. Figure 3-3 shows these unfilled samples after grinding and polishing.

3.2.4 Materials for filled samples

The CY225/HY925 system was used as the base dielectric for filled epoxy systems due to its lower viscosity and longer gel time when the temperature is high [146]. The embedded filler for this research was fumed silica powder which is commonly used in concrete applications due to its excellent improvement of mechanical properties and thermal resistance. Both the micro- and nano-size silica was provided by Sigma-Aldrich with a particle size range of 1 -
10 µm and 20 nm according to their manufacturing datasheets. The mixing of nanocomposites is always difficult because nano-sized particles readily aggregate during storage. The surface energy of the nano particle is very large [147], which directly leads to agglomerations especially in the presence of moisture in the air. With agglomerations, some intrinsic properties of the composite may be significantly different [100, 148, 149] so particle dispersion is very crucial for sample preparation. The two most often used methods to make nanocomposites are sonication and high-speed shearing. High-speed shearsers such as batch and inline mixers are widely used throughout industries and can effectively disperse these fillers into composites. Advantages of shearing mixers include short processing times and easy setup. Sonication is achieved by using ultra-sonic waves to agitate particles into liquids and it is commonly used in nanotechnology as it is helpful to break up agglomerations.

In order to find out which method was more appropriate to the nanocomposites required here, some samples made from these two methods were compared. Micro-sized fillers were not compared in this case because of the totally opaque feature of the microcomposite. In this research, the high-speed mixing was achieved by IKA T25 Ultra-Turrax® at 6000 rpm for 20 minutes and kept a steady solution temperature at 70°C. Qsonica® Q700 was used to disperse nanoparticles into epoxy composites with 700 W/20 kHz. The whole sonication process was divided into several cycles because of high levels of thermal energy released, so a temperature probe was used for temperature monitoring. Once the temperature inside of the epoxy rose up to 80 °C, the facility shut down and then restarted only when the temperature dropped down to 30 °C. In practice every sonication cycle lasted 30 seconds and the shutdown time was 270 seconds with water bathing. An observation of transparency between samples made from shearing and sonication are shown in Figure 3-4. It can be found both the 1 wt% and 3 wt% sonicated samples showed better transparency than the shear mixed samples. Generally better light penetrability suggests better dispersion of embedded filler according to the mechanism of light scattering. So it can be considered that sonication is the better choice to prepare nano-filled samples in this case. Further study of particle dispersion carried out by using SEM is presented later in chapter 5.3.
The surface treatment of fillers was carried out to generate primary amine groups at the interface between the epoxy matrix and the filled nanoparticles. The process of silanisation was achieved using 3-Aminopropyl triethoxysilane (APTES) which is a commonly used aminosilane for silica fillers. A proofing Ninhydrin test was carried out to check silanisation. Figure 3-5 shows two solutions dissolved with SiO₂ and the transition from colourless to dark blue which illustrates the presence of primary amines group (R-NH₂). Figure 3-6 shows an illustration of the modified chemical features on the particle surface.

Figure 3-5: Ninhydrin test: (left) with untreated SiO₂; (right) with treated SiO₂ - goes dark blue in presence of primary amines(R-NH₂).
Chapter 3: Experimental description

The whole compounding procedure for the filled epoxy material was carried out by the following step-by-step process:

1. Araldite CY225 has a high viscosity at room temperature (8500-15000 mPa·s). To obtain a lower viscosity for compounding, the epoxy resin was pre-heated to ~60 °C for 5 minutes.

2. All fillers used were dried in advance to remove moisture and avoid further aggregation. Then the fillers were mixed with the epoxy manually firstly by a glass rod until there was no visibly distinct particle observed by eye.

3. The hardener HY925 which had the lower viscosity (250-450 mPa·s) at room temperature was directly added into the composite and mixed by a magnetic mixer with a heating base at 60 °C. The mixing ratio for epoxy and hardener was 100:80 by weight. To keep a low viscosity, the temperature was controlled at 60 °C. At the same time, the same cylindrical PTFE moulds as shown in Figure 3-2 were pre-heated at 100 °C.

4. Once the colour of the composite became uniform, a Q700 sonicator was used to continue mixing with water bathing at room temperature (~20 °C). The sonic intensity was set at 60% and the whole agitating time was 30 minutes. Once the temperature of the mixture was above 80 °C, the sonication immediately shut down and restarted again until the temperature was back to 30 °C.

5. After that the mixture was degassed in a vacuum oven at 60 °C for 30 minutes to remove air from the composite.

6. When the first-step degassing was finished, the composite was casted into the PTFE mould. The needle was set on the cap fixer in advance to give a 2-mm

Figure 3-6: A schematic of the silanisation taking place on the silica surface.
distance to the PTFE base. After locating the cap, the mould was returned back into the vacuum oven for another 15 minutes degassing.

7. Finally the composite then was placed into the oven for curing. The first cure took 6 hours at 80 °C and was then followed by the post-curing at 130 °C for 12 hours. Figure 3-7 shows the cured samples with micro- and nano-silica. There is one more sample (reference sample) without the needle prepared for each patch for the basic material characterization (i.e. SEM).

![Figure 3-7: Epoxy resin sample filled with: (left) 5% microsilica; (right) 3% nanosilica.](image)

### 3.3 Electrical tree creation

Electrical trees were grown and observed in an experimental cage in the University of Manchester [3] and the experimental circuit is showed in Figure 3-8 and Figure 3-9. The experiment in the cage was energized by the signal generated from a NI PCI-5412 Arbitrary Waveform Generator. The output signal can be any type as long as the peak voltage is below 12 V and the maximum bandwidth is within 20 MHz. An HV amplifier TREK 30/20 was used to amplify the voltage with the fixed ratio 3000:1 so the maximum output voltage available was 30 kV DC or 20kV peak AC. A resistor of 10 MΩ was set in the circuit to limit the current when failure occurred. In order to protect facilities during the long-term treeing experiment, the trap function of the amplifier was enabled and the trap threshold was set at 10% of the peak current. During the test, all samples were immersed in silicon oil to avoid surface discharges. The silicone oil with working temperature from -40 °C to 200 °C was provided from Alfa Aesar® and before the test, the oil was firstly filtered to remove impurities and then degassed in a vacuum oven at room temperature for a whole day. A Marlin CCD camera with ×50...
magnification lens was placed to capture the tree image and the camera could provide a picture size of 1392×1038 pixels with a field of view of 4.65×4.65 µm.

A MPD-600 Omicron system was installed to measure PD events during the tree growth. This system included:

1. **MPD 600**: the PD detector which measured PD signals by amplifying the leakage current. The centre frequency of integration of the system was 32 MHz with a bandwidth of 9 kHz to 3 MHz. Following the IEC 60270 Standard, in order to collect PD signals for electrical trees in this case the detector frequency was set at 250 kHz ± 150 kHz.

2. **MCU 502**: It is an accessory for PD measurement and it acted as the transmitter between the MPD 600 and the terminal (a desktop computer in this case). Signals from MPD 600 are transferred by using optic fibres.

Figure 3-8: Experimental circuit designed for electrical treeing tests.

Figure 3-9: Photograph of hardware assembly in the experimental cage prepared for treeing test.

A MPD-600 Omicron system was installed to measure PD events during the tree growth. This system included:

1. **MPD 600**: the PD detector which measured PD signals by amplifying the leakage current. The centre frequency of integration of the system was 32 MHz with a bandwidth of 9 kHz to 3 MHz. Following the IEC 60270 Standard, in order to collect PD signals for electrical trees in this case the detector frequency was set at 250 kHz ± 150 kHz.

2. **MCU 502**: It is an accessory for PD measurement and it acted as the transmitter between the MPD 600 and the terminal (a desktop computer in this case). Signals from MPD 600 are transferred by using optic fibres.
3. **Balanced circuit**: as recommended in IEC 60270, a balanced circuit is used to help to mitigate background noise [150]. The circuit is shown in Figure 3-10. It contained two samples: one was the test sample and the other was the dummy sample. The dummy sample had needle electrode but with a blunted tip with the same radius of the Ogura® needle. Ideally there should be no treeing phenomenon in the dummy sample. In real case, there was no visible tree observed after all experiments reported in this thesis. As a result, the measurement could be considered to accurately represent the electrical PD caused by partial discharges from the needle tip in the test sample. The sensitivity of the measurement system is important for correct interpretation. Some potential causes for the unwanted noise are: sharp points, tube connections and electromagnetic interference. In order to optimize the background noise, all copper tubes were well connected and wrapped by insulating tapes and the needle end was fixed into a spherical electrode by a grub screw to ensure a corona free condition. All facilities for PD measurement were separately place together and away from charging cables. Then the background noise was estimated when the whole system was run at a low voltage (< 1 kV) and it was found that there was still some discrete signals generated in the range of 0.2 pC to 0.8 pC. However, according to the other PD measurements especially concerning the treeing phenomenon, the PD sensitivity is usually in the range of 0.5 pC to 5 pC and most analysis also revealed more representative features for the tree growth at the higher magnitude [3, 24, 25, 151]. As a result, considering the main objective of this research is to study the tree growth instead of the aging in the tree initiation stage, the background noise was acceptable in this case and the threshold for the PD analysis in the later sections were all set at 1 pC.

![Balanced circuit used in PD measurements](image.png)

Figure 3-10: Balanced circuit used in PD measurements [12].
The whole system was connected to a computer and PD activity was visualized with Omicron software. The MPD600 system provided a highly responsive display and configurable view for partial discharge and voltage inputs, as shown in Figure 3-11 the software showed the phase resolved partial discharge (PRPD) pattern which yields both magnitude and the corresponding voltage phase. For further PD analysis, the PD dataset could be imported into Matlab with the same PD plot.

Figure 3-11: Omicron recording window showing phase resolved partial discharge data during the treeing progress in an unfilled epoxy resin sample at 18.4 kV peak.

Table 3-1: Description of tested samples.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Number of samples</th>
<th>Description</th>
<th>Base material</th>
<th>Voltage (kV&lt;sub&gt;rms&lt;/sub&gt;)</th>
<th>Imaging technique</th>
</tr>
</thead>
<tbody>
<tr>
<td>U</td>
<td>1</td>
<td>Unfilled (room-temperature curing)</td>
<td>HY5052/LY5052</td>
<td>10+6+6</td>
<td>Optical + XCT (lab)</td>
</tr>
<tr>
<td>UF</td>
<td>6</td>
<td>Unfilled (hot curing)</td>
<td>CY225/HY925</td>
<td>13</td>
<td>Optical</td>
</tr>
<tr>
<td>MF</td>
<td>7 (5 wt%) +7 (15 wt%) +7 (30 wt%)</td>
<td>Microsilica filled (hot curing)</td>
<td>CY225/HY925</td>
<td>13</td>
<td>XCT (synchrotron)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>UT</td>
<td>7 (1 wt%) + 7 (3 wt%) + 6 (5 wt%)</td>
<td>Untreated nanosilica filled (hot curing)</td>
<td>CY225/HY925</td>
<td>13</td>
<td>XCT (synchrotron)</td>
</tr>
<tr>
<td>ST</td>
<td>6 (1 wt%) + 6 (3 wt%) + 6 (5 wt%)</td>
<td>Treated nanosilica filled (hot curing)</td>
<td>CY225/HY925</td>
<td>13</td>
<td>XCT (synchrotron)</td>
</tr>
</tbody>
</table>
A summary of all samples tested for electrical treeing is presented in Table 3.1. It should be noted that there was only one unfilled sample (U1) tested but there were three energizing voltages (10 kV$_{\text{rms}}$, +6 kV$_{\text{rms}}$, +6 kV$_{\text{rms}}$) and imaging stages for this sample.

3.4 XCT imaging

For X-ray imaging, samples containing electrical trees needed to be machined smaller to meet the needs of the XCT system. There were two micro-XCT systems applied in this treeing study: ZEISS Xradia Versa 520 from MXIF and a synchrotron beamline I13 from Diamond Light Source.

Figure 3-12 shows two samples after machining and polishing prepared for XCT imaging. For the laboratory source Versa 520 system, the volume of the epoxy sample containing the needle tip and electrical trees was machined down to a smaller cylinder of 4 mm diameter and 8 mm in height. Because of the risk of damaging the sample, this was the smallest size achieved by using the mechanical process. For Synchrotron XCT, the sample was firstly cut down and then manual polished to the required size (~3 mm in diameter), thereby constraining the whole treeing area in a smaller region.

![Figure 3-12: Processed sample for (left) multi-stage imaging with laboratory micro-XCT (unfilled epoxy sample); (right) Tree imaging with synchrotron micro-XCT (15 wt% micro-silica filled epoxy sample).](image)

3.4.1 Lab-source Micro-XCT

The Versa 520 system has a cone beam geometry that has been applied several times previously to image electrical trees in unfilled epoxy materials [14]. The system was fitted with a 40X magnification lens and it provided the capability of achieving a spatial resolution of 0.238 µm. In-line phase contrast was applied in the Versa system to increase contrast while imaging these low absorbing specimens, typical of polymeric
materials. As the schematic shown in Figure 3-13, the sample was placed at 25 mm after the X-ray source ($D_{s-s}$) and 10.6 mm behind the detector ($D_{s-d}$). There were 2001 projections taken during the X-ray scanning, which lasts 34 hours under the 110 keV beamline energy.

![Schematic of Micro-XCT Versa 520 imaging system in MXIF](image)

Figure 3-13: Setup for the Micro-XCT Versa 520 imaging system in MXIF.

According to previous experience, the lab-source Micro-XCT beam is too weak to image tree structures in filled samples so in this research it was mainly used to image electrical trees in unfilled epoxy resin and for the multi-stage treeing and imaging. Table 3.2 shows imaging settings for the multi-stage imaging experiment in laboratory.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Description</th>
<th>Object</th>
<th>Exposure time (s)</th>
<th>Energy (kV)</th>
<th>Number of projections</th>
<th>Scan time (h)</th>
<th>Voxel size ($\mu$m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>U1.1, U1.2, U1.3</td>
<td>Multi-stage imaging</td>
<td>40X</td>
<td>60</td>
<td>110</td>
<td>2001</td>
<td>34</td>
<td>0.238</td>
</tr>
</tbody>
</table>

### 3.4.2 Synchrotron XCT

Previous attempts at tree imaging in filled system were not satisfactory [115]. The essential challenge was that the X-ray energy was attenuated too much after passing through silica particles. To counter these problems, several solutions were explored:

- Enhance the X-ray energy;
- Decrease the filler concentration;
• Reduce the sample thickness so as to avoid the beamline frequently countering microsilica;

During the experiment, all these three ideas were achieved in practice. The synchrotron micro-XCT is further developed by focusing on the absorption of silica fillers and also the weak contrast of internal tree channels. The ‘pink beam’ system was applied on the beamline in the synchrotron source to image the electrical trees in filled systems. The ‘pink beam’ is a monochromatic beam which can provide XCT images with higher signal-to-noise in even shorter exposure times, and using this system the X-ray flux could be intensified by at least one order [152]. As a result, the ‘Pink-beam’ synchrotron XCT provided faster tree imaging with better quality so in the research it was used to capture the tree structure in both micro- and nano-filled systems. Within the beamline, each sample was scanned at the spectral energy of 17.51 keV for only 2 minutes yielding 2160 radiographs, showing an 844.8 µm × 844.8 µm field of view in each slice and saving 4 more hours than the previous attempt [105]. Phase contrast imaging was also applied in this synchrotron imaging system. Figure 3-14 reveals the schematic of the synchrotron imaging system in I13 beamline which has much longer source-to-sample distance (D_{s,s}) than the lab-source imaging system.

![Diagram](image)

Figure 3-14: Setup for the synchrotron XCT imaging system at I13 beamline in Diamond Light Source.

### 3.5 Scanning electron microscope (SEM)

As introduced in Section 2.3.2, SEM is generally used to investigate the surface topography and composition of a sample. SEM can be also used to investigate tree structures [7, 99], but morphological information gained is limited and in 2D dimensions only. The previous section has shown that the XCT technique has the great ability to
reveal a treeing feature completely, so in this thesis SEM was mainly used to investigate
the silica dispersion. Electron Microscopy (EM) usually demands high-quality sample
preparation so the samples were firstly machined to a small cone shape and then cut to a
smaller plane with the dimension of 2 mm×2 mm using a microtome, as shown in Figure
3-15. The processed sample had a smooth surface and it was then attached on a support
platform by a sticky carbon-filled layer to ensure a good electrical connection to ground.

![Figure 3-15: Schematic of the processed sample for SEM imaging.](image)

Epoxy resin is a non-conducting material so the raw surface will accumulate charges
from the electron beam [65]. The charging effect can greatly influence the brightness and
contrast of a SEM image so it is necessary to eliminate the effect. There are several ways
are available to eliminate this charging as shown in Figure 3-16, such as:

a) Using a low accelerating voltage;
b) Coating with a conductive material to remove surface charge (as shown in Figure
3-16);
c) Using a relatively high gas pressure (still much lower than atmosphere) to
remove surface charge;
d) Building a conductive connection between the sample surface and the earth.

![Figure 3-16: Illustration of electron emission on non-conducting specimens.](image)
Metal coating is widely used to help the imaging of surface topography on polymeric materials. It increases signal to noise ratio while it also tends to cover the original elemental characteristics on the surface. For this reason, low-vacuum environments are used in this research and the sample was not metal coated. The pressure inside the chamber was adjusted to about 0.3 mbar to eliminate most of the charging effects and retain tomographic information as much as possible.

SEM facilities have different gun types such as heated tungsten wire and crystals but in order to observe nano-sized particles, the Field Emission Gun Scanning Electron Microscopy was applied in this research. FEI Quanta FEG 250 in School of Materials, University of Manchester was applied to carry out the SEM which can provide nano-sized resolution to 1.4 nm. For imaging types, secondary electron (SE) and backscattered electron (BSE) are the two commonly used detection modes. Secondary electrons are emitted from samples with lower energy (<50 eV) and can provide very high resolution which can reach to less than 1 nm. By contrast, backscattered electrons mainly come from the beam and are reflected by elastic scattering. Its resolution can be a little bit poorer while it is able to see the distribution of particles or fillers on the surface. In order to get more information about the dispersion of filled micro- and nano-sized silica, BSE was mainly used in this research and also with the Energy Dispersive X-ray (EDX) analysis to study the elemental characteristics. EDX is achieved by collecting specific spectrum peaks generated from the interaction of X-ray excitation with the different element because each element having a unique atomic structure.

3.6 Evaluation of X-ray irradiation

As mentioned in Section 3.1.3, both mechanical and chemical properties of the epoxy resin need to be estimated to assess the damage caused by X-ray irradiation. The tensile test is the most commonly used method to measure the sample’s response to aging. In this case, this test was applied to compare the change of mechanical properties between samples before and after X-ray scanning. Figure 3-17 shows the Instron 8802 facility and the dumbbell geometry designed for the test, which is recommended by ASTM-E8-E8M standards. The material used in this case the LY 5052/ HY 5052 epoxy system and the sample preparation is the similar with the procedure introduced in Section 3.2.2. In this case a metal dumbbell mould with depth of 3 mm was used and all mould surfaces were brushed with silicon oil to prevent adhesion. Some samples were firstly exposed to X-
rays. The irradiated area was a square with the dimension of 13 mm × 13 mm. Two extra jags are included in the mould and they are designed to make sure the breaking could always happen at the middle of the irradiated region.

Four groups of dumbbell samples were divided according to different X-ray sources and exposure times. The control Group A comprised 15 samples, which were not exposed to radiation. Each irradiated group B, C, and D consisted of 5 samples. Samples from Group B and C were exposed in a 320 Nikon Custom Bay system for 80 mins and 160 mins respectively. Samples from Group D were exposed in a TH 225 WIR for 80 mins. All radiated samples were exposed to X-ray sources at 150 keV and 125 W with 30 mm source-to-detector distance. The voltage and power set here ensured the similar X-ray dose in a shorter period, compared with the laboratory XCT system introduced in Section 3.4.1.

Fourier Transform Infrared Spectroscopy (FTIR) is an analytical technique to study chemical features of a material. It measures the absorption at different wavelengths and then identifies corresponding molecular components and structures. In this thesis FTIR was carried out to estimate the change of chemical composition in the epoxy samples after they were scanned by X-rays. Samples after the tensile test were then studied by comparing different areas. As shown in Figure 3-18, there were two regions compared: the broken area with discolouration and the unchanged epoxy area for comparison.
3.7 Summary

This chapter has described the development of the experimental set up and equipment used for all experiments reported in this thesis. Electrical trees were developed by a conventional energizing method: needle-to-plane geometry under 50 Hz AC voltage and measurement systems recorded experimental data synchronously during the tree growth. There are two 3D imaging techniques used in this thesis (lab-source micro-XCT and Synchrotron micro-XCT) and each of them has advantages for different purposes. 3D imaging for the real electrical trees then was achieved using both systems: Laboratory-source XCT has a flexible setup and is relatively accessible in most of time so it was applied for tree imaging in multiple stages; The synchrotron source XCT has higher X-ray energy and the ‘pink beam’ system helps to dramatically increase the contrast of different objects so it was more suitable to distinguish electrical tree structures in silica filled systems.

The image processing and segmentation are also crucial for the successful reconstruction and they will be explained in detail in the next chapter.
4 Processing of SEM and XCT images

4.1 Introduction

The review of literature has shown the unique structural features of electrical trees and also various methods used for tree characterisation in both 2D and 3D. This chapter includes the general methodology applied to analyse 3D imaging datasets and the particle dispersion in filled dielectrics from SEM images. Image processing and segmentation of XCT images in Avizo software are described step by step as it is the optimization to reveal the real tree structure as accurately as possible. After the well-controlled reconstruction, some parameters used for quantification are introduced to describe 3D structures. The particle dispersion studies are based on SEM images and quantified analysis is achieved by introducing ideas of the skewness and neighbouring distance.

4.2 3D renderings of electrical trees

4.2.1 XCT reconstruction

The stack of XCT images is exported from the Versa system and to regenerate the 3D tree replica, both imaging processing and segmentation are needed. The main purpose of imaging processing is to improve the visualization of image data, for example to adjust the brightness and contrast. But at the same time, too many artificial modifications can change the original geometry so the image should be processed very carefully. Figure 4-1 is a projection slice selected from a 5 wt% micro-filled sample scanned at the Diamond synchrotron source and it shows how the raw XCT data is processed by using Avizo software.

For the first-step of processing, the original image was adjusted by changing brightness and contrast, which made different objects recognizable. For example in this case the black circle was the tree channel and the white ones corresponded to silica particles. After extracting the treeing area from the whole bulk material, the 3D Median algorithm...
is used to filter the critical interfaces. The filter was not only helpful to moderate the background with high signal-to-noise ratio but also effectively faded the shining artefacts caused by phase contrast imaging. These processing operations were conducted by using Avizo and ImageJ software.

![Figure 4-1](image1.png)

Figure 4-1: An example of intensity adjustment and filter application to a slice of 5 wt% sample. (Left) Original slice collected from a microsilica filled sample; (middle) the partial region after adjusting the contrast and brightness of the image; (right) the same region after median filtering.

![Figure 4-2](image2.png)

Figure 4-2: Both electrical tree channels (red outlined) and silica particles (black outlined) were extracted by using 'Magic Wand' tool.

After processing, useful information should be retained and the bulk epoxy material needs to be eliminated so as to reconstruct the internal voids and structures. Imaging segmentation is regarded as a separating operation to label the same material according to similar pixel intensities. Generally the segmentation should be as automatic as possible so as to maintain the most accurate structural information. In practice both the pixel intensity and volume connectivity should be considered as the major factor to identify
required objects. The electrical tree is actually hollow tubules and may be surrounded by a wall of degradation (generally with low molecular weight) so most of dark pixels should be regarded as electrical tree channels. To achieve the segmentation, several methods are applied step by step:

- The ‘Top Hat’ method aims to extract small features by defining the size of the structuring element. In this case the disk shaped element is used to select circular cross-sections of electrical tree channels. Then by limiting the range of threshold value, the ‘Top Hat’ method (also called ‘Black Top Hat’) can effectively extract suspected tree channels that are smaller than the defined element and have lower intensity value than their surrounding area. This is the first-step segmentation to remove background and correct non-uniform illustrations. By the end of this step, the tree structure should be observable but with variable levels of noise.

- The ‘Magic Wand’ tool is then used to select the whole treeing area and remove most of unwanted structures away from the main tree body. The tolerance of the ‘Magic Wand’ tool should be adjusted to an appropriate range so that only the real tree channel can be chosen and disconnected structure will not be removed by mistake. This is the step to identify the main tree body and also check the disconnecting level of the whole reconstruction as shown in Figure 4-3.

- After the tree structure is clearly extracted and there are only few disconnections or little noise exists, the ‘Remove Island’ technique can be used to further improve the image quality. This technique is an automatic operation aiming to remove small and isolated features. It also has the merging function which tends to attach isolated elements into the main structure if they are within a certain distance. It should be noted that this segmentation sometimes can modify the real intensity feature during the merging and isolating operation so the principle is to gradually adjust the cell size so that the modification can be compared and judged.

- Silicon dioxide (SiO$_2$) has largest unit mass (64 g/mol) in the dielectric system so it usually shows bright white according to the mechanism of X-ray tomography. A similar ‘Top Hat’ method (also known as ‘White Top Hat’) is used for the segmentation of silica fillers. Sometimes the cross-section of a silica particle may appear to be hollow due to the significantly attenuated energy when the X-ray penetrates through the filler so additional manual compensations are applied such as ‘Hole Filling’ (for small pixels) and the ‘Brush’ tool (for large clusters).
Figure 4-3 shows the comparison of processed projections before and after segmentation. After intensity classification, tree channels and filled particles are separately selected by using the ‘Magic wand’ tool. All projection slices are then stacked up together so as to generate the 3D view including all degradation paths and silica fillers as shown in Figure 4-4.

![Image showing processed projections before and after segmentation.](image)

**Figure 4-3:** Image segmentation process. (Left) the slice after intensity adjustment and median filter; (right) the segmented slice with only tree channels (in red) and silica particles (in grey).

![Image showing reconstructed electrical trees and fillers.](image)

**Figure 4-4:** Reconstructed electrical trees (brown) and fillers (grey) by stacking up radiographs collected from synchrotron micro-XCT. The electrical tree was grown in a 5 wt% micro-filled epoxy sample under 13 kV<sub>rms</sub> for 1.2h.

### 4.2.2 Quantitative characterisation

The 3D imaging technique provides the opportunity to observe the complex tree structure. Most studies about electrical tree structures have been based on 2D optical methods, which usually gave structural information of projected images of electrical trees such as tree length [80], tree diameter [86] and fractal dimension [104]. However, with the help of XCT imaging, 3D visualisation of the electrical tree can involve more general
structural features for example the volume and surface area of the tree, bifurcation divergence angles and tortuosity [14]. By doing 3D analysis, a more thorough understanding of electrical trees can be achieved.

In terms of the measurements, parameters are classified into two types:

- **Directly measurable parameters**: these parameters can be directly measured by Avizo and ImageJ software. The 3D reconstruction of an electrical tree in this case does not need to be translated and so these parameters can accurately explain real and accurate structural information. The directly measurable parameters used in this thesis include: surface area, volume, tree length, channel diameter.

- **Indirectly calculable parameters**: these parameters are mainly developed by analysing the whole structure and may sometimes change in time. These parameters tell the general structural characteristics of the electrical tree. The indirectly calculable parameters used in this thesis include: number of tips, fractal dimension and tortuosity.

Generally the directly measurable parameters present ‘how big the electrical tree is’ and the indirectly calculable parameters suggest ‘what the electrical tree looks like’. In order to collect these indirectly calculable parameters, the whole tree structure is firstly translated into skeleton. By doing this translation, the tree structure can be presented by only nodes and connecting lines, where the radii of the tree channels are ignored.

Tortuosity is introduced in [14] and it is a parameter to describe the straightness of tree channels. As shown in Figure 4-5(a), it can be calculated as the ratio of the tree length to the straight line distance between nodes:

$$ T = \frac{d_{12} + d_{23}}{d_{13}} $$

(6)

Where $d_{12}$, $d_{13}$, $d_{23}$ are the shortest distance between every two nodes shown in Figure 4-5(a). By using the methods explained, the tortuosity explains how straight a curved branch is, which basically refers to specific points on the main trunk. One of the disadvantages of this method is that it ignores the varying curvature of branches while this can be crucial information to study the interaction between barriers and the tree.
growth in filled systems, so in this thesis, the tortuosity is calculated by the ratio between two nodes as shown in Figure 4-5(b):

\[ T = \frac{c_{12}}{d_{12}} \]  

where \( c_{12} \) is the curvature length (or real branch length) of the branch and \( d_{12} \) is the shortest distance between node 1 and 2. This ‘tortuosity’ describes how curving the tree branch is and will be mainly compared in different filled systems.

![Tortuosity calculation](image)

Figure 4-5: Tortuosity calculation by using (a) three-nodes exemplification shown in [14]; (b) two-nodes simplification considering the curvature length.

4.3 Particle dispersion

Studies in filled composites should always pay great attention to the dispersion, especially for nanocomposites. SEM and TEM images are widely used to present the corresponding particle dispersion in a sample. However, in most of time the dispersion is only described by objective judgement and not statistically derived from experimental observation. As a result, the use of quantitative interpretation for characterizing the dispersion in composites is necessary to avoid user’s bias. This section introduces how the quantification method is applied to SEM images.

4.3.1 Theoretical dispersion model

Before imaging, a simplified particle model is developed to get a quick overview about the separation of the micro-size particles. Figure 4-6 assumes all silica fillers are well
dispersed and arranged in a body-centred cubic (bcc) disposition in the bulk epoxy resin [153, 154]. In the bcc structure the particle at the central lattice point is surrounded by 8 corner points so the closest distance is defined as the surface-to-surface distance between the central particle and either particle at each corner ($D_{s-s}$). The theoretical equation is:

$$D_{s-s} = \sqrt{5}R \times \sqrt[3]{\frac{12\pi [w\rho_{ep} + (100 - w)\rho_{si}]}{w \cdot \rho_{ep}}} - 2R$$  \hspace{1cm} (8)

Where $R$ is the radius of the particle, $w$ is the weight fraction of embedded fillers, $\rho_{ep}$ is the density of epoxy resin (1.20 g/cm$^3$) and $\rho_{si}$ is the density of SiO$_2$ (2.65 g/cm$^3$).

4.3.2 Quantification methods

In order to define the particle dispersion in SEM images, quantification is carried out. Before calculation, SEM images were firstly processed by using ImageJ software. Figure 4-6 shows a SEM image of 5 wt% nanocomposite, the raw image was firstly digitalized by using threshold tools and then any particle with the area less than 314 nm$^2$ (a 20 nm particle has a surface area of 314 nm$^2$) was considered as noise and removed. Sometimes the centre of gravity is needed to measure the distance between particles so in order to
collect the coordinate of gravity centres, the segmented particles shown in Figure 4-8 were approximately transformed to ellipses for simplification.

Based on processed SEM images, skewness could be used to evaluate the uniformity of the spacing of particles and agglomerates [155]. This parameter was mainly used to measure the asymmetry of variables’ distribution by dividing the sampling area into small quadrats (square areas with equal size) and then counting the quantity of included particles in every quadrat. So if all particles are well separated and distributed, these equal-sized quadrats should contain similar number of the particles, which leads to a smaller skewness. In contrast if there are some aggregates, they will result in large number of empty or less populated quadrats so as to lead a poor asymmetry which shows
higher skewness. As shown in Figure 4-9, the sand box method was applied on the segmented image in Figure 4-7. The quadrat size can significantly influence the result and it always varies according to different composites. Curtis et al [156] and Kim et al [102] suggested that approximately 2 to 4 times of the mean area of the single particle was suitable for analysis so in this case, an area of 2.5 times (800 nm$^2$) larger than the minimum area of a single particle (314 nm$^2$) was selected as the quadrat size and this generated was a 140×140 grid.

![Image](image.png)

Figure 4-9: (left) The particle dispersion after imaging processing and segmentation to a SEM images of a 5 wt% nanosilica filled samples; (right) the image separated with a 140×140 grid (800 nm$^2$ quadrat size).

Then by counting the number of fillers in each cell, the skewness can be defined as:

$$\beta = \frac{q}{(q-1)(q-2)} \sum_{i=1}^{q} \frac{N_{qi} - N_{q_{mean}}}{\sigma}^3$$  \hspace{1cm} (9)

where $q$ is the total number of cells studied, $N_{qi}$ is the number of particles in the $i^{th}$ cell, $N_{q_{mean}}$ is the mean number of particles per cell, and $\sigma$ is the standard deviation of the $N_q$ distribution.

The nearest neighboring distance was also considered. The idea is to describe the mean distance between every pairs of neighboring particles (or aggregates) by assuming all aggregations are spherical. The centroid of each particle in the SEM image was determined by ImageJ software and then the mean distance between centroids could be calculated by the equation:

$$d_{ij} = \frac{\sum \sqrt{(x_j - x_i)^2 + (y_j - y_i)^2}}{n-1}, \quad i, j = 1, 2, ...$$  \hspace{1cm} (10)
where \( n \) is the total number of agglomerates or particles in the image, \((x_j, y_j)\) is the coordinates of the particle of interest, and \((x_i, y_i)\) is the coordinates of every other agglomerate or particle. A smaller distance indicates better particle dispersion and it suggests more nanoparticle surfaces available for contact with the bulk polymer in a unit area. With the consideration of the body-centred cubic structure introduced in Section 4.3.1, \( n \) is chosen to be 9 here.

All calculated parameters shown in later chapters are mean values which consider skewness and nearest neighboring distance based on 5 SEM images in each batch.

### 4.4 Summary

This chapter has introduced the methods used for reconstruction of electrical trees and the quantification of 3D trees models, as well as the particle dispersion from SEM images. The parameters used to describe the 3D tree structure such as tree volumes cannot be collected by using normal 2D optical methods and so they can now be further applied to the study of real treeing phenomenon. The indirect parameter tortuosity was re-defined so as to pay more attention to the curvature of tree branches, which may be an important factor in micro-filled system.

The quantification methods applied to SEM images will allowed the particle dispersion to be evaluated statistically. Based on that, it is also possible to link the abstract mixing degree with the results from other tests, which would help to determine the sensitivity of the filled system to the dispersion state. This will provide a better understanding on how to improve the dielectric property in practice.
Chapter 5: Correlation between PD and tree growth

5 Correlation between PD and tree growth

5.1 Introduction
This chapter describes the multi-stage treeing experiments, the purpose being to develop a technique which generates three-dimensional images of a tree as it grows. The aim of this case study is to capture very slight structural changes and then investigate its mechanism. The micro-XCT system was used to ensure the whole tree pattern was captured, and to allow growth of the tree between imaging events. Such a technique requires repeated exposure to high X-ray doses, so an assessment of the impact of X-ray irradiation on epoxy resin was also carried out. The reconstructed renderings of a single electrical tree at three treeing stages in its development are introduced and discussed. This work was carried out collaborating with Dr. Zepeng Lv in the University of Manchester.

5.2 The impact of X-rays on epoxy resin
Experimental details are described in Section 3.6. As shown in Figure 5-1, the irradiated areas in all four group samples turned from light yellow to amber and the discoloration degree was seen, by the naked eye, to be deeper with longer irradiation time. It is well documented that discoloration of polymers can result from the oxidation of high-intensity light or thermal damage [157, 158] so in order to determine whether these chemical changes would influence subsequent treeing phenomenon, the mechanical strength of 15 unexposed samples and 15 exposed samples were tested. The result is shown in Figure 5-2. The mean breakdown stress for unexposed group is 43.13 MPa and the deviation of the three irradiated groups from this is -13.43%, -8.27% and +9.5% respectively. Considering sample to sample variation for example the variation of the sample thickness is ±0.32 mm (±10.25%), these changes do not show a meaningful difference in terms of mechanical properties. Significant reduction in mechanical strength, which might be expected if there is damage in the form of cross-linking increasing tensile strength or
Chemical properties were characterized by FTIR (experimental details are introduced in Section 3.6) and the result is shown in Figure 5-3. Typical peak positions do not have a shift in wavenumbers because some key chemical groups were not modified. From comparison, an additional appearance of the absorption peak was found in the (B) and (C) groups at 1720 cm\(^{-1}\) corresponding to the C=O Carbonyl group. A weak absorption at 1658 cm\(^{-1}\) was also found in both irradiated sample groups (B) and (C), which suggested the formation of the C=O stretch of carboxylic acid groups or the C=C alkene stretch. For photo-oxidation, the formation of some UV-sensitive groups such as carbonyl groups
could be significant in most systems [159] so the result here suggested a pre-stage oxidation may have started.

![Figure 5-3: FTIR transmittance spectra of samples: (a) unexposed sample from group A; (b) epoxy sample irradiated by X-ray for 80 minutes; (c) epoxy sample irradiated by X-ray for 160 minutes. The dashed line is at 1658 cm\(^{-1}\) and 1720 cm\(^{-1}\).]

Overall, for the purpose of the three-step tree imaging, after the first X-ray scanning only two further treeing experiments were needed. Discoloration did occur but according to the limited evidence for the change in the mechanical properties, ageing was most likely restricted on the polymer surface. Because the early tree structure was in the most interior region of the epoxy sample and was \(\sim 1\) mm distance away from the surface so in this case the tree-step imaging could be considered reflective of ‘normal’ tree growth. However, it is possible that the material damaged by PD in the vicinity of the tree, after a period of tree growth [160], may be more susceptible to x-ray damage than other epoxy regions so when the tree structure is continuously spreading, the macroscopic properties may need to be revaluated.

### 5.3 Multistage treeing and imaging

Multistage testing was designed to investigate small changes to the tree structure after very short periods of ageing, so each step was carried out at the early treeing stage for few minutes. The whole treeing test was observed by both optical imaging and PD measurements to follow tree growth by conventional means. The tree structure was initiated at 10 kV AC for 10 mins and then imaged using micro-XCT. The same procedure was repeated twice to get the 2\(^{\text{nd}}\) and 3\(^{\text{rd}}\) tree but at 6 kV for 3 mins and 2 mins
respectively to ensure only slight a change to the tree. Optical images shown in Figure 5-4 revealed the tree in all three steps. It was found that there was only little tree growth at tree tips on the image from the left side to the middle and until the 3rd stage the top tips continued growing and the whole tree structure seemed to be thicker and darker.

Figure 5-4: Optical images of tree growth at (a) 10 kV for 10 mins, (b) 6 kV for a further 3 mins and (c) 6 kV for a further 2 mins.

By using the XCT settings introduced in Table 3-2, a sub volume of 135.7 μm × 138.1 μm × 111.8 μm was extracted around the treeing area for detail processing. As shown in Figure 5-5, virtual replicas for each stage were reconstructed with the same processing methods (as introduced in Section 4.2.1) and all processing and reconstruction were completed in Avizo software and with the same settings in all steps. These tree models have been normalized and relocated so that new characteristics grown in the next two stages could be easily identified. The general structure shown in 3D images were consistent with optical observations while there is an apparent discontinuity in the channel after the first bifurcation point. No obvious discontinuity was found in Figure 5-4 but overlapping tree structures may hide these defects in the optical projections.

Figure 5-5: 3D reconstruction of the three-step tree growth in epoxy resin after (a) 10 kV for 10 mins; (b) 6 kV for 3 mins; (c) 6 kV for 2 mins. The corresponding optical images are shown in Figure 5-4.
From the comparison between Figure 5-4 and Figure 5-5, it was also found that some small tubules branching on the main tree trunk was invisible in these 3D reconstructions. The cause might be either the too small diameter (less than 0.24 µm) or the low contrast with the epoxy due to the similar material constituent.

It should be noted that the cone beneath the tree structure was not the metallic needle tip. Figure 5-6 shows what appears to be an air gap covering the needle tip and this feature is rarely seen by normal optical methods. As measured, the tip radius of the needle shape void presented by the air gap was 1.85 µm which is smaller than the needle (3 µm). This air gap could be regarded as a result of the sample preparation because of its needle-like geometry and the size of the air gap did not change during the treeing process. It can be inferred that as the electrical tree started to grow there was no further damage to the polymer nearby, which suggests the majority of the PDs could freely pass through the air gap and into tree channels. The importance of such a gap and movement of gas between the needle and polymer has been discussed previously [161] and it was also proposed that the size of the air gap could significantly influence the tree initiation [162]. However in practice, observing this is quite challenging due to the difficulty of observation of air gaps optically and the difficulty in making reproducible samples of this type.

![Figure 5-6: The air gap (black triangle) covering on the needle tip (white triangle). A small tree structure started from the tip of the air gap.](image)

One of the most helpful contributions of the 3D imaging is that the real tree structure can be accurately quantified. Some global and structural parameters are shown in Table 5-1. In the first-stage model, 13 bifurcations were found and the divergence angle varies from 34.2° to 74.5°. And for the second-stage, 6 more bifurcated channels were generated and the maximum diverging angle increased to 93.4°. All newly generated tree tips were branches and located at the main tree trunk from the 1st step. From 1st to 2nd stage and 2nd...
to 3rd stage, the largest tree increment appeared at the same top branch. There are four branches growing less than 1% in length and all of them were located at what appear to be disconnected branches (left-hand side). The surface area increased 90.7% from the 1st to 3rd step while it was enlarged by 154.9% for the volume. The increased surface area and volume could be traced in two aspects: the widened tree channels and the newly extended channels (tree tips). It was found that between 1st and 2nd stage 73.5% of the volume increment (1759 μm³) was from the extension of tree tips while from 2nd to 3rd treeing, it was 33.6% of 2165 μm³. The maximum diameter was located at the bifurcation near the needle tip and it increased 65.13% and 2.80% in each respective step. The 3D fractal dimension is measured by Avizo software and it was changed very slightly when the tree structure became longer. However the tree growth among all these three steps was very slight so may not be expected to change the fractal dimension.

Table 5-1: Directly measurable and indirectly calculable parameters collected from the three-step treeing and imaging experiment.

<table>
<thead>
<tr>
<th></th>
<th>1st step</th>
<th>2nd step</th>
<th>3rd step</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Number or tree tips</strong></td>
<td>16</td>
<td>24</td>
<td>27</td>
</tr>
<tr>
<td><strong>Longest branch Length</strong> (μm)</td>
<td>112.11</td>
<td>122.70</td>
<td>144.79</td>
</tr>
<tr>
<td><strong>Surface area</strong> (μm²)</td>
<td>4180.12</td>
<td>6553.34</td>
<td>7970.28</td>
</tr>
<tr>
<td><strong>Tree Volume</strong> (μm³)</td>
<td>2533.01</td>
<td>4292.20</td>
<td>6457.47</td>
</tr>
<tr>
<td><strong>Mean Diameter</strong> (μm)</td>
<td>2.65</td>
<td>3.00</td>
<td>3.21</td>
</tr>
<tr>
<td><strong>Maximum Diameter</strong> (μm)</td>
<td>6.28</td>
<td>10.37</td>
<td>10.66</td>
</tr>
<tr>
<td><strong>3D Fractal dimension</strong></td>
<td>1.70</td>
<td>1.76</td>
<td>1.78</td>
</tr>
<tr>
<td><strong>Minimum diverging angle (°)</strong></td>
<td>34.2</td>
<td>34.2</td>
<td>27.8</td>
</tr>
<tr>
<td><strong>Maximum diverging angle (°)</strong></td>
<td>74.5</td>
<td>93.4</td>
<td>93.9</td>
</tr>
</tbody>
</table>

From the comparison of the tree structure in Figure 5-5 by eye, only the growth on tree tips is obvious. From Table 5-1, it can be found that the average width of tree channels
only had a total enlargement of 13.2% and 7.0% in diameter between stages while the longest branch grew 9.46% and 18.01% in length respectively.

The corresponding PD data were collected simultaneously with the treeing experiment. The total ejected energy was measured by summing the PD energy from all apparent charges and the expression used was:

$$\text{Total PD energy} = \sum_i q_i \times U_i$$  \quad (11)

where $q_i$ is the apparent magnitude of the discharge and $U_i$ is the instantaneous voltage when the discharge occurred. All of these data were recorded by the Omicron system. Figure 5-7 shows accumulated PDs during the energization between stages. It was found that the magnitudes of these PDs linearly increased with the point on voltage waveform and all extinguished around 108°. By using Equation 11, the total released PD energy calculated from all PD clusters from the 1st to the 2nd image and the 2nd to the 3rd was $6.25 \times 10^{-4}$ J and $372 \times 10^{-4}$ J respectively. If the major role of the energy consumed is to evaporate the base material, the released PD energy could be assumed to be transferred into evaporation energy of the epoxy resin material. A previous study [14] has estimated the required energy for the evaporation of unit epoxy resin volume ($E_{\text{mol/V}}$) to be $7.56 \times 10^{-8}$ J·µm$^{-3}$. The increment of the entire tree volume between stages are 1759.19 µm$^{-3}$ and 2165.27 µm$^{-3}$ so the corresponding energy required for vaporisation is $1.32 \times 10^{-4}$ J and $1.64 \times 10^{-4}$ J respectively. The increased volume contained two components: the widen structure of the previous stage and the newly-grown parts from tips. By calculating the conversion rate of released PD energy, it can be found that from the 1st to 2nd stage 19.68% of PD energy contributed to the evaporation of epoxy resin, while from the 2nd to the 3rd stage, only 0.44% of PD energy was transferred. Both transferred ratios were found to be out of the estimated PD-to-vaporization ratio range (1.4% to 5%) shown in [42].
By considering the linear relationship of the maximum PD magnitude and the length of the tree channel [3, 64] and also referring to the characterization of PD activities during the tree growth [3], the PD activity could be further associated with the structural extension. PD patterns in Figure 5-7 could be regarded as the typical ‘wing-like’ patterns according to their phase dependence feature and they were closely linked with the tree extension [3]. The PD pattern shown in Figure 5-7(a) could be separated into two isolated clusters by magnitude and these two contained $1.23 \times 10^{-4}$ J discharge energy (19.68% of the total released energy from 1st to 2nd stage). As the tree grows longer the maximum PD magnitude would also increase, so for the 3rd stage, PDs with higher magnitudes than the maximum magnitude in the 2nd stage (~11 pC in the positive cycle and ~9 pC in the negative cycle) were selected as shown Figure 5-7(b) and they contained $0.55 \times 10^{-4}$ J of PD energy. Figure 5-8 shows the new tree growth based on the differential structure identified by XCT imaging and the increased volume at tree tips was measured to be 1292 $\mu$m$^3$ and 727 $\mu$m$^3$ respectively. The corresponding evaporating energy of the epoxy resin in these treeing parts could be estimated to be about $0.98 \times 10^{-4}$ J and $0.53 \times 10^{-4}$ J. It was found that the evaporation energy calculated in these two periods was very near to the released PD energy marked by red squares shown in Figure 5-7 ($1.23 \times 10^{-4}$ J and $0.55 \times 10^{-4}$ J).
The quantified analysis shown above suggests a potential correlation between PD signals and the corresponding tree growth, which provides a new way to predict the theoretical treeing volume only based on PD measurement. The conclusion found in this case can be also applied in opaque materials such as filled dielectrics where the internal tree growth is difficult to be observed and it is supposed to finally benefit to the treeing estimation in the real insulation system.

### 5.4 Summary and conclusion

For the three stages considered, it has been shown that the X-ray radiation for 3D imaging does not significantly damage the core area of the epoxy resin being studied. However some surface oxidation may indicate early signs of ageing is present. It is concluded that the multi-stage XCT imaging and reconstruction can be considered reflective of ‘normal’ tree growth, and it is not unduly affected by the imaging process.

The XCT imaging of an electrical tree three times at close intervals during its growth disclosed detailed structural variation as the tree grew. Parameters reflecting the tree size, such as length, volume and surface area were shown to increase. Parameters reflecting morphology, such as fractal dimension did not change a lot, but that is to be expected given the small increment of growth examined. Some important information was observed in X-ray imaging such as the existence of air gaps between the needle and polymer. Discontinuities in tree structure may be not interpreted as isolated tree channels but instead suggested the limitation of the micro-XCT used in the tree study. This was
previously noted, for example, in the observation of the sub-micro details such as fine

tree growth [24]. A recent study [116] about DC trees in LDPE also revealed the similar
discontinuities in the tree structure by using nano-XCT technique and these features were
regarded as a ‘pre-tree’ stage of degradation. However, due to the very weak contrast of

sub-micron tree channels presented, it is still difficult to identify whether the detail has

been damaged by subsequent processes. More work is required to understand the

fundamental quality of X-ray imaging and image processing to ensure a clear

interpretation of such data.

The 3D reconstruction of electrical trees have been analysed together with PD

measurements. In the multistage treeing study, it was proposed that PDs with higher

maginudes could directly contribute to the structural extension in tree length and the rest

can contribute to the widening of the tree channel. This provides increased quality of

interpretation of PD data.
6 The impact of inorganic fillers on electrical tree growth

6.1 Introduction

This chapter presents electrical treeing tests in both unfilled and filled epoxy systems using needle-to-plane samples. The aim of these tests is to find out how trees grow in the dielectric and if, and why, inorganic fillers enhance the treeing resistance. The first section analyses the characteristics of the electrical tree in unfilled epoxy resin. Then the treeing phenomenon in micro-filled materials is discussed including its PD features and tree structures revealed by synchrotron XCT imaging. Finally, particle dispersion and surface treatment in nanocomposites are compared and discussed according to their impact in the tree tests. Several factors such as the inception time, time to breakdown and the tree structure are used to describe the electrical tree in different dielectric batches.

6.2 Unfilled epoxy resin

Electrical treeing was carried out in unfilled epoxy resin (CY225/HY925) to investigate the degradation procedure. This group was considered as the reference to the other treeing results in filled dielectrics, so the aim of this section is to develop knowledge about the way of tree growth and some intrinsic treeing properties.

6.2.1 Treeing breakdown tests

Electrical tree growth in the unfilled system was observed by using optical methods. Table 6.1 shows the time to breakdown in 6 samples tested. All samples failed at 13 kV_{rms} and most of the trees were quickly initiated (the initiation time was less than 1 seconds) and then led to breakdown. The overall tree structures in all tested samples were similar but detailed characteristics of the tree growth could be different among samples. However, it was believed that although the lifetime might be different, the classification of treeing stages should be the same in all samples. As a result, one sample was selected as an example to explain the whole treeing procedure.
Chapter 6: The impact of inorganic fillers on electrical tree growth

Table 6-1: Initiation and breakdown times for unfilled epoxy resin energized at 13 kV_{rms}.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Initiation time* (mins)</th>
<th>Breakdown time** (mins)</th>
</tr>
</thead>
<tbody>
<tr>
<td>S1</td>
<td>2</td>
<td>73</td>
</tr>
<tr>
<td>S2</td>
<td>0</td>
<td>44</td>
</tr>
<tr>
<td>S3</td>
<td>0</td>
<td>41</td>
</tr>
<tr>
<td>S4</td>
<td>20</td>
<td>66</td>
</tr>
<tr>
<td>S5</td>
<td>0</td>
<td>23</td>
</tr>
<tr>
<td>S6</td>
<td>0</td>
<td>46</td>
</tr>
<tr>
<td>Mean</td>
<td>3.7</td>
<td>48.7</td>
</tr>
</tbody>
</table>

* Initiation time was estimated from optical images. ‘0’ means the tree was initiated immediately after applying the high voltage.

** Breakdown time = initiation time + time for current to runaway.

Figure 6-1 to Figure 6-4 shows the whole tree growth in the unfilled sample S2. By identifying the different treeing features from optical images, the growth of an electrical tree can be divided into four stages:

- **Tree initiation stage**: This is the period leading up to the first observation of an electrical tree after energization. Generally this stage can last from a few seconds to several hours depending on the different insulation materials and voltage levels [14, 16]. In this case the tree was immediately initiated (less than the exposure time of the CCD camera) at 13 kV_{rms}. Optical images of this period are shown in Figure 6-1.

![Figure 6-1: Images of the electrical tree growth in the epoxy sample S2 at 13 kV_{rms}, 50 Hz in the initiation stage from 0 s (left) to 1 s (right).](image)

- **Early tree growth stage**: After the initiation stage, the tree structure started to propagate further. In this period the tree structure is simple. There are two distinguishable types of tree branch found by using optical imaging: the first one
looks dark under the backlight and usually with the diameter of 2 to 10 µm; the other one is much brighter and has very small tubes (usually < 1 µm). By referring to the description introduced by Zheng et al [24] and Iddrissu et al [25], for all the texts below, the darker channel is called ‘dark tree’ and the tubule channel is called ‘fine tree’. The growth of the ‘dark tree’ seems to be consistent to the typical ‘branch tree’ growth in the literature while the ‘fine tree’ is rarely mentioned probably due to the insufficient imaging accuracy [1, 11, 17]. In the same time, with the consideration of the tree conductivity introduced in Figure 2-2, the ‘dark tree’ may be consider as conducting while the ‘fine tree’ is non-conducting but this will be further discussed in the Discussion chapter. It was also found that all tested samples contained these two tree types and they usually occurred at the same time. In this stage, both ‘fine tree’ and ‘dark tree’ grew from the beginning. Optical images of this period are shown in Figure 6-2.

- **Tree extension stage**: The growth of the ‘dark tree’ structure almost stopped after the early treeing stage. The similar slowed growth of the ‘dark tree’ was also found in [24] and it was named as an ‘initial dark tree’. The lighter tubule channels (‘fine tree’) with smaller diameters extended further than the ‘dark tree’. After several minutes without growing the dark channel restarted to grow by following the existing small tubules and the whole structure become visibly denser and more complicated. This period corresponds to the period in Figure 6-3.
Chapter 6: The impact of inorganic fillers on electrical tree growth

- **Pre-breakdown**: Tree channels continually extended further until reaching the planar earth, but the insulation material did not immediately fail. It was found that some ‘dark tree’ channels appeared, following those paths of previous ‘fine tree’ channels. A dark bubble was observed in the later stage and this might be interpreted as localised breakdown occurring. This period corresponds to the period in Figure 6-4.

![Images of the electrical tree growth in the unfilled epoxy sample S2 at 13 kV_{rms}, 50 Hz in the extension stage from 435 s (left) to 745 s (middle) to 2015 s (right).](image)

- **Breakdown**: a large black breakdown channel appeared in the material and the leakage current immediately increased. The insulation material has lost its dielectric properties in this stage and becomes conducting.

![Images of the electrical tree growth in the unfilled epoxy sample S2 at 13 kV_{rms}, 50 Hz in the extension stage from 2015 s (left) to 2450 s (right).](image)
These treeing stages in the sample S2 were then identified according to their quantified growth in the vertical direction, as shown in Figure 6-5. Treeing in this case was initiated at the same time as energizing so the tree initiation stage was ignored in the plot. The early tree extension stage could be identified from 0 to about 420 seconds where ‘dark tree’ grew simultaneously with ‘fine tree’ and its growth rate was almost constant at about 0.5 µm/s. After that when the ‘fine tree’ structure started to spread further, the growth rate of ‘dark tree’ stopped and after about 500 seconds it restarted to propagation with a faster growth rate at 2.3 µm/s. Around 1980 seconds, the ‘fine tree’ channel firstly reached the earthed plane and after another 180 seconds the ‘dark tree’ approached the earth as well but without breakdown. Until that, the phenomenon stepped into the pre-breakdown stage and after about 480 seconds, the material finally failed.

6.2.2 PD measurement

An overview of PD activities in the unfilled batch (6 samples) is quantified in Table 6-2 and it will be used to compare the different PD phenomena when silica fillers are involved later as shown in Section 6.3.4 and Section 6.4.4. In order to understand how PD activities impact or reflect the tree morphology in the unfilled epoxy, a case study on PD phenomenon was also carried out in sample S2. Figure 6-6 shows a plot of the maximum PD magnitude and the PD number per voltage cycle collected during the treeing. It was found that changes in these two parameters are almost simultaneous and they were also associated with the treeing stages shown above.
Chapter 6: The impact of inorganic fillers on electrical tree growth

Table 6-2: Statistical results of PD number and PD energy of all 6 unfilled samples.

<table>
<thead>
<tr>
<th></th>
<th>Unfilled</th>
</tr>
</thead>
<tbody>
<tr>
<td>Number of PDs</td>
<td>1.21×10^5</td>
</tr>
<tr>
<td>Average PD rate (s)</td>
<td>54</td>
</tr>
<tr>
<td>Total PD energy (J)*</td>
<td>0.021</td>
</tr>
<tr>
<td>Average PD power (W)</td>
<td>8.04×10^-6</td>
</tr>
</tbody>
</table>

* Total PD energy was calculated by Equation 11 introduced in Section 5.3.

Before the first 420 seconds, regarded as the early tree growth stage, the PD magnitude gradually increased to ~40 pC, which corresponded to the early tree growth stage identified in Figure 6-5. The PD number per cycle in this period quickly increased to about 80 per cycle at 240 seconds and then varied between 50 and 80 per cycle. After the early tree growth stage, when the fine tree started to extend further but the dark tree did not, there was no PD detectable and both the PD magnitude and number immediately dropped to 0. Around 700 s, some sporadic PDs re-ignited with a magnitude of 20 pC but the PD number still remained at a very low level. Even in the later extension stage (~1500 s) the sporadic PD appeared with a higher magnitude up to 52 pC but the number of PD per cycle was still low (1 to 2 per cycle). During the pre-breakdown stage from about 2000 s, the trend in PD number started to increase again and had a small increment with the time. Some significant bursts in magnitude happened in this stage and when the tree was approaching the breakdown stage, the PD magnitude became the highest value recorded at 210 pC and the PD number reached 480 per cycle.

Figure 6-6: Maximum discharge magnitude (red) and number of discharges (black) in each cycle during the tree growth in the unfilled epoxy sample S2. The whole treeing period is divided by 3 stages: (1) early tree growth stage; (2) extension stage and (3) pre-breakdown stage.
As shown in Figure 6-5 and 6-6, it was found that both the tree growth rate and maximum PD magnitude linearly increased from 0 s to 430 s and the overall maximum magnitude also showed an increasing trend. According to the theory stated in [3, 66] and findings in Section 5.3, the maximum PD magnitude is considered to occur in the channel with the longest length. The correlation between these two parameters is shown in Figure 6-7. It was found that at the early stage, the maximum PD magnitude and the tree length seems to be linear and the fitted function could be expressed by:

\[ Q_{\text{max}} = 0.147 \times L + 2.796 \]  

where \( Q_{\text{max}} \) is the maximum discharge magnitude in each voltage cycle with the unit of pC and \( L \) is the tree length in \( \mu m \). A new parameter \( dQ/dL \) could be considered to describe the dark tree growth in the early tree growth stage, which physically suggested the value of PD magnitude required for a certain tree length and in the previous case as shown in Figure 6-7, it was calculated to be 0.147 pC/\( \mu m \). After the tree length arrived about 250 \( \mu m \), the maximum PD magnitude showed less dependence to the tree length for a while. The similar linearity and slope value was found again at later treeing stage where the tree length is about 1300 \( \mu m \). It should be noted that the maximum tree length at this moment did not correspond to the original ‘dark tree’ structure in the early tree growth stage. One of the potential mechanism of this special PD characteristics revealed in this case may be the conductivity of the tree channel which is thought to be varying during the tree growth. Further understanding about the tree conductivity is explained in the Discussion chapter (Section 7.1).

Figure 6-7: Maximum discharge magnitude against the tree length (‘dark tree’) in the unfilled epoxy sample S2 at (1) early tree growth stage, (2) extension stage and (3) pre-breakdown stage; The red line shows the linear relationship between the maximum PD magnitude and the tree length at different periods.
Chapter 6: The impact of inorganic fillers on electrical tree growth

The optical images taken during the tree growth could be linked with the corresponding PD changes at different stages. To obtain better contrast, the tree image of sample S2 was further processed and as shown in Figure 6-8, the original ‘dark tree’ is now observed in bright white and the ‘fine tree’ is shown in grey. Two transitions of the tree structure from 435 s - 745 s and 745 s – 1500 s are compared in Figure 6-8.

![Figure 6-8](image)

Figure 6-8: The tree structure in the unfilled epoxy sample S2 imaged at (a) 435 s, (b) 745 s and (c) 1500 s. The colour of all images was inverted.

When the tree was mature at 435 s, both ‘dark tree’ and ‘fine tree’ could be identified by the difference in diameter. After just entering into the extension stage from 435 s, only a few PD signals could be detected as shown in Figure 6-6. From the optical image from Figure 6-8(a) to Figure 6-8(b), the extension of ‘fine tree’ channels was still observable but the original ‘dark tree’ stopped growing. So it can be concluded that the growth of ‘fine tree’ is not associated with PD phenomena or the corresponding PD signal is below the sensitivity of the detection system. Then from Figure 6-8(b) to Figure 6-8(c), the growth of ‘dark tree’ reappeared in the tree structure and more PD signals were also detected. Figure 6-9 shows an example of PD magnitude and number behaviour between 752 s and 755 s. Some sporadic PDs are found with varying magnitudes from 1 pC to 40 pC but the number per cycle is only 1 to 5 in this case. In contrast to the PD magnitude trend from 0 s to 435 s shown in Figure 6-6, the PD behaviour here was not continuous and there were several extinction periods found in this stage and the interval time was from 0.02 s (one voltage cycle) to 1 s (50 voltage cycles). Referring to the treeing image shown in Figure 6-8, it was found that the growth of ‘dark tree’ was following with the extension of ‘fine tree’ and the newly developed ‘dark tree’ parts followed the ‘fine tree’ path, which means the growth of a ‘fine tree’ might be the pre-condition of the
occurrence of a ‘dark tree’. As a result, it can be proposed that these sporadic PDs may directly point to the ‘fine-to-dark’ transition (a transition from ‘fine tree’ to ‘dark tree’). The experimental results shown in [24] also revealed similar sporadic PD magnitudes during the ‘fine tree’ and ‘dark tree’ growth but its characteristics were not so deeply considered.

During the ‘fine-to-dark’ transition the number of PD per cycle could be very few in the early treeing stage but it increased and the trend of both magnitude and number could become steady and continuous in the later stage. Figure 6-10 shows the very late extension stage from 2253 s to 2256 s. The PD is also shown to be sporadic with the magnitude above 55 pC while there is no extinction period. There are many troughs containing only PDs of low magnitudes about 3 pC. The PD number also becomes steady in this case, which means many PDs occurred in each voltage cycle. The physical change of the tree structure in the later stage became very complex so it was difficult to identify newly developed channels when PD occurred. This will be discussed later in the Discussion chapter.
Figure 6-11 illustrates typical PRPD patterns within 10 second periods in different treeing stages. During the initiation stage, two typical shapes of PD clusters can be identified: a ‘wing-like’ pattern with higher magnitude and dependent on the point on voltage waveforms; ‘turtle-like’ patterns with small magnitude and all amplitudes independent of the point on wave [10]. When the treeing was in the extension stage an ‘upright’ pattern was found. The PD number was small and these PDs only occurred in a narrow phase range as shown in Figure 6-11(b). In addition, the ‘upright’ pattern with little magnitude showed less dependence on the point on the voltage wave. After several minutes until the pre-breakdown stage, the PD pattern developed as shown in Figure 6-11(c), in which there were still some ‘upright’ PD patterns found with higher magnitude but superimposed upon many low-magnitude PDs. Finally when approaching the breakdown, PDs occurred with similar features as the one in Figure 6-11(d) but with 5-times higher magnitudes.
6.2.3 Summary

The whole treeing process in an unfilled epoxy sample has been characterized by analysing both optical images and PD data, and the growth of ‘fine trees’ and ‘dark trees’ were identified in experimental results. It was found that there were no discharges detected during periods of only ‘fine tree’ growth, but this may be also interpreted as the result of insufficient sensitivity (1 pC) in this case. The associated PD activities of ‘dark tree’ was found to be closely associated with the tree length especially at the early stage and it was consistent to result reported in the literature [3, 64].

The electrical treeing process was described and characterized in several stages. The initiation stage was defined as the time interval between energization and the appearance of the first tree channel but in most samples it was too quick to be detected according to optical images. In the early tree growth stage both ‘fine trees’ and ‘dark trees’ grew with time but the growth of the ‘dark tree’ stopped at the end of the stage. At the early
extension stage, only the ‘fine tree’ was observed to extend along with a reduction of both PD magnitude and number. After several minutes the ‘dark tree’ restarted to extend and the transition from the ‘fine tree’ to the ‘dark tree’ could be very rapid, which resulted in the occurrence of sporadic PD at the same time. The growth of the ‘fine tree’ and ‘dark tree’ could be simultaneous and when the treeing approached the pre-breakdown stage, both PD magnitude and number became larger and continuous.

During the pre-breakdown stage, there were several ‘fine tree’ channels connecting the electrode and the earth but they did not result in the final dielectric failure. Both PD trends and PRPD patterns became complicated because there are more channels and lots of phenomena may occur at the same time.

The characterization of the electrical tree growth in the unfilled sample is a reference to compare the treeing phenomenon in filled systems described in next two sections. Some further characterization and discussion about the treeing and breakdown in unfilled epoxy system is included in the Discussion chapter (Section 7.1).
6.3 Micro-filled epoxy resin

In this section, microsilica-filled samples were prepared for the treeing tests. Before the electrical experiments, SEM was used in this case to investigate the particle dispersion. Due to the opaque feature of micro-filled epoxy resin, the only way the breakdown time can be measured is by using the leakage-current detector from the HV amplifier. Treeing information was mainly extracted from PD data due to the lack of optical imaging in the opaque material. Synchrotron XCT was applied in three samples with different loads and the tree structure in the micro-composite was successfully reconstructed for the first time. Some detailed characteristics such as the treeing path and the interaction between tree channels and fillers was also revealed.

6.3.1 Dispersion of microsilica

Several SEM images were captured on reference samples (described in Section 3.2.3). Figure 6-12 compares the particle distribution from 5 wt% to 30 wt%. Objectively the denser population of particles found in (b) and (c) suggests the reduced separation as the load increases. The theoretical surface-to-surface distance could be calculated by using the Equation 8 and the result is shown in Figure 6-13. The calculated distance between neighbouring particles by using the method introduced in Section 4.3.2 was smaller than the theoretical value shown in Section 4.3.1, which may be the result from non-uniform particle sizes, shapes and imperfect dispersion. Also it should be noted that SEM images only considers 2D images, which resulted in a larger distance between particles. All SEM images of micro-composites are shown in the Appendix 1.

Comparing these measured distances with the diameter of the electrical tree channel (normally between 2 µm and 5 µm) measured from both optical and X-ray images, more individual micro-size particles in 15 wt% and 30 wt% increases the possibility of appearing in the path of the tree propagation and may block the tree growth physically [27].
6.3.2 Treeing breakdown tests

Table 6-3 gives the information for all the tested samples. Samples were energized with a 13 kV$_{\text{rms}}$, 50 Hz AC voltage in the same circumstance as the unfilled test shown in Section 6.2.1. In each batch there was one sample selected for XCT imaging. In these imaged samples, only an early-stage tree was grown so only initiation time was recorded for these samples. The breakdown time, $T_{BD}$, is the time measured from application of the high voltage to the electrical failure (initiation time + time for current to runaway). The treeing process could not be observed by optical methods for those samples so in this case the initiation time was estimated from the first observed PD signal.

It was found that the initiation time needed for the PD occurrence in most samples was very short. There were 5 samples (5MF5, 5MF6, 15MF4, 15MF6 and 30MF1) which had an initiation time above 10 mins and one of them did not breakdown after energizing for 36 h. No PD was observed in that sample.
By comparing with the unfilled results, it can be seen that the lifetime in all filled groups was effectively enhanced by the presence of the microsilica. Figure 6-14(a) presents the box chart of the breakdown times in different batches. It was found that the breakdown time $T_{BD}$ increased linearly with the square root of the filler level. The line of linear fit shown in Figure 6-14(b) is:

$$T_{BD} = 93,900 \cdot w^{1/2} + 2550 \quad (13)$$

where $w$ is the weight fraction of silica, and $T_{BD}$ is in seconds. This relationship suggests the limited effect of continuously adding fillers into the dielectric (improvement from +4000 s/wt% to +1333 s/wt%). During the compounding, it was found that ~30 wt% was the physical limit to the amount of this micro filler which could be added into the epoxy resin used in this case.

Figure 6-14: (a): Time to breakdown at 13 kV$_{rms}$, 50 Hz. For each set of results the box shows the range between the upper quartile and the lower quartile, the small square and the middle line are the mean and median value respectively. The dotted line is given in Equation 13; (b): fitted line of breakdown time against the square root the weight fraction of fillers.
Table 6-3: Initiation and breakdown time for micro-filled samples.

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>Initiation time* (mins)</th>
<th>Breakdown time** (mins)</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>5 wt%</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
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<td>481</td>
<td></td>
</tr>
<tr>
<td>5MF2</td>
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<td>572</td>
<td></td>
</tr>
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</tr>
<tr>
<td>5MF4</td>
<td>0</td>
<td>203</td>
<td></td>
</tr>
<tr>
<td>5MF5</td>
<td>12</td>
<td>322</td>
<td></td>
</tr>
<tr>
<td>5MF6</td>
<td>24</td>
<td>287</td>
<td></td>
</tr>
<tr>
<td>Mean</td>
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<td>366</td>
<td></td>
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<tr>
<td>5MF7</td>
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<td>/</td>
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<td></td>
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</tr>
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</tbody>
</table>

* Initiation time was estimated at the moment when the first PD generated with the magnitude above 1 pC. ‘0’ means the tree was initiated immediately after applying the high voltage.

** Breakdown time = initiation time + time for current to runaway.

*** No PD was detected in the next 36 hours after initiation. This sample was not included in the mean value calculation.
6.3.3 XCT tree imaging

The reconstruction of composites required identifying and selecting the range of pixel intensities needed. As shown in Figure 6-15, a line profile of the pixel intensities along a length of 80 pixels was plotted across three materials present in the sample: epoxy resin, silica filler and tree channel (regarded as air). Materials with different mass densities could be distinguished by the grey level values in XCT images. According the mechanism of the phase contrast imaging [110], the intensity plot in Figure 6-15(b) can be explained in detail. The image was firstly simplified from 16 bits to 8 bits so the maximum grey value was compressed from 65025 to 255. The large range between the 20th to 45th pixel giving the higher intensity from 125th to 250th pixel corresponded to the silica material and also there should be two fringes generated from the phase contrast effect (highlighted in circles at 15 and 52). Similarly the tree channel could be identified from the 66th to 70th pixel with the intensity of 0. In this way, the required objects could be accurately located with artificial noise removed as much as possible.

![Image of XCT tree imaging](image.png)

Figure 6-15: (a) An intensity meter along a length of 80 pixels across the microsilica and tree channel in the 5 wt% sample 5MF7; (b) Profile of pixel intensity along the red line. Pixel size: 0.33 µm.

In reality, the reconstruction work was not always easy, for example the tree channel and fillers were usually overlapped. Figure 6-16 shows an example of the challenge during the reconstruction. When the tree channel propagated near the agglomeration objects, the fringe outside usually contained dark fringes and they could overlap the tree cross-section and consequently, merge with a channel giving an illusion of an enlarged diameter. In order to reduce the influence of these fringes, the identification of different materials is partly achieved by judging continuity in consecutive slices. Due to the X-ray
absorption by the silica, some details within large particle clusters could not be fully resolved. So in this study, most agglomerations were identified by their size rather than constitution. It was found that more than 90% of filler particles are between 1 µm and 10 µm which matches the information from the supplier. As a result, those apparent particles with sizes between 20 to 80 µm were considered as agglomerations of microsilica.

![Figure 6-16: The situation where electrical tree overlapped with fringes from silica particles in the 5 wt% microsilica-filled sample 5MF7. Yellow outlined: cross-sections of the electrical tree.](image)

3D data was then reconstructed shown in Figure 6-17. Both silica and tree channels could be identified and the epoxy resin was removed in the reconstruction. XCT is not the usual way to investigate filler’s distribution because the X-ray energy can be attenuated when encountering high density objects such as silica. As a result, filler deeper in the material might be hidden. For that reason, SEM imaging is used instead for dispersion analysis in later sections.

Before describing the tree geometry, it should be emphasised that these 3D reconstructions do not present the general tree structures in micro-filled samples and their repeatability should be further examined. However, the 3D tree structures shown below do reveal the different geometry caused by the addition of fillers in epoxy systems so they are still very worthy to study individually to understand the treeing phenomenon with the impact of micro-size fillers.

As shown in Figure 6-17, the general shapes of electrical trees in 5MF7 and 15MF7 looked similar, but the tree structure in the 30MF7 was much denser. By using the generally adopted descriptions of tree shapes [11], in the 5MF7 and 15MF7 case the electrical trees had distinguishable main trunks and few discrete branches so they could be regarded as typical ‘branch trees’. The highly dense tree structure in 30MF7 could be better characterised as a ‘bush tree’. But it could also be observed in figure 6-17(c) that it has a double-bush feature, which differs from typical bush tree growth found in unfilled
Chapter 6: The impact of inorganic fillers on electrical tree growth

epoxy systems. Generally bush trees have a spherical structure with the diameter of hundred micrometres, while in the case seen here it appeared a second bush structure extended and grew out of an initial, more typical bush tree of only 70 µm in length. It was also found that there were some large disconnections between branches. The cause of these disconnections may be similar to the situation found in the unfilled sample (shown in Section 5.3) where some tubule channels were presumed to exist in an area but were too narrow to be detected.

![3D rendering of electrical trees in microsilica-filled sample](image)

Figure 6-17: 3D rendering of electrical trees in microsilica-filled sample (a) 5MF7 energized at 13 kV\textsubscript{rms} for 73 mins, (d) 15MF7 energized at 13 kV\textsubscript{rms} for 116 mins and (c) 30MF7 at 13 kV\textsubscript{rms} for 199 mins.

The ‘double bush’ feature of the 30MF7 can be more readily identified in Figure 6-18. This shows the number of tree channels in a slice taken every 0.33 µm in the direction of tree growth. Both tree structures in 5MF7 and 15MF7 blends have few channels around
needle tips while the double-bush type in 30MF7 has over 30 increasing to 45 within 50 µm of the needle tip. This represents a relatively low branch-density bush tree [105]. The two bush structures can be distinguished at about 60 µm from the needle tip, where the number of branches growing away from the needle is reduced from 58 to 5 before it increases up to 33 again.

Quantitative parameters from the 2D image and 3D models are given in Table 6-4. The fractal dimensions in 5MF7 and 15MF7 were lower than that in the 30MF7. The tree structure in the 5MF7 has the largest diameter channels, even though it was grown for the shortest time, but the growth rate and fractal dimension are similar to the 15MF7. The fractal dimension value of 2 is commonly considered as a threshold between branch and bush trees [84]. The trees had only crossed 20% of the insulation at the time of imaging, so the trees might evolve as they develop. The double-bush tree in 30MF7 had a growth rate of about half the lesser-filled materials, and a 3D fractal dimension of about 2. This value is consistent with other observations of bush trees [105]. As described in Section 4.2.2, tortuosity is a measure of the straightness of branches determined from the ratio of the total branch length and the shortest distance between the start and end points. Branches in 30MF7 had a similar tortuosity to trees in the less-filled materials, so the slower growth rate, and longer lifetimes is not because of significantly greater meandering in branch growth paths in the highly filled polymer. As the imaged tree structures in these filled system had tubule diameters between 1 µm and 4 µm, all tree

Figure 6-18: Number of tree channels in slices taken perpendicular to the direction of growth along the tree length in the imaged microsilica-filled samples 5MF7, 15MF7 and 30 MF7.
channels could be considered as the ‘dark tree’ according to the basic classification between ‘fine tree’ and ‘bush tree’ introduced in Section 6.2. It should be noted that the ‘fine tree’ channels are too thin to be imaged in this case so it does not imply they do not exist.

Interactions between tree channels and filler particles can be examined in the XCT tree replicas. There are three specific situations found in the models illustrated in Figure 6-19. Figure 6-19(a) shows a tree branch deviating from its path in front of a single silica particle of about the same diameter as the branch width (2 to 4 µm) in 5MF7. It is found that the tree branch will not grow to touch or envelop a silica particle. This deviation away from individual silica particles suggests that branches may become more tortuous in filled materials, but this is not reflected in the data in Table 6-4. This is perhaps because the deviation is not at a large angle from the original direction. Figure 6-19(c) shows a tree channel passing through a large ~80 µm agglomeration. The agglomeration is much larger than the tree branch diameter. As the released PD energy is insufficient to directly damage the inorganic silica particles we can assume the tree grows through epoxy or pre-existing low density regions in the agglomeration [163]. The XY slice in Figure 6-19(d) shows the tree channel maintains its size and integrity within the agglomeration (the channel has a mean diameter of 3.46 µm within the agglomerate and 3.62 µm outside). So it is speculated that a tree channel can be diverted by single filler particles but not by an agglomeration. Further interpretation will be given in the Discussion chapter (Section 7.2).

Table 6-4: Quantified tree parameters in micro-filled samples 5MF7, 15MF7 and 30MF7.

<table>
<thead>
<tr>
<th></th>
<th>5 wt%</th>
<th>15 wt%</th>
<th>30 wt%</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Vertical Length (µm)</strong>&lt;sup&gt;(a)&lt;/sup&gt;</td>
<td>122.12</td>
<td>228.65</td>
<td>186.01</td>
</tr>
<tr>
<td><strong>Fractal Dimension (3D)</strong></td>
<td>1.71</td>
<td>1.65</td>
<td>1.94</td>
</tr>
<tr>
<td><strong>Average Growth Rate (µm/min)</strong>&lt;sup&gt;(b)&lt;/sup&gt;</td>
<td>1.70</td>
<td>1.98</td>
<td>0.96</td>
</tr>
<tr>
<td><strong>Mean Diameter (µm)</strong></td>
<td>3.05</td>
<td>1.89</td>
<td>2.31</td>
</tr>
<tr>
<td><strong>Tortuosity (3D)</strong></td>
<td>1.33</td>
<td>1.08</td>
<td>1.02</td>
</tr>
</tbody>
</table>

<sup>(a)</sup> The length was calculated by multiplying the number of slices by the voxel size (N×0.33 um);

<sup>(b)</sup> The growth rate was calculated by using vertical length divided by the total time of tree growth;
Figure 6-19(e) illustrates one of the apparently disconnected structures seen in the 30MF7. A free-standing low density section exists on the side of a filler particle, and is not associated with any nearby tree branch in the reconstruction. Figure 6-19(f) shows the cross-section in which the channel can be observed as isolated and some 20 µm away from a tree branch. There are only few cases which have been found and most of these isolated features are about ten micrometres in length. It is likely that the disconnected branches and voids are linked by narrow channels which the XCT is not yet capable of observing. Over interpretation, and assuming all features are observed would be incorrect.

Figure 6-19: Examples of interaction between tree branches and filler particles and agglomerates from XCT tree replicas. (a) A tree channel growing around a single microsilica particle in 5 wt% sample 5MF7 and (b) the corresponding XY cross-section of the bent tree channel; (c) A tree channel penetrating through an agglomeration in the 5 wt% sample 5MF7 and (d) the corresponding XY cross-section showing the penetrating channel; (e) The area where an apparently disconnected degradation area is seen in 30 wt% sample 30MF7 and (f) the corresponding XY cross-section.
6.3.4 PD measurement

Table 6-5 summaries the PD phenomenon in all tested microcomposites shown in Table 6-3 and then compares with unfilled samples described in Section 6.2.2. With the modification of microsilica, it was found that both total PD number and average PD rate significantly increased but as the concentration increased, fewer PD occurred in each time unit. Higher PD energy was measured during the tree growth in 30 wt% samples but because the average PD power gradually decreased from 0 wt% to 30 wt%. So generally, it could be concluded that the addition of micro-size fillers would promote partial discharge in number but slow down the releasing speed of PD energy.

<table>
<thead>
<tr>
<th></th>
<th>Unfilled</th>
<th>5 wt%</th>
<th>15 wt%</th>
<th>30 wt%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Number of PDs</td>
<td>1.21×10^5</td>
<td>1.43×10^7</td>
<td>1.68×10^7</td>
<td>9.90×10^6</td>
</tr>
<tr>
<td>Average PD rate</td>
<td>54</td>
<td>821</td>
<td>581</td>
<td>152</td>
</tr>
<tr>
<td>Total PD energy</td>
<td>0.021</td>
<td>0.15</td>
<td>0.23</td>
<td>0.24</td>
</tr>
<tr>
<td>(J)*</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Average PD power</td>
<td>8.04×10^-6</td>
<td>7.84×10^-6</td>
<td>6.77×10^-6</td>
<td>4.63×10^-6</td>
</tr>
<tr>
<td>(W)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

*Total PD energy was calculated by Equation 11 introduced in section 5.3.

Then PD data was selected from these samples imaged by X-ray (5MF7, 15MF7 and 30MF7) in order to find out how the channel/filler interaction impacts PD characteristics. As shown in Figure 6-20, for the 5 wt% case, the PD signal was collected in a 70-minute period which generated the ~122 µm branch tree shown previously in Figure 6-17 (a). Referring the PD data in the unfilled sample shown in Section 6.2.2, it was found that at a similar tree length ~ 200 µm, the maximum magnitude had a ~50 pC increment in the 5 wt% sample. At the early stage the PD magnitude was relatively consistent but then there was an extinction periods found from 740 s to 780 s. Some typical sporadic PD signals were detected from 1300 s to 2000 s with the maximum magnitude below 70 pC and PD numbers below 10. Peaks of magnitude showed an increasing trend until 2850 s where the PD magnitude reached maximum at 110 pC. After that the PD rate maintained at a high level until the breakdown at 4200 s.
Figure 6-20: Maximum discharge magnitude (red) and the number of discharges (black) in each cycle during the tree growth in 5% micro-filled epoxy sample 5MF7.

Figure 6-21 shows the plot of PD magnitude and PD number for the tree structure in the sample 15MF7. There were lots of variation in the magnitudes even through the tree structure did not have obvious changes. These bursts of high magnitude PD occurred from the beginning of the tree growth with a peak of 75 pC but with very low number of PDs per cycle. With time, peaks of magnitude showed increases until 170 pC around 4500 s and after a period of reduction, the PD magnitude reached 280 pC at 4900 s. Few periods in this case had no PD activity but there were some with the minimum PD value of 1 to 3 pC. Generally speaking, the overall PD magnitude in this case was much higher than that in 5 wt% and unfilled sample. There were only few PDs found in the first 90 mins where each voltage cycle only contained less than 20 PDs. At around 5700 s and 5900 s, there were two temporary bursts up to 320 in number and then they dropped below 20 again until the breakdown.

For the sample with 30 wt% micro fillers, while still sporadic, PD activities with high magnitudes covered the whole treeing period as shown in Figure 6-22. Discharges with the magnitude of ~60 pC occurred from the beginning and these peaks had a relatively steady increase to 150 pC from 800 s to 5400 s. There was an extinction period found from 7300 s to 7600 s and then the maximum PD magnitude jumped to the maximum value 170 pC around 8000 s. The number of PDs showed burst characteristics as well in
this case but most of them contained less than 25 PD. The highest number appeared at 7200 s where the voltage cycle at that moment contained 150 PDs in every 0.02 seconds.

As previously shown in Figure 6-9 and Figure 6-11, the PD in unfilled epoxy formed ‘upright’ patterns especially at the moment when the sporadic PD occurred. A similar finding can be also found in micro-filled samples. As shown in Figure 6-23, some typical sporadic PDs were selected from the PD trends shown above (i.e. 1840 s to 1900 s in
5MF7). In all three filled samples, the corresponding PRPD pattern of the selected time interval matched the characteristics of the ‘upright’ patterns: occurring in a narrow phase range and magnitude showing little or no dependence on the point on the voltage wave. The main difference between these three samples with different loadings is the PD magnitude: most PDs in 5 wt% are at the range of 1 pC to 4 pC and the maximum value is 17.6 pC; 15 wt% contained more PDs with the magnitude 20 pC to 40 PC; for 30 wt%, the majority of PDs are below 15 pC. These values only present the early treeing stage and may change in different intervals in later time.

![Figure 6-23: Phase-resolved partial discharge (PRPD) patterns of typical sporadic PD collected from the tree growth in the (a) 5MF7 from 1840 s to 1900 s; (b) 15MF7 from 1380 s to 1420 s; (c) 30MF7 microfilled sample from 1290 s to 1320 s.](image)
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It should be noted that even though the theoretical treeing stages identified in Section 6.2.1 may be also applicable to for filled samples, the PRPD pattern collected here can not directly suggest which stage the treeing is in, due to the unknown growth pattern of the tree structure with time.

6.3.5 Summary

The opportunity afforded by the Synchrotron XCT technique to be used on micro-composites has been successful, and the result reveals the capability to observe the electrical tree structure in the opaque samples. From these reconstructed replicas, electrical treeing phenomena have been further characterized especially concerning tree propagation in the presence of micro-size fillers.

Structural features of electrical trees in different micro-composites have been identified from 3D images. The tree shape in micro-filled systems at 5 wt% and 15 wt% could be identified as branch trees while at 30% a bush-like tree grew from the needle firstly and then another bush shape extended from the first bush tree. The formation of different tree types at the same voltage suggests that the loading level of micro-size particles could result in structural change of the electrical trees. In [32] it is suggested that the micro-size filler particles could attract the tree branch propagation due to their high permittivity and work as a barrier to influence the treeing direction physically. However from the reconstructed 3D tree images, it was found that the propagation was not indicting that branches were attracted to embedded filler locations. It was also found when there were huge agglomerations existing on the tree path, the tree channel could directly penetrate through these agglomerated clusters. Generally both of these two interactions could exist together in the micro-filled epoxy system.

From the comparison of breakdown tests between unfilled and micro-filled epoxy, the addition of microsilica has shown dramatic enhancement for the treeing breakdown time by at least eight times at the loading level from 5 wt% to 30 wt%, which is qualitatively consistent with the results in other reports [27, 123]. It was also found that the treeing breakdown time increased almost linearly with the square root of the filler level, which suggested the decreasing level of improvement as the loading goes higher. The comparison of PD features revealed that the addition of micro-size fillers could not resist the frequency of PD occurrence which is contradictory with the result shown in [164]. 5 wt% samples contained the highest average PD rate, which is 15.2 times higher than that
in unfilled samples. However, the PD power was found to be effectively suppressed when more fillers were filled into the epoxy and this seems to be consistent with the opinion expressed in other work [164]. Typical sporadic PD signals were also detected in micro-filled epoxy samples and their PRPD patterns were consistent to those in unfilled systems. As introduced in Section 6.2.2, the sporadic PD suggests the ‘fine-to-dark’ transition but so far there is little evidence supporting the growth of ‘fine trees’ in micro-filled materials.
6.4 Nano-filled epoxy resin

This chapter shows the experimental results from the nanosilica filled epoxy resin. The silane treatment introduced in Section 3.2.3 was applied in three batches to compare the dielectric properties of materials prepared in different conditions. Besides the breakdown test, the dispersion of nano particles was further analysed by comparing both skewness and nearest neighbouring spacing. Electrical trees in most of samples were hard to observe by optical methods so some treeing features were also estimated by PD measurement. Synchrotron XCT was applied in two samples (1 wt% untreated and 3 wt% untreated) in this case to investigate structural features.

6.4.1 Dispersion of nanosilica

Figure 6-24 shows some SEM images on the particle dispersion in different samples. By comparing SEM images, it can be seen that larger agglomeration occurs much more often with increased loading especially in those samples without the surface treatment while the treated particles were observed to be dispersed more homogeneously. Both skewness and nearest neighbouring spacing can be calculated by using the methods introduced in Section 4.3.2. Table 6-6 compares calculated results with the theoretical surface-to-surface distance (introduced in Section 4.3.1) in both untreated system UT and silane treatment system ST.

Table 6-6: Skewness and nearest neighbouring spacing measurement in nano-filled samples.

<table>
<thead>
<tr>
<th></th>
<th>Skewness</th>
<th>Nearest neighbouring distance (experimental) (nm)</th>
<th>Surface-to-surface distance (theoretical) (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1% UT</td>
<td>5.40</td>
<td>568.02</td>
<td>77.11</td>
</tr>
<tr>
<td>1% ST</td>
<td>5.07</td>
<td>432.32</td>
<td>77.11</td>
</tr>
<tr>
<td>3% UT</td>
<td>4.24</td>
<td>513.41</td>
<td>46.88</td>
</tr>
<tr>
<td>3% ST</td>
<td>3.98</td>
<td>398.60</td>
<td>46.88</td>
</tr>
<tr>
<td>5% UT</td>
<td>3.95</td>
<td>765.11</td>
<td>36.01</td>
</tr>
<tr>
<td>5% ST</td>
<td>3.76</td>
<td>198.24</td>
<td>36.01</td>
</tr>
</tbody>
</table>
It was found that the calculated nearest neighbouring distance is at least five times larger than the theoretical surface-to-surface distance. The cause of that could be aggregation because the formation of the agglomerates could consume many particles leading to less concentration of particles in the bulk. On the other hand, the theoretical distance was calculated based on the particle size of 20 nm but in practise the particle size was not uniform and could be much bigger, which lead to the higher particle-to-particle distance. It could be also found that the measured spacing between neighbouring nano particles was at least 5 times smaller than that in micro-composites.

Additionally, SEM captured tomography inside of the tree channel. As shown in Figure 6-25, a cross-section of the tree channel was exposed and imaged, it can be found that the inside of the channel was more rough than the outside which had been microtomed and there were lots of white spots found. EDX showed that there were more areas identified with Si in the treeing channel area than outside, which suggested that silica particles were
exposed on the rougher inner surface. This result is consistent with findings in other literature [18] where denser particle dispersion was found on the tree channel surface.

Figure 6-25: (a) SEM image showing the cross-section of the tree channel in the 5 wt% nanosilica filled epoxy sample 5UT1; (b) Si element map of the same image collected from EDX.

### 6.4.2 Treeing breakdown test

Tables 6-7 and Table 6-8 give the information for all the tested samples. The 13 kV\textsubscript{rms}, 50 Hz AC voltage was applied in all nanocomposites using the same circuit as for the unfilled samples. As shown in Section 3.2.3, nanocomposites made by sonicator have good transparency. However in the treeing experiments, it was still found to be a challenge to directly observe the tree structure by the CCD camera, so similarly, there were also two samples selected for the synchrotron XCT imaging. The experiment was stopped before breakdown of these two samples to get a complete tree structure. The samples were labelled in short as shown in Figure 6-26.

Table 6-7 shows a summary of the breakdown times (from energizing to complete electrical failure) obtained from untreated nano-composites with the loading range from 1 wt% to 5 wt%.

![Sample nomenclature for different nano-filled epoxy samples.](image)

Figure 6-26: Sample nomenclature for different nano-filled epoxy samples.
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Table 6-7: Tested samples without surface treatment.

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>Initiation time* (mins)</th>
<th>Breakdown time** (mins)</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 wt% untreated</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1UT1</td>
<td>0</td>
<td>121</td>
<td></td>
</tr>
<tr>
<td>1UT2</td>
<td>0</td>
<td>102</td>
<td></td>
</tr>
<tr>
<td>1UT3</td>
<td>0</td>
<td>73</td>
<td></td>
</tr>
<tr>
<td>1UT4</td>
<td>0</td>
<td>99</td>
<td></td>
</tr>
<tr>
<td>1UT5</td>
<td>0</td>
<td>113</td>
<td></td>
</tr>
<tr>
<td>1UT6</td>
<td>3</td>
<td>86</td>
<td></td>
</tr>
<tr>
<td>Mean</td>
<td>0.5</td>
<td>99</td>
<td></td>
</tr>
<tr>
<td>1UT7</td>
<td>&lt;1</td>
<td>\</td>
<td>Prepared for XCT – voltage applied for 1.8 h</td>
</tr>
<tr>
<td>3 wt% untreated</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3UT1</td>
<td>12</td>
<td>248</td>
<td></td>
</tr>
<tr>
<td>3UT2</td>
<td>2</td>
<td>197</td>
<td></td>
</tr>
<tr>
<td>3UT3</td>
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<td>154</td>
<td></td>
</tr>
<tr>
<td>3UT4</td>
<td>0</td>
<td>167</td>
<td></td>
</tr>
<tr>
<td>3UT5</td>
<td>5</td>
<td>142</td>
<td></td>
</tr>
<tr>
<td>3UT6</td>
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<td>112</td>
<td></td>
</tr>
<tr>
<td>Mean</td>
<td>3.17</td>
<td>170</td>
<td></td>
</tr>
<tr>
<td>3UT7</td>
<td>0</td>
<td></td>
<td>Prepared for XCT – voltage applied for 2.2 h</td>
</tr>
<tr>
<td>5 wt% untreated</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5UT1</td>
<td>0</td>
<td>98</td>
<td></td>
</tr>
<tr>
<td>5UT2</td>
<td>3</td>
<td>215</td>
<td></td>
</tr>
<tr>
<td>5UT3</td>
<td>0</td>
<td>158</td>
<td></td>
</tr>
<tr>
<td>5UT4</td>
<td>0</td>
<td>121</td>
<td></td>
</tr>
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<td>5UT5</td>
<td>0</td>
<td>115</td>
<td></td>
</tr>
<tr>
<td>5UT6</td>
<td>78</td>
<td>\</td>
<td>Did not breakdown ***</td>
</tr>
<tr>
<td>Mean</td>
<td>0.6</td>
<td>141.4</td>
<td></td>
</tr>
</tbody>
</table>

* Initiation time was estimated at the moment when the first PD was generated with a magnitude above 1 pC. ‘0’ means the tree was initiated immediately after applying the high voltage.

** Breakdown time = initiation time + time for current to runaway.

*** No PD was detected in the next 36 hours after initiation. This sample was not included in the mean value calculation.
Table 6-8: Tested samples with silane treatment.

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>Initiation time* (mins)</th>
<th>Breakdown time** (mins)</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 wt% treated</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1ST1</td>
<td>0</td>
<td>\</td>
<td>Did not breakdown ***</td>
</tr>
<tr>
<td>1ST2</td>
<td>3</td>
<td>65</td>
<td></td>
</tr>
<tr>
<td>1ST3</td>
<td>0</td>
<td>49</td>
<td></td>
</tr>
<tr>
<td>1ST4</td>
<td>7</td>
<td>93</td>
<td></td>
</tr>
<tr>
<td>1ST5</td>
<td>0</td>
<td>123</td>
<td></td>
</tr>
<tr>
<td>1ST6</td>
<td>5</td>
<td>178</td>
<td></td>
</tr>
<tr>
<td>Mean</td>
<td>2.5</td>
<td>101.6</td>
<td></td>
</tr>
<tr>
<td>3 wt% treated</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3ST1</td>
<td>2</td>
<td>245</td>
<td></td>
</tr>
<tr>
<td>3ST2</td>
<td>4</td>
<td>935</td>
<td></td>
</tr>
<tr>
<td>3ST3</td>
<td>29</td>
<td>1109</td>
<td></td>
</tr>
<tr>
<td>3ST4</td>
<td>0</td>
<td>794</td>
<td></td>
</tr>
<tr>
<td>3ST5</td>
<td>15</td>
<td>1006</td>
<td></td>
</tr>
<tr>
<td>3ST6</td>
<td>0</td>
<td>554</td>
<td></td>
</tr>
<tr>
<td>Mean</td>
<td>8.33</td>
<td>773.83</td>
<td></td>
</tr>
<tr>
<td>5 wt% treated</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5ST1</td>
<td>8</td>
<td>1342</td>
<td></td>
</tr>
<tr>
<td>5ST2</td>
<td>204</td>
<td>1684</td>
<td></td>
</tr>
<tr>
<td>5ST3</td>
<td>30</td>
<td>1087</td>
<td></td>
</tr>
<tr>
<td>5ST4</td>
<td>52</td>
<td>1441</td>
<td></td>
</tr>
<tr>
<td>5ST5</td>
<td>0</td>
<td>962</td>
<td></td>
</tr>
<tr>
<td>5ST6</td>
<td>97</td>
<td>\</td>
<td>Did not breakdown ***</td>
</tr>
<tr>
<td>Mean</td>
<td>58.8</td>
<td>1303.2</td>
<td></td>
</tr>
</tbody>
</table>

* Initiation time was estimated at the moment when the first PD was generated with a magnitude above 1 pC. ‘0’ means the tree was initiated immediately after applying the high voltage.

** Breakdown time = initiation time + time for current to runaway.

*** No PD was detected in the next 36 hours after initiation. This sample was not included in the mean value calculation.
Chapter 6: The impact of inorganic fillers on electrical tree growth

The electrical trees in most untreated samples were initiated very quickly so the increasing loadings in this batch only contributed to the enhancement of the breakdown time. Sample 5UT6 had the longest initiation time (78 minutes) but after the initiation, no further PDs were detected in the next 36 hours. There were two samples 1UT7 and 3UT7 imaged by Synchtron XCT and the imaging result is shown in the next section.

For samples with treated particles, there were two samples (1ST1 and 5ST6) that did not breakdown after 36 hours even though PD activity was detected. It was found that all samples with 1 wt% fillers had very short tree initiation time which was less than 7 minutes while as the load went higher to 3 wt% and 5 wt%, the initiation time was enhanced up to 204 minutes.

![Figure 6-27: Treeing breakdown time of nanosilica filled epoxy samples under 13 kVrms. For each set of results the box shows the range between the upper quartile and the lower quartile, the small square and the middle line are the mean and median value respectively.](image)

![Figure 6-28: Comparison of treeing breakdown time and initiation time of nanosilica filled epoxy samples (surface treated) under 13 kVrms.](image)
Chapter 6: The impact of inorganic fillers on electrical tree growth

The plot in Figure 6-27 and Figure 6-28 reveals how the treeing breakdown time and initiation time varied with the addition of nano-size particles. It was found that no matter whether the particle surface was treated or not, epoxy filled with nanosilica can enhance the treeing strength in comparison to the unfilled one. By comparing with the unfilled group shown in Section 6.2.1, the mean breakdown time was enhanced 103.3% and 249.1% in 1 wt% and 3 wt% untreated samples respectively. While at 5 wt%, the breakdown time showed a very similar level to the 3 wt% and there was only 30 minutes (17.6%) difference in terms of the mean breakdown time. Samples with treated nanosilica showed greater improvement from 1 wt% to 5 wt%. Although the breakdown time at 1 wt% ST was still similar to that in 1wt% UT, the mean breakdown time in 3 wt% ST prolonged dramatically and it was more than 4.5 times the 3 wt% UT value. The breakdown time of surface-treated samples continuously increased at 5 wt% and it had a further 68.4% improvement in comparison to 3 wt% ST.

It was found that the tree initiation was improved by introducing surface treated particles especially at 3 wt% and 5 wt% concentrations. Figure 6-28 presents a comparison between the treeing time after initiation (breakdown time – initiation time) and initiation time of each sample in ST batches. It was found that the longer initiation time can be also associated with a longer time to failure after initiation. This relationship between the initiation time and the breakdown time is consistent with the result revealed by Niedernhuber et al [165] in the nano-filled (12 nm) epoxy system at 20 kV but in his experiment the 1 wt% nanocomposite showed the best performance and the 5 wt% group was even worse than the unfilled sample, which is reversed to the result shown in this case.

6.4.3 XCT tree imaging

The tree structure in 1UT7 and 3UT7 was imaged with the same X-ray source in the Diamond Light Source with the exactly same settings as the microfilled samples. Figure 6-29 shows one projection of the treeing area in 3UT7. There were no obvious particles observed under the 0.33 um pixel size so unfortunately any interaction between tree channels and filled nanoparticles was not apparent.

As reported in the micro-filled materials (shown in Section 6.3.3), imaging noise (white areas) were also observed around the tree channel (black) while, in this case, some white spots were found in the centre area of the tree channel. That could happen in two
situations: (1) there were nano particles aggregated in the tree channel; (2) a fringe is caused by phase contrast imaging. If the first situation was confirmed, those particles clusters attached on the tree wall could be much denser than others outside so it could be considered that there were huge number of nano particles accumulated within the micro-size tree channel. However if the second situation was true, according to the mechanism of phase contrast and also referring to Figure 6-15 in Section 6.3.3, these white fringes would be generated between two heterogeneous materials, which means there are truly some other objects inside the tree channel and with very high mass density. Both of these two assumptions revealed the loose nano particles might attach on the tree wall or just drop down into the channel void. This is consistent with the finding from SEM and EDX shown in Figure 6-25.

Figure 6-29: (a) Intensity along the length of 34 pixels across the tree channel in a 3 wt% nano-filled sample 3UT7; (b) profile of pixel intensity along the length of the red line. Pixel size: 0.33 μm.

Figure 6-30 shows the reconstructed tree structure in 1UT7 and 3UT7. Only two samples with untreated fillers were imaged by synchrotron XCT during the project due to limited beamline time. During the imaging it was found that the tree structure was actually much longer than expected and far beyond the field of view for the synchrotron imaging system so as a result these reconstructed data only presented partial structures. These two revealed tree structures looked consistent with the concept of the ‘branch’ tree. As shown in Table 6-9, some quantified parameters were summarized. It can be found that in the same epoxy volume (2.4×10^8 μm^3) the deteriorated region in the 1 wt% sample is about 2.5 times bigger than that in 3UT7. Both of these tree structures can be considered as a ‘branch-type’ tree according to their fractal dimensions which are less than 2. The mean
diameters of the electrical trees in these two nanocomposites are found to be 2.76 µm and 2.01 µm respectively and they were a little bit less than the tree diameter measured from 3D models of most unfilled and micro-filled samples (as shown in Section 5.3 and Section 6.3.3). It was also found that the treeing region around the electrode in Figure 6-30(a) was much denser than that in Figure 6-30(b) and this perhaps means that the material around the needle in 1UT7 was damaged more severely than 3UT7.

Table 6-9: Quantified tree parameters in the 1 wt% and 3 wt% nanocomposite 1UT7 and 3UT7.

<table>
<thead>
<tr>
<th></th>
<th>1 wt%</th>
<th>3 wt%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Volume (µm$^3$)</td>
<td>15489.54</td>
<td>6256.92</td>
</tr>
<tr>
<td>Surface area (µm$^2$)</td>
<td>38501.34</td>
<td>20917.51</td>
</tr>
<tr>
<td>3D fractal dimension</td>
<td>1.73</td>
<td>1.57</td>
</tr>
<tr>
<td>Mean diameter (µm)</td>
<td>2.76</td>
<td>2.01</td>
</tr>
</tbody>
</table>

Figure 6-30: 3D rendering of electrical trees in (a) 1 wt% nano-filled sample 1UT7 energized at 13 kV$_{rms}$ for 1.8 h and (b) 3 wt% nano-filled sample 3UT7 energized at 13 kV$_{rms}$ for 2.2 h.

6.4.4 PD measurement

Table 6-10 shows the quantified PD activities during the treeing breakdown test in both untreated and treated nanocomposites. By comparing with unfilled samples, all nano-filled samples contained much more PDs during the treeing phenomenon but the 5 wt% treated group showed the even lower average PD rate than the unfilled one. Higher total
PD energy and average PD power were found in all untreated samples while with the silane treatment, these two parameters reduced. Both 3 wt% and 5 wt% treated samples had even lower PD power than the unfilled samples.

<table>
<thead>
<tr>
<th></th>
<th>Unfilled</th>
<th>1 wt%</th>
<th>3 wt%</th>
<th>5 wt%</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>untreated</td>
<td>treated</td>
<td>untreated</td>
</tr>
<tr>
<td>Number of PDs</td>
<td>1.21×10^5</td>
<td>4.66×10^7</td>
<td>2.21×10^7</td>
<td>5.11×10^7</td>
</tr>
<tr>
<td>Average PD rate</td>
<td>54</td>
<td>6561</td>
<td>347</td>
<td>6132</td>
</tr>
<tr>
<td>(s)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Total PD energy</td>
<td>0.021</td>
<td>4.2</td>
<td>0.31</td>
<td>5.1</td>
</tr>
<tr>
<td>(J)*</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Average PD power</td>
<td>8.04×10^-6</td>
<td>8.11×10^4</td>
<td>5.91×10^-5</td>
<td>8.21×10^-4</td>
</tr>
<tr>
<td>(W)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

*Total PD energy was calculated by Equation 11 introduced in section 5.3.

It seems that the PD performance in 1 wt% samples (both treated and untreated) is contrasting with 3 wt% and 5 wt% samples, so the case study below only selects four comparable PD patterns (1UT5, 1ST5, 5UT3 and 5ST4) during the tree growth in 1 wt% and 3 wt% batch. These aim to illustrate how the addition of nano fillers and the surface treatment affects PD activities.

Firstly for the sample 1UT5 as shown in Figure 6-31, the partial discharge was ignited immediately on voltage application and the corresponding PD magnitude continuously increased in the form of bursts of activity. At about 1500 s, the maximum magnitude dropped down from 200 pC to 40 pC and then rose up again until breakdown at 600 pC. At the later stage, bursts of PD activity were observed frequently and both the PD magnitude and number were high. Most of time the occurrence of PDs was frequent and steady so no PD extinction period was identified in the whole treeing time.
As for the 1 wt% treated sample 1ST5, the overall PD magnitude was much lower than that in the untreated one as well as the PD number except for a few PD with extremely high number up to 1000 per cycle. After the PD was initiated and underwent a steady growth from 0 pC to 30 pC for 250 s, some sporadic PDs appeared. These sporadic signals contained steady magnitudes from 30 pC to 40 pC but at 2400 s the magnitude
dropped to ~ 5 pC and remained within this level until 4000 seconds. Then after a 50-minute period of PD activities from 10 pC to 125 pC, the dielectric finally got breakdown.

When the loading of untreated nanosilica was increased to 5 wt% as shown in Figure 6-33, the overall PD magnitude and PD number of 5UT3 were lower than those of 1UT5. The maximum PD magnitude 450 pC happened at about 5500 s where the maximum PD number also happened nearby at 600 per cycle. Most PD magnitudes in later treeing case were about 400 pC which was a little bit lower than that in 1 wt% UT. However, there were more troughs appeared with relatively higher PD magnitude from 2000 s and then continuously grew up to more than 100 pC until the breakdown.

As shown in Figure 6-34, very different PD activity was found in the treated sample 5ST4. Firstly, differing from the other samples, as shown in the window of Figure 6-34, the electrical tree was initiated at least 50 minutes after energizing and the initiation PD started with the typical sporadic signals (described in Section 6.2.2) with the magnitude of 1-2 pC. Most of extinction periods were found to be at least 10 times longer than seen in unfilled samples and the longest extinction period was almost 2 hours. The overall maximum magnitude still showed an increasing trend to 550 pC while only in the last hour before failure there was an increase in frequency of typical mid-sized discharges (from 80 pC to 110 pC) between the larger events.
Due to its translucent nature, electrical trees were not clearly observable in nanocomposites. The synchrotron XCT applied in this case revealed part of tree structures around the needle in the 1 wt% and 3 wt% epoxy sample 1UT7 and 3UT7. Both fractal index and quantified structural features suggested less degradation regions were generated in the 3UT7. It was also found that the tree diameter in these two nanocomposites was relatively smaller than in unfilled and micro-filled epoxy systems shown in Section 5.3 and Section 6.3.3. These XCT tree images in nanocomposites need further check about their repeatability but the imaging result here is mainly considered as the evidence to prove that nano-size filler particles (20 nm) do not act similarly as microsilica (1 - 10 µm) shown in Section 6.3.3.

It was found that the addition of nano-size fillers could enhance the lifetime of the epoxy resin system by at least 2.06 times in 1 wt% untreated system and up to 27.1 times in 5 wt% treated samples. The breakdown time of 5 wt% UT showed a similar performance with the 3 wt% sample and this may be associated with the similar ‘crossover’ phenomenon found in epoxy/alumina composite [18]. The main reason for this may be the poor particle dispersion as shown in Section 6.4.1 when the load level was too high. The introduction of these inorganic particles in the epoxy matrix could disturb the
material quality, and enhance the electrical field in the insulation system due the high permittivity of the silicon dioxide. This may do harm to the dielectric property so it can be reasonable to expect both increase and decrease of dielectric strength in nano-filled polymers as stated in literature depending on material preparation quality [166, 167].

The silane treatment significantly improved particle dispersion and PD suppression especially according to the 23.9%, 22.4% and 74.1% decrease of the neighbouring distance as shown in Table 6-6. It has been well documented that the silane coating can effectively reduce the surface energy on the particle and improves the adhesive capability with epoxy resin. Additionally, the surface treatment decreases the appearance of weaker interfaces the dielectric and the stronger chemical bonds could also contribute to the PD resistance [168]. The experimental result here was consistent with that conclusion by comparing the PD number and energy between untreated and treated samples. It was also found that the PD phenomenon in treated samples could be clearly suppressed compared to the unfilled epoxy system especially at 5 wt% loading.

The surface treatment was found helpful to delay the initiation time of the electrical tree in 3 wt% and 5 wt% samples while this was not so obvious in 1 wt%. The suppression of nano particles on tree initiation was also identified in the literature [151, 169] but according to the experimental result in other reports, both untreated and treated helped while in this case only nanosilica particles with silane treatment showed the consistent conclusion. Discussion will be continued in the next chapter.
Chapter 7: Discussion

7 Discussion

7.1 Treeing and breakdown in unfilled epoxy resin

Two distinct types of tree are observed to grow in epoxy resin samples and these are referred to as ‘fine tree’ and ‘dark tree’. The reason why only few publications [24, 25] have ever identified them may be: Firstly a ‘fine tree’ is thin and slight so in most of semi-crystalline materials such as XLPE and silicon rubber, the growth of a ‘fine tree’ may be not observable due to poorer transparency. Even in this thesis, the imaging shown in Section 5.2.1 only provides blurred images of it, which limits the further study on the characteristics of ‘fine tree’ structure. On the other hand, some experimental results also suggest the formation of the fine channel may take place at high electric field while at low electrical field only broad or degraded tree channels can be found [18], which also explains the reason why the ‘fine tree’ structure is rarely observed.

From the experimental results shown in Section 6.2.1, the most straight-forward difference between ‘fine tree’ and ‘dark tree’ is the diameter of the tree channels: generally < 1 µm for a ‘fine tree’ and > 1µm for a ‘dark tree’. The mechanism of the appearance of a ‘fine tree’ is still unknown but due to its independence from the final breakdown and the possible PD-free features as shown in Section 6.2.2 and other publications [24, 25], the growth of ‘fine tree’ may be ascribed to a kind of propagation of mechanical cracks. The possible mechanism of this electro-mechanical process has been proposed as the result from electrostatic energy [170, 171] or the pressure change caused by newly generated gases within the tree channel [33]. It was also proposed that the energy needed for a tubular or Filamentary channel growth (radius of the order 0.1 to 1 µm [171]) is not from the gas discharge but the electrostatic energy given by the Maxwell stress tensor [170].

The ‘fine tree’ growth does not directly result in electrical breakdown but precedes the growth of a ‘dark tree’, which can eventually lead to the electrical breakdown in dielectric systems by bridging two electrodes. As a result, the growth of a ‘fine tree’ can be regarded as the pre-treeing phenomenon and needs to be also considered to improve.
the lifetime of dielectric systems. An important mechanism of the tree propagation is the transition from ‘fine tree’ to ‘dark tree’. From experiment results and PD analysis, during this ‘fine-to-dark’ transition at the early treeing stage, PD activities can be identified by sporadic signals (illustrated in Figure 6-9) where a bursty PD magnitude periodically occurred in individual voltage cycles (small PD number per cycle). Tanaka et al [162] found that when the void size was less than the mean free path of electrons in gases (1000 Å = 100 nm), no partial discharge (the detection sensitivity was 0.05 pC) occurred. It can be found that the ‘fine tree’ channel measured in Section 6.2.1 is also with the size of sub-micron. Hence it can be proposed that these ‘fine tree’ channels contain insufficient space to accommodate PDs so that no obvious PD activities were detected in the experiment with the detecting sensitivity of 1 pC. However the injection and extraction of electrons around the needle tip will continuously damage the polymer nearby by the scission of chains or the formation of cavities by Maxwell stress [172]. Eventually the ‘fine tree’ channel is widened so PDs start to ignite and propagate in the channel and also erode the inner surface to generate the observed ‘dark tree’ structure. So generally the ‘fine-to-dark’ transition can be regarded as a widening process of a hollow channel from the sub-micron to micron.

As suggested in Section 6.2.1, the whole treeing phenomenon can be divided into five stages according to the different propagation features measured by optical images and PD measurement. With the consideration of the ‘fine tree’ and ‘dark tree’, a developed treeing model can be also characterized by different treeing stages:

- **Tree initiation stage**: the tree initiation stage is generally regarded as being the period before a tree is visible. Electron injection and extraction take place repeatedly between the electrode and the surrounding material [53]. The initiation of a ‘fine tree’ is also thought to be preceding to a ‘dark tree’ initiation which usually includes an incubation period [18]. However there is no evidence to support this point and the optical images shown in Section 6.2.1 is too blurry to identify them clearly.

- **Early tree growth stage**: this is the stage where the initial ‘dark tree’ appears. As illustrated in Figure 7-1(a) and Figure 7-1(b), it is proposed that after the ‘fine tree’ is created in the dielectric, the region around the needle tip is firstly widened by the repeated process of electron injection and extraction, which eventually results in the void formation or chain scission around the needle tip [49, 58]. The
size of the evaporated or deteriorated region around the needle tip should be at least 1 µm [58] so as to support further discharges. Figure 7-1(c) shows the image in which the ‘fine tree’ channel is enlarged, which leads to the growth of the visible ‘dark tree’. This ‘fine-to-dark’ transition in this stage can be very fast and continuous according to the experimental results shown in Section 6.2.2. The linear correlation between the maximum PD magnitude and the tree length shown in Section 6.2.1 is consistent with the conclusion suggested from the multi-stage treeing and imaging experiment in Section 6.3: only PDs with higher magnitudes can reach the tree tip and so as to increase the tree length. On the other hand, the ‘wing-like’ PD pattern revealed in Section 6.2.2 suggests that at least initially the ‘dark tree’ is non-conducting [3, 173]. Experimental results from other publications [24, 25] also found that the growth of the initial ‘dark tree’ can suddenly stop and the corresponding PD activity is almost extinct at the end of the stage. However the reason this phenomenon is still unknown. A possible explanation can be that conductivity of the initial ‘dark tree’ is increased quickly, which eliminate PDs near the needle tip [17]. But in the same time, the local electric field at the end of the tree branch is still too low to form new PDs at the tree tip, which lead to the extinction of PDs along the whole channel.

![Diagram of the treeing process in the early growth stage](image)

**Figure 7-1:** Illustration of the treeing process in the early growth stage: (a) a ‘fine tree’ is initiated from the needle tip; (b) The region near the needle tip enlarged by repeated charge injection and extraction and the initial ‘dark tree’ appears; (c) The ‘fine tree’ channel was enlarged by the damage of partial discharges.

- **Tree extension and pre-breakdown stage:** these two stages contain all the periods after the early tree growth stage until the breakdown. At the beginning of this period, only the ‘fine tree’ can continue growing while the initial ‘dark tree’...
Chapter 7: Discussion

stops according to imaging results shown in Section 6.2.1. Small branches appear from the needle tip with the form of the ‘dark tree’ again. As suggested before, these newly developed ‘dark tree’ channels can be regarded as non-conducting tree. At the beginning of this period, the newly developed ‘dark tree’ channel may be still too short so that PDs cannot propagate far from the needle tip quickly enough so the formed space charges would reduce the electric field around the needle tip [171, 173]. As a result, the transition from the ‘fine tree’ to ‘dark tree’ becomes sporadic and the number of PD charge per cycle also becomes low at the beginning of the tree extension stage as shown in Section 6.2.2. But when the tree length is long enough to allow most injected charges to migrate far away from the needle tip, the space charge effect can be greatly decreased increasing the field around the needle tip and so the PD number and magnitude are increased later. Some low-energy PDs which cannot propagate to the end of the channel still occur and may gradually erode the tree wall, widening them further and generating energy enabling chemical degradation processes in the channel walls [1, 3]. These PDs with relatively small magnitude therefore not only result from the formation of new short branches but also suggest the transition from non-conducting tree to conducting one [17]. The evolution of conductivity can result from carbonized deposits [7] and the conducting tree also acts to reduce the electric field around the needle tip but concentrate the local field around the channel tip so once tree develops, most PDs within the tree channel fall and they gradually become mainly ignited from the conducting tree tip, which has been previously suggested by the luminescence pictures shown in Figure 2-7 [173]. Figure 7-2 shows the three situations of the ‘dark tree’ growth with the consideration of the channel conductivity. As described in the early-tree growth stage, a ‘dark tree’ channel derived from a ‘fine tree’ is initially non-conducting. When the channel length is long enough, i.e. L in Figure 7-2(a), a non-conducting tree will not reduce the field at the needle tip, in which case charge injection into the polymer will continue and in particular the needle tip will discharge into the tree structure. According to the result shown in Figure 6-5, L should be much bigger than the initial ‘dark tree’ channel, which is about 250 µm in the experiment result of S2. At some point, when PDs can hardly propagate to the tree tips, the tree length stops growing [173] and a crucial factor can be the applied voltage. However, PDs can be still ignited from the needle tip damaging
the tree walls and form carbonaceous deposits [1, 17], which finally increase the conductivity. As shown in Figure 7-2(b), partial discharges in a conducting tree channel mainly happen at the tree tip instead of the needle tip (the distance is \( \Delta L \)) so the region of high field driving the ionisation process is pushed forward. At this moment the distance between the conducting tree tip and the old tree tip is decreased (\( L - \Delta L \)) so the PD magnitude can be also temporarily suppressed [24, 66], which may explain the cause of many small PDs in the later treeing stage. When the conductivity near the needle tip is high enough, a conducting tree channel can be regarded as an extension of the needle tip and new channels may continue to grow further with until the same length \( \Delta L \) as shown in Figure 7-2(c).

- **Breakdown stage:** the growth of a ‘fine tree’ does not bridge electrodes and result in the failure but a ‘dark tree’ does. In this case ‘fine tree’ may be always considered as non-conducting because there is no reason to assume at any stage that it has lost its dielectric properties as there is no chemical degradation from a cracking process and it does not immediately lead to the electrical breakdown [24]. The transition of the tree conductivity effectively increases the propagation range of the ‘dark tree’ so until it eventually reaches the earth electrode, flashover happens.

As shown above, the whole treeing progress can be identified into four stages when both the ‘fine tree’ and the ‘dark tree’ are considered. On the other hand, according to the different PD features, three different tree states can be supposed if the conductivity of the ‘dark tree’ is considered:
• **Insulating tree**: most of discharges are emitted from the needle tip and their propagation path is all along the tree channel. This state mainly occurs at the early treeing stage, i.e. early tree growth stage.

• **Weakly-conducting tree**: as the carbonization caused by PD energy affects the tree channel, the conductivity of insulating tree gradually increases. It acts to reduce the field stress around the needle tip but in the same time it is still not conducting enough to transfer higher potential to the tree tip to generate further discharges. As a result, no PDs along the whole tree channel. This state mainly occurs at the intermediate treeing stage, i.e. at the beginning of the extension stage.

• **High-conducting tree**: once the conductivity increases to a certain threshold, high electric field is transferred from the needle tip to tree tips which act as new electrodes to re-ignite PDs and promote further tree growth. This state mainly occurs at later treeing stage, i.e. from the middle of the extension stage to the end.

In summary, a more general identification between ‘fine tree’ and ‘dark tree’ is summarized in Table 7-1.

### Table 7-1: Classification of ‘fine tree’ and ‘dark tree’.

<table>
<thead>
<tr>
<th></th>
<th>Diameter</th>
<th>Conductivity</th>
<th>Characterization</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Fine tree</strong></td>
<td>&lt; 1 µm</td>
<td>Non-conducting</td>
<td>• May be an electro-fracture mechanics;</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>• Does not lead to breakdown;</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>• Guides the path of a subsequent ‘dark tree’;</td>
</tr>
<tr>
<td><strong>Dark tree</strong></td>
<td>&gt; 1 µm</td>
<td>• insulating in early stage;</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>• Partially Conducting from intermediate stage;</td>
<td>• Caused by PD;</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>• Leads to breakdown;</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>• Follows the path of ‘fine tree’;</td>
</tr>
</tbody>
</table>

### 7.2 Treeing and breakdown in micro-filled epoxy resin

From the comparison of breakdown tests between unfilled and micro-filled epoxy (shown in Section 6.3.2 and Section 6.2.1), the addition of microsilica has dramatically
enhanced on the treeing breakdown time by at least 10 times at the loading level from 5 wt% to 30 wt%. The linearly increased breakdown time with the square root of the filler level suggests the reducing effectiveness of continuously adding filler concentrations. As shown on SEM images in Section 6.3.1, the increased possibility of agglomeration may be an important factor influencing the dielectric property of the filled composite if the different interactions between the tree propagation and the filler particle are considered.

The synchrotron XCT showed excellent capability to reveal the tree structure even in the opaque sample, which provided a new chance to further study the tree growth in filled dielectrics. Based on the 3D reconstruction revealed in Section 6.3.3, interactions between tree channels and filled micro-size particles can be grouped in two main types as shown in Figure 7-3:

- **Bending:** the tree channel tends to grow around the micro-sized particle. It is the most common theory found in other treeing models in microcomposites and consistent with the optical images of thin samples [122, 174]. Due to the high mechanical strength and degradation resistance, the micro-size silica filler changes the original propagation path of the electrical tree.

- **Penetrating:** when there is a large agglomeration (> 10 µm) in front of the tree structure, the tree channel around the cluster can directly penetrate through the big cluster instead of the tortuous growth. This has not previously been reported because of the difficulty of the imaging and the no-agglomeration condition people usually assume.

![Figure 7-3: Illustration of the tree propagation affected by microsilica fillers (assuming each single filler has the same size as the tree channel) in the form of (a) uniformly dispersed; (b) huge agglomerations.](image-url)
It has been reported by Kurnianto et al [27, 122] that the higher concentration of fillers can lead to more obstruction so as to create more branches and torturous channels, which results in higher fractal dimension but lower growth rate. However the quantified results shown in Section 6.3.3 seems to be contradictory because the 15 wt% sample has the lower fractal dimension (1.65) but higher growth rate (1.98 µm/min) than the 5 wt% sample (1.71, 1.70 µm/min). To explain this, the first noteworthy thing is the particle size. In Kurnianto’s experiments, most of filler particles are with a diameter of 50 µm that is more than 5 times than the silica particle used in this thesis (1 - 10 µm) and also about 10 times larger than the diameter of electrical tree channels (~ 3 µm). The larger particle size increases the possibility of bending tree channels, which can effectively lead to the more torturous tree shape. However in this thesis, the bending interaction is not the only predominant factor but the penetrating interaction should be also considered. In contrast to the bending interaction, the tree propagation tends to be promoted by the penetrating interaction. The formation of these agglomerated clusters may be due to the presence of moisture which acts as a binder on the particle surface and generate inter-particle bridges [175]. The possible paths for tree propagation might be some weakly-connected interfaces or de-bonded regions between the filler and resin caused by Maxwell-Wagner interfacial effect [176-178]. Within agglomerations these interfaces with poor adhesion may be even overlapped, creating more weak regions for the tree growth. In practice, both of the bending and penetrating interaction exist in the microcomposite and offset the effect caused by each other. This may explain the reason why these quantified parameters (fractal dimension, growth rate and tortuosity) shown in Table 6-4 are not significantly changed as the increased concentration and even show contradictory result comparing with the literature [27]. On the other hand, it should be noted that the tree structure imaged in Section 6.3.3 only presents the early treeing stage (less than 300 µm) that may not suggest the features of the whole treeing path.

The main reason why the direction of treeing propagation is changed is most frequently explained by the distorted local electrical field caused by the relatively higher permittivity of the silica and the tree propagation was considered to be guided along the silica/epoxy interface [30, 122]. However, according to the imaging result revealed in Section 6.3.3, the tree channel seems to be distant from the particle. Because the matrix material is unaltered, filled composites very likely contain ‘fine-trees’ as described in Section 7.1. So the role of the micro-size fillers may be reconsidered in two key
processes: the growth of ‘fine tree’ and the transition from ‘fine tree’ to ‘dark tree’. As suggested before, the growth of the ‘fine tree’ may be not PD-driven and one of the possible mechanisms can be the fracture mechanics [24, 170]. It has been well documented that with the modification by filled particles, an increased fracture toughness is mainly associated with the crack deflection mechanism [179]: the extension of the crack on the neat epoxy is directional and straight so the roughness of the fracture surface is low; more twisting cracks appear on the material with the modification of fillers and the surface roughness is increased. The cause of deflection may be explained by a tangential compressive strain generated near the epoxy/filler interface, which can divert the crack front when approaching the particle [180]. It can be speculated that the propagation of a ‘fine tree’ may be also follow the crack deflection mechanism as shown in Figure 7-4.

![Figure 7-4: Schematic of the ‘fine tree’ growth in microfilled material following the crack deflection mechanism.](image)

The ‘fine-to-dark’ transition in micro-filled material can be similar with the progress introduced in Section 7.1 because the typical sporadic PD is also found as reported in Section 6.3.4, and displays a similar PRPD pattern. The particle dispersion model shown in the Section 6.3.1 suggests the particle-to-particle distance in low loadings (5 wt%) can be larger than the diameter of tree channels so even through the ‘fine tree’ channels are widened by PD degradation, sometimes they are normally still distant from the nearby micro-silica and that main explain why the tree channel a not along the particle surface. A higher concentration of fillers means a higher like-hood and frequency that the tree channel meets with single particles so more active interactions can occur between the tree and the filler. The weak interface between the epoxy matrix and the filler may act as the extra free volume for discharges. As shown in Figure 7-5, when the tree channel merges
with the filler/epoxy interface, the channel diameter can be suddenly increased and that leads to an immediately burst in the PD magnitude and number, which is frequently found in the 15 wt% and 30 wt% samples.

The isolated elements of tree structure observed in Figure 6-19(e) cannot be surely defined yet due to the insufficient resolution of the micro-XCT technique, but if the ‘fine tree’ model is considered in the microfilled system, it may be more likely a part of ‘dark tree’ structure growing near the particle. The immediately increased tree diameter results in the faster PD damage towards the nearby epoxy matrix so the widening speed can be faster than normal ‘fine tree’ and makes this part observable in advance.

7.3 Treeing and breakdown in nano-filled epoxy resin

The proposed mechanism for the tree propagation in nanocomposites was considered as similar as that in microcomposites according the literature [141, 142]. However, there is an essential difference between nanocomposites and microcomposites: the diameter of the filler in microcomposites is similar or larger than the tree channel while the nano particle is at least one order of magnitude smaller. So the nano-size particle cannot in itself act as a physical barrier to impact the tree growth nor can it essentially influence the field at the tree tip. Furthermore, SEM images in Section 6.4.1 and other publications [18, 181] suggest that the nano particle can be enveloped by the tree channel, which is impossible in microcomposites.
Similarly, the tree propagation in nanocomposite may be re-explained with the ‘fine tree’ model. The different filler particle dimension could significantly influence the transition progress from a ‘fine tree’ to a ‘dark tree’. When the PD degradation starts, the extending ‘dark tree’ appears by eroding the epoxy devoid of fillers. First of all, the nano-structuration can effectively resist PD damage on the epoxy surface and this mechanism has been widely revealed by other publications [26, 182]. Based on Tanaka’s multi-core model [28], it is believed that the damage of partial discharges propagates through weak regions (inter-filler matrices or third layer) an forms zigzag paths so resulting in strong PD resistance. However, the circumstance can be a little bit different if the ‘fine tree’ growth is considered because the particle-to-particle distance measured in Section 6.4.1 is tens or hundreds of nanometres which can be even smaller than the visible ‘fine tree’ channel. In nano-filled materials then, the interaction is continuous as the tree growth and the material can be considered virtually homogenously modified, in contrast to the micro-filled samples which are inhomogeneous for the tree growth. Figure 7-6 shows what may happen during the ‘fine-to-dark’ transition. It is proposed that when the epoxy is split and the ‘fine tree’ appears, these nano fillers at the original location will be still adhesive on the tree wall. When the ‘fine-to-dark’ transition starts, discharges evaporate the epoxy matrix and some nanosilica particles are de-bonded. These nanosilica may be either retained on the tree wall due to the very rough inner surface observed in nanocomposites [18] or just drop into the tree channel because of gravity. Hence a great number of nanosilica can be grouped on the tree wall, for example, the EDX image in Section 6.4.1 suggest that the area fraction of silica in the treeing area (17.2%) is 2.8 times higher than outside (6.2%), which is also consistent with the SEM image shown in Figure 2-45. But it should be noted that the quantification also contained particles at different depth so in practice the real distribution may be not so dense. Just like a protective skin on the inner surface of the tree wall, nanosilica with higher permittivity and PD resistance prevent the tree channel to be further widened, which can inhibit both the ‘fine-to-dark’ transition and carbonization. This may explain why the mean diameter of the tree channel in nanofilled materials revealed by XCT in Table 6-9 was smaller than unfilled and micro-filled samples even though there were more PDs with higher magnitude, and energy detected in some nanocomposites. Another assumption is that after the epoxy near the tree tip is vaporized, some de-bonded nanosilica particles can accumulate at the tree tip. These nano fillers worked as a silica barrier with relatively high permittivity causing an increased tangential component of the electric field along
the barrier interface [183], which also prevents the tree channel to propagate further. This phenomenon may only occur in the later treeing stage where enough nano particles are accumulated as a barrier in front. Some reports also proposed the similar ideas but there is still no sufficient evidence for those characteristics.

![Figure 7-6: Illustration of the ‘fine-to-dark’ transition in nanocomposites. (a) A ‘fine tree’ is just initiated from the needle tip; (b) The ‘fine tree’ channel is widen by partial discharges and these de-bonded nano particles are attaching on the inner tree wall and the tree tip to prevent further PD erosion.](image)

The quantified results of particle dispersion in Section 6.4.1 show the great capability of nano particle with the silane treatment. It is reasonable because the silane coating can effectively reduce the surface energy on the particle and improves the adhesive capability with epoxy resin, which significantly reduces the possibility of agglomeration formation during the storage and compounding process. Additionally, the surface treatment decreases the appearance of weaker interfaces and the stronger chemical bonds can be regarded as the improved first layer according to the multi-core model and so contributes to the improved PD resistance [28, 184]. The much lower PD power and PD number of 5 wt% treated samples shown in Table 6-11 proved the improved PD resistance at a relatively higher loading. On the other hand, the bond strength between fillers the epoxy matrixes was found to be increased by silane coating [185], which may also act against the growth of ‘fine tree’ channels.

The different tree initiation time found in treated samples as shown in Section 6.4.2 may be explained by the increased inception field caused by suppressed charge injection effect [169, 186] in nanocomposites. In nanocomposites these embedded fillers could be considered as traps and more electrons tend to accumulate around the needle tip during
the tree initiation stage. Trapped electrons with relatively high density decrease the local field around the electrode and make electron injection more difficult [151]. The reason why most samples in untreated batches do not reflect the suppression on tree initiation may be explained by the incompatibility between the nanosilica and the epoxy matrix, which will generate weakly bonded interfaces and may change the charging tendency. When the tree is about to initiate, these layers may act to help the formation of voids around the needle tip [58, 162] so as to skip the stage of charge injection or extraction and then generated enough PD energy for the further tree growth. That may be the reason why the XCT image shown in Section 6.4.3 has severe degradation found near the needle tip.
8 Conclusions and further work

8.1 Conclusions

The contribution of this thesis includes the development of X-ray imaging techniques for the study of electrical treeing phenomenon and the comparison of the treeing mechanism in different dielectric systems. By introducing the 3D imaging tool, several electrical trees were represented in three dimensions and fully characterised by quantitative parameters, which helps to clearly describe different tree structures.

The intrinsic characteristics of the tree growth in unfilled epoxy system were further investigated by the multi-stage treeing and imaging tests where the tree growth in three stages was imaged separately by using laboratory Micro-XCT. Resulting 3D models have helped further understanding about tree degradation in unfilled system by quantitatively associating with PD energy.

Electrical tree imaging in filled polymer dielectric systems was challenging according to previous attempts, but in this project the full tree structure was successfully reconstructed by applying ‘pink-beam’ Synchrotron XCT imaging. Moreover, for the first time this has enabled the physical interaction between tree channel and filler to be revealed. The interaction between tree channels and embedded microsilica was the key evidence to study the role of fillers influencing the tree propagation.

Treeing tests in different dielectric systems have shown characteristics of the tree growth by combing imaging and PD results. Two distinct types of tree: ‘fine tree’ and ‘dark tree’ have been clearly identified by structural information and PD features. The ‘fine-to-dark’ growth model was proposed to describe the relationship between these two tree types and also revealed the treeing and breakdown mechanism. The role of inorganic fillers was also discussed by introducing a growth model of ‘fine tree’ and ‘dark tree’.

The conclusions from this research are:

- Laboratory XCT is capable of imaging electrical tree structures in most unfilled polymeric materials. But in consideration of multiple treeing and imaging tests,
the influence of X-ray irradiation should be carefully evaluated so as to make sure the dielectric is not additionally degraded. Experimental results of the tensile test and FTIR suggest that the X-ray irradiation may only oxide on the sample surface but not in internal regions so at least the three-stage imaging is acceptable. XCT imaging in filled materials is achievable by applying a synchrotron source with ‘pink beam’ system. Tree structures in microsilica filled samples (5 wt%, 15 wt% and 30 wt%) can be fully represented and interactions between the electrical tree and filler particles can be also revealed for the first time.

- The multi-stage treeing and imaging test is able to investigate the treeing features in a small-step of tree growth and by linking with the released PD energy, it was found that not all the PDs will result in the increment of the tree length. By quantitatively comparing the released PD energy and the theoretical vaporization energy of the newly developed tree structure, it is found that between every two stages, only those PDs with high magnitudes (contains 15.7% and 0.15% of total PD energy respectively) can reach tree tips and so contribute to the tree growth in length. PDs with lower magnitudes may increase the tree channel width.

- By optically observing the treeing progress in an unfilled epoxy sample, the growth of ‘fine tree’ and ‘dark tree’ are identified as different but closely associated. Partial discharge monitoring is not able to detect the growth of a ‘fine tree’ below 1 pC but the transition from ‘fine tree’ to ‘dark tree’ can be identified by the occurrence of sporadic PD. Simultaneous imaging and PD measurement suggest five typical stages of tree growth which are: initiation, early growth, extension, pre-breakdown and breakdown. On the other hand, three different states are also proposed in terms of the conductivity of ‘dark trees’: insulating tree, low-conducting tree and high-conducting tree.

- Insulation lifetime of the dielectric system can be prolonged by adding inorganic fillers. Experimental results suggest the addition of microsilica has increased the treeing breakdown time from 7.5 times to 18.2 times as the concentration adding from 5 wt% to 30 wt%. The mean treeing breakdown was found to be linearly increased with the square root of the microsilica concentration. Nanosilica fillers with and without silane treatment showed similar effect at 1 wt% where both batches performed 2.03 and 2.08 times longer breakdown time comparing with unfilled samples. While at 5 wt%, treated fillers provided 9.2 times longer
lifetime than untreated. Silane treatment also helped to increase the initiation time at 3 wt% and 5 wt% loadings by 2.25 and 15.7 times respectively comparing with unfilled samples, which may be explained by its better compatibility with the epoxy matrix.

- The propagation direction of an electrical tree branch can be essentially changed by the introduction of individual micro-size fillers that lead to the change the tree propagation but when there is a huge agglomeration in front, the tree channel can directly penetrate through the aggregated clusters, which is thought to accelerate tree growth. The interaction in nanocomposites is not observable but both of XCT and SEM imaging suggest the appearance of nano particles in tree channels which may act as a protecting skin on the inner tree wall to prevent further damage from discharges.

8.2 Further work

This research has investigated the characteristics of electrical tree growth in different dielectric systems and developed methodology helpful to understand treeing mechanisms. There are still some related areas necessary for further research.

- XCT imaging is a powerful tool to study the treeing phenomenon in different dielectric systems. For the further application, the multi-stage treeing and imaging should be carried on in the future with well controlled X-ray doses. In this research, the three-step treeing and imaging test successfully revealed the characteristics of a typical early ‘branch tree’ so the ‘bush tree’ can be the next objective. A higher level of accuracy and care are required to represent the growth of dense ‘bush tree’ and a more comprehensive analysis should be considered to determine the impact of X-ray irradiation on the polymer.

- The interaction between tree channel and filled microsilica has been well revealed but the change of some structural parameters such as tortuosity and 3D fractal dimension do not well match the expectation. It is important to design a better comparison test to identify the different tree morphology in filled materials. An ideal is to reduce the needle-to-plane distance to meet the maximum view field (~500 µm) of the XCT imaging. By doing this, trees with the same length could be compared after breakdown. The limiting resistance should be increased to avoid the too huge breakdown channel.
Chapter 8: Conclusions and further work

- For further imaging in micro-filled systems, more typical samples should be selected. For example, the micro-size fillers used in this case was about 0.5 to 10 \( \mu \text{m} \) which was still in the same order as the tree channel so sometimes their interactions may be not so obvious. Particles with larger size, such as 50 to 100 \( \mu \text{m} \) may be more meaningful to investigate the structural change caused by the modification of a large barrier. On the other hand, more samples should be imaged in order to examine the repeatability of the similar tree structure, i.e. 5 samples in each micro-filled batch with different concentrations.

- As described in Section 6.4.3, only two untreated samples were imaged by synchrotron XCT in this thesis and even though the tree structure could be revealed in nanocomposites, all interactions were lost due the limited resolution of micro-XCT system. A nano-XCT synchrotron system may be required to study the 3D tree growth in nanocomposites in the future. Additionally, samples with silane treated fillers should be also included into the imaging plan in order to check whether the tree structure is changed in the treated nanocomposites.

- This research paid particular attention to tree growth while little work was done on the tree initiation. It is important to understand in which circumstance the initiation can be eliminated or speeded up. Ramping voltages may be essential to investigate the initiation and there are two situations should be mainly considered: how long it takes at a low voltage and what is the voltage for quick initiation. It is also meaningful to investigate the needle/dielectric interface. Different treatments on the interface may help understand the initiation mechanism. For example a comparison on the tree initiation time of a needle with conducting coating and another with non-conducting coating.

- The treeing model in micro-filled system speculate the changed propagation of the ‘fine tree’ channel so it will be meaningful to study the morphology of ‘fine trees’ in filled polymers. A possible method is to place few microsilica particles manually into the material, which will affect the tree propagation locally but still remain enough transparency for optical observation. Alternatively nano-XCT may be also capable to find the growth of ‘fine tree’ in filled systems. Additionally, the propagation of electrical trees in filled composites is proposed by assuming the similar ‘fine-to-dark’ transition also exists and this also needs more evidence to prove it.
• ‘Fine tree’ and ‘dark tree’ have been described in the epoxy system but it is still unsure whether these two tree types are common in other materials like XLPE. It is necessary to find out why they are only found in epoxy resin if there is no ‘fine tree’ confirmed in other materials. More characterisations should be determined especially on the nature of ‘fine tree’ such as applying chemical analysis.

• The parameter $dQ/dL$ mentioned in Section 6.2.2 needs a more general definition with the consideration of the whole treeing procedure. It will be helpful to start from the DC tree because of its simple tree structure [2] so that the linear relationship could be further examined. It will be much more meaningful if this parameter can be also applied in opaque filled samples, which provides a way to estimate the tree length from PD magnitudes even though the tree structure is not visible.

• The silane treatment of nano fillers shows great help on the treeing resistance but it is worthy to exploring novel dielectrics with excellent performance. One of the popular topic is the self-healing materials and here are some available options that has been used in the study of dielectric properties: superparamagnetic nanoparticles [187], microcapsules [188], Voltage Stabilizer [189] and the addition of functionalized boron nitride nanosheets (BNNSs) [190]. The long-term treeing experiment on the self-healing material may contribute to the more flexible insulation system in the future.
References


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References


Appendix 1: SEM images of micro fillers in epoxy resin

SEM images of the particle dispersion in different batches are shown in Appendix 1 and Appendix 2. All images were taken under BSE imaging mode in a FEGSEM facility FEI QUANTA 250. Due to the significant difference of mass density, silica particles are represented as white objects in the bulk epoxy material. All images shown below were captured on surfaces with the same process (only ultra-microtomy) and based on these images the particle dispersion could be quantified as introduced in chapter 5.3.2.

5 wt%

15 wt%
Appendix 1: SEM images of micro fillers in epoxy resin

30 wt%
Appendix 2: SEM images of nano fillers in epoxy resin

1 wt% UT

1 wt% ST
Appendix 2: SEM images of nano fillers in epoxy resin

3 wt% UT

3 wt% ST
Appendix 2: SEM images of nano fillers in epoxy resin

5 wt% UT

5 wt% ST

2 μm
Appendix 3: List of publications


