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Prebreakdown phenomena in hydrocarbon liquids in a point-plane gap under step voltage. Part 1: behaviour at positive polarity

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Abstract

This study addresses the dielectric performance of nonpolar hydrocarbon liquids and mineral oils under positive polarity stress. It is of interest to improve knowledge on how functional properties of dielectric liquids vary, as new brands arrive in the market, and existing standards are unsuited for documenting the dielectric functional parameters of these new liquids. Stopping length for non-breakdown streamers, breakdown voltages and velocities for various pre-breakdown streamer modes have been studied for a selection of model liquids (cyclohexane and white oils), for a gas to liquid oil, and a refined naphthenic transformer oil. Studies of propagation modes were done using an 80 mm point to plane gap and a step voltage with a 0.5 μs rise time. Light emission and pre-breakdown currents have been recorded and instantaneous velocities have been derived from images of propagating streamers. There are clear differences in streamer stopping lengths and mode occurrence and mode velocities between these liquids. The differences seem to be influenced by molecular sizes governing evaporation energy for streamer formation and by concentration of aromatics which can be coupled to electron avalanche processes in the streamer heads.

1. Introduction

Electric insulating liquids are used in various types of electric power apparatus, with transformer applications dominating totally in terms of market volume. The most important functional property of a transformer insulating liquid is its ability to withstand high voltage stresses over large open oil volumes. In the market for electric insulating liquids mineral oils are totally dominating. The composition and possibly the performance of these oils change with time due to changes in refining processes and environmental requirements. There is a recent trend that highly refined inhibited oils take over after uninhibited oils, which were more dependent upon crude oil origin. Lately, new liquids like esters and gas to liquid hydrocarbon liquids are increasingly used, also for the higher voltage ranges. It is therefore important to learn more about their performance, how it may vary and what governs these variations.

In gases, the functional properties of an electric insulation are determined by electronic avalanche processes that can be modelled based on gas discharge physics [1]. Breakdown occurs as a result of an ionized, conductive plasma channel that propagates across the insulation. These channels are denoted streamers or leaders, depending on e.g. plasma temperature. Practical considerations for use in gas insulated substations are widely documented, see e.g. [2–4].

Like in gases, breakdown in liquids also results from an ionized plasma channel crossing the insulation, a phenomenon also called a streamer. However, even if nomenclature and observations indicate similarities with gas discharges there are profound differences; the most significant being that to form a plasma channel in a liquid the liquid must be heated and evaporated in front of the streamer in order to form a phase change. The processes involved in this evaporation are still not understood, therefore models for calculating the dielectric performance of a liquid are still unavailable, and thus other engineering tools have to be stablished. In the past,

when mineral oils that exhibited similar behaviour were exclusively used, dimensioning of insulation systems was partly based on experimentally established statistical criteria derived from breakdown initiation at highly stressed volumes or surfaces [5], and partly on experience gained in test rooms. In addition, dimensioning criteria mimicking gas discharge theories were also proposed [6, 7].

On the academic side, an extensive number of studies have been published on small gap breakdown, [8, 9]. To study the propagation of a streamer in oil, larger gaps and therefore higher voltages are needed. Most of these studies are done using impulse or step voltages. For mineral oil insulation it has been demonstrated that streamer propagation velocities and hence time to breakdown vary with voltage [10–12] and with chemical composition of the liquid [13–16]. It was also documented that other liquids performed differently from mineral oils [17–20]. Important observations are that there can be wide variations in the streamer speeds, and that by increasing the applied voltage beyond breakdown, to the so-called acceleration voltage, a sudden increase by a factor of 100 in the propagation velocity can occur for positive streamers. The ratio between acceleration and breakdown voltage has been observed to vary depending on the type of liquid, and is dependent upon the gap distance [17]. The streamer velocity will have a large influence on the breakdown level under lightning impulse stress as propagating streamers could be quenched and stop once the voltage at the tail of the impulse drops below what is required to sustain propagation [21].

The fact that neither analytical models for calculating dielectric functional properties of a dielectric liquid, nor adequate test methods for revealing their function exist, calls for a careful investigation of how functional properties may vary from liquid to liquid.

Liquid breakdown depends on polarity, where positive polarity in general shows higher velocities. Different propagation modes that are characterised by their velocities can be distinguished [22]. The different modes have different shapes (propagation patterns) which also differ between liquids. For positive polarity there are 4 distinct propagation modes: 1st mode streamers that are very slow (0.1 km s^{-1}), 2nd mode streamers that propagate at some 1–3 km s^{-1} (comparable to the speed of sound in the liquid), 3rd mode streamers propagating up to about 10 times faster and 4th mode streamers propagating 100 times faster than the 2nd mode. Velocities up to several hundred km s^{-1} are observed for positive streamers of 4th mode. 1st mode is of little interest in long gap breakdown as it only occurs from very sharp anode tips, and it is not present in this study with the low curvature point electrode used. For the higher streamer modes, we see that as voltage increases branching generally increases to a bush shaped structure. Further increase introduces new modes: streamers may start in 3rd mode and later fall back to 2nd and then again switch to 4th mode towards the end of the propagation. When increasing the voltage further, the 4th mode may start from a 3rd mode streamer. Acceleration voltage as a parameter describing average velocity is therefore a rather vague parameter, but nevertheless important from a functional point of view. During propagation reilluminations in plasma channels and light from the streamer tips can be observed. The mechanism of the propagation seems to be at the tips where the field is high, and phase transition occurs. The electric field at the streamer tip will be controlled by applied voltage, potential drop along the streamer channel from electrode to tip, and from charge and charge separation formed by ionisation at the tips. Many observations converge to a hypothesis that tip processes are in fact electron avalanches [23]. However, no final conclusions can be drawn on this matter.

The present study focuses on hydrocarbon liquids; from pure cyclohexane via isoparaffinic liquids synthetized from gases and white oils to commercial inhibited transformer oils. These liquids are relevant for mineral oils in various stages of refining; their molecular sizes and their content of polyaromatics vary. This study presents images from propagation, the measured currents and light emission, together with global observations like stopping lengths, average velocities, breakdown and acceleration voltages. Occurrences of the various propagation modes are discussed in detail.

2. Experimental setup, materials and procedures.

2.1. Test cell, impulse circuit and registration

A schematic of the setup is shown in figure 1.

The point-plane gap was 80 mm long since this was the maximum which could be imaged with the lens which was optimal for the camera. There was a ground-potential plane (φ = 340 mm) and a high voltage wire electrode. The wire was changed for different oils: 0.7 mm tin plated copper wire with cyclohexane, 0.15 mm tungsten wire with the oils Marcol 52 and Primol 352, and 0.20 mm tungsten wire with Nytro 10XN and Diala S4 ZX1. The wire ends became rounded to a near-hemispherical shape due to breakdown conditioning. These electrodes had so large radius of curvature that 1st mode positive streamers were never seen. The electrodes were placed in a test cell (originally glass, later fibreglass) possessing viewports and containing 100 litres of oil.

The development of the streamers should be caused by the physics of the propagation, not by a continually changing voltage. As a simple example, consider a slow streamer under standard lightning impulse voltage

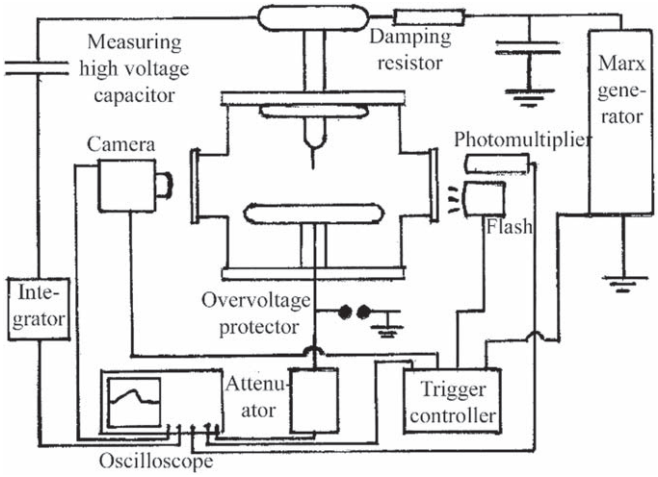

Figure 1. Schematic of the setup, excluding oil filtering/degassing system.

(1.2 μs rise time, 50 μs to half value of tail, conventionally written 1.2/50). The velocity of most streamer modes is not very dependent upon voltage. A slow mode streamer may have time to cross a short gap before the voltage drops too much for propagation but may not manage to cross a larger gap even under increased voltage. With further increased voltage, crossing will occur, but only after switching to a faster mode [24]. With an ideal step voltage, the same phenomena should be observable at all practical gap lengths, but at increasing voltage when increasing distance. This has been observed at least for some phenomena in mineral transformer oil in gap ranges from 10 mm to more than 150 mm. Even the ratios between minimum voltages where at least some investigated phenomena appear may stay close to constant in some but not all liquids [12, 22, 25, 26]. The step voltage was simulated with an impulse voltage of 0.5 μs rise time and decay time 1700 μs to half-maximum. This waveshape causes a 3% voltage drop relative to crest voltage for the maximum development time of 80 μs observed for positive streamers, thus avoids streamer quenching due to decaying voltage. The rise is not fast enough to avoid inception during the rise, at a voltage lower than the crest voltage when working above the minimum inception voltage. At the end of the voltage rise, there was an 8% overshoot with a duration of about 1 μs. There was a 76 Ω series resistor in the impulse supply to dampen high frequency oscillations. The voltage was measured via an oil-filled high voltage capacitor 1.5 metres from the test cell and a passive integrator mounted directly on the oscilloscope input [27]. The integrator was designed for 10 pF oscilloscope input, however the actual one used was 13 pF, causing the recorded voltage curve, after the overshoot, to contain a long duration, low amplitude 'hump' which is not factual.

The time development of streamers was imaged with 6 or 7 frames in an image converter camera (DRS Imacon 468). Either shadowgraphs (streamer backlit with a flash) or images of the streamers' emitted light were taken, in the latter case an additional streak image was taken with the same camera, and the time development of the overall light output was registered with a photomultiplier tube (Philips 56 AVP, 2 ns rise time). The timing, exposure time and image intensifier amplification could be set independently for all the frames and the streak. The camera and flash were synchronized with the impulse voltage. All streamer images shown in this paper show the entirety of the 80 mm gap length.

Gap current was measured by connecting the plane electrode to a 50 Ω cable with the necessary attenuators. Overvoltage protectors were installed to divert the breakdown current. Above approximately 310 kV the gap current could not be measured because the capacitive current from the rising impulse flank exceeded the overvoltage protection trigger level.

The four-channel oscilloscope used was a Tektronix DPO 4104 with 1 GHz bandwidth and 5 Msamples/s per channel, and a record length of 1 Msamples was usually used. Current, photomultiplier output, voltage waveshape and a monitor signal indicating the camera's frame and streak time positions were sampled by the oscilloscope. When not using the photomultiplier, current was sometimes registered on two separate vertical scales simultaneously. The capacitive current pulse from the impulse rise saturated the channel used with the most sensitive setting, however it recovered within 5–6 μs.

The time between subsequent impulses was not fixed, as it depended on the time needed for storing captured data files, measuring streamer lengths in the images taken and logging down measurements. This resulted in more than 5 min between impulses, with 10–15 min being the norm.

With Marcol 52, 10 impulses were applied at each voltage level. With the other liquids, 10 impulses were applied at each voltage level where significant changes took place (e.g. breakdown, acceleration or the occasional

Table 1. A number of properties of the liquids used. From manufacturers' data sheets or safety data sheets unless otherwise noted. Cyclohexane: From Wikipedia.

Unit	Kinem. viscosity @ 40 °C cSt	Density @ 20 °C kg/l	Carbon distri-bution Paraff./Napht./Arom. %	# C
Cyclohexane	1.3 (@ 17 °C)[a]	0.778	0/100/0‡	6[b]
Nytro 10XN	8 (typ.)	0.877	40/54/6 [29]	
Diala S4 ZX1	9.6	0.805	92/7/1 [30]	17–30 [30]
Marcol 52	7–8	0.830	68/32/0	
Primol 352	65–75	0.865	68/32/0	34(average) (5% < 25)

[a] Recalculated from dynamic viscosity.
[b] Implied by chemical formula.

appearance of a new mode), in all other cases 3–5 impulses were applied (typically if the only change was a few per cent shorter time to breakdown).

2.2. Liquids

Experiments have been conducted in three liquids, while results from previous research using two other liquids are included for comparison. For these two liquids, the main interest in the older studies was the effect of additives. That will be touched upon only in passing in this paper.

Cyclohexane [28] is included because it has become a standard liquid for research into the effect of additives, being a homogeneous liquid with only one type of molecule, (representative for a naphtenic oil molecule), only one type of C–C -bonds and one type of C–H -bonds. Purity was 99.5% with less than 0.01% benzene and cyclohexene. Neither gap current nor light output was measured in cyclohexane.

The medicinal grade white oils Marcol 52 (previously reported in [16]) and Primol 352, both produced by Exxon Mobil, are included mainly for comparing the effect of molecular size, which influences macroscopic parameters like boiling point (not well-defined for a complex oil) and viscosity. It is probably difficult to find a pair of chemically more similar oils with significantly different physical properties. Both are purported to be virtually aromatic-free. No oxidation inhibitor has been declared by the manufacturer.

Nytro 10XN from NYNAS is a much used highly refined inhibited naphthenic mineral transformer oil, with typically about 6% aromatics. Inhibitor content is 0.3%. Results from studies in the 1990's conducted in its forerunner, Nytro 10X [11, 12], were not included since the experimental conditions were different and the images obtained had a low resolution with only emitted light recorded, making streamer shape comparison difficult. New experiments were therefore considered necessary.

Shell Diala S4 ZX1 is a commercial transformer oil manufactured in a gas-to-liquid process. Using this process gives more control of the resulting molecules than refining of a mineral oil does. It is included because it is both commercially available for transformer use and is low in aromatics. This oil has been the subject of a comparison to a Shell mineral transformer oil [18].

Some properties of the liquids are listed in table 1.

2.3. Liquid treatment

Equipment for filtering and degassing of the liquids was available. The filtering was mainly for removal of carbon particles produced by breakdown. When used, the transparency of the liquids seemed unchanged, no visual signs of soot were seen, and oil performance did not change over time. This equipment was not yet installed when cyclohexane was tested. This liquid eventually became filled with soot from the numerous breakdowns. This was at the time not considered to influence the results as it was reported that soot had little influence on the propagation of 2nd and 3rd mode streamers [31]. A repeated breakdown test after the end of the experiments showed no change from clean to contaminated state. Degassing by means of evacuation could not be done due to the high vapour pressure and low boiling point of the liquid. It was subsequently found out that carbon particles may reduce initiation and breakdown voltage [15].

Marcol 52 and Primol 352 were filtered and degassed every 5–10 breakdowns. Nytro 10 XN was usually filtered and degassed after every breakdown, and Diala S4 ZX1 was filtered and degassed after each breakdown. This frequent filtering may not have been necessary for positive streamers, but certainly was for *negative* ones. This is described in detail in part 2 of this paper [32], which focuses on negative streamers.

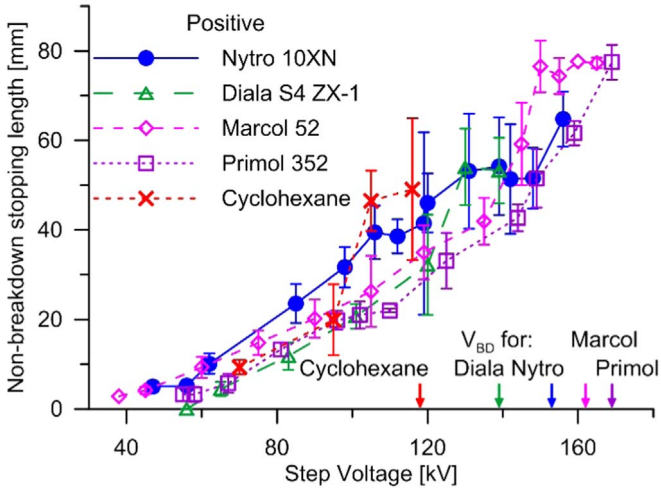

Figure 2. Stopping length of non-breakdown streamers, with standard deviation in the form of error bars. Arrows indicate 50% breakdown voltage (V_{BD}) for all liquids.

3. Results

3.1. Stopping length, breakdown and overall velocity

There are several ways of defining average velocity. One way is the average propagation velocity of a streamer which may vary in speed through time due to mode shifts. Another way is by averaging the complete collection of streamers from all impulses registered at a given voltage. This paper uses the term 'overall velocity' for the average of the velocity from inception to stop or breakdown for a single impulse application, while 'average velocity' is the average of the 'overall velocity' of the streamers of all the impulses registered at a given voltage.

Stopping length is the maximum observed length of streamers not leading to breakdown. The overall velocity (i.e. the average velocity for one streamer) is calculated from the time stamp and length at stopping, as observed from frame image sequences for non-breakdown streamers, or from time to breakdown, as observed from the ensuing voltage collapse. Time is measured from a fixed position early on the rising voltage edge. This time point is always earlier and at lower voltage than the actual streamer inception, leading to a measured overall velocity that is too low. For a slow streamer taking long time to breakdown this may cause an error of approximately 1%. For a fast streamer at high voltage though, with breakdown occurring in less than 1 μs, the error may be up to 30%.

If voltage is below a certain level, streamers will stop as shown in figure 2. The 50% breakdown voltage (V_{BD}) is also indicated in the figure. It is found from plots of experimental relative breakdown rate versus voltage. The voltage where a 50% line intersects a curve of straight lines from data point to data point is taken to be V_{BD}. In the 80 mm gap it was observed that if streamers in cyclohexane, Nytro10 XN and Diala S4 ZX1 propagate beyond a 50–60 mm point, they will always cross the gap resulting in breakdown and are thus not included in the figure. With Marcol 52 [33] and Primol 352 at about 50% breakdown voltage, there is a fair proportion of streamers reaching the plane without causing breakdown, and the stopping length average includes these. That phenomenon has not been observed in cyclohexane, Diala S4 ZX1 or Nytro 10XN, however has been observed only once in Nytro 10X [12].

Overall velocities versus voltage for all liquids are shown in figure 3. The velocity curves show that for all liquids examined, there is a voltage above which the velocity increases much faster with voltage than below. This voltage is known as 'acceleration voltage', V_a. The true voltage V_a lies somewhere between the highest experimental voltage where no obvious velocity increase has taken place and the lowest experimental voltage where the increase obviously has taken place. It is common practice that the former of these two voltages is reported as V_a. It is therefore important that the experimental voltage steps are small in that range. V_a is higher than V_{BD}. At an even higher voltage, the velocity increase reaches a saturation point. Below V_a, streamers grow mainly in what is known as 2nd mode. Above V_a, there are mainly 3rd mode streamers shifting to 4th mode ones, with the mode switch taking place closer to the point electrode as applied voltage increases, and the velocity levelling-off happening when there is only 4th mode propagation.

The overall velocity of non-breakdown streamers is generally very low at the lowest voltages, well below 1 km s^{-1}. This is caused by most of the streamer growth occurring at normal 2nd mode velocities (0.8–2.5 km s^{-1} depending on liquid and voltage), however this propagation stops and a considerably slower propagation (less than

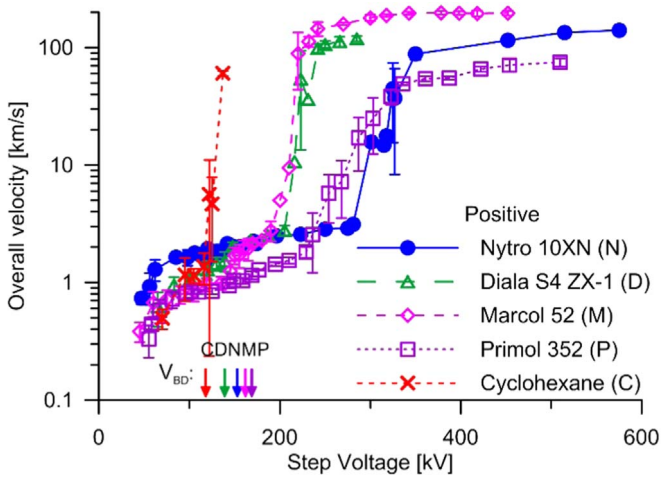

Figure 3. Overall streamer velocity, with curves showing the average velocity, and standard deviation markers. Arrows indicate 50% breakdown voltage (V_{BD}).

100 m/s) follows for up to half the propagation time while the streamer starts to die out. As the overall velocity includes this slow propagation phase, it is thus very low.

Cyclohexane has very low breakdown voltage and has V_a just 7% higher than V_{BD}, despite having slow and short streamers at the lowest voltages. Diala S4 ZX1 has similarly slow and even shorter streamers at the lowest voltages however has the second lowest breakdown voltage and a relatively low acceleration voltage. Despite Nytro 10XN having the fastest streamers at low voltage, breakdown voltage is higher than in Diala S4 ZX1 and it has the highest V_a of all oils examined. Velocity in Marcol 52 is very similar to velocity in Diala S4 ZX1, however the streamers propagate less for higher voltages applied compared to Diala S4 ZX1 while V_{BD} is higher even compared to Nytro 10XN. Streamers in Primol 352 which contains the largest molecules are the slowest and shortest of all oils and it accordingly shows the highest V_{BD} and the second highest V_a. It is interesting to note that the medicinal grade white oils have higher breakdown voltage than the insulating oils.

3.2. Mode velocities and mode existence ranges

Except at very low and very high voltages, streamers in all the liquids exhibit changes between different propagation modes during propagation. Velocities of the three observed modes are shown in separate plots in figure 4 for each liquid. At low voltages we see only 2nd mode, in the range 0.4–3 km s^{-1} depending on liquid and voltage, but most typically seen in the range 0.8–2.5 km s^{-1}. 3rd mode appears from the point electrode when the field is sufficiently high, with velocity usually within the range 3–50 km s^{-1}, however being lower in some liquids and occasionally higher as in the case of cyclohexane. The lowest voltage for 3rd mode to appear is marked in the plots in figure 4, denoted V_3. Propagation continues in the very fast 4th mode when the voltage is high enough, with a minimum velocity of 25 km s^{-1} registered in Primol 352 and a maximum of up to 400 km s^{-1} in^{-1} most other liquids tested. The voltage for 4th mode to appear is also marked in the plots, denoted V_4. For some liquids, voltage for 4th mode termination is above V_a. In the other liquids, streamer propagation may end in a short 4th mode streamer at voltages well below V_a. In this case, the length propagated in 4th mode is so short that it contributes little to the overall velocity despite its high value.

For the 2nd and 3rd mode, each point in the graphs shows the maximum velocity of that mode observed during a single impulse application. For the 4th mode, both minimum and maximum velocity is included, except for cyclohexane where only the maximum is included. For some liquids, V_3 and V_4 are lower than the start voltage of the corresponding velocity curve for those modes. This occurs due to pictures being captured at points that these mode velocities could not be measured independently. To clarify this, if in the time period between the last frame up to breakdown a streamer velocity of 8 km s^{-1} is calculated for a streamer which propagated at 2 km s^{-1} in 2nd mode up to and including the last frame, this is a clear indication of a 4th mode ending with unknown velocity and length since this fast 2nd mode streamer or this slow 4th mode streamer have never been observed independently, i.e. last frame positioned close enough to breakdown to see part of a 4th mode streamer having not yet reached the plane electrode.

Below acceleration, the propagation is dominated by the 2nd mode even when it starts in 3rd mode and ends in a short duration 4th mode (i.e. above V_3 and V_4 when these are below acceleration), and the overall propagation velocity is still equal to or lower than the maximum 2nd mode velocity. Sometimes the overall velocity and thus the average 2nd mode velocity is quite lower than the maximum, as is the case of cyclohexane,

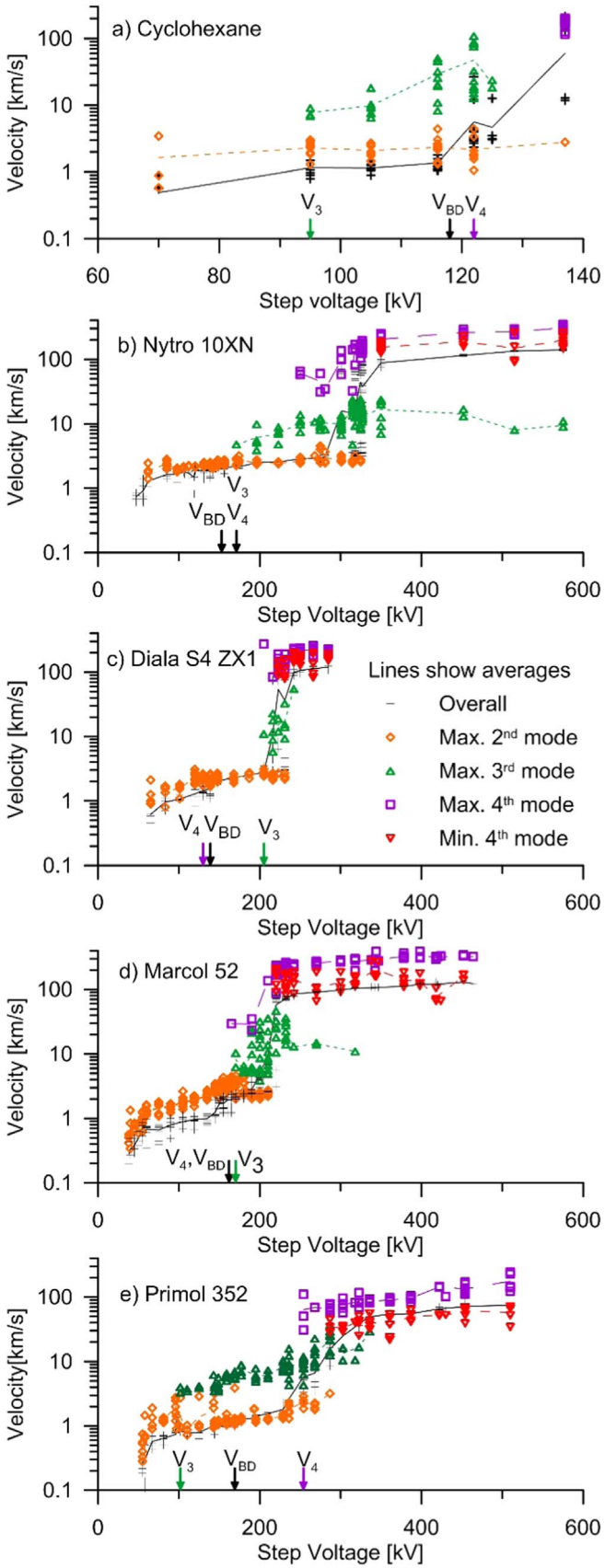

Figure 4. Velocities: overall, maximum 2nd, 3rd and 4th mode and minimum 4th mode (excluding cyclohexane). Minimum observed voltage for appearance of 3rd and 4th mode (V_3 and V_4) are shown with arrows, which are black if equal. 50% breakdown voltage (V_{BD}) is also shown with black arrows. Legend in (c).

Marcol 52 and to some extent Diala S4 ZX1. Typical 2nd mode streamers propagate at 1–2.5 km s^{-1} depending on voltage and liquid. In Primol 352 most propagate at 0.8–1.5 km s^{-1}.

The rapid overall velocity increase above acceleration voltage does not occur due to one of the modes gradually becoming faster, however because a larger part of the initial propagation occurs in the comparatively fast 3rd mode and also a substantial amount of the late propagation occurs in the very fast 4th mode. Eventually 2nd mode totally disappears, while further voltage increase causes the 3rd mode to shorten, being replaced by the 4th mode. The overall velocity stabilizes when the entire propagation takes place in 4th mode except for a very short 3rd mode (less than 5 mm) that takes place at the initial stages of streamer propagation. The 3rd mode appearing initially is due to the finite rise time of the voltage curve, causing streamer initiation to take place during the voltage rise phase at a considerably lower voltage than the rest of the propagation.

Minimum velocity of the 4th mode is substantially higher than the overall velocity in the region totally dominated by 4th mode. As mentioned in subchapter 3.1, overall velocity is measured from a fixed and very early position on the voltage curve while the mode velocities are measured from frame sequences, or in cases calculated by using remaining distance and the time from the last image frame to the observed breakdown occurrence. Above the levelling-off of the average velocity, minimum 4th mode velocity is typically 130–200 km s^{-1} while maximum 4th mode velocity is usually 300–400 km s^{-1}. A clear exception is Primol 352 where 4th mode velocity, in line with 2nd mode velocity, is lower than in the other liquids tested, with minimum and maximum 4th mode velocity usually in the ranges of 35–70 km s^{-1} and 60–200 km s^{-1}, respectively.

Relative change of 2nd and 4th mode velocities with voltage is small. 3rd mode displays a much wider relative range of velocities. Mode distinction is not always straightforward; In cyclohexane, Marcol 52 and Primol 352, there is not much difference in velocity between the fastest 3rd mode and the slowest 4th mode streamers. If the initial fast streamer is followed by 2nd mode, the fast streamer is considered to be 3rd mode. Classification is more difficult if a fast 3rd mode switches directly to 4th mode, however the 4th mode will display higher luminosity compared to the 3rd mode. In Primol 352, all streamers, even those at the lowest voltages, had an initial velocity which was much higher than the subsequent 2nd mode propagation velocity. This might indicate that the streamer initiation is always in 3rd mode, however at the lowest voltages the shape of the fast, early streamer was more typical for 2nd mode thus the streamers were classified as 2nd mode. Undoubtedly, this makes V_3 for Primol 352 somewhat subjective. With increasing voltage however, the shape and velocity difference between 2nd and 3rd mode became much clearer.

Differences in streamer propagation were also observed between the liquids. In both Diala S4 ZX1 and Marcol 52, 3rd mode streamers are clustered around the rapid average velocity increase above acceleration and exhibit a very wide range of velocities. In all five liquids, there exists a voltage range where at least some of the streamers start in 3rd mode, grow in 2nd mode and terminate in 4th mode. This is depicted for each liquid in figure 4 as a voltage range where all three modes exist however the average velocity is almost identical to the 2nd mode velocity. This is easily noticeable for Nytro 10 XN, where this voltage range is very wide. In Diala S4 ZX1, 4th mode termination is common even at fairly low voltages, even though 3rd mode start is not, while 3rd mode start without 4th mode ending happens over a wide voltage range in Primol 352.

3.3. 3.3 Streamer shapes

The velocities indicated in this subchapter are 'instantaneous', calculated from the difference in streamer length between the frame shown and its preceding one.

Shapes of propagation patterns of the 2nd mode streamers below breakdown voltage are shown in figure 5. At these voltages, streamers in Nytro 10XN and Diala S4 ZX1 show similar patterns, while streamers in Marcol 52 and Primol 352 exhibit more fine branching at the tips. Cyclohexane shows less branching than any of the other liquids tested.

At low overvoltages, i.e. voltage above however close to breakdown, cyclohexane typically shows one or two stems with a low degree of further branching, see figure 6. Streamers in the other four liquids show numerous channels radiating from the point, with a higher density of such channels in Nytro 10 XN and Diala S4 ZX1. As the streamers grow, they branch further. Some of those channels do not develop and disappear as the rest of the channels elongate. When more than ¾ of the gap has been traversed, a much denser set of branches may develop, see figure 6. This is most pronounced in Nytro 10 XN and Primol 352, with streamers in the latter showing the highest branching density. The density of branches is much lower in Diala S4 ZX1 and Marcol 52. Dense fine branching at the streamer tips will cause mutual shielding between the channels, reducing the field in front of the streamer [34]. This shielding is thought to suppress the conversion to 4th mode, however the branching needs to be very dense for the shielding to be effective [34].

2nd mode streamer shapes at higher overvoltage are shown in figure 7. Cyclohexane and Marcol 52 are omitted because in those liquids only slightly higher voltage from the one in figure 6 causes acceleration and 2nd mode being replaced by 3rd and 4th mode. This is probably due to the lack of mutual shielding, and the next to

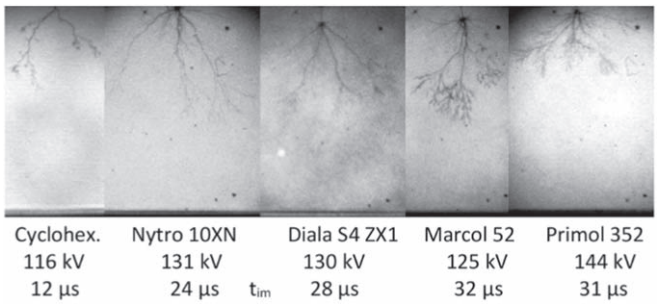

Figure 5. 2nd mode non-breakdown streamers.

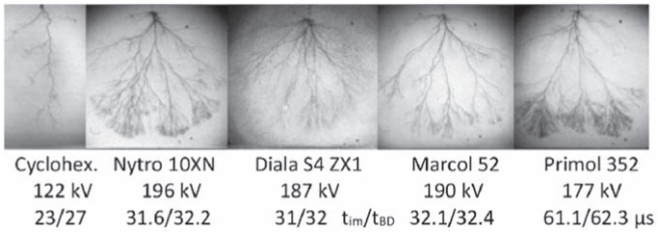

Figure 6. 2nd mode breakdown streamers, low overvoltage.

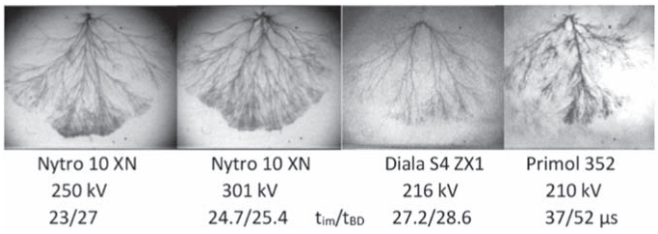

Figure 7. 2nd mode breakdown streamers, high overvoltage. Cyclohexane and Marcol 52 are not shown as these voltages are above acceleration in those liquids. Diala S4 ZX1 and Primol 352 are just below acceleration.

show acceleration is Diala S4 ZX1, at 216 kV. Streamers do not develop finer branching patterns with increasing voltage above the voltage applied in figure 6 neither in Diala S4 ZX1 nor in Primol 352. The fine branching becomes denser at much elevated voltages in Nytro 10 XN, probably contributing to its high acceleration voltage. The very fine branching does not occur closer to the point electrode with increasing voltage in neither Nytro 10 XN nor Primol 352.

3rd mode streamers are found in a wide velocity range. The slower 3rd mode streamers may be considered as more pointed examples of 2nd mode ones, with very few channels (often only one) radiating from the point electrode and with hardly any fine branching at the tips, as shown in figure 8. The shape is most branched in Nytro 10XN with Primol 352 exhibiting the second most branched pattern of the oils tested. The resemblance with 2nd mode streamers decreases with increasing propagation of the 3rd mode and is therefore closer for streamers that are shorter than those shown in figure 8. This figure shows 3rd mode streamers at voltages where 3rd mode exist for all the liquids (see figure 4), thus the images recorded correspond to different voltage levels for each liquid.

Most of the faster 3rd mode streamers (>10 km s^{-1}) differentiate from the 2nd mode, as depicted in figure 9, with an exception of the more branched streamers in Nytro 10XN, in which the fast 3rd mode is marginally slower compared to the other liquids. The typical pattern consists of one or two main channels with very short side branching, appearing as a slightly 'hairy' channel. Cyclohexane exhibited the fastest 3rd mode streamers reaching 109 km s^{-1} for the one shown in figure 9. Both velocity and luminosity are close to what is expected in 4th mode. Therefore, it could have been classified as 4th mode. However, after the frame shown it continued as 2nd mode at just above 1 km s^{-1}, and 4th mode switching to a slower mode has never been observed in any other liquid studied in our laboratory.

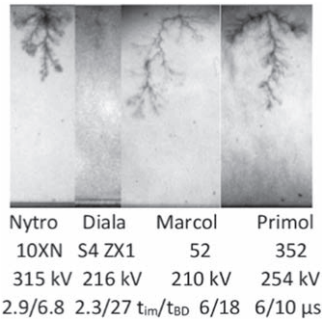

Figure 8. Slow 3rd mode streamers (<10 km s^{-1}).

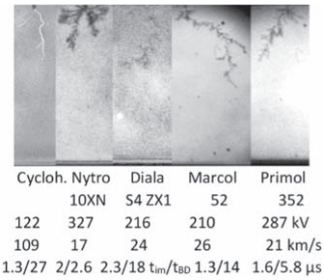

Figure 9. Fast 3rd mode streamers (>10 km s^{-1}).

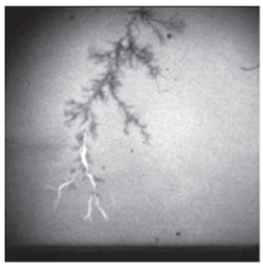

Figure 10. A 4th mode streamer growing out of a 3rd mode streamer. Primol 352, 287 kV.

A short 4th mode streamer terminating an otherwise long 2nd mode streamer consists of only a single, very luminous channel. Both images of 2nd mode streamers in Nytro 10 XN in figure 7 show small extensions from the 'spherical section' surface. These are probably on the verge of switching to 4th mode, however they are not sufficiently luminous yet. After these images were taken, the rest of the gap is bridged with a velocity of 98 and 108 km s^{-1} respectively. A 4th mode streamer (78 km s^{-1}) growing directly out of a rather long 3rd mode streamer (15 km s^{-1}) without any intervening 2nd mode is shown in figure 10. This is a typical transition observed on the steep part of the average velocity versus voltage curve above acceleration voltage.

Figure 11 shows 4th mode streamers at higher voltages where the entire propagation is in 4th mode except for possibly the initial 3–5 mm where the 3rd mode could exist. The very luminous channels are easily seen despite the images being flash lit shadowgraphs. The rather slow 4th mode streamers in Primol 352 show extensive short side branching which have such low luminosity that they appear dark in the flashlit image. Similar pattern, with less extensive side branching, is sometimes observed in the early propagation stages in Marcol 52.

With further increase in voltage, 4th mode may develop extensive branching almost resembling 2nd mode, however without the superfine branching seen at the tips of 2nd mode in some of the liquids tested. Some examples are shown in figure 12. In Primol 352 low luminosity short side branches persist, and a longer apparently dark branch is also visible. This branch has been luminous earlier in the propagation. Almost spherical examples have been reported in Marcol 52 [16] and Exxsol D140 [15]. With cyclohexane and Diala S4 ZX1 the voltage was not increased to a level where this extensive 4th mode branching might be seen.

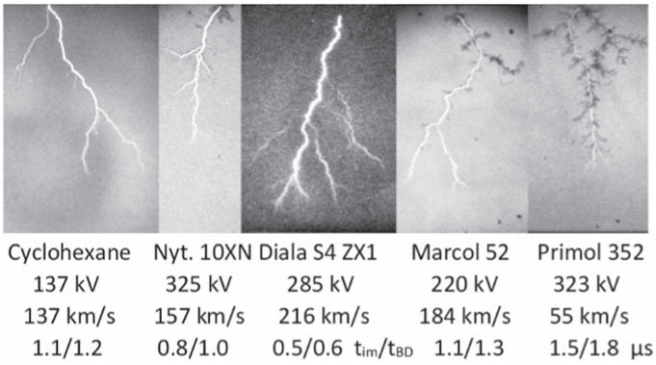

Figure 11. 4th mode streamers at the flattening out of the average velocity above acceleration.

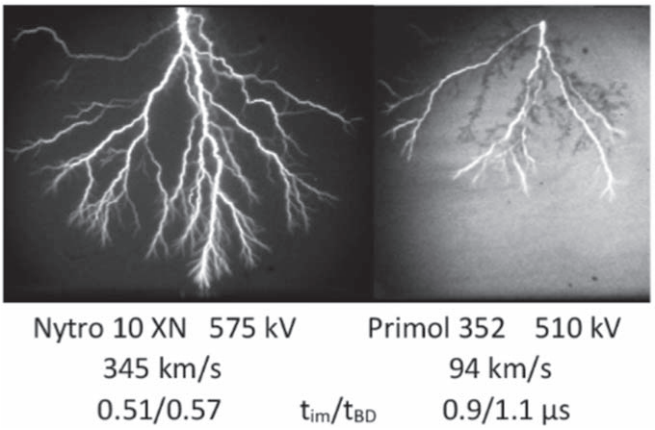

Figure 12. 4th mode streamers at voltage high above the point where velocity saturates above acceleration.

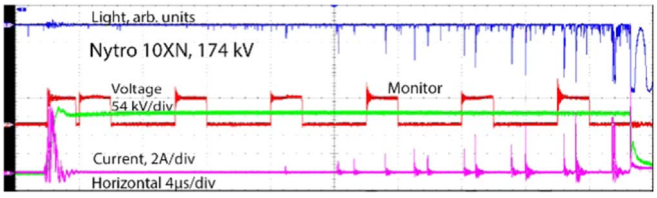

Figure 13. Reilluminations during 2nd mode in Nytro 10XN.

3.4. Reilluminations

Reilluminations are brief light flashes usually emitted from the entire length of one streamer channel. Channel reilluminations may include some of the finer branching near the tip. They are all accompanied by a current pulse of amplitude typically 0.5–10 amperes and sometimes exceeding 15 amperes with duration typically 10–15 ns. All oscillograms in this chapter show the current pulses and in some the light pulses are also shown. Some of the largest current pulses occurring during reilluminations have been clipped by overvoltage protectors. The large current pulse seen in the start of the oscillogram, is due to capacitive coupling to the high voltage parts and the fast-rising step voltage, and its amplitude and duration may mask smaller current pulses from the streamer. For cyclohexane neither gap current nor light output was measured.

2nd mode breakdown streamers in Nytro 10XN generate a lot of reilluminations (figure 13), although not in the beginning of the propagation, and they tend to grow in amplitude as propagation progresses. Reilluminations are rare in *non-breakdown* streamers above 120 kV and absent below. In 2nd mode in the other liquids there are seldom more than one to five reilluminations, at the end of propagation just before breakdown. Figure 14 shows 2nd mode reilluminations at a voltage higher than breakdown for all the oils. Streak images are included for two oils. Except in Nytro 10 XN, there are very few reillumination events recorded from 2nd mode.

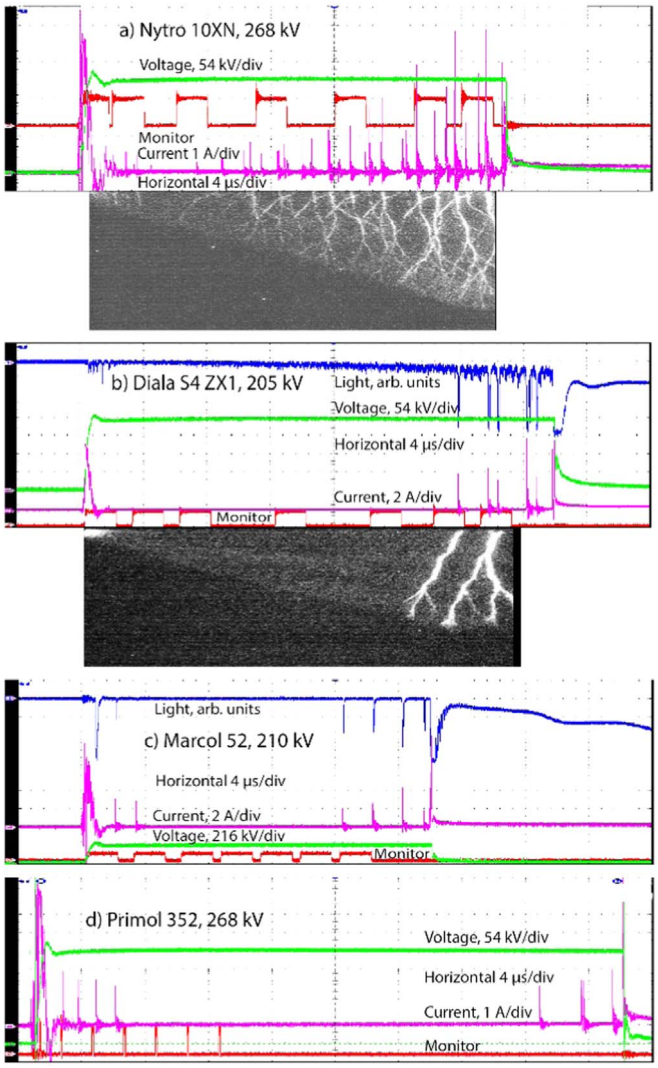

Figure 14. Reilluminations when propagation is initially in the comparatively slow 3rd mode and the rest is in 2nd mode. Streak images are included for Nytro 10XN and Diala S4 ZX1.

At the voltages shown, the streamers start in 3rd mode which is responsible for the small group of reilluminations in the start or the sometimes apparently continuous bright light as seen for Diala S4 ZX1 in figure 14(b). The reilluminations in this early 3rd mode are often of high brightness, however they do not usually register with high amplitude from the photomultiplier, probably because the streamers are still very short and high brightness does not correspond to much total light output compared to later when there are long channels reilluminating. The early reilluminations continue through much of the transition to 2nd mode and then cease, beginning a pulse-free interval before reilluminations from 2nd mode commence. The duration of this interval decreases with increasing voltage and disappears in the case of Nytro 10 XN. There is a weak continuous light between reilluminations. Often this can be hard to register in streak images because the sensitivity is set low enough to avoid overexposure of the reilluminations.

When the streamers have only 3rd and 4th mode, most of the *time* they are in 3rd mode although rarely much more than half the gap distance is traversed before transition to the 4th mode. 3rd mode streamers reilluminate frequently during their propagation (figure 15), sometimes with little variation in current amplitude while in cases with increasing amplitude. There is still continuous weak light between the reilluminations, however as the reilluminations are now brighter on average than at lower voltage, it becomes even more difficult to register, unless the sensitivity of the streak recording is set specifically for detecting that weak light. The transition to 4th mode with its almost exponential current increase usually takes place immediately after what becomes the last reillumination. This is observed for Nytro 10 XN and more easily for Primol 352 in figure 15, although it may be hard to distinguish because of the short time spent in 4th mode, usually less than half a microsecond. No pulsed phenomena have been observed within 4th mode. All

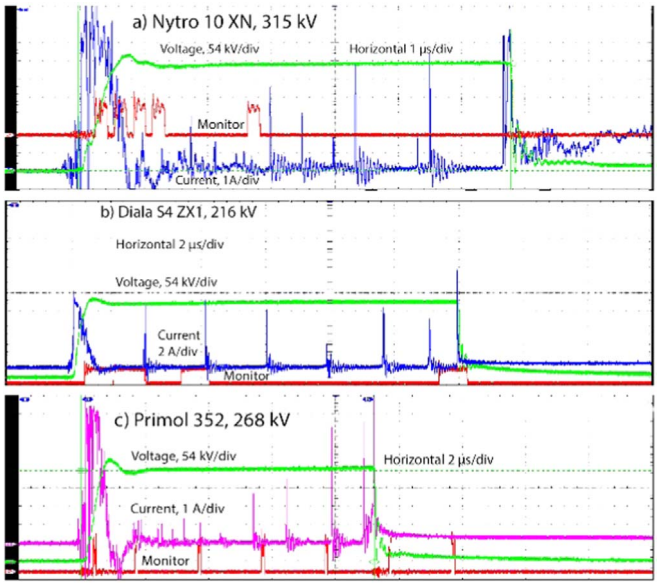

Figure 15. Current pulses of reilluminations for 3rd mode streamers terminating directly in 4th mode for three of the liquids tested.

oscilloscope traces of 4th mode current were chopped by the overvoltage protection, like the subsequent breakdown current.

3.5. Background current

The oscilloscope current traces showing the reilluminations may give the impression that no current is flowing except during the reilluminations. Nonetheless, a small background current can be registered if the sensitivity is set high enough, as shown in figure 16 for non-breakdown streamers and figure 17 for a breakdown streamer.

Up to 6 μs after impulse start, there are oscillations while the oscilloscope recovers from overload due to the large capacitive current pulse caused by the fast-rising slope of the impulse. Some minor, rapid pulses may be superimposed on the continuous current, however they are rare except in Primol 352. Some oscillations which look more like current instabilities may appear in Nytro 10 XN above 120 kV, with amplitudes of the order of 10–30 mA. The current of non-breakdown streamers generally shows a declining trend and apparently ends in a small step. Light emission grows up to this step, then starts a rapid decline. There may be a small, declining current of the order of 0.5 mA or lower immediately after the step, however there was not sufficient resolution to reveal its presence. The instabilities in Nytro 10 XN very often appear at or slightly after that step. Comparatively small instabilities are seen at the step in figure 16(a).

With breakdown streamers, the background current shows a weakly increasing trend (figure 17). Reilluminations will saturate the oscilloscope channel and recovery takes a few microseconds. When reilluminations are infrequent, the background current is also observed between them. Figure 17 shows some small oscillations or instabilities in the current. In Diala S4 ZX1 and Primol 352 such instabilities were often preceding the small group of reilluminations. It is not known if this phenomenon is present in Marcol 52 because sufficient sensitivity was not used.

The background current measured at 6 μs after the impulse start is shown in figure 18 for three of the liquids tested. At low voltages, the differences between the liquids are small. The current is smallest in Primol 352, higher in Diala S4 ZX1 with the highest recorded in Nytro 10 XN, all differences being marginal. This trend however diverges more above approximately 150 kV, with the most noticeable feature being that the current suddenly jumps to a much higher value in Nytro 10XN compared to the other two liquids. Up to approximately 200 kV the branching seems to be as extensive in Diala S4 ZX1 as in Nytro 10XN, therefore branching causing higher capacitance can be ruled out.

For non-breakdown streamers, there is a rough correspondence between the time marking the end of measurable background current and the time that the streamer stops growing, i.e. when it has reached its maximum length, as shown for Nytro 10 XN in figure 19. Light emission lasts marginally longer.

3.6. Light pulse at streamer initiation

In Primol 352, there was a short duration light pulse at the streamer initiation, even at a voltage level below breakdown (figure 20). Its magnitude is however comparable to the maximum overall light emission. In the

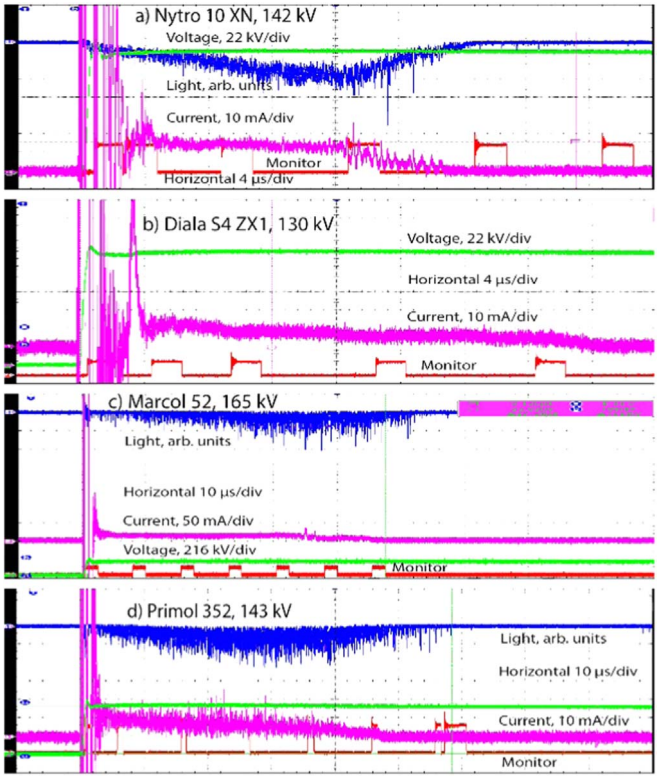

Figure 16. Background current for non-breakdown streamers. Light output is shown for three of the liquids.

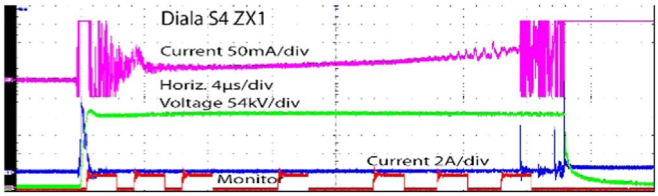

Figure 17. Background current example for breakdown streamer in a liquid where there are few reilluminations.

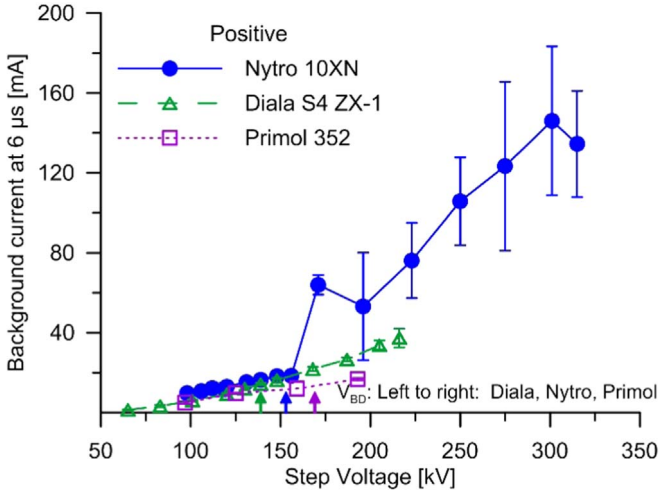

Figure 18. Background current at 6 μs after impulse start for three of the liquids. Arrows indicate the 50% breakdown voltages (V_{BD}).

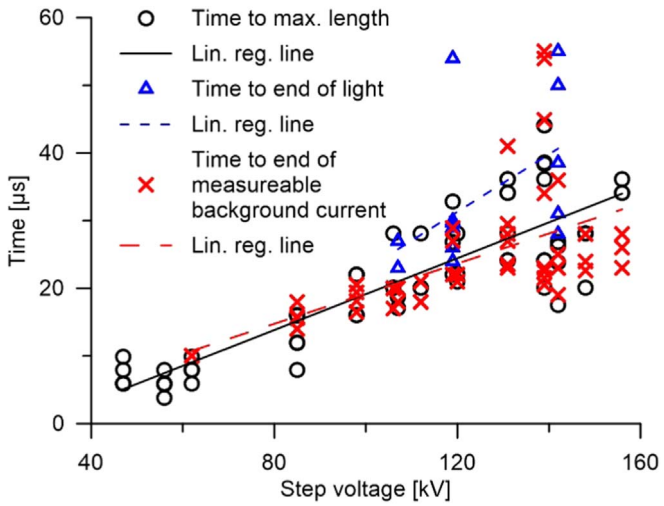

Figure 19. Time to max. length, end of light and end of measurable background current in Nytro 10 XN.

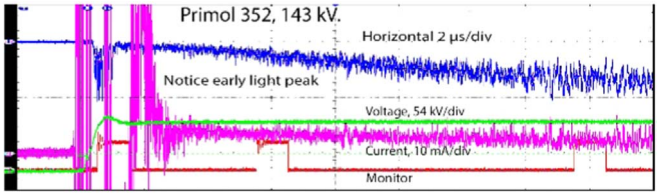

Figure 20. Light pulse at initiation of streamer in Primol 352.

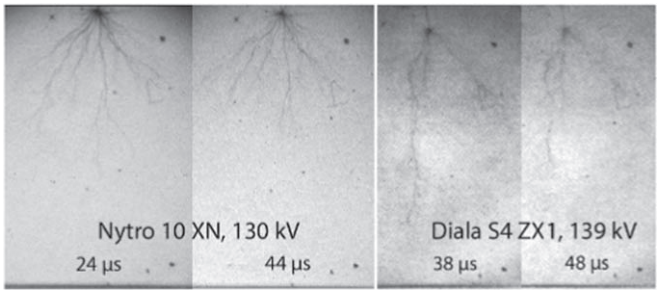

Figure 21. Initial stages of positive non-breakdown streamer channel decay. The first frame in each pair was taken approximately at the time the streamer reached its maximum length.

other liquids where oscillograms of light emission have been recorded (i.e. Nytro 10XN, Diala S4 ZX1 and Marcol 52) there is a luminous start at elevated voltages where the streamer start is clearly in 3rd mode, and is often due to a rapid series of reilluminations (figures 14(b) and (c)). It is tempting to associate the pulse in Primol 352 with the fact that all streamers were fast in the start at all voltage levels, compared to the rest of the propagation (figure 4(e)) although at the lowest voltages the initial velocity was more comparable to the 2nd mode velocities in the other liquids rather than typical 3rd mode velocities. A similar or larger pulse could be expected with 3rd mode start in cyclohexane because 3rd mode is very fast, and images taken indicate that it could be very luminous (figure 9).

3.7. Decay of non-breakdown streamer channels

The channels of a non-breakdown streamer will eventually decay after propagation stops. For positive streamers this starts at the streamer tip and decays backwards towards the point electrode (figure 21). The decay is not very fast, and since the experiment was focusing on streamer propagation, not decay, only early stages of the decay have been imaged.

4. Discussion

4.1. Cyclohexane as a model oil

Cyclohexane has been much used as a model oil because it consists of one type of molecule having only two different types of chemical bonds (covalent C–H and C–C). Having such molecular structure and uniform composition makes it particularly well suited for investigations of the effect of additives, however the pure liquid has a breakdown voltage far below that of any of the other liquids in this study. Cyclohexane has a low boiling point of just 80.7 °C. Forming and maintaining channels may therefore be much easier in cyclohexane than in most oils. Even its acceleration voltage is far below the breakdown voltage of the other liquids. Streamers in the pure liquid also have a very low degree of branching compared to the other oils. An additive with low ionization potential, e.g. dimethylaniline (DMA) [28], leads to increased branching and a high acceleration voltage. However, this additive does not raise the very low breakdown voltage. Low ionization potential additives clearly do not convert cyclohexane into an acceptable insulating oil. There are more properties required from a good insulating oil than just its low ionization potential constituents. In a few recent studies, molecular additives with low 1st excitation energies in addition to low ionization potential were added to natural ester liquids. In these formulations both breakdown and acceleration voltage were improved [35, 36]. Another candidate in this case might be molecule size. Cyclohexane has small molecules that are easy to evaporate, therefore forming and sustaining a channel in cyclohexane should require comparatively little energy.

4.2. Streamer velocity

In the voltage range where 2nd mode streamers dominate, Nytro 10XN has the fastest streamers. This is a result of its low ionization potential constituents, i.e. the polyaromatics. In both cyclohexane [28] and Marcol 52 [16, 37] it has been observed that the addition of 1% (=0.064M) dimethylaniline, which has low ionization potential, led to faster 2nd mode streamers. This did not occur in Exxsol D140 which already had 1% aromatics content from its production [15].

The slowest 2nd mode streamers were observed in Primol 352. In addition to not having any aromatic content, its large molecules are harder to evaporate, making the formation of a gaseous channel (low density, non-thermal plasma) difficult and thus require more energy per length unit of channel formed compared to the other liquids including Marcol 52 which is chemically similar to Primol 352 except for the molecule size. Since no reason exists for more energy per time unit to be available at the streamer tips in Primol 352 than in the other liquids, slower streamers are expected. Shorter non-breakdown streamers should also be expected, and this is what is experimentally observed. The same effect is probably the reason for the slow 4th mode streamers in Primol 352, while there is comparatively little difference between 4th mode velocity in the other oils.

4.3. Breakdown voltage

It is a little surprising that the oils designed for high voltage insulation (Diala S4 ZX1 and Nytro 10 XN) have lower breakdown voltages in this kind of experiment than the medicinal grade white oils Marcol 52 and Primol 352. It is known that low ionization potential additives lower the BD voltage because they cause increased streamer velocity and length [15, 16]. It is therefore not expected that Nytro 10 XN with its low ionization constituents (polyaromatics) would have the highest BD voltage, however the BD voltage of Diala S4 ZX1 is substantially lower despite the streamers being slower than in Nytro 10XN in the same voltage range. Primol 352, with its large molecules, exhibits the highest breakdown voltage, even higher than Marcol 52. This is in line with it having the shortest stopping length of non-breakdown streamers (subchapter 4.2). This argument is in good agreement with cyclohexane with its small molecules having the lowest breakdown voltage. However, a natural ester was tested in the same setup [38], having a high viscosity and thus comparatively large molecules. Quite low breakdown voltage was registered, in the mid-range between that of cyclohexane and Diala S4 ZX1. Hence, large molecules cannot be the only driving factor for making a good dielectric liquid, either.

4.4. Voltage for appearance of third mode

An analysis of the raw materials for [15, 16] shows that the addition of a low ionization potential additive (DMA) to Exxsol D140 and Marcol 52 increases the onset voltage for third mode, V_3. With Nytro's content of polyaromatics having low ionization potential, it was expected that Nytro 10XN would have the highest third mode onset voltage. This was not the case. It is higher in the oils where third mode is clustered around the rapid average velocity increase just above acceleration voltage.

As shown in [22], the onset voltage for 2nd mode streamers, V_2, can be found by extrapolating the stopping length versus voltage curves to zero stopping length. According to [22], V_3/V_2 is 2–3 in hydrocarbon liquids. All liquids in the present study are hydrocarbon ones, and the extrapolation has been done based on figure 2. The result can be found in table 2, and in 60% of the liquids this ratio is significantly higher than 3.

Table 2. Overview of inception voltages for 2nd and 3rd mode streamers and breakdown and acceleration voltages for the tested liquids.

	Liquid types				
	Cyclohexane	Nytro 10XN	Diala S4 ZX1	Marcol 52	Primol 352
V_2 - 2nd mode initiation	51	43	56	31	48
V_3 - 3rd mode initiation	95	171	205	170	102
V_{BD} - Breakdown voltage	118	153	139	162	169
V_a - Acceleration voltage	122	291	205	210	236
V_a/V_3	1,3	1,7	1,0	1,2	2,3
V_3/V_2	1,9	4,0	3,7	5,5	2,1

4.5. Acceleration voltage

Acceleration voltage is by far the highest in Nytro 10 XN. This is not surprising as its polyaromatics have low ionization potential, and low ionization potential additives increase branching and acceleration voltage in cyclohexane [28], Exxsol D140 [15] and Marcol 52 [16], despite also increasing 2nd mode velocity. This is theorised to be caused by the extensive branching lowering the field in front of the streamers by mutual shielding [25, 34]. The other oils have similar branching to Nytro 10XN below breakdown voltage, however with further voltage increase, the fine branching develops most in Nytro 10XN. The second highest acceleration voltage is found in Primol 352. This could be caused by the large molecules that have the effect of keeping velocities generally low [subchapter 4.2]. Primol 352 also shows the second most extensive fine branching, therefore some mutual shielding effect between tips is expected to contribute. The branching is not quite as extensive as in Nytro 10XN, however Primol 352 nonetheless demonstrates that some fine branching can take place in a liquid with virtually no low ionization potential constituents. Whatever causes the fine branching in Primol 352 probably has more influence on the acceleration voltage than the molecule size alone, since the natural ester [38], which also has large molecules, has a very low acceleration voltage almost equal to its very low BD voltage and only marginally higher than the BD and acceleration voltages of cyclohexane.

Table 2 shows that acceleration voltage is to varying degrees higher than breakdown voltage, with V_a/V_{BD} being by far highest in Nytro 10 XN with its polyaromatic constituents. Because V_a increases and V_{BD} typically slightly decreases when adding low ionization constituents to liquids having little or no of them, like cyclohexane, Exxsol D140 and Marcol 52, V_a/V_{BD} also increases. Cyclohexane is again an outlier with V_a being almost the same as V_{BD}. For traditional mineral transformer oils with polyaromatic constituents, the V_a/V_{BD} has been found to vary little with gap length [22], and this has also been observed in the mineral transformer oil Nytro 10X from the raw data in [12]. For a natural ester the ratio decreases and eventually equals unity with increasing gap length although both V_a and V_{BD} increase [22]. The gap length variation of this ratio is not known for this study's liquids that do not contain polyaromatics.

Acceleration requires the presence of modes faster than 2nd mode. The rapid increase in overall velocity is caused by faster modes taking over from 2nd mode with increasing voltage. However, an initial 3rd mode streamer in a streamer otherwise dominated by 2nd mode is not sufficient for the acceleration to take place. It has been stated that V_3 is significantly lower than V_a in mineral transformer oils [22]. This is confirmed for Nytro 10 XN in this study (table 2), however is also the case for Primol 352 with its very low V_3 and fairly high V_a. In contrast, in Marcol 52 and Diala S4 ZX1 the 3rd mode first occurred around acceleration voltage.

It has been demonstrated that in large gaps cyclohexane, Marcol 52 and Exxsol D140 switch to a behaviour more like mineral transformer oil when adding electronic active compounds [14–16]. For mineral oils their behavior is linked to the content of polyaromatics. Small scale experiments showed that one could rank V_a/V_{BD} for various liquids in smaller gaps [39]. Unpublished results showed that an effect like that observed by adding pyrene to cyclohexane [14], could be also observed by adding 0,4% by weight of the oxidation stabilizer BHT (Butylated Hydroxytoluene) to cyclohexane. This changed the ratio V_a/V_{BD} from 1.26 to 1.8, however also reducing the V_{BD}. Increasing the content of BHT increased these effects. This is a clear indication that additives like oxidation inhibitors, which are not necessarily declared for all liquids, should be considered in addition to the basic composition of a liquid itself.

4.6. Reilluminations

2nd mode reilluminations are much more frequent in Nytro 10 XN compared to the other oils where it is limited to a few reilluminations during the late propagation stage. An analysis of the raw data for [15, 16, 28] showed that the addition of a low ionization potential additive (dimethylaniline) or an electron scavenger (trichloroethylene) caused a large increase in the number of reilluminations in cyclohexane, Exxsol D140 and Marcol 52, with trichloroethylene having a strong effect at fairly low voltages (while also reducing the acceleration voltage). Thus,

it is made uncertain which property of the polyaromatics causes the frequent reilluminations in Nytro 10XN: low ionization potential, electron scavenging, or possibly both.

In [22] 2nd mode reilluminations of a stopping streamer in a 100 mm gap in a natural ester is depicted. There are frequent current pulses, with their amplitudes being initially below 10 mA and growing gradually to about 100 mA. These low current amplitudes (of reilluminations) were not observed in Nytro 10XN, Diala S4 ZX1 or Primol 352, where sufficiently high sensitivity to detect such pulses was used, although single pulses in that range were observed on rare occasions. In addition, according to [22], third mode streamers grow only during the brief reilluminations. However, as explained in subchapter 3.4, this is most likely due to the sensitivity setting of the streak recording. In the same paper an apparently stepped 3rd mode streamer with long steps is depicted, during few reillumination events in the same natural ester. These reilluminations have a current pulse duration of 200 ns, and a velocity exceeding 100 km s^{-1} is calculated by the extension from one reillumination to the other. In the oils examined in the present study, current pulses from reilluminations in any mode lasted approximately 10 ns and in rare cases up to 20 ns. Based on the typical number of 3rd mode reilluminations observed in this study and the duration of the current pulses, the velocity of the steps calculated would exceed 500 km s^{-1}, which is higher than any 4th mode streamers. In fact, older studies using streak imaging on Nytro 10 X show a continuous growth between reilluminations for 3rd mode streamers. Thus, more experiments with streak imaging are needed to investigate this phenomenon further.

The high current seen at reilluminations implies a sudden movement of charge, most probably negative charge which has become trapped behind the positive streamer head. It is undisputed that there is no abrupt extension of the channels at 2nd mode reillumination, therefore it is unlikely to be associated with an explosive increase in ionization at the head. It can be assumed that the trapped charge can cause an elevated field along the channel with some subsequent internal breakdown taking place. However, this elevated field along the channel should be associated with a reduced field somewhere closer to the plane electrode, in all probability at the streamer head, reducing the tip field. Decreased head ionization and resulting quenching of streamer growth might be expected until the trapped charges have been removed during the reillumination, however that is not observed. The nature of the trapped charges is also unknown. Since reilluminations become much more frequent with an electronegative additive in oils which have little of such components naturally, and also being frequently observed in Nytro 10 XN which contains electronegative polyaromatics, it can be presumed that the negative charges get trapped because of the formation of negative ions. However, increased frequency of reilluminations also occur with the addition of dimethylaniline which has a low ionization potential while not being electronegative. Thus, it is difficult to draw any satisfactory conclusion.

The voltage-drop per centimeter for the 2nd mode streamer channel is usually found as the slope of stopping length versus voltage for non-breakdown streamers. It is typically listed as being 20 kV cm^{-1} [40] which would fit most of the liquids in this study (figure 2) up to about 50 mm length. The slope becomes steeper (fewer kV/cm, more conductive) for longer streamers. This drop has also been assumed to be constant for most of the propagation, however, this seems unlikely for reilluminating channels. The drop should depend on the channel conductivity which would be different just before and just after a reillumination, even though the value may be correct as a time average. Reilluminations appearing or becoming significantly more frequent may possibly be the reason why the slope versus voltage of the stopping length increases in some liquids when approaching breakdown voltage.

Usually, only a few of the main channels of 2nd mode streamers reilluminate. As observed in [33], there does not seem to be any difference in propagation velocity for reilluminating and non-reilluminating channels.

The driving force behind reilluminations seems to be present for the faster 3rd mode in all the oils tested. The higher velocity compared to 2nd mode ought to correspond to more charge generated per time unit per channel. Possibly the channels on average between reilluminations has less conductivity in the 3rd than in the 2nd mode, leading to a charge build-up in all liquids which causes the frequent internal channel breakdowns (i.e. reilluminations).

No reilluminations occur in the 4th mode. This mode has continuous high current and is luminous. Conditions in the channel, can be likened to that of a continuous, long-lasting reillumination.

4.7. Background current

Since there is a background current, and simultaneously a continuous light exists, movement of the charges separated during continuous ionization is expected. However, this is very small compared to the current amplitudes registered during reilluminations. After the step down in the current of non-breakdown streamers, there may still be a low residual current with barely any illumination, indicating that there may still be a low and decaying ionization despite the propagation having stopped or becoming very slow.

The background current increases with voltage, and above breakdown level it is notably higher in Nytro 10 XN than in the other liquids. An argument considering capacitances might explain this: There is a capacitance

between the streamer and the plane electrode. The channels are expected to be neutral plasma, and there is also charge separation at the streamer tips with immediate neutralization of what was previously at the tip. The channels are conductive, though not highly. Thus, a growing streamer can be theorized of as a charge layer on an electrode moving towards the plane, which causes a capacitive current. The faster a streamer grows and the wider the frontal area it has towards the plane, the higher should this current be. This would be a fitting explanation on why the current is higher in Nytro 10 XN than in Marcol 52. When it comes to Primol 352, the low speed of its 2nd mode streamers should contribute to the low current, however there is a substantial amount of branching (figure 6), therefore it is difficult to explain the substantially large difference between Primol 352 and Nytro 10XN. There is a clear step in the curve for Nytro 10XN. There could possibly have been a similar step in Primol 352 if measurements had been extended to higher voltages. This hypothesis of a moving charge layer will also be in line with a streamer model where propagation is driven by the field created by the space charge itself, something that also agrees with the fact that in Marcol 52, 2nd mode light and dark channels move equally fast.

The branching increases as the plane is approached, increasing the area of the 'electrode'. This should increase the background current, which is also observed (figure 17) although the increase is smaller than one might have expected from the above argument.

4.8. Decay of non-breakdown streamers

Conventional understanding of the decay of 2nd mode streamers that have stopped growing suggests that the channels initially break down into a string of bubbles [41]. That is not obvious here, however the imaging resolution may be too low to resolve this phenomenon properly. Apparently, the newest parts of the channels decay first, therefore there is a gradual decay backwards toward the point electrode. This is different from what has been described for positive 1st mode streamers in short gaps where the decay grows outwards from the electrode [42].

4.9. Summarizing discussion

The principal properties of a liquid as a dielectric insulation material for transformers are the breakdown voltage and the velocities of the streamers leading to breakdown. If velocity is high, then short duration over-voltages like lightning impulses may result in breakdown. Hence, the focus on acceleration voltages and mode transitions are of great interest. It was observed that the naphthenic mineral oil like Nytro 10 XN has the highest acceleration voltage, however it is expected that changes in refining processes with heavy hydrogenation that saturates aromatic molecules would change it towards a white oil like Marcol 52. In contrast, Marcol 52, although being free from aromatics, has a higher acceleration voltage than cyclohexane. A possibility is that nondeclared oxidation inhibitors like DBPC (di-tert-butylparacresol) govern Marcol's performance, but cyclohexane's poor performance compared to Marcol 52 is more likely caused by its low boiling point.

The present study investigates the functional properties of several liquids including mineral oils under step voltages, using captured images, light emission and measured currents as descriptors to support distinction between the different modes. We observe differences in velocities together with differences in shapes; some of the characterizations have an element of subjective interpretation. Overall, what is observed in this study is in line with the proposed characteristic modes documented in earlier studies [12, 22]. Light measurements can prove to be problematic as light yield varies considerably from liquid to liquid, and aromatic compounds are known to emit more light than e.g. cyclohexane does. However, it is shown that both background light and reilluminations are coupled to a continuous background current and current pulses. These measurements support each other, thus there is little risk that invalid conclusions on reilluminations can be drawn from differences in light yield.

In the ongoing discussion on driving mechanisms behind streamer propagation no clear conclusions can be drawn. The general picture is that there is a heating mechanism at the streamer tips that evaporates liquid to establish a conductive plasma channel in its wake. The fact that Primol 352 with large molecules and an expected high evaporation energy has the shortest propagation distance for a given voltage below breakdown and the lowest velocities both for 2nd and 4th mode streamers is in line with this. Contrary to this however, 2nd mode streamers move faster in Nytro 10XN than in any of the other liquids, while the level-off of 4th mode velocity was slower than in Marcol 52. Possibly the most significant difference in molecular composition between Nytro 10XN and Marcol 52 is the content of easily ionizable polyaromatics. Compared to Marcol 52, the acceleration voltage is also significantly higher in Nytro 10XN. A similar observation was made when adding DMA to respectively Marcol 52 and Exxsol [15, 16]. It is therefore probable that the differences between the Marcol 52 and Nytro 10XN seen here are caused by the easily ionizable compounds in Nytro 10XN, namely its polyaromatics. The considerable large voltage range of stable 2nd mode can be caused by the dense fine branching that is observed in Nytro 10XN that produces a field guarding effect around propagating streamers. All these observations are in line with the hypothesis that electronic properties govern 2nd mode streamer

propagation, while electron avalanching is a potential candidate as the driving mechanism. For the 4th mode streamers, an avalanche mechanism assisted with a forward coupling from light excitation of the molecules in front of the streamer head could explain their very high velocities. Polyaromatics are known to have a low ionization potential while also emitting light and having a low first excitation level. There are indications that velocity of 4th mode streamers correlate to 1st excitation level of liquid molecules [39] and that constituents with low 1st excitation level can also lower the 2nd mode velocity and enhance both breakdown and acceleration voltage [38]. It should be noted that a low 1st excitation level will probably not help if the cross section for the excitation is low.

Altogether there are many factors influencing the propagation to various degrees and some of them are listed below:

1. Molecular size, most likely mainly through the energy required for evaporation for forming and maintaining the channel.
2. Constituents with low ionization potential. They make 2nd mode streamers faster, however they also cause extensive fine branching together with a higher acceleration voltage, probably because the resulting field lowering in front of the streamer suppresses conversion to 4th mode.
3. Constituents with low 1st excitation level. They lower 2nd mode velocity and increase breakdown and acceleration voltages. The mechanism is uncertain, and three hypotheses exist [38].
4. Electronegative constituents have not been considered in this paper however they have been known of having a varying and usually small effect on positive streamers, depending on the main liquid. They have been observed to marginally increase BD voltage and acceleration voltage [17] or in other cases lower both [18] or even to have virtually no effect [27]. The polyaromatics in Nytro 10 XN may have an electronegative effect.

5. Conclusions

All tested liquids show a similar development of propagation velocities with increasing applied voltage. At lower voltage, equal to BD or less, subsonic and sonic velocities that gradually increase to supersonic with voltage are dominating and this is known as 2nd mode. With increasing voltage new, faster propagation modes occur (3rd and 4th mode). In a small voltage range above a certain voltage threshold—the acceleration voltage - the average velocities increase by a factor of 50 - 100 to velocities in the range of 100 km s^{-1}. Above this voltage, the velocities seem to saturate and become little dependent on voltage.

A high content of polyaromatics results in a high acceleration voltage despite also resulting in fast 2nd mode streamers, while high molecular weight gives low 2nd and 4th mode velocities. Polyaromatics and to some extent high molecular weight lead to considerable 2nd mode branching at higher voltages. The current and light pulses known as reilluminations are considerably more frequent in 2nd mode in Nytro 10 XN, which contains polyaromatics, than in the other oils, however reilluminations in 3rd mode do not differ to a great extent between the oils.

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