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Pre-breakdown phenomena in hydrocarbon liquids in a point-plane gap under step voltage. Part 2: behaviour under negative polarity and comparison with positive polarity

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Abstract

This study addresses the dielectric performance of nonpolar hydrocarbon liquids and mineral oils under negative polarity stress. 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 0.5 μ s rise time. Light emission and pre-breakdown currents have been recorded and instantaneous velocities have been derived from images of propagating streamers. Compared to positive polarity, there are less differences in streamer behaviour in the oils examined under negative polarity. Breakdown voltages and acceleration voltages are higher for negative streamers than for positive ones, while their propagation velocities are lower. While propagation modes for positive voltages are quite distinct, the mode changes for negative ones are more gradual. The behaviour of both positive and negative streamers is in line with the hypothesis that the propagation is governed by electron avalanches and quantum chemical properties of liquid components.

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. Lately, new liquids like esters and gas to liquid hydrocarbon liquids are increasingly used, also for the higher voltage ranges. It is important to learn more about their performance, how it may vary and what governs these variations. Pre-breakdown behaviour has already been investigated for a number of mineral oils and a gas to liquid oil under positive voltage stress [1]. Both from a practical and from a scientific perspective, it is of interest to investigate the similarities and differences in behaviour depending on the polarity of the voltage stress applied.

In gases, the functional properties of an electric insulation is determined by electronic avalanche processes that can be modelled based on gas discharge physics [2]. Breakdown occurs as a result of an ionized, conductive plasma channel that propagates across the insulation with a driving so-called corona region in the tip, where avalanches and charge separation take place. These channels are denoted streamers or leaders, depending on e.g. plasma temperature. Standard textbooks as e.g. [3] describes how this behaviour depends on polarity effects.

Like in gases, breakdown in liquids also results from an ionized plasma channel crossing the insulation, a phenomenon also called a streamer. The streamer channels are gaseous, or plasma filled, and are formed by an energetic evaporation process or phase change, taking place at the tips of the propagating channels. There are many visual similarities in both positive and negative polarity: both form treelike structures that emit pulsed light, however also differ in propagation stopping lengths, breakdown voltages, velocities and their dependence

on applied voltage [4, 5]. The dependence on electrochemically active additives is strongly dependent upon polarity [6–8].

To study the dielectric performance for design relevant dimensions, 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 [4, 9, 10], polarity and the chemical composition of the liquid [6–8, 11, 12]. It has also been documented that other liquids behave differently from mineral oils [13–16]. While there are quite discrete propagation modes for positive polarity depending on the applied voltage [17], the propagation behaviour of negative streamers is more difficult to categorize. For positive polarity the ratio between acceleration and breakdown voltage has been observed to vary depending on the type of liquid, and may depend on gap distance [13].

The propagation mechanism seems to be active at the tips where the field is high, and the phase transition occurs. The electric field at the streamer tip will depend upon the voltage applied, the potential drop along the streamer channel (from electrode to tip), and from the charge and the charge separation formed by the ionisation in the high field regions in front of the tips. Many observations converge to a hypothesis that the tip processes are in fact electron avalanches [5, 18–20], maybe less controversial for negative than for positive polarity.

The present study focuses on the behaviour of hydrocarbon liquids under negative polarity stress; from pure cyclohexane via isoparaffinic liquids synthesized 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, material and procedures

2.1. Set-up and methodology

The test set-up was described in detail in the previous paper on positive polarity [1]. Tests were performed in an 80 mm point to plane gap with a thin wire electrode at the high voltage side placed in a 100 litres test cell. The wire was thin enough so as not to influence the streamer lengths and breakdown voltages. Unless otherwise stated, the oil was circulated regularly through degassing and filtering equipment.

The applied voltage was approximating a step voltage having 0.5 μs rise time and a long fall time of 1700 μs to half-maximum. This ensures that all streamers except very long slow streamers propagate under almost constant voltage so that the phenomena taking place are caused by the physics and not by a constantly changing applied voltage like e.g. a standard lightning impulse (1.2 μs rise time, 50 μs to tail half-value). Some slow streamers propagated for up to 400 μs , and then the voltage had fallen to 84% of the maximum. Like for positive streamers, the results under step voltage are expected to scale well to other gap distances but at a voltage which is expected to scale monotonically but not necessarily linearly with gap distance. This has been shown for some negative streamer phenomena in mineral oil for gaps 60 mm and 150 mm [4] and 50–350 mm [21].

Pictures were taken using a fast shutter time image converter camera, either as shadow-graphic images with a back illumination or by using a streak camera module where a continuous image of the light emission in the gap could be observed. Current was measured at the low voltage side and a photomultiplier was used to measure light emission in cases where back illumination was not used.

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 appearance of a new mode), otherwise 3–5 impulses were applied (typically if the only change was a few per cent reduced 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.

Cyclohexane [22] is included because it has become a standard liquid for research into the effect of additives, being a simple liquid with only one type of molecules, 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 [6]) and Primol 352, both produced by Exxon Mobil, are included mainly for comparing the effect of molecule size, which influences macroscopic parameters like boiling point (not well-defined for a complex oil) and viscosity. No oxidation inhibitor has been declared by the manufacturer.

Table 1. A number of properties of the liquids used. From manufacturers' data sheets or safety data sheets except for cyclohexane: Wikipedia.

Unit	Kinem. viscosity @ 40 °C cSt	Density @ 20 °C kg/l	Paraff/napht/arom. % by weight	# C
Cyclohexane	1.3 (@ 17 °C) ^a	0.778	0/100/0	6 ^b
Nytro 10XN	8 (typ.)	0.877	40/54/6 [23]	
Diala S4 ZX1	9.6	0.805	92/7/1 [24]	17–30 [24]
Marcol 52	7–8	0.830	67/33/0	
Primol 352	65–75	0.865	66/34/0	34(average) (5% < 25)

^a Recalculated from dynamic viscosity.

^b Implied by chemical formula.

Nytro 10XN from NYNAS is a much used naphtenic mineral transformer oil, with typically about 6% aromatics and 0.3% inhibitor.

Shell Diala S4 ZX1 is a commercial transformer oil manufactured in a gas-to-liquid process. Using this process, chemical composition and molecular size range of the oil can be better controlled compared to refining a mineral oil. It is included because it is both commercially available as a transformer oil and is low in aromatics. This oil has been the subject of a comparison to a Shell mineral transformer oil [14].

Some properties of the liquids are listed in table 1.

2.3. Liquid treatment

A plant for filtering and degassing the liquids was used. The unit could either be operated alone, containing 100 litres of oil, or connected to the test cell. The filtering was done mainly to remove carbon particles.

The plant was not yet installed when cyclohexane was tested. Under test, this liquid became eventually full of soot because of the numerous breakdowns. This was at the time not considered to influence the results. A repeated breakdown test after the end of the experiments showed no change from clean to contaminated state.

Nytro 10 XN, Diala S4 ZX1 and Marcol 52 were filtered and degassed after every breakdown because of the experience from testing Primol 352 (and Exxsol D140 [7]). Primol 352 was initially filtered after every 10 breakdowns, but it soon became apparent that the soot had a profound effect on the streamers. Circulation for one hour through the test cell and filtering plant was attempted, however the high viscosity made this ineffective. Therefore, it was necessary to empty the test cell into the filtering plant, filter the oil and refill the test cell. This procedure took more than an hour. Thus, the number of shots available for breakdown streamers in Primol 352 is low, also because many shots initially thought to be valid proved to be doubtful at closer inspection after completion of the lab work.

3. Results

3.1. Stopping length, breakdown and overall velocity

Stopping length is the maximum observed length of streamers not leading to breakdown using frame image sequences. For non-breakdown streamers, the overall velocity is measured from time to stopping and stopping length as observed on images from the Imacon camera. Because there are only 7 image frames, the recorded stopping time may sometimes include as much as 20% error. For breakdown streamers, overall velocity is calculated from time to breakdown divided by gap length. The time to breakdown is measured from a fixed position early on the rising voltage edge. Because the setting of start time is always earlier and at lower voltage than the actual streamer inception, the calculated overall velocity will be lower than the actual one. For a slow streamer taking long time to breakdown this may cause an error of around 1%. For a fast streamer at high voltage with time to breakdown of about 4 μ s, this error may be up to 5%.

Stopping length is shown in figure 1. Arrows indicate the 50% breakdown voltage (V_{BD}) which is found from plots of experimental relative breakdown rates versus voltage. V_{BD} is the voltage where straight lines through the experimental points cross the 50% line. At a given voltage, cyclohexane has by far the lowest V_{BD} , while Nytro 10XN has the second lowest V_{BD} . This is also reflected in the stopping length, being longest by far in cyclohexane and second longest in Nytro 10XN. For voltages below 250 kV the stopping length difference in the other liquids is small, being shorter than 25% of the gap length. At higher voltages the stop length difference diverges more with further voltage increase. This is also reflected in differences in V_{BD} .

The overall velocity is shown in figure 2. There is an 'acceleration voltage', V_a , as is the case for positive streamers in all liquids in the test [1], above which the velocity increases much faster with voltage than below. The reported acceleration voltage is the value where this visually happens in overall velocity plots, and that is the highest voltage where rapid velocity increase has not taken place. At the voltage where velocity reaches the new

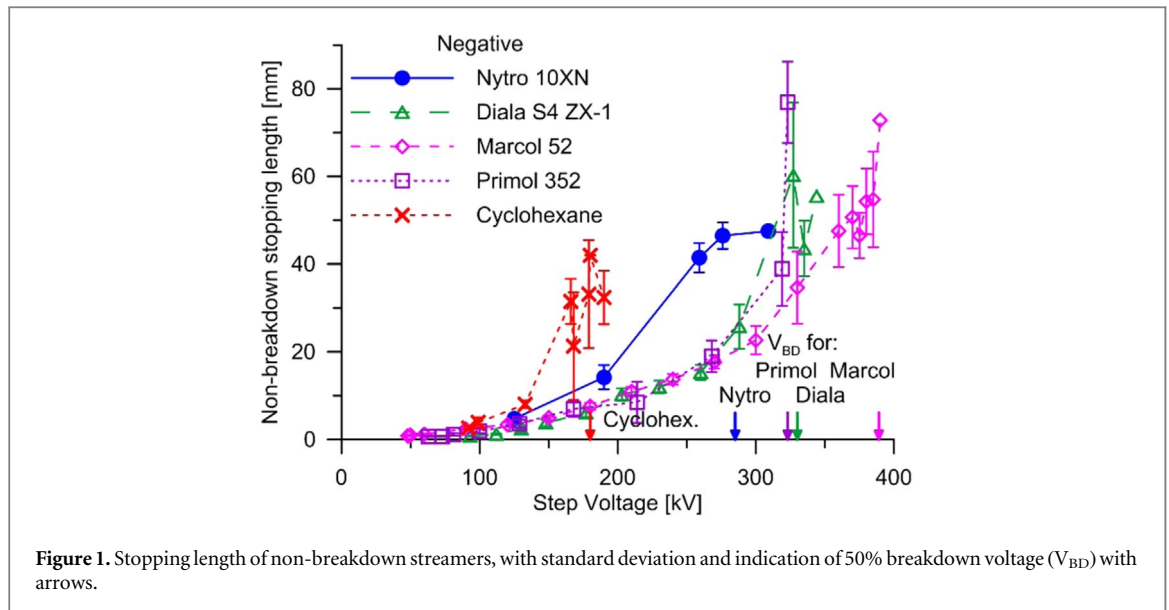


Figure 1. Stopping length of non-breakdown streamers, with standard deviation and indication of 50% breakdown voltage (V_{BD}) with arrows.

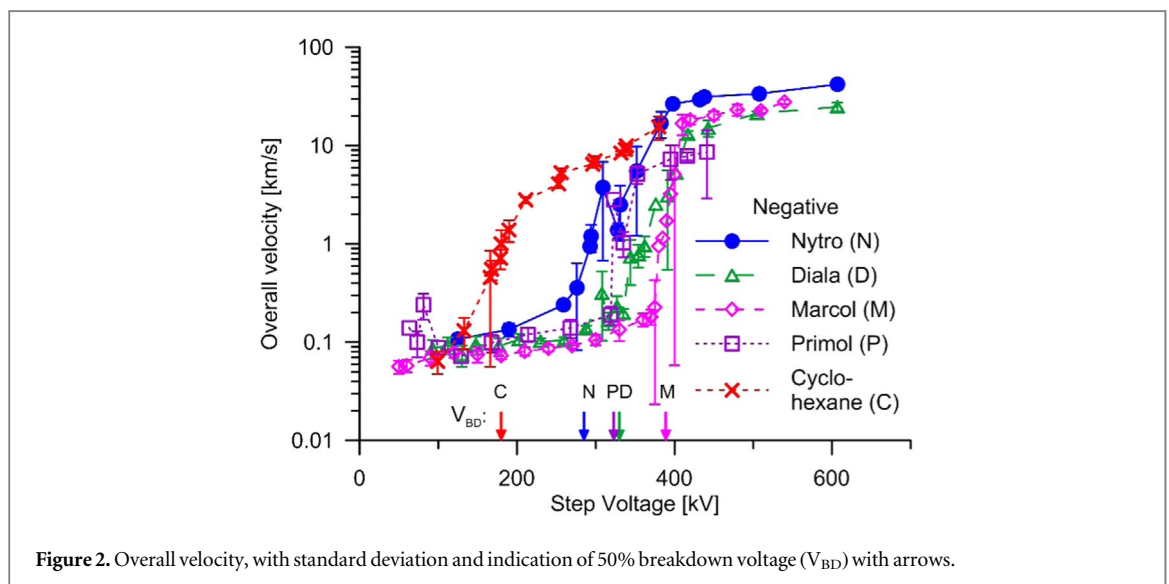


Figure 2. Overall velocity, with standard deviation and indication of 50% breakdown voltage (V_{BD}) with arrows.

plateau the relative velocity increase is comparable to what it was below acceleration. V_a is close to or even below V_{BD} , and the velocity just below V_a is typically $150\text{--}200\text{ m s}^{-1}$. At a given voltage, streamers are faster in Nytro 10XN and cyclohexane compared to the other liquids, and especially so in cyclohexane. Streamers in Primol 352 are generally faster than streamers in Marcol 52 which is chemically similar however has smaller molecules. This was unexpected as it shows a different trend to what was observed for positive streamers [1]. Primol 352 still has the slowest streamers at the high velocity plateau, though.

3.2. Mode velocities and mode existence ranges

Positive streamers exist in four significantly different modes having different shapes and velocities, with streamer velocity clearly increasing when shifting from one mode to a higher one. For negative streamers, the mode distinctions are not as clear, however, it was possible to use small differences in shape combined with small gaps in velocity ranges of the apparent modes to group the streamers into four modes. In this work, what can be considered as the separate 1st and 2nd mode had almost identical shapes and thick channels, however the 2nd mode was faster having higher velocity in the range of $300\text{ m s}^{-1}\text{--}2.4\text{ km s}^{-1}$. 3rd and 4th modes were clearly different from and faster than 1st and 2nd modes. The shapes and channel thickness of 3rd and 4th modes were similar to each other, except for the developed 4th mode main channels which were more luminous. The 3rd and 4th mode had much thinner channels than 1st and 2nd modes. Their shapes are treated in more detail in the next chapter.

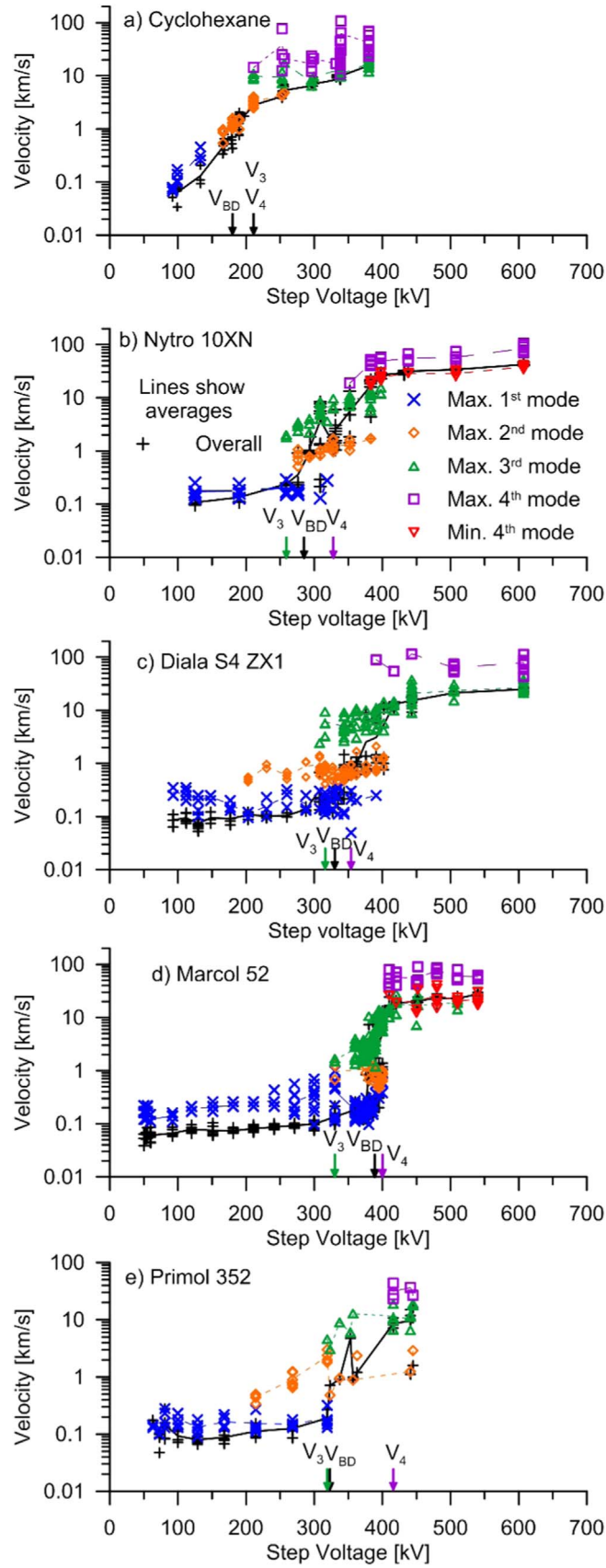


Figure 3. Mode, overall velocities, and minimum observed voltage of 3rd and 4th mode appearances (V_3 and V_4); 50% breakdown voltage marked with arrows. For cyclohexane, V_3 and V_4 are identical. Legend in (b).

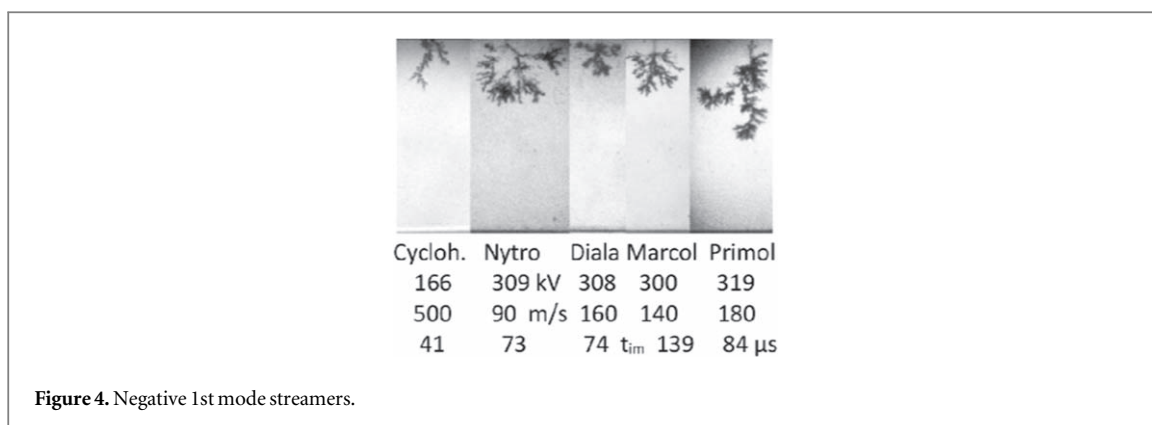


Figure 4. Negative 1st mode streamers.

Figure 3 shows the occurrence of the different modes for each of the liquids versus applied voltage. At the lowest voltages, 1st mode dominates and has usually average velocities in the range $70\text{--}200\text{ m s}^{-1}$. Velocities as low as 40 m s^{-1} were observed. 1st mode could bridge the entire gap and cause breakdown, with overall velocity below 300 m s^{-1} . This mode could be faster with velocities more than 500 m s^{-1} depending on the liquid, usually during the first microseconds after initiation. At higher voltage, initiation is taken over by a faster mode, first only occasionally with 2nd mode in Diala S4 ZX1 and Primol 352 and with 3rd mode at a higher voltage denoted V_3 in all oils. Typical 2nd mode velocities are $0.7\text{--}1.5\text{ km s}^{-1}$, however up to 2.4 km s^{-1} were observed in Primol 352 and 4.8 km s^{-1} in cyclohexane. Typical 3rd mode velocities are $2\text{--}20\text{ km s}^{-1}$, depending on the voltage and liquid. A streamer starting in 2nd mode will usually continue in 1st mode, while streamers starting in 3rd mode are followed either by 1st or 2nd mode propagation, depending on voltage. In Primol 352 2nd mode is always short lived and quickly switches to 1st mode.

3rd mode is restricted mainly to the ‘acceleration range’, where velocity versus voltage changes very rapidly, however, it may also extend into the plateau range dominated by the 4th mode. Cyclohexane is different with 2nd mode taking totally over from 1st mode while what appears to be 3rd mode is found only in the same voltage range as 4th mode, above the acceleration range, with 4th mode seen only in a final jump starting from approximately mid-gap position even at the highest voltage investigated (380 kV).

In the higher voltage part of the acceleration range, a streamer starting in 3rd mode and continuing in 2nd mode will often continue with the last part of the gap crossing in 3rd mode. This was never seen for positive streamers [1]. At a voltage denoted V_4 , fast 4th mode termination starts to appear. With increasing voltage, 4th mode termination will eventually always occur, and at even higher voltage 4th mode will take over almost the entire propagation. Average 4th mode velocities are typically $20\text{--}50\text{ km s}^{-1}$ depending on voltage and liquid, with the fastest part just before final contact with the plane and being up to 100 km s^{-1} .

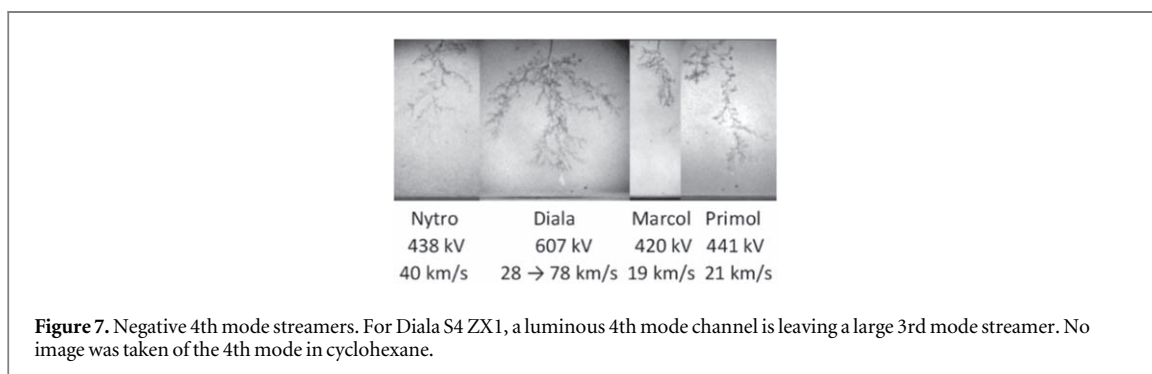
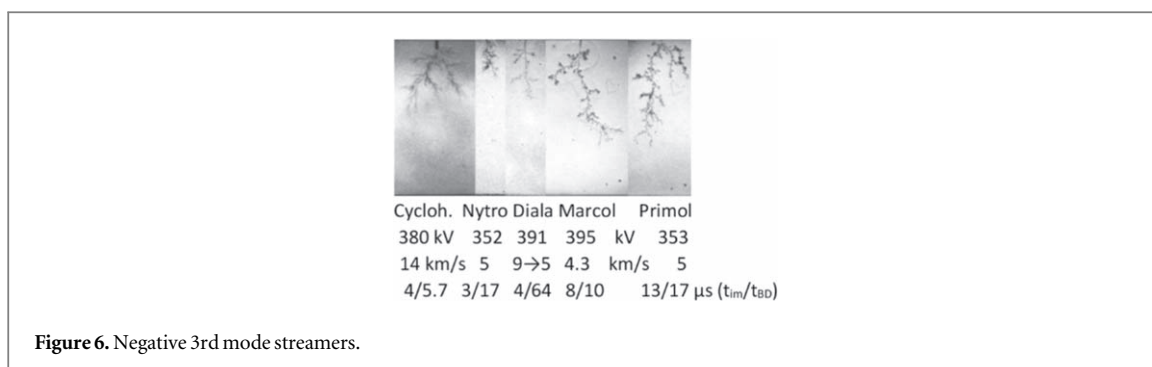
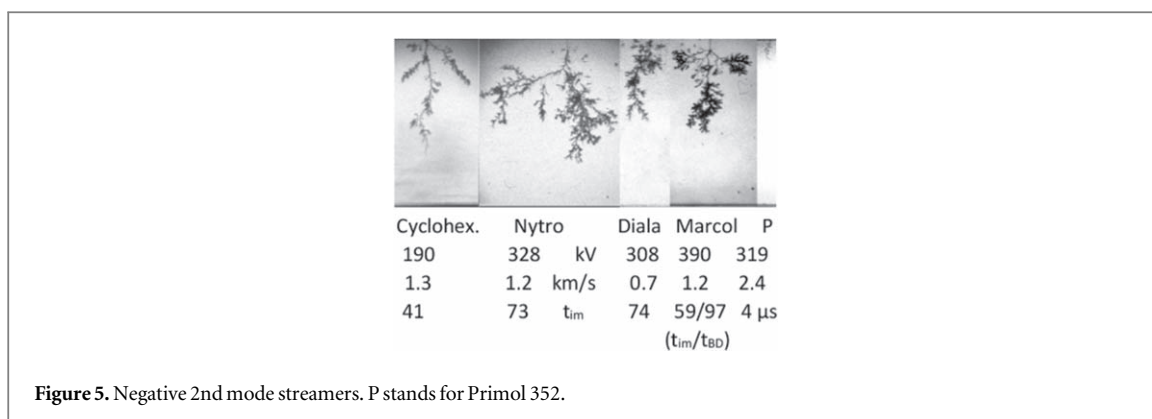
3.3. Streamer shapes

The velocities indicated in this subchapter are ‘instantaneous’, calculated from the difference in streamer length between the frame shown and its preceding one. The time at which image frames were taken are indicated in the figures. For breakdown streamers, time to breakdown is also listed.

A first observation is that 1st and 2nd mode streamers are thicker compared to the more filamentary 3rd and 4th mode ones. 1st mode (figure 4) and 2nd mode streamer shapes (figure 5) are almost identical for all liquids, with some exceptions: Primol 352 has a short and less branched 2nd mode, while cyclohexane has a less branched 2nd mode than Nytro 10 XN, Diala S4 ZX1 and Marcol 52. None of the 1st mode streamers have the classical lobed bubble shape which literature [25, 26] indicates as typical for the 1st mode.

3rd mode streamers look fairly similar in all liquids, however the faster ones in cyclohexane are bushier with longer side branches than the ones in the other liquids (figure 6). 4th mode streamers (figure 7) are also similar in the three liquids (Nytro 10XN, Marcol 52 and Primol 352) where satisfactory images were obtained and were not substantially different from the 3rd mode. Notice that the Diala S4 ZX1 image in figure 7 shows a 3rd mode streamer which has just switched to 4th mode in the form of a single luminous channel leaving the 3rd mode structure. A careful inspection shows that the channel is luminous all the way back to the point electrode but is partially hidden by the other channels.

At high voltage where there is no 2nd mode, 3rd mode streamers may propagate across most of the gap before switching to 4th mode, similar to what is seen for Diala S4 ZX1 in figure 7. Positive 3rd mode streamers rarely grew much longer than the mid-gap point level before switching [1].

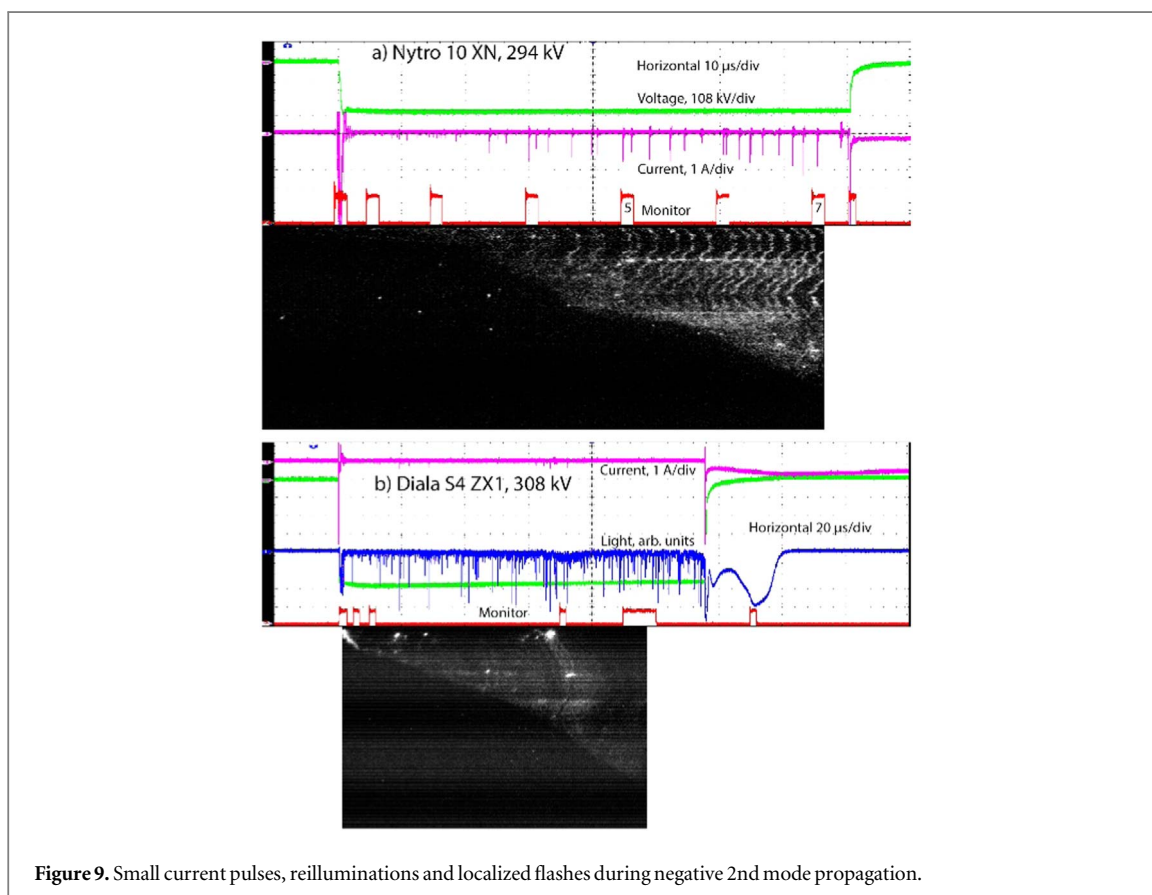
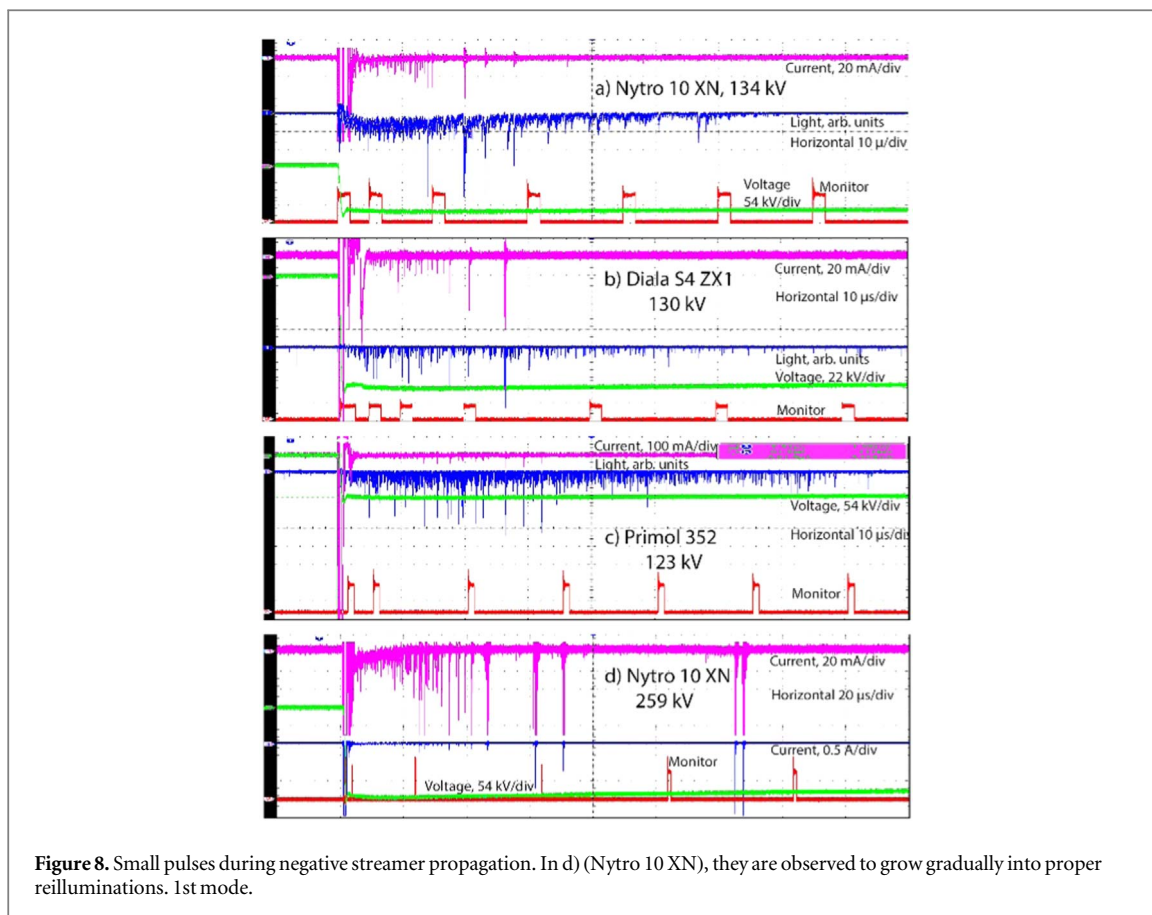


3.4. Small current pulses, reilluminations and localized flashes

As for positive streamers, pulsed phenomena in the form of light and current can be seen. Reilluminations were observed, i.e. brief light flashes accompanied by current pulses, where the duration of the current pulse is typically 10–15 ns and the amplitude ranges from approximately 0.1 A up to several amperes. Small pulses were also present in great numbers, in most cases matching in light and current, with current amplitudes ranging from about 10 mA and up to 0.1 A, as shown for 1st mode streamers in figure 8. The light flash of a reillumination is in most cases emitted from the entire length of a channel, as seen for 2nd mode streamers in Nytro 10XN in the streak image of figure 9(a) and for 1st mode in Marcol 52 in figure 10(c). Several channels may reilluminate, but their reilluminations are usually separated in time with this separation being sometimes down to 10 ns. The camera has too low sensitivity to detect the small pulses, therefore it was impossible to pinpoint their location. It is plausible they are weak reilluminations since the light and current amplitude ranges seem to grow seamlessly into the ranges of ‘proper’ reilluminations that can be observed with the camera.

No significant differences were found in the frequency and amplitude of small current pulses and reilluminations between the oils. The behavior could be rather erratic even for one oil: In figure 9(a) Nytro 10XN shows numerous reilluminations in 2nd mode, while in figure 10(a) none are seen during 2nd mode and only a few during 3rd mode. Generally, there were small pulses and/or reilluminations in 1st, 2nd and 3rd modes, with higher voltages favouring more reilluminations and fewer small pulses.

For negative streamers localized flashes could be observed, which was not observed for positive polarity. The streak image of figure 9(a) shows spots being particularly bright during reilluminations, and in figures 9(b) and 10(c)



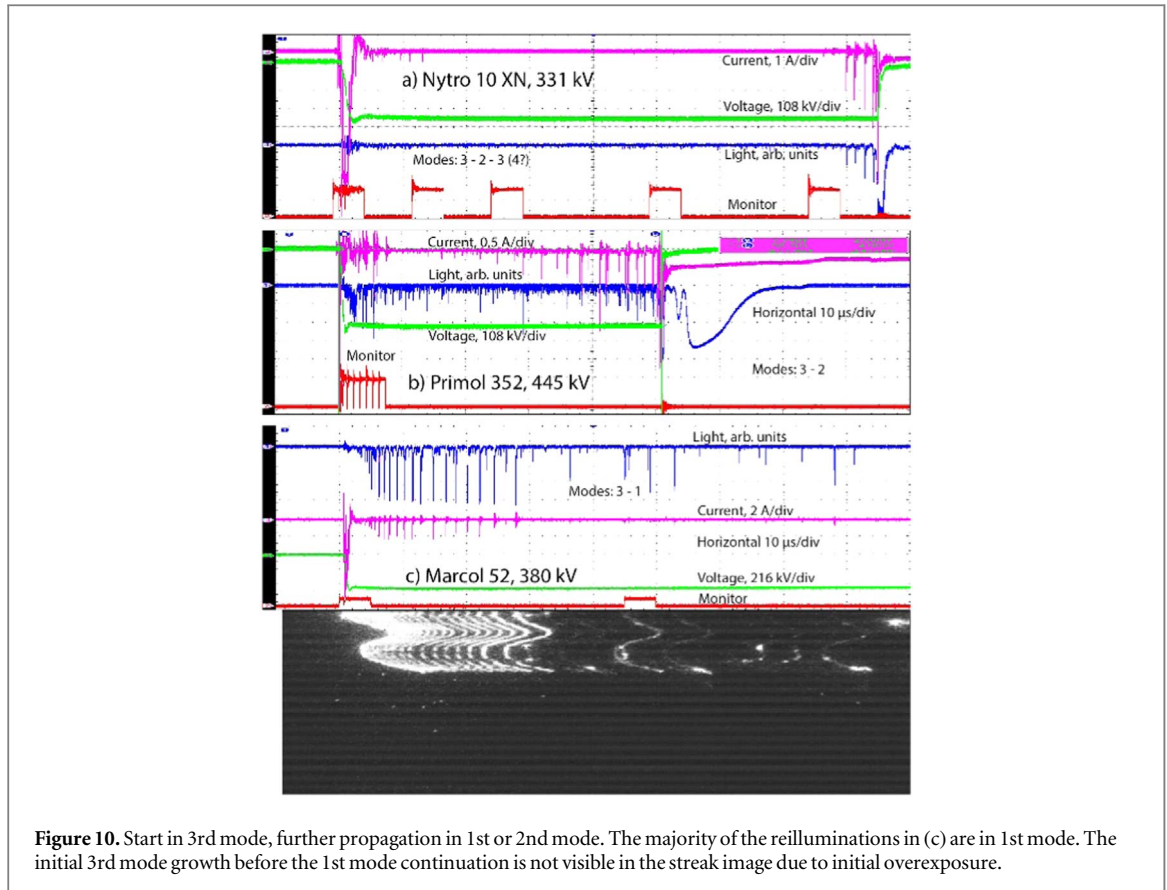


Figure 10. Start in 3rd mode, further propagation in 1st or 2nd mode. The majority of the reilluminations in (c) are in 1st mode. The initial 3rd mode growth before the 1st mode continuation is not visible in the streak image due to initial overexposure.

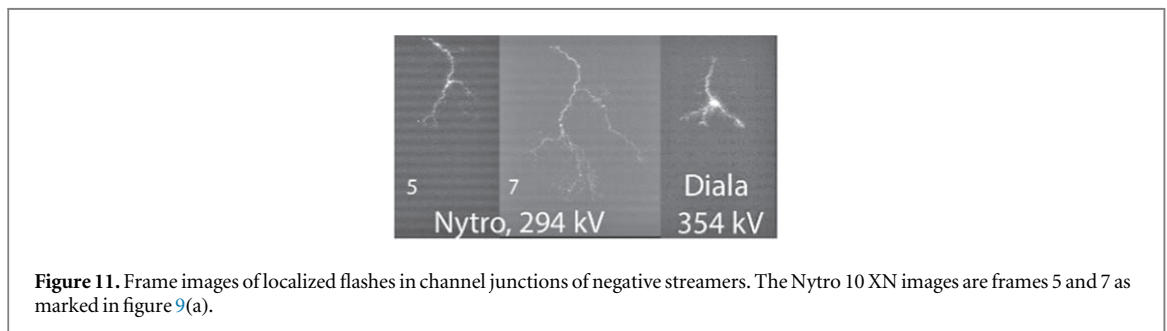


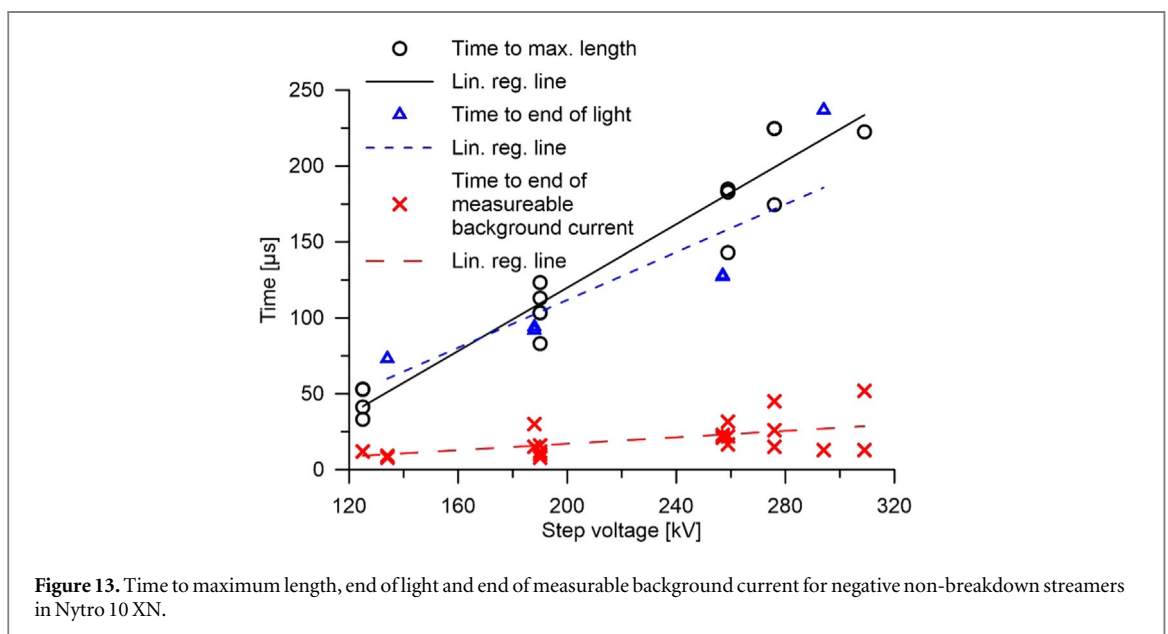
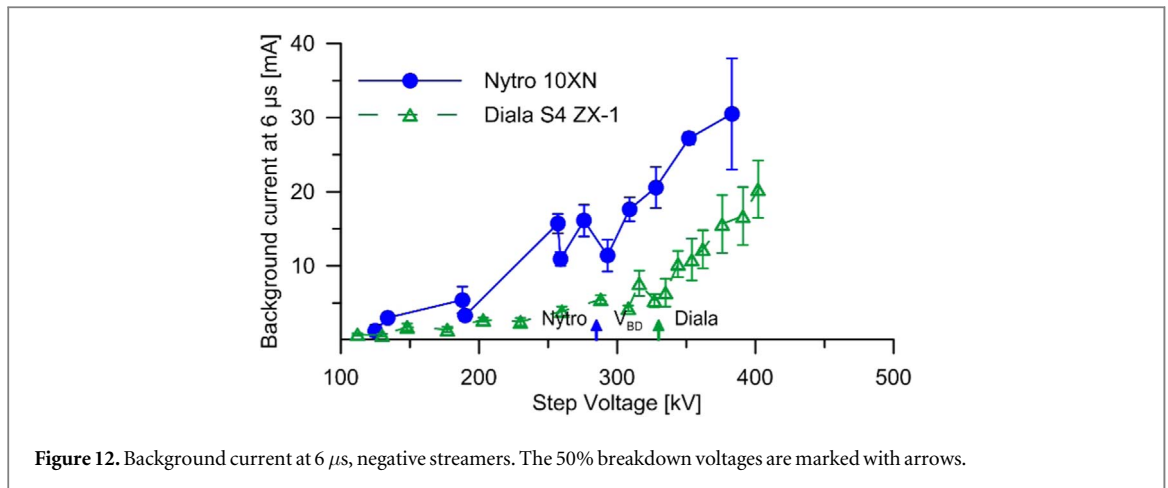
Figure 11. Frame images of localized flashes in channel junctions of negative streamers. The Nytro 10 XN images are frames 5 and 7 as marked in figure 9(a).

there are spots that are much brighter than the weak reilluminations that could have appeared simultaneously. These bright spots predominantly appear in already existing branching junctions as shown in figure 11.

At voltages above 250–300 kV often a rather luminous pulse is observed at the beginning in Diala S4 ZX1 and Primol 352, usually lasting less than 1–2 μs (figure 9(b)), however in some cases a little longer (figure 10(b)). This is not a reillumination. It could happen when the streamer started in 3rd mode. In the streak image of figure 9(b) the fast, luminous start can be observed.

3.5. Background current

The oscilloscope current traces showing the reilluminations may give the impression that no current is present except during reilluminations. Nonetheless, a small background current can be registered if the sensitivity is set high enough. The current generally decreases with time for non-breakdown streamers. This decrease is not observed for breakdown streamers, and current may often increase abruptly just before breakdown. The capacitive current pulse during the impulse front overloads the oscilloscope at the high sensitivity setting making detection of the smaller currents impossible. Therefore, comparative measurements of the current were done at 6 μs , at which time the oscilloscope had recovered. Measurements were conducted only for Nytro 10XN and Diala S4 ZX1 and are shown in figure 12. The current magnitude is much lower in Diala S4 ZX1 than in Nytro 10XN, reflecting the overall lower velocity of streamers in Diala S4 ZX1 than in Nytro 10XN (figure 2).



For non-breakdown streamers in Nytro 10 XN, the duration of light and the duration of the background current at negative polarity was measured (figure 13). While light emission lasts approximately for the entire duration of streamer propagation, measurable background current disappears very early on. This differs from positive streamers where there was background current during the entire propagation and light lasted a little longer.

3.6. Decay of non-breakdown streamer channels.

All non-breakdown streamers appeared to experience channel collapse at random locations near the end of their life (figure 14), but nonetheless they continued propagating (83–113 μ s). The apparent channel collapse proved to be not complete. The thick negative channel collapsed to a thin channel resembling positive streamer channels. Eventually the streamer stopped growing and gradually disappeared. Non-breakdown streamers were mainly of 1st mode. Streamers propagating for any significant length in higher modes did usually lead to breakdown and did not show this kind of collapse.

Even 1st mode breakdown streamers could show this apparent collapse, as shown in figure 15. In a) some collapse can be seen already at 103 μ s. At 183 μ s the left main channel which has previously apparently collapsed has become much more visible. It has been ‘rejuvenated’ by a reillumination at 148 μ s, i.e. 5 μ s after the third frame shown. Such late-stage reilluminations in long 1st mode streamers are rare, and in b) no ‘rejuvenation’ by reillumination has taken place. Nonetheless the streamer has propagated for more than 160 μ s after showing early signs of collapse at 94 μ s, and it continued up to breakdown.

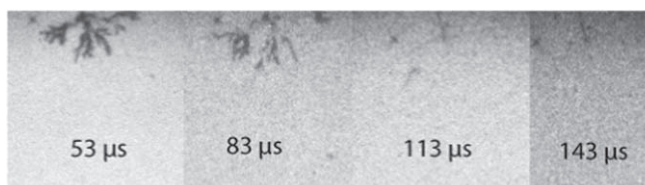


Figure 14. Collapsing non-breakdown negative streamer continues growing. Nytro 10 XN, 190 kV. Only 30 mm of the 80 mm long gap is shown.

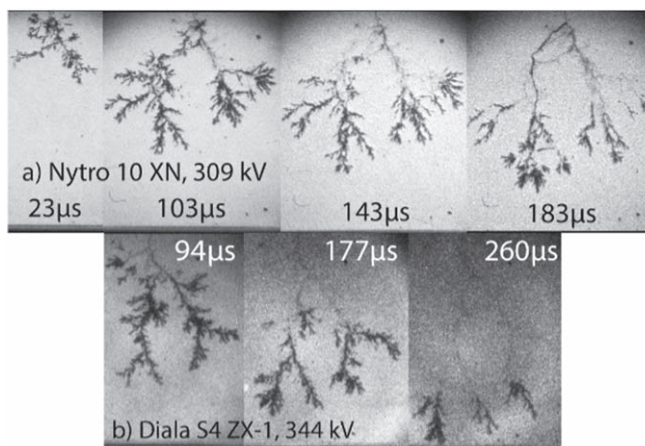


Figure 15. Apparent collapse of breakdown streamers' channels. Time to breakdown (a) 274 μ s and (b) 282 μ s.

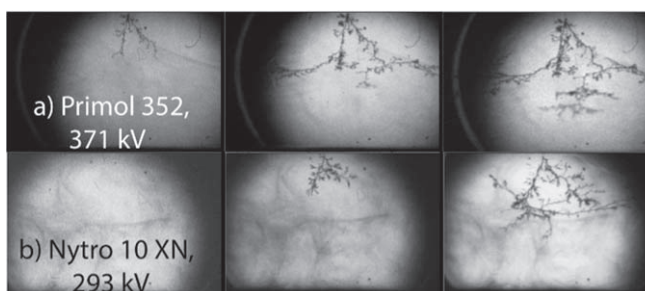


Figure 16. Negative breakdown streamers following soot clouds.

3.7. Effect of soot clouds

Working with Primol 352 with high viscosity and hoping to clean the oil after every 10 breakdowns, it was discovered that breakdown streamers, typically 3rd mode, would follow the non-dispersed soot clouds left in the stagnant oil in the gap by a previous breakdown. Therefore, it became obvious that filtering was necessary after each breakdown. However, because of the oil's high viscosity, filtering was inefficient, and even the thin soot streaks remaining after one hour of filtering were enough to deflect the streamer (figure 16(a)). Initially it was believed that this occurred because the soot particles would not disperse in the high viscosity liquid, however, in experiments that followed, the same problem appeared in the rather low-viscosity Nytro 10XN (figure 16(b)).

Because soot clouds would usually cause the streamer to follow a very long and tortuous path, time to breakdown in these experiments were longer with than without soot clouds.

This is in stark contrast to positive streamers, which showed no sign of following soot clouds [1], although as mentioned in chapter 2.3, it has been shown that carbon particles in the oil may shift onset, breakdown and acceleration voltage even for positive streamers [7, 8].

Table 2. Breakdown voltage and onset voltages for propagation of various streamer modes, in kV.

	Cyclohexane		NYTRO		Diala S4 ZX1		Marcol 52		Primol 352	
	Pos	Neg	Pos	Neg	Pos	Neg	Pos	Neg	Pos	Neg
	V_2	51	166	43	190	56	203	31	330	48
V_3	95	211	171	259	205	308	170	330	102	319
V_{BD}	118	180	153	285	139	330	162	389	169	323
V_{BD-}/V_{BD+}	1.5		1.9		2.4		2.4		1.9	
V_A	122	140	291	331	205	308	210	380	236	320
V_{A-}/V_{A+}	1.1		1.1		1.5		1.8		1.4	
V_A/V_{BD}	1.0	0.8	1.9	1.2	1.5	0.9	1.3	1.0	1.4	1.0
V_A/V_3	1.3	0.7	1.7	1.3	1.0	1.0	1.2	1.2	2.3	1.0
V_3/V_2	1.9	1.3	4.0	1.4	3.7	1.5	5.5	1.0	2.1	1.5

Table 3. Typical velocities for various propagation modes, in km/s.

	Cyclohexane		NYTRO		Diala S4 ZX1		Marcol 52		Primol 352	
	Pos	Neg	Pos	Neg	Pos	Neg	Pos	Neg	Pos	Neg
	Velocity ₁		0.1		0.13		0.11		0.09	
Velocity ₂	1.3	2.6	2	1	2.1	0.6	1.7	0.9	1.3	0.9
Velocity ₃	25	12	11	6	15	7	13	4	9	9
Velocity ₄	178	28	225	43	185	45	235	51	80	30
Velocity ₄ /Velocity ₂	137	11	113	43	88	75	138	57	62	33
Velocity ₄₊ /Velocity ₄₋	6.4		5.2		4.1		4.6		2.7	
Velocity ₃₊ /Velocity ₃₋	2.1		1.8		2.1		3.3		1.0	
Velocity ₂₊ /Velocity ₂₋	0.5		2.0		3.5		1.9		1.4	

4. Discussion and comparison with positive streamers

4.1. Cyclohexane as a model oil

Like for positive streamers, cyclohexane is an outlier in the study. It has by far the lowest negative breakdown and acceleration voltages of all the liquids tested, probably because its low boiling point of 80.7 °C makes forming and maintaining channels easy. It is used in studies because having only two types of atom-atom bonds makes it convenient to compare the effect of additives with specific properties. Neither electronegative nor low ionization potential additives improve its negative breakdown and acceleration voltages [22]. It is by no means a typical model for oils, although varying additives in it can indicate what molecular properties cause the streamer properties of various oils.

4.2. Streamer shape

While for positive streamers the shape of the different modes varied with the oil [1], there is hardly any oil-dependence for modes' shape for negative streamers. What has been observed for the negative ones is that when switching from 2nd to 3rd mode streamers the channels became more filamentary and thinner. This may, at least partly, be explained from effects of internal pressure in the streamer channel relative to propagation velocity: slow growth will allow more time for lateral expansion. The speed increased a factor 5–10 with this mode shift.

4.3. Streamer velocity, breakdown and acceleration

Table 2 shows the voltages for the appearance of streamer modes, breakdown and acceleration and relations between them. Table 3 shows *typical* velocities of the observed modes. Figure 3 (which shows *maximum* mode velocities) shows that for a given liquid, the velocity of a given mode can have a large variation, and what has been selected as 'typical' in table 3 is roughly the average velocity of that mode at a voltage in the middle of that mode's existence range. The same approach was used for the positive mode velocities. Italics is used in table 3 when the 'typical' velocity is less representative.

For negative polarity the streamer velocity at low voltages dominated by 1st mode is quite similar in all liquids. The faster 2nd and 3rd modes have up to a factor of 5 variation in 'typical' velocities although the velocity

ranges in the different oils are large causing a considerable overlap between the oils, particularly for 3rd mode (figure 3). Acceleration occurs at a voltage very close to breakdown in all the liquids. 4th mode velocity is low in the large-molecular Primol 352.

Except for the outlier cyclohexane, Nytro 10XN exhibits the longest stopping length and the lowest breakdown and acceleration voltages. This is because of electronegative constituents present [6, 7, 22]. The polyaromatics of Nytro 10XN have an electronegative effect as well as low ionization potential [26]. However, an electronegative additive in Marcol 52 [6] and Exxsol D140 [7] did also cause 1st mode to disappear and be replaced by the almost ten times faster 2nd mode, however the 1st mode is very much present in Nytro 10 XN and it is not noticeably faster than in the other oils examined. The low ionization potential of the polyaromatics is not expected to have any significant effect on the negative streamers, since a low ionization potential additive had little to no effect on negative streamers in Marcol 52 [6] and Exxsol D140 [7].

The breakdown voltage of negative streamers is twice that of positive streamers [1], and stopping lengths at a given voltage are shorter. This is similar to what was earlier found for Nytro 10X [4], however is contrary to another study on a mineral oil (Voltesso 35) which found roughly the same breakdown voltage in both polarities [21]. The order of the negative breakdown voltages for different liquids, except cyclohexane, is also distorted compared to positive streamers. In particular, Nytro 10XN had one of the highest V_{BD} s for positive streamers however has the second lowest one for negative ones, only surpassed by cyclohexane, while Diala S4 ZX1 exhibited the opposite tendency.

For positive streamers, the large molecules of Primol 352 appeared to slow down the streamers considerably, and they were much slower and shorter than in the chemically similar Marcol 52 consisting of smaller molecules [1]. It was assumed that this was because the processes in the head could not supply enough energy per time unit to evaporate the large-molecular oil in the channel formation process. No such effect was observed for negative streamers which are faster in Primol 352 than in Marcol 52 except for the 4th mode.

In general, the overall velocity of positive streamers and the velocities of the observed individual modes were higher than the corresponding velocities of negative streamers. The velocities typically increased by a factor of 2 from negative to positive for the 2nd and 3rd mode and by a factor of up to 5 for the 4th mode. In Primol 352 these differences were much reduced or non-existent. There was considerable overlap of the 3rd mode velocity ranges between the two polarities. Like for V_{BD} , V_A for the different liquids was also different for the positive and negative case. V_A was almost the same in both polarities for Nytro 10XN and cyclohexane, while for the others it was 50%–90% higher for negative than for positive streamers. In addition, velocity at voltages just below acceleration is typically $150\text{--}250\text{ m s}^{-1}$ for negative streamers but $1.5\text{--}2.5\text{ km s}^{-1}$ for the positive ones.

4.4. Reilluminations

As for reilluminations in positive streamer channels, there is a large current running for a short time. The head of the streamer leaves behind a substantial amount of positive charge which the channel has not been sufficiently conductive to neutralize (by electrons from the point electrode). 1st and 2nd mode streamer channels are often not very conductive, which has been documented for 2nd mode positive streamers [27]. Eventually the charge build-up causes the field along the channel to become large enough for a breakdown along the channel to occur, with conductivity increasing considerably to neutralize the built-up charge in a mere 10–15 ns with electrons from the point electrode. It is unlikely that the current can be caused by moving the comparatively heavy positive ions to the negative point electrode in such a short time. The enhanced conductivity persists after a reillumination, again documented for positive 2nd mode streamers reaching the plane electrode [27]: at 50% breakdown voltage, they will cause a breakdown, but only if there has been a reillumination shortly before it reaches the plane.

Positive charge built up in the channel near the streamer head increases the average field between that charge and the point electrode and therefore the field in front of the head decreases. This decrease does not become large enough to halt the propagation of the head though.

On average, negative reilluminations of a particular mode have smaller current amplitudes than the corresponding positive reilluminations in Part 1 [1].

The small pulses, probably weak reilluminations, with current amplitudes ranging from a few milliamps to 100–200 mA and increasing with increasing voltage were not seen in positive streamers [1] except for occasional single pulses in some oils. Such pulses have the same duration as ‘proper’ reilluminations. These small current pulses correspond to weak light pulses. Because there is a current when these pulses occur, they must represent charge redistribution in the gap like for ‘proper’ reilluminations. Lacking photographic evidence, it is not known if this happens along entire channels or just in parts of them. The small charge build-up which happens here too, must be able to induce some kind of breakdown along the channel, and thus be neutralized with a small current pulse.

As liquid breakdown has borrowed the term ‘streamer’ from gas discharge physics, the term ‘leader’ was also used for high speed and luminous channel phenomena [9]. The frequent reilluminations seen particularly in refined mineral transformer oils containing some aromatics are reminiscent of the light flashes in ‘stepped leaders’ in long air gaps, where the elongation of the channel happens in a sudden light flash. However, the analogy is far from perfect. In most of the liquids used in this study, reilluminations are rare in positive 2nd mode and in much of the propagation of negative 1st or 2nd mode. Even in Nytro 10XN, reilluminations in the shorter positive non-breakdown streamers well below breakdown voltage are too infrequent (or even nonexistent) to be the main channel elongation mechanism. Only in 3rd mode, in both polarities, do reilluminations seem to be ubiquitous in all the oils and the propagation may be compared to that of ‘stepped leaders’.

4.5. Background current

The background current can be viewed in two different ways: the ‘electrotechnical’ capacitance view where the streamer front is considered to be a capacitance surface moving towards the plane electrode, which constitutes the opposite capacitance surface [28, 29], or the more physical view that the current comes from ionization events at the head of the streamer.

The background current is much higher in Nytro 10XN than in Diala S4 ZX1. There is little difference in the shape of the streamers in the two oils, therefore trying to explain the current difference based on differences in capacitance between the plane electrode and the conductive streamer structure is not possible. Streamers are much faster in Nytro 10XN than in Diala S4 ZX1 and in the capacitance view, this means that the capacitance increases more rapidly in Nytro 10 XN since the streamer structure approaches the plane electrode faster than in Diala S4 ZX1. In the ionization view it means that the ionization is larger in Nytro 10XN, permitting the streamers to grow faster, and with more charges necessarily moving per time unit, which means higher current.

Compared to positive streamers [1], negative streamers have much smaller background current at a given voltage. In the capacitance view, this can be explained by the velocity difference between negative and positive streamers. In addition, the difference in shape may also contribute. In both Nytro 10XN and Diala S4 ZX1, positive streamers will grow a fine branched almost hemispherical structure towards the plane electrode, and more so in Nytro 10XN than in Diala S4 ZX1. This structure has a larger apparent surface toward the plane than any negative streamer has and can either be viewed as being a larger capacitance surface moving toward the plane, or as a much larger number of streamer tips feeding current into the streamer structure by ionization.

4.6. Decay of non-breakdown streamers

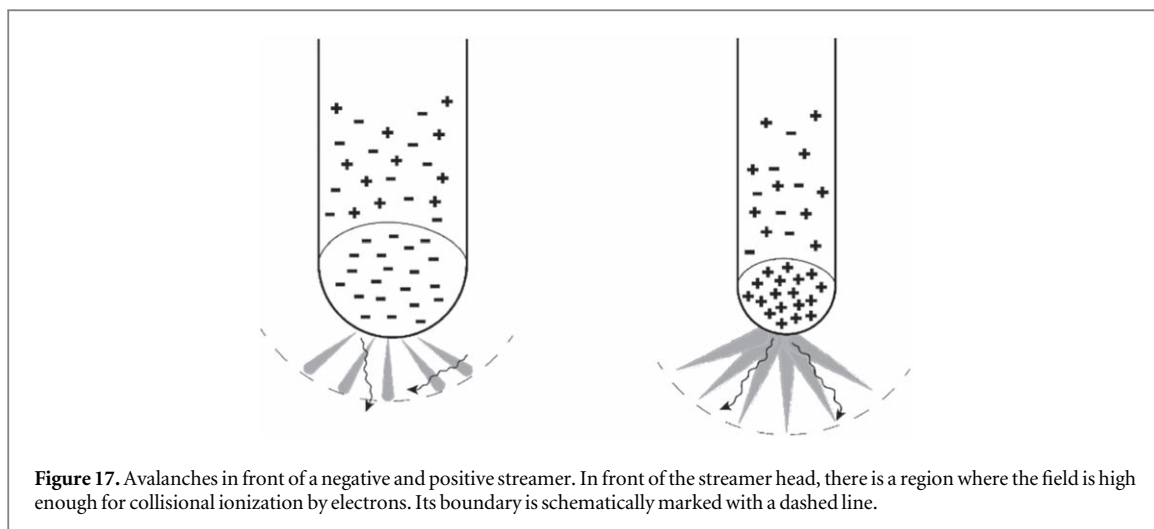
Negative non-breakdown streamers will eventually decay, but the virtual collapse of the channel at more or less random locations along the 1st mode streamers is not a true decay but some sort of channel collapse to a very much thinner channel, more resembling a positive 2nd mode channel than the thick negative channels. This channel is sufficiently conductive for the streamer to continue propagation towards the plane for some duration. At sufficiently high voltage propagation can continue for several hundred microseconds, long enough for the streamer to reach the plane and initiate breakdown. Unless the voltage is sufficiently high, the streamer will eventually die and disappear.

This apparent partial collapse was not seen for positive streamers [1], which were never of 1st mode because of the large diameter of the point electrode. Stopping streamers were always 2nd mode and their final collapse appeared to be initiated at the streamer tip and moving backwards towards the point.

Neither of these streamer decays seem to fit very well with the Rayleigh bubble collapse typically used to describe the collapse of 1st mode streamers in the literature, in short gap experiments of typically 5 mm gap length [30]. Neither the negative nor positive propagating channels seen in this study or in [1] look very much like the dented bubbles of those small scale experiments, though, except the negative 1st mode streamers in the early stages when they are up to 1 mm long.

4.7. General discussion

Even if many observations on streamer breakdown in liquids exist and physical models are proposed the driving mechanism for streamer propagation is not yet fully understood. For positive 1st mode streamers satisfactory models explaining initiation from conductive and electrohydrodynamic processes exist [31]. The results presented here and in the previous paper [1] are discussed based on the hypothesis that for the higher order propagation modes both for positive and for negative polarity the mechanism behind the propagation are electron avalanches in a ‘corona’ region in front of and forming the streamer tips. Some of the electron collisions in this region cause electron multiplication and other collisions excite molecules and thus supply sufficient energy to evaporate the liquid and create a plasma filled streamer channel. Simulations of positive streamers based on this mechanism give realistic results [32, 33]. Light from relaxation of some of the excitations can be a source for creating free seeding electrons by photoionization, generating a feed-forward mechanism [5, 17]. This



kind of feed-forward mechanism is necessary for propagating positive streamers but is not required for negative streamers since there is ample electron supply from the electrode via the channel. There are several observations that support this hypothesis:

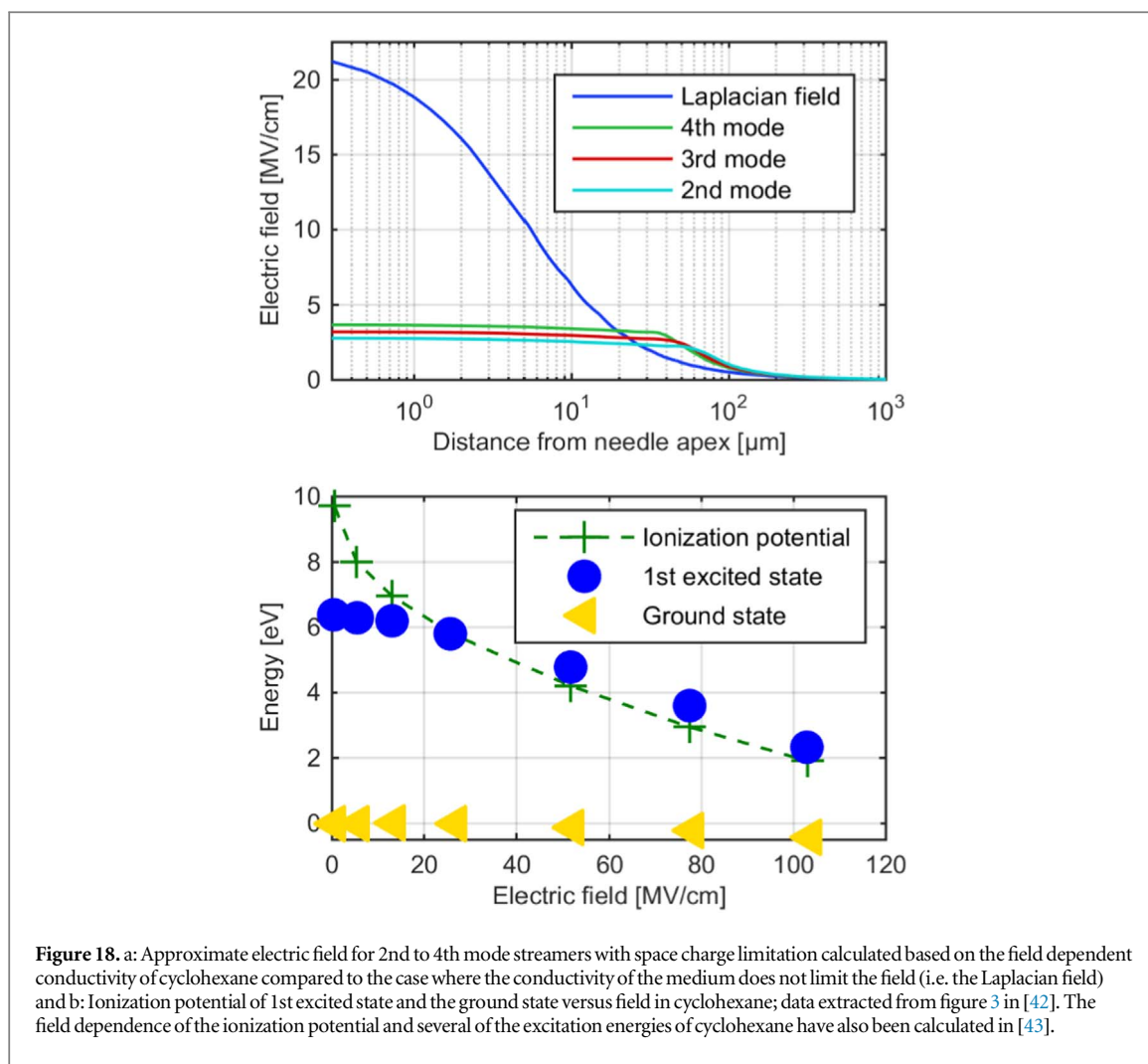
- For negative streamers it is shown that electron avalanches initiate 1st mode bubbles in pentane and Cyclohexane [20].
- It is documented that additives with specific electronic properties influence streamer propagation [6, 12, 26].
- Time delay of positive streamer initiation after voltage application is in line with expectations in the case of scarce seeding electrons [4, 34].
- Very high 4th mode speeds $>100 \text{ km s}^{-1}$ substantiate the presence of very fast processes as e.g. feed-forward photoionization.
- There is light emanating from streamer tips, and chemometric analysis indicates correlation between 1st excitation level and positive 4th mode speed [35].
- The fact that streamer propagation speed is not influenced by high pressure indicates that processes take place in the liquid phase [6, 36].

Regarding the importance of electron avalanches, there is the often used argument that since liquids are much denser than gases, the avalanche processes with electron multiplication in gases cannot be supported in a dense hydrocarbon due to the mean free path of electron travel being too short to cause new ionizations [37].

To be effective as a heating source for evaporating and converting liquid to a plasma channel the active region must have a field strength above which electron multiplication occurs. This region must have a size within which avalanches can grow to a sufficient size to deposit energy for liquid evaporation to occur. For positive streamers, avalanches starting in the liquid must therefore be initiated a minimum distance away from the streamer head. The longer this area within which avalanches may occur are, the faster the streamer will be. To get an efficient propagation one needs abundant electrons, high field strength on a large volume, and a process to allow the energy of electrons to be transferred to heat.

The main differences between positive and negative streamers are that the positive ones are faster, have a longer stopping length at a given voltage, are composed of thinner channels and that the mode shifts are more well-defined than for the negative ones. It has been proposed that electron avalanches directed away from the channel tip will be more diffusive than avalanches directed towards the tip as indicated in figure 17 [5]. Avalanches from negative streamers ought to start roughly at the outer end of the channel and multiply outwards, so that the majority of collisions take place some distance from the channel. Avalanches in positive streamers must start some distance away from the channel end and multiply towards the channel, having the majority of collisions taking place close to the channel. In negative streamers, heating will subsequently take place over a larger volume, resulting in negative streamer channels being orders of magnitude thicker than the positive ones. Produced heat per volume will be smaller and results in a lower velocity.

For positive streamers the seeding electrons have to come from the liquid. It is likely that at the high fields occurring at the tip of the electrode or in a filament the conductivity will be higher than in low field conditions [38, 39], leading to the formation of space charges and a space charge limitation regime in the vicinity of the tip. This



results in a reduced field however the field will stay constant over a longer distance depending on the time it took to establish the space charge. A rough estimate could be given from the relaxation time constant calculated from the permittivity and the high field conductivity of the liquid. Typically, the high field conductivity increases exponentially with electric field, thus a relatively modest change in the electric field may give a large change in the conductivity (and thus the time constant of the medium) [40]. Therefore, reducing the rise time by a factor of ten or even a hundred (representative of the transition from 2nd to 3rd/4th mode) would not result in a very large increase in the space charge limited field. Electric field simulations with voltage rise times of 1 ns, 0.1 ns and 0.01 ns shows that the electric field would be limited to around 4.5 MV/cm for 2nd mode and 6.5 MV cm^{-1} for 4th mode (see figure 18). The conductivity would have to increase by a factor of 100 to allow for this limitation of the field at the faster modes. While the high field conductivity for the 2nd mode streamers are in the range measured experimentally [38, 39], the higher conductivities at the higher modes cannot be verified based on existing experimental results. At these voltages the electrons will probably not have mobility high enough in the liquid phase to cause an efficient charge separation on the time scale required (an electron terminal velocity in hydrocarbons of approximately 41 km s^{-1} is suggested in [41]), or there may not be a sufficient amount of weakly bound electrons in the liquid to warrant the large increase in conductivity required to limit the field at these high speeds. In either case, the conductivity would no longer be able to limit the field in the high field region effectively, and the resulting field would lie somewhere between the Laplacian field and the calculated space charge limited field. Thus, fast phenomena may drastically alter the field distribution in the high field region, increasing the maximum field in this region by several orders of magnitude.

The source of seed electrons for initiating positive streamers could be provided by detachment of negative ions, field ionization of the base liquid or additives or photoionization from the light formed from relaxation of excited molecules. Ionization from light emission because of quantum chemical processes in the streamer region will act as a feed forward mechanism. Ionization can start new avalanches as long as the background field is above the critical level that is similar for positive and negative streamers.

It is documented that at high field stress the ionization potential of a molecule is reduced (figure 18(b)), therefore with increasing field, electron energy removal by excitation, relaxation and formation of metastables

are greatly diminished [42–44]. The field strength shown in figure 18(b) is the local field around the molecule that is higher than the macroscopic field of figure 18(a) by a factor of 2–10 depending on the type of molecules in the liquid [45]. This indicates that while the macroscopic field might not be high enough to significantly influence the ionization level or cause removal of excited states, the local field may very well be.

For avalanches to be able to heat and evaporate the liquid, the more concentrated the process that takes place are, the more efficient the process will be. A high electron affinity will support inelastic collisions. A high light absorption for energetic radiation may also help to concentrate ionization and start avalanches. A further factor will be the amount of energy needed to evaporate the liquid.

These are all processes that can take place in the region with space charge limited field present. However, if the space charge formation becomes unable to support the constant field region, there will be a significant increase in field strength, and other processes like direct field ionization can start to dominate. This could explain the mode shifts.

All tested liquids acted in a way as described above. Chemical differences and molecule size resulted in differences in acceleration voltages. It looks as if compounds as e.g. aromatics can explain the differences. Nytro 10XN, having more aromatics, has a high positive acceleration voltage compared to Diala S4 ZX1 and Marcol 52, and with the electronegative properties of aromatics it shows a lower acceleration voltage for negative voltage. However, it is impossible to draw more conclusions until more detailed characterization of the chemical composition is conducted.

Typical velocities for most of the 4th mode propagation in negative streamers is in the range 20–50 km s⁻¹, however the maximum negative 4th mode velocity is usually 50–80 km s⁻¹, and may occasionally even exceed 100 km s⁻¹ in some liquids, occurring when the streamer approaches the plane electrode. This is close to twice as fast as the *approximate* electron limiting velocity of 41 km s⁻¹ suggested in [41], however it is much slower than the maximum positive 4th mode velocity. If this value is considered as a very rough approximation, this may indicate that the negative 4th mode does not rely on any feed forward mechanisms like e.g. photoionization, and its velocity is therefore limited by the limiting electron velocity. This is different from the positive 4th mode case, where some kind of rapid feed forward mechanism is necessary for avalanche formation.

5. Conclusions

Velocities of streamers for negative polarity varies little between the tested oils, compared to what was seen for positive polarity. The propagation modes are less well-defined than for positive streamers and are generally thicker and slower. Branching is also different from the positive ones.

The observed differences between the various mineral oils in breakdown voltage, acceleration voltage and 2nd mode velocity for both positive and negative polarity can to some extent be explained by differences in concentration of aromatic molecules. Large oil molecules also have an effect, resulting in low 4th mode velocity for negative streamers while for positive streamers all observed modes (2nd–4th) were slower. Negative streamer shapes varied little between the tested oils, while there was considerable variation in positive streamer shape.

The behavior of higher mode positive and negative streamers is in line with what can be expected from a breakdown mechanism governed by electron avalanches and quantum chemical effects.

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