

Article

The Influence of Diethylaniline and Toluene on the Streamer Propagation in Cyclohexane between a Point-Plane Gap under Positive Impulse Voltage Stress

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Abstract: Liquid insulation is used in high voltage equipment such as power transformers as both dielectric medium and coolant. Breakdown in liquid insulation tends to be governed either by streamer initiation under more uniform fields, or by streamer propagation under more non-uniform fields. A model streamer propagation study, which screens the effectiveness of additives based on cyclohexane and mixtures with diethylaniline (DEA) and toluene, is presented in this paper. The effect of additives, at different concentrations, on streamer propagation velocity in cyclohexane under an applied lightning impulse voltage of positive polarity is studied. Cyclohexane (ionisation potential 9.88 eV) was chosen because, being a hydrocarbon, it shares similarities with the constituents of common insulating liquids. Previous studies have also shown how, in general, the addition of additives of lower ionization potential than the bulk liquid can slow down streamer propagation in insulating liquids. A point-plane electrode configuration of 70 mm gap with a 5 μm tip radius is used and subjected to an applied positive polarity impulse of 1.2/50 μs . A high velocity imaging system is also used to capture streamer images to validate a Time-To-Breakdown (TTB) measurement approach used in inferring approximate streamer velocity. The DEA (ionisation potential 6.98 eV) was found to be an effective additive to slow down positive polarity streamers in cyclohexane in the applied voltage range (≈ 220 – 280 kV peak) in concentrations above approximately 0.33% (by volume). Toluene (ionisation potential 8.82 eV) was found not to significantly slow down streamers in cyclohexane, even at 10% concentration, for the same voltage range. This is postulated to be due to the fact that toluene does not have a low enough ionisation potential (with respect to that of the cyclohexane) to change the streamer branching characteristics sufficiently during propagation.

Keywords: streamer; propagation; cyclohexane; additives; high-speed imaging



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1. Introduction

Insulating liquids are used in liquid-immersed electric equipment such as power transformers [1] where the liquid is both the insulation and cooling medium. The most common types of insulating liquid used in such equipment are “mineral oils” which refer to a broad a range of complex hydrocarbons that are predominantly based on hydrotreated crude oil distillates [2]. Specifications of mineral insulating oils are in standards such as IEC 60296 [3] and ASTM D3487 [4]. Other hydrocarbon-based insulating liquids, such as Gas-To-Liquid (GTL)-based liquids as well as bio-based hydrocarbons, are also used. Ester-based insulating liquids such as synthetic esters and natural esters are specified in standards such as IEC 61099 [5] (synthetic esters) and IEC 62770 [6] (natural esters). An important

characteristic of an insulating liquid is the ability to suppress streamer initiation and/or slow down propagation under divergent electric field stresses. Further improvement in liquid insulation technology, to effectively slow down streamer propagation, as well as suppressing streamer initiation, is an ongoing research endeavour. The common approach in such studies often uses pure liquids such as cyclohexane as a representative model base and as a host to evaluating additive behaviour [7–10]. The use of model and pure liquids (such as cyclohexane) makes it possible to more precisely control the variables that influence the streamer propagation speed. The fundamental knowledge generated from this approach can then be transferred to the engineering of more appropriately viable insulation liquids.

In the present work, the concept of slowing down streamer propagation velocity is explored by mixing a base liquid (cyclohexane) with additives of lower ionisation potential than that of base liquid. In the following paragraphs, the key aspects around streamer phenomena in liquids are described to provide contextualisation.

The intrinsic dielectric behaviour of an insulating liquid can be characterised through pre-breakdown and streamer studies. An important generalisation from earlier research in this field, such as that by Lesaint [9–16] and others [17–29], is the concept of classifying streamers into “modes”. The pre-breakdown phenomena can be characterized by significant changes in the average velocity of the streamer. Whilst several factors (such as the electric field distribution, polarity, wave-shape, and duration of the applied voltage) may have influence, the streamer propagation modes are also dependent on the liquid chemistry. The concept of streamer modes, originally proposed by Hebner [30] and Lesaint [11], is based on categorising the propagation velocity into four modes. This classification of streamer modes is adopted in the present work, where V_{avg} is the average streamer propagation velocity.

- i. 1st Mode, V_{avg} in the order of 0.1 mm/ μ s.
- ii. 2nd Mode, V_{avg} in the order of 2 mm/ μ s.
- iii. 3rd Mode, V_{avg} in the order of 10 mm/ μ s.
- iv. 4th Mode, V_{avg} in the order of 100 mm/ μ s.

Dielectric breakdown processes are primarily related to ionisation phenomena, driven by electric field effects. Therefore, the electric field distribution is an important factor influencing the streamer breakdown phenomena in liquid dielectrics. The Schwaiger factor (η) [2,31] is a useful expression to characterize the degree of homogeneity of an electric field distribution described by (1).

$$\eta = \frac{E_{mean}}{E_{max}} \quad (1)$$

where η is the Schwaiger Factor, E_{mean} is the mean electric field strength in kV/mm, and E_{max} is the highest electric field strength in the same region where the mean was calculated, in kV/mm. In uniform and quasi-uniform electric field distributions, once initiated, streamers typically lead to a complete breakdown whereas, in non-uniform electric field distributions, an initiated streamer may not necessarily lead to breakdown, as the nature of the propagation becomes a dominating factor. Consequently, streamer breakdown mechanisms in liquids can be classified into two main categories that are dependent on the electric field distribution (described by the Schwaiger Factor, η).

Initiation governed breakdown is expected when the electric field distribution is fairly uniform (for example $\eta > 0.5$). This is a situation where the electric field stress required to initiate the streamer is higher than that to ensure the streamer propagates in the gap. In such a situation, once initiated, the streamer typically leads to complete breakdown [2,11,32].

Propagation governed breakdown is expected when the electric field distribution is significantly non-uniform (for example $\eta < 0.1$). In this situation, the electric field stress required to initiate the streamer is much lower than that for it propagate to breakdown [11]. Therefore, factors such as the liquid chemistry, which determines the propagation in the electrode gap, become more influential [2,11,16,33].

In liquid immersed equipment, there may be situations where non-uniform fields exist (for example, $\eta < 0.1$). In such cases propagation governed breakdown can occur and the expected streamer velocity, for the applicable voltage range, has design implications. There

is therefore a relationship between breakdown probability and the duration of the overvoltage stress [2]. The understanding of which factors influence the propagation velocity in insulation liquids is thus essential. In this regard, in the present work using model additives, the concept of slowing down streamer propagation velocity using additives is explored.

In laboratory-based experimental work, to achieve a suitable non-uniform electric field distribution (with a η less than 0.1) for streamer propagation velocity studies, point-plane electrode geometries are used [34]. The precise value of η depends on the specific electrode and gap dimensions.

The streamer propagation velocity under a given applied voltage range has been shown to differ substantially between liquids with different chemical properties, and notably is sensitive to the presence of additives [9,12,18,33,35]. Differences in streamer propagation velocity between differing liquids are normally evaluated using point-plane studies using impulse test voltages. A particular phenomenon observed in insulating liquids is that there is usually a threshold for the applied voltage after which the streamer propagation velocity “accelerates”. This is a phenomenon that is particularly distinct for positive polarity.

It has been demonstrated that additives can be used to change the streamer propagation velocity and acceleration threshold of base liquids, including cyclohexane, medical white oils, and ester liquids [8,9,35,36]. It has also been found that if the additive has an ionisation potential (IP) lower than that of the base liquid, a streamer velocity slowing effect can be expected [36]. The postulation is that additives of lower IP values slow down the streamer by increasing the degree of streamer branching. This increased branching leads to shielding effects on the neighbouring tips, and consequently an overall reduction of the field stress at the streamer’s tips [11,35].

Furthermore, in addition to the IP value of the additive, it has been shown that the relationship between the different excitation states of the additive and the base liquid is likely a key factor in streamer branching mechanisms [37]. The concentration of the additive also has an impact [9]. Since various aspects of an additive are expected to effect streamer behaviour in different ways, the modelling of streamer propagation is highly complex [37,38]. Therefore, establishing laboratory test procedures that use model base liquids and additives allows comparison of the streamer propagation behaviour with practical insulation liquids.

In the present work, an experimental screening method to evaluate the possibility of slowing down positive polarity streamers in cyclohexane using diethylaniline (DEA) and toluene has been undertaken.

Examples of additives that have been shown to successfully slow down and increase the acceleration voltage threshold of streamers in cyclohexane include dimethylaniline (DMA) and pyrene [8,9,35]. In the present work, DEA was first evaluated since it has a similar chemical structure to DMA and was readily available. Both DMA and DEA have significantly lower IP values than cyclohexane and because DMA has been shown to be effective in the literature, there is a high likelihood of DEA also being effective. The other additive candidate considered in the current work, toluene, is chosen, as it is an aromatic hydrocarbon (resembling those aromatics contained in mineral oils [2]), is readily available, and also has a lower IP than cyclohexane (see Section 2.2). DEA is used at an initial concentration of 0.9% *v/v* based on the relative molar concentration of DMA used in other studies [11]. The nominal concentration for toluene was chosen to be 1% *v/v*.

As the purpose of the investigation is to observe whether there is a streamer velocity slowing effect caused by the additive, the base liquid (in this case, cyclohexane) must be stressed at a voltage level, in a non-uniform field distribution, which is above its original acceleration threshold. This will allow a comparison of the effect of the additive relative to the base liquid under practically equivalent electric field stress.

The rest of this paper is structured in the following way. Section 2 deals with the materials and methods, Section 3 presents the results, Section 4 provides the discussion, and Section 5 is the conclusion.

2. Materials and Methods

2.1. Test Setup and Test Cell Design

A point-plane electrode gap was used to study the streamer propagation velocity of liquids under a non-uniform electric field distribution [2]. A Perspex test cell, inspired by the designs stated in IEC 60897 [39] and IEC TR 61294 [40], was designed and constructed. The cell volume was 900 mL and housed the point-plane gap as depicted in Figure 1. Perspex was chosen due to its durability while also having good chemical resistance (to facilitate solvent cleaning) and to allow observation of the breakdown phenomena. The point electrode was a needle from OGURA Jewel Industry, Tokyo, Japan, Type X-253-4, of 60 mm long, with a tungsten tip of radius of $5 \pm 1 \mu\text{m}$. The plane electrode was a brass disc with smoothed edges and a diameter of 70 mm. The liquid gap distance between the point tip and plane was set at 70 mm and falls within the range of other reported studies [8,9,16,18,19,35,36,41] ($\approx 50\text{--}150 \text{ mm}$). The tip radius of $5 \mu\text{m}$ is sharper than those reported in the abovementioned studies, but still allows the observation of both slow and fast streamers (i.e., between those of 2nd, 3rd, and 4th mode). The tip radii can, however, influence whether streamers will initiate as 1st or 2nd mode streamers [9,35]. The needle was not replaced after every measurement, raising the possibility of needle tip erosion due to repetitive streamer activity. However, the present work focusses on the effects of streamer propagation, and the voltage range is significantly above the inception voltage. The electric field at the streamer channel's tips is more influential to the streamer propagation speed than that at the needle tip [35]. Repeat experiments in the present work with a new needle further confirm the independence of results from possible erosion of the needle tip, presented in Appendix B.

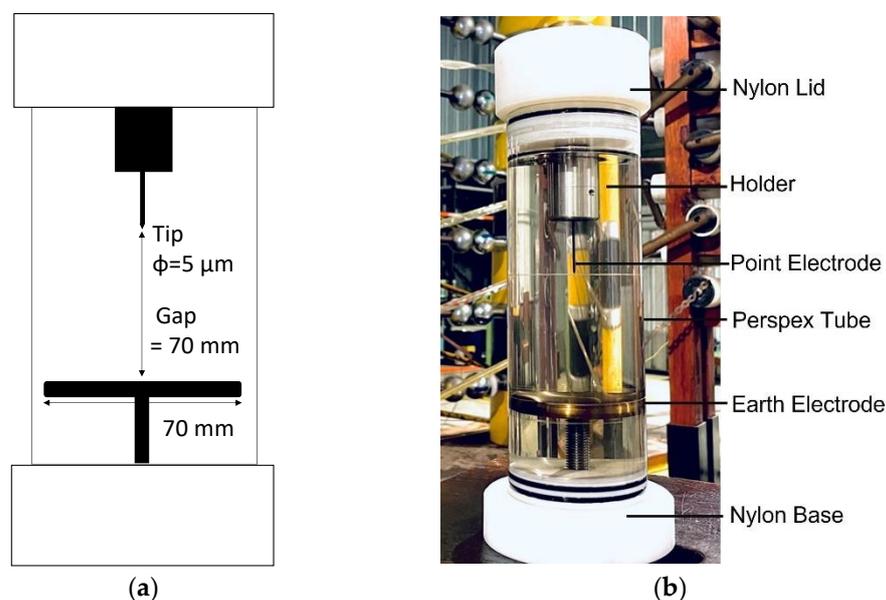


Figure 1. The Perspex test cell: (a) The electrode configuration is point-plane. The point electrode is 50 mm long, with a tungsten tip of radius $r = 5 \pm 1 \mu\text{m}$. The plane electrode is made of brass with a diameter of 70 mm and is earthed. The gap size is 70 mm.; (b) Image of test cell with annotations.

Efforts were made to minimize the ingress of moisture and external contamination of the liquids. These include use of a sealed test cell (with O-rings) and minimizing exposure time to the atmospheric air when filling the liquid.

The test cell length of 280 mm was able to withstand up to 290 kV before external flashover would start and therefore 290 kV (lightning impulse peak, positive polarity) is the upper voltage limit in the tests. Positive polarity impulses of $1.2/50 \mu\text{s}$ were applied to the test cell using a Marx generator and the waveforms are measured through a voltage divider and RIGOL (Suzhou, China) DS1064B digital oscilloscope.

2.2. Test Liquid Preparation and Experimental Procedure

The liquids tested in the study are cyclohexane, and mixtures of cyclohexane containing different concentrations of the two additives, toluene and DEA. Pure anhydrous cyclohexane of 99.5% purity from Sigma Aldrich (Merck Group, Johannesburg, South Africa) is used. All concentrations reported in the study are percentage volume by volume (% v/v). Tests were performed at room temperature (20 to 30 °C). Table 1 lists the IP values for the cyclohexane and additives.

Table 1. The liquids and additives used in the study. For reference the ionization potentials are also listed [42].

Substance	Ionization Potential [eV]
Cyclohexane	9.88
Toluene	8.82
Diethylaniline (DEA)	6.98

The cell was rinsed with cyclohexane before being filled with the liquid to be tested. After a settling time of 5 min the cell was inspected for any bubbles and the electrical connections to the cell were made. A single impulse was then applied, in the case of a withstand or breakdown the waveform was checked, recorded, and then a subsequent impulse shot could be applied after a brief settling period.

Liquids were used in their received state and were not filtered. Use of point-plane geometry establishes a strong field enhancement at the tip of the needle. Streamer initiation is therefore expected to be governed by the tip radius more than the presence of impurities. Linhjell and Nguyen [8,43] both report that the impact of whether a liquid is filtered or degassed on the acceleration voltage of a liquid is not as significant as the addition of additives (in point-plane geometry). Notwithstanding, as breakdowns occur in the test cell, carbon particles are created and increase with subsequent applied impulses. During the initial tests in the present work, the approach was to test a fixed number of impulse applications and see whether the time-to-breakdown changed significantly with subsequent impulse applications. Nguyen [35] reports that streamers of positive polarity are reported to be less sensitive to carbon particles than with negative polarity. However, some influence on positive polarity cannot be completely ruled out. Nguyen postulates that the reason that carbon particles have more impact on negative streamers is because the carbon particles can lead to further field enhancement at the streamer tips. For the case of positive streamers, however, the field enhancement caused by the streamer itself is understood to be sufficiently effective such that the carbon particles have little additional effect. Interestingly, it has been also reported that carbon contamination is likely to slightly reduce the positive acceleration voltage in hydrocarbon liquids, rather than increasing it [43].

Streamer breakdown in liquid insulation has time durations in the order of microseconds. Therefore, to infer the velocity of streamer propagation, accurate time measurements in microsecond resolution are necessary. In the present work, the incorporation of high-speed imaging was used to correlate observed events with the applied voltage waveform.

2.3. High Speed Imaging and Validation of the Time-to-Breakdown (TTB) Measurement Approach

The approach to inferring streamer velocity is based on analyzing the Time-To-Breakdown (TTB) determined from the applied time-domain voltage waveform [18,19,26]. The TTB is defined as the time elapsed, after application of the impulse, until breakdown occurs and is evidenced by a collapse of the voltage waveform.

A 12-bit depth monochrome Phantom (Vision Research, New Jersey, USA) v2012 high-speed camera was used. The Phantom Ultra-High Speed (UHS) series of cameras are designed for very high frame rate applications using a CMOS technology. A trade-off between resolution and frames per second is necessary, that is, the maximum frames per second will determine the resolution limit. The camera functions with an electronic

shutter that opens and closes after the exposure time duration. All physical processes that happen during this time will be shown in the frame. At the end of each exposure a sensor dead time is used to transfer information. The camera operates with a frame rate between 180,00 and 430,000 Frames Per Second (FPS) with exposure times between 4.28 and 1.75 μs , respectively, and time intervals between 5.5 and 2.32 μs , respectively. Image spatial resolution used varies from 128×256 pixels and 128×128 pixels, respectively.

Each event is captured using a camera feature that allows auto-trigger based on motion detection of the live image. The camera uses a Nikon 50 mm lens $f/1.8$ and is placed 2 m away from the cell. This f -number specification is chosen to allow a clear observation of the discharge (higher f -number lenses did not allow enough luminosity through them).

The images for 1% DEA and 1% toluene in cyclohexane, for similar applied voltage levels (230–233 kV), are shown in Figures 2 and 3, respectively. The streamer images shown for 1% DEA in cyclohexane (Figure 2, 230 kV applied voltage peak) represent a “slow” streamer. They are captured with a 4.82 μs exposure time, and 5.55 μs elapsed between frames. Frame (a) represents the first sign of activity from the captured images. Frame (h) represents the frame where the image is fully saturated, signifying the point where breakdown occurred; there is energy follow-through after the breakdown channel is made. The calculated elapsed time between Frame (a) and (h) was 38.85 μs , while the time calculated using the TTB approach (for slow streamers, peak to knee point) was a value of 39 μs . There is therefore reasonable agreement between the TTB approach and the propagation images of the streamer. However, there is some uncertainty derived from the exposure time of the image. A good illustration of this (in Figure 2) is Frame (g), where the breakdown event of Frame (h) is partly visible in the previous frame. In Frame (g), the start of the intense current due to breakdown is registered at the bottom of the image. Frame (h) captured the moment of maximum luminosity of the event and all image pixels saturate. In this instance, the streamers are predominantly of 2nd mode as the overall velocity is inferred to be 1.79 mm/ μs .

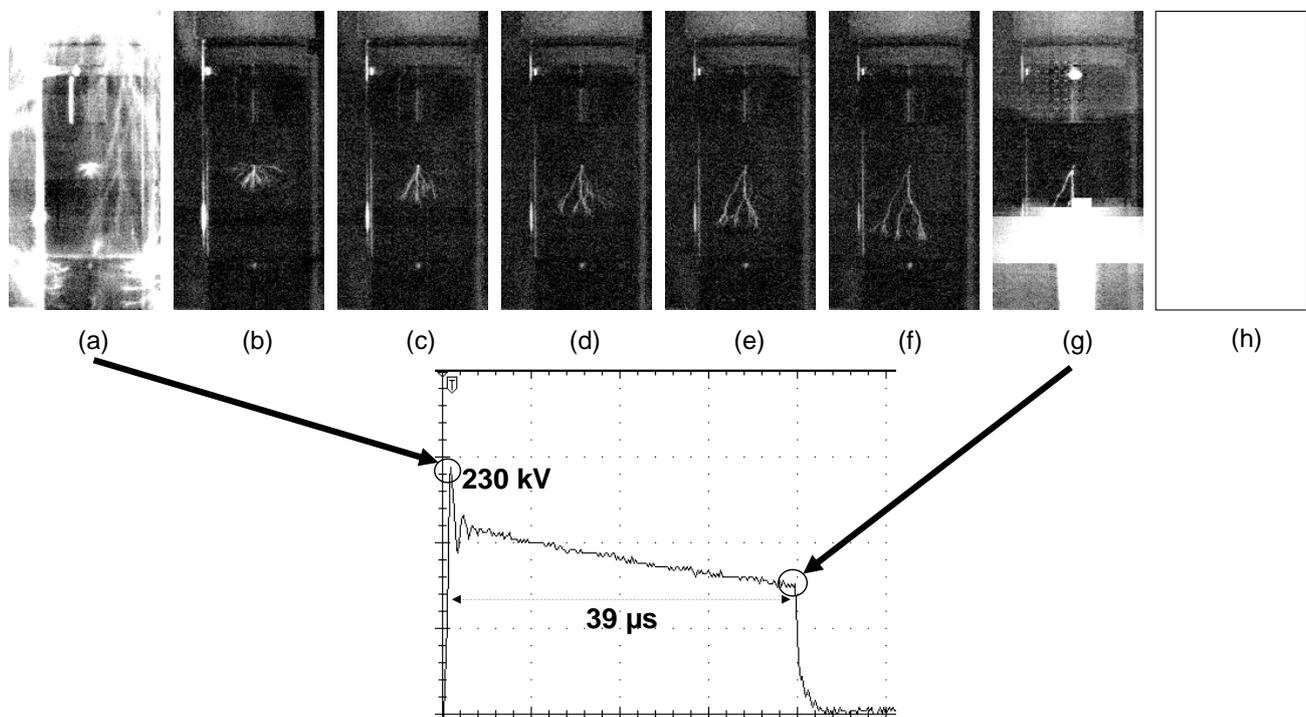


Figure 2. Images of 1% DEA in cyclohexane, positive polarity (230 kV peak, 70 mm gap). Frame rate of 180,000 FPS, 5.55 μs between frames, exposure time is 4.82 μs , TTB from waveform is 39 μs . From frame (a–h) the elapsed time is 38.85 μs .

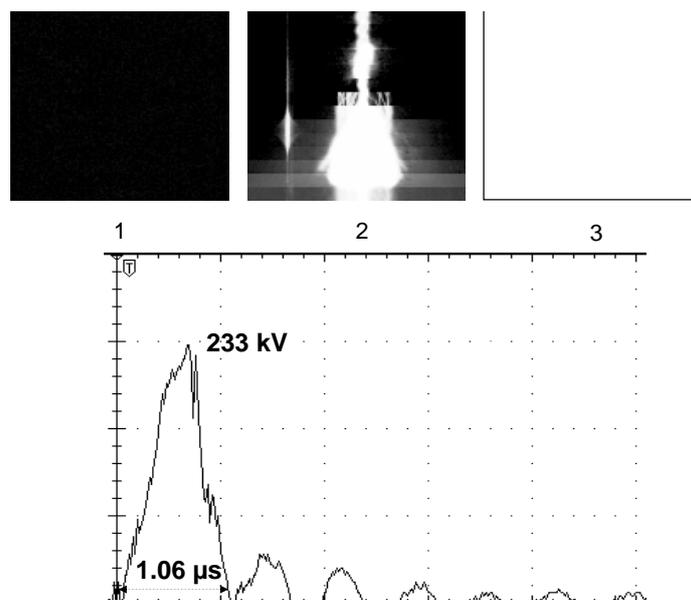


Figure 3. Images of 1% toluene in cyclohexane, positive polarity (233 kV peak applied, 70 mm gap). Frame rate of 430,000 FPS, 2.32 μs between frames, and 1.75 μs exposure time. The TTB from the waveform is $<1.06 \mu\text{s}$. The annotations “1 to 3” above refer to the frame number.

An example of a “fast” streamer, in this instance for 1% toluene in cyclohexane (233 kV applied voltage peak), is shown in Figure 3. It is notable that the imaging system is not fast enough to clearly capture the streamer propagation with a TTB in the order of 1 μs and below, as the maximum frame rate is 2.32 μs . In Figure 3, the whole propagation process happens in one frame (Frame 2) until the complete image saturation (the energy follow-through of the breakdown event). Frame 2 captures the whole discharge event, and the electronic shutter (during the test) was able to close just prior to the intense luminosity. Frame 3 captures the maximum intensity of the attachment process and breakdown event. The TTB from the waveform is $<1.06 \mu\text{s}$, the streamers are therefore at least faster than 66 mm/ μs and are therefore likely predominantly 4th mode streamers.

The accuracy in quantifying the TTB is dependent on whether the breakdown is “fast” (i.e., TTB of $\leq 1 \mu\text{s}$) or “slow” (TTB of 20 to 40 μs). For fast streamers, like that shown in Figure 3, the original approach (results in Section 3.1) was to take the TTB to be the full duration from the start of the waveform to its first zero crossing i.e., $\approx 1 \mu\text{s}$. This, however, leads to underestimation of the streamer velocity as voltage collapse (signifying commencement of breakdown) has already begun shortly before reaching the peak (as the applied waveform has a nominal wave front of 1.2 μs). Therefore, for all subsequent measurements where fast streamers are measured for TTB values $< 1.1 \mu\text{s}$, the nominal TTB is taken to be $0.75 \mu\text{s} \pm 0.25 \mu\text{s}$, based on the resulting streamer velocity range being 70–140 mm/ μs (for the 70 mm gap). This is consistent with the approximate velocity range of the 4th mode streamers [9,11].

The TTB for slow streamers is determined by measuring the time between the peak and the “knee” point (just before voltage collapse), rounding to the nearest μs . The overall streamer velocity is then inferred from the TTB and the gap size (70 mm). A tolerance of $\pm 1 \mu\text{s}$ is applied to the TTB for slow streamers to account for uncertainties in the exact time initiation and termination of the streamer occur.

The experimental error, in inferring streamer speed from the TTB approach, is therefore estimated to be (considering the TTB tolerances stated above and a gap size uncertainty of $\pm 5 \text{ mm}$) $\pm 0.3 \text{ mm}/\mu\text{s}$ for slow streamers (of nominally 2 mm/ μs) and $+57 \text{ mm}/\mu\text{s}$ and $-29 \text{ mm}/\mu\text{s}$ for fast streamers (of nominally 93.3 mm/ μs).

Regarding the terminology of streamer velocities, Linhjell et al. [18] use the definition of “overall velocity” which is defined as the mean velocity from inception to breakdown

for a single impulse application. In the present work, therefore, “approximate streamer velocity” is used to describe the overall streamer velocity inferred by the TTB. Thus, even if the streamer mode switches during propagation, the streamer velocity that is derived from the TTB reflects the average value during the entire propagation.

3. Results

The experimental tests were conducted in two phases. The initial study (Section 3.1) aimed to evaluate whether the test setup and procedure provided repeatable results. The second phase (results presented in Sections 3.2–3.5) focussed on comparing the effects of the additives at different concentrations and the validation of the TTB measurement approach.

3.1. Initial Study

Measurements were first made on pure cyclohexane to establish the baseline for comparison. The voltage range of 230–240 kV was the initial level for evaluation, as the acceleration voltage of cyclohexane (for a similar electrode configuration) was reported to be below this range [18]. This was confirmed to be appropriate as the measured TTB values for cyclohexane indicated fast streamers. A mixture of 0.9% DEA (by volume) in cyclohexane was then tested. DEA was used at an initial a concentration of 0.9% *v/v* based on the relative molar concentration of DMA used in other studies [11]. Five lightning impulse shots per single fill of liquid were applied without changing the applied voltage, to indicate measurement repeatability. Results are shown in Table 2. The 0.9% DEA increases the mean TTB of cyclohexane from 1.28 μs to 38.1 μs and the approximate streamer propagation velocity was therefore slowed down from $>55 \text{ mm}/\mu\text{s}$ to $\approx 1.8 \text{ mm}/\mu\text{s}$. The first impulse application for cyclohexane (2.13 μs) is likely to be an outlier attributed to the measurement system (oscilloscope connection) that was still being fine-tuned in the initial experiments.

Table 2. The time to breakdown for cyclohexane with and without 0.9% DEA when tested with a positive lightning impulse (1.2/50 μs) with a 70 mm gap (point-plane arrangement). The peak voltage magnitude for each impulse application was between 235 to 240 kV.

Liquid	Impulse Application	Time to Breakdown [μs]
Cyclohexane	1	2.13
	2	1.01
	3	1.00
	4	1.10
	5	1.15
	Mean	1.28
	Std. Deviation	0.43
Cyclohexane + 0.9% DEA	1	38.0
	2	38.3
	3	37.9
	4	37.7
	5	38.53
	Mean	38.09
	Std. Deviation	0.33

3.2. Investigation of the Effect of the Concentration of DEA on the Time to Breakdown

The influence of additive concentration was investigated by testing different concentrations of DEA in cyclohexane to establish the minimum dosage that causes streamers to slow down (for the voltage range in question). Table 3 presents the TTB and calculated approximate streamer velocity for different peak voltages (positive polarity impulse of 1.2/50 μs). The acceleration voltage of cyclohexane lies between 192 kV and 213 kV. At DEA concentrations of 0.33% and above the TTB is increased (streamers are slowed, from predominant 4th mode to 2nd mode) for the voltage range in question (≈ 220 –280 kV). The mixtures containing 0.11% and 0.22% DEA resulted in fast streamers, as was the case for

pure cyclohexane when measured in a similar voltage range. The increase of DEA above 0.33% does not further slow streamers. All concentrations above 0.33% yield streamer velocities in the order of 2 mm/ μ s (2nd mode).

Table 3. The time to breakdown and inferred overall streamer velocity for cyclohexane and different concentrations of DEA in cyclohexane. Applied voltage was a positive polarity lightning impulse (1.2/50 μ s) with a 70 mm gap (point-plane arrangement).

Liquid	Peak Voltage [kV]	Time to Breakdown [μ s]	Approximate Streamer Velocity [mm/ μ s]
Cyclohexane	157	22	3.2
	192	25	2.8
	213	0.75	93.3
	225	0.75	93.3
	263	0.75	93.3
Cyclohexane + 0.11% DEA	244	0.75	93.3
	246	0.75	93.3
	247	0.75	93.3
	277	0.75	93.3
Cyclohexane + 0.22% DEA	236	0.75	93.3
	242	0.75	93.3
	261	0.75	93.3
Cyclohexane + 0.33% DEA	265	32	2.2
	265	30	2.3
	274	40	1.8
	280	40	1.8
Cyclohexane + 0.35% DEA	238	38	1.8
	239	33	2.1
Cyclohexane + 0.45% DEA	227	34	2.1
	228	35	2.0
	238	30	2.3
	252	31	2.3
	253	31	2.3
	258	33	2.1
Cyclohexane + 1% DEA	231	35	2.0
	236	36	1.9
	242	32	2.2
	246	33	2.1
	250	35	2.0
	265	34	2.1
Cyclohexane ¹ + 1% DEA	179	36	1.9
	206	42	1.7
	220	42	1.7
	227	40	1.8
	227	41	1.7
	227	41	1.7
	228	40	1.8
	230	40	1.8
	235	30	2.3
	235	40	1.8
	246	30	2.3

¹ The second set of data for 1% DEA was from a separate testing instance.

3.3. Investigation of the Effect of the Concentration of Toluene on the Time to Breakdown

The effect of DEA on slowing down streamers in cyclohexane is consistent with the theory that the use of additives of lower IP than the base liquid increase streamers branching and therefore slows down the streamer propagation velocity. To test the continuity of this

theory across different additives, a similar test was done using toluene. Toluene has an IP of 1.06 eV lower than cyclohexane but 1.85 eV higher than DEA (see Table 1).

Table 4 below presents the data for 1% toluene and 10% toluene. For 1% toluene, only fast streamers were recorded for the voltage range 216 kV to 233 kV. It may be that between 209 kV and 216 kV 1% toluene may have a small effect (increasing the acceleration voltage slightly) but, as shown in Table 3, cyclohexane itself has an acceleration voltage of somewhere between 192 kV and 213 kV (for this arrangement).

Table 4. The time to breakdown and inferred overall streamer velocity for 1% and 10% (by volume) of toluene in cyclohexane. Applied voltage was a positive polarity lightning impulse (1.2/50 μ s) with a 70 mm gap (point-plane arrangement).

Liquid	Peak Voltage [kV]	Time to Breakdown [μ s]	Approximate Streamer Velocity [mm/ μ s]
Cyclohexane + 1% Toluene	224	0.75	93.3
	250	0.75	93.3
	268	0.75	93.3
	271	0.75	93.3
	274	0.75	93.3
Cyclohexane + 1% Toluene ¹	208	36	1.9
	209	30	2.3
	216	0.75	93.3
	219	0.75	93.3
	233	0.75	93.3
Cyclohexane + 10% Toluene	246	0.75	93.3
	271	0.75	93.3

¹ The second set of data for 1% toluene was from a separate testing instance.

A 10% mixture of toluene was then tested to see if a relatively very high concentration may have a notable effect on the streamers. For both concentrations toluene does not appear to slow streamers for the similar voltage range where DEA was effective.

3.4. Analysis of the Effect of DEA and Toluene

The data in Sections 3.2 and 3.3 are plotted in Figure 4 below. The error bars represent the uncertainty in the approximate overall streamer velocity. The main source of uncertainty is in the exact TTB, as the streamer can start to propagate at any point on the wavefront (see Section 2.3). There is apparent clustering of the slow streamers (predominantly second mode, around 2 mm/ μ s) and fast streamers (likely fourth mode, around 90–100 mm/ μ s). When the DEA concentration is 0.33% and above, slow streamers occur for the entire voltage range in question. Neither 1% nor 10% toluene are effective at slowing streamers above \approx 210 kV.

Additional measurements were done using 2% toluene, 0.33% DEA, and pure cyclohexane to verify the repeatability of the key findings and the data are presented in Appendix B. When comparing Figures 4 and A5, the effectiveness of the 0.33% DEA in slowing streamers is confirmed. The repeat measurements were conducted using a new needle with a new batch of the base cyclohexane.

3.5. Streamer Imaging, Effect of DEA Concentration on Streamer Branching

Streamer images for some individual impulse applications of 0.35% DEA and 1% DEA in cyclohexane are shown in Appendix A. Figure A1 shows 1% DEA in cyclohexane using an applied peak voltage of 228 kV, and Figure A2 shows a second application at 227 kV. In comparison, Figure 4 (1% DEA, 230 kV) exhibits a similar degree of branching. For comparison, images of the streamers at a lower concentration of 0.35% DEA for 239 kV (Figure A3) and 238 kV (Figure A4) are presented. The four figures in Appendix A are comparable for the voltage range \approx 230–240 kV. However, the degree of branching of the

streamers appears to be higher for the 1% DEA than the 0.35% DEA, but this effect is difficult to quantify. Notwithstanding, both concentrations resulted in very similar TTB values and overall streamer velocity. A comprehensive study of the streamer branching images as a function of the additives could be a possible extension of the present work.

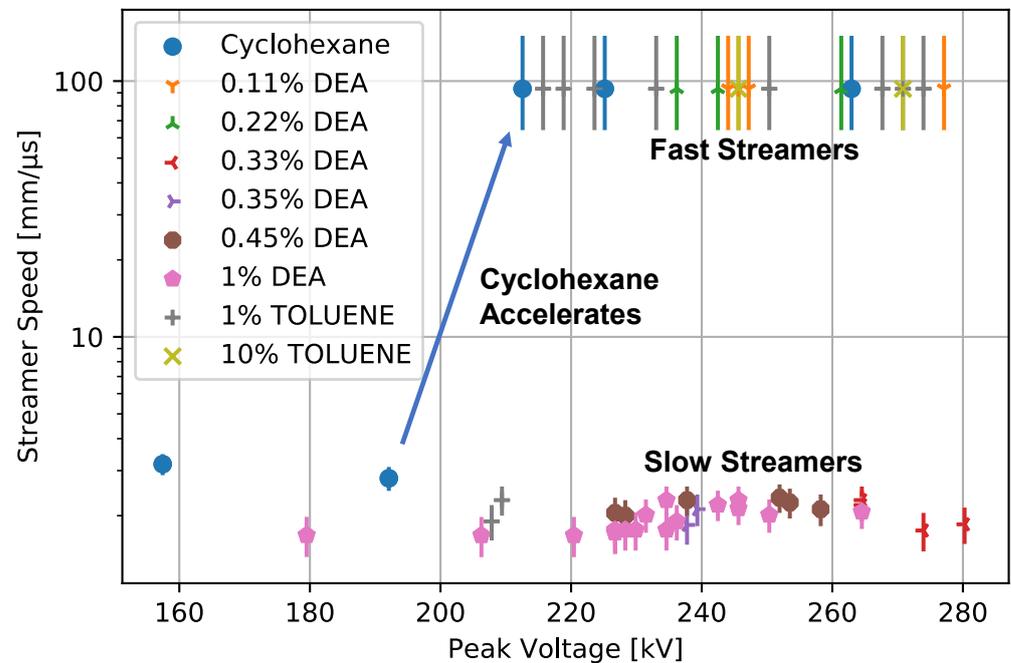


Figure 4. The approximate overall streamer velocity of streamers for various DEA and toluene mixtures in cyclohexane. Pure cyclohexane also shown for reference. Applied voltage was a positive polarity lightning impulse (1.2/50 μ s) with a 70 mm gap (point-plane arrangement).

4. Discussion

The time-to-breakdown method validation data presented in Section 2.3 provides evidence that the approach of using the voltage waveform profile to infer on the overall streamer velocity is reasonable. Whilst the TTB approach has accuracy limitations, it can be used to discriminate between fast streamers (4th mode dominant, with velocities greater than ≈ 70 mm/ μ s) and slow streamers (2nd or 1st mode, with velocities less than 5 mm/ μ s) [18]. As is evident in Figure 2, Frame (g), capturing the image at the exact moment of streamer termination on the opposite electrode is difficult due to pixel saturation from the resultant high intensity light emitted by the attachment process. Consequently, considering the images alone, about 5.55 μ s uncertainty exists between the two frames capturing the streamer termination event. However, the corresponding voltage waveform collapses at the time of breakdown, as the insulation gap becomes short circuited by the resultant plasma channel. The voltage collapse point is therefore an accurate measure of the end point of the TTB.

In the present work, the first evidence that, as an additive, DEA effectively slows down streamers was obtained from the initial measurements (Section 3.1). At around 240 kV applied peak voltage, pure cyclohexane gave a mean TTB of 1.28 μ s with a standard deviation of 0.43 (from 5 impulses). In the case of cyclohexane containing 0.9% DEA, the mean TTB increased to 38.1 μ s with a standard deviation of 0.33. The results give a clear distinction between “slow” streamers (with 0.9% DEA) and “fast” streamers in pure cyclohexane.

The work has also demonstrated the effect of DEA as an additive at different concentrations in slowing down streamer velocity in cyclohexane. Toluene as an alternative additive in cyclohexane was also investigated. The results show that for DEA in cyclohexane, below a concentration threshold (in the present work this threshold is approximately at 0.33%

DEA by volume) had no influence in the overall streamer velocity in cyclohexane. However, above the threshold concentration, the streamers were slowed from predominantly 4th mode (fast) to 2nd mode (slow). Furthermore, concentrations above the threshold (i.e., up to 1%) showed no further significant change in the overall streamer velocity. The effectiveness of DEA in cyclohexane, as found in the present work, is similar to that of dimethylaniline (DMA) in cyclohexane and white mineral oils, as investigated by Linhjell [8] and Nguyen [35]. With an ionization potential of 7.14 eV for DMA, the similarity on influencing streamer velocity in cyclohexane is expected for DEA (with an ionization potential of 6.98 eV), which is even lower than that of cyclohexane at 9.88 eV. Other research has also shown that pyrene [9] is effective at slowing streamers in cyclohexane. The theory advanced in the literature to explain the phenomenon of additives slowing down streamer velocities in a base liquid is based on the shielding effect of increased streamer branching [35]. If the IP of the additive is sufficiently lower than the bulk liquid, streamer branching is promoted (as illustrated in Figure 5) by introducing an electric field shielding effect on the tips that leads to an overall slowing of the streamer [11,35]. The streamers in the case of 1% DEA in cyclohexane, as shown in Figures 2, A1 and A2, are evidently significantly branched.

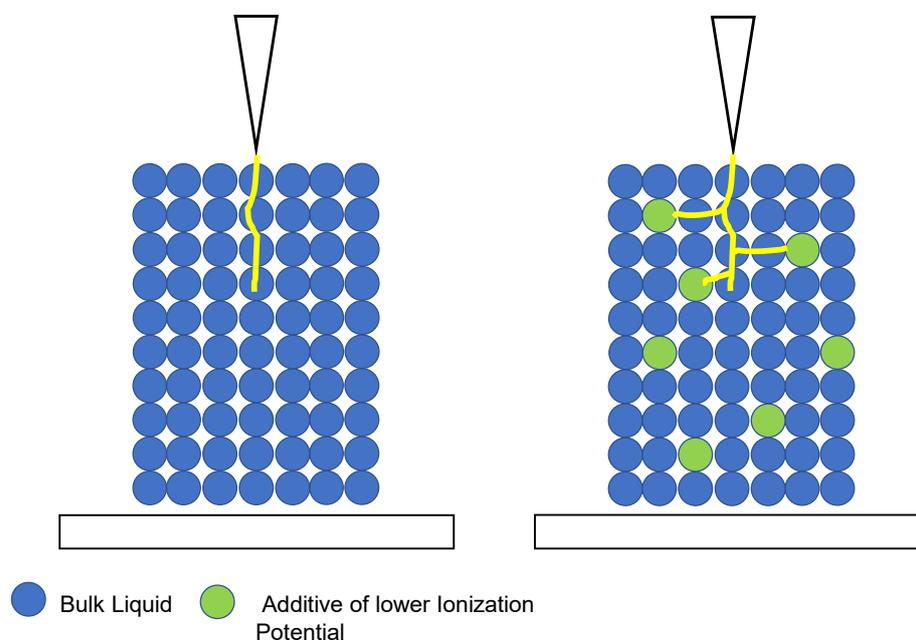


Figure 5. A simplified illustration postulating how a lower IP additive may promote streamer branching.

The extent to which the IP of the additive is lower than that of the base liquid could be an important variable influencing the degree of streamer branching. Toluene, with an IP of 8.92 eV (9.7% lower than cyclohexane compared to 29.4% for DEA), did not lead to slow streamers in the same way (for the same voltage range) as did the DEA. It is however noted that the use of IP values, alone as a proxy, is probably insufficient to explain the phenomenon of streamer propagation being affected by additives. The relationship between the first excitation energies of the additive and base liquids [33,37] may allow more accurate predictions, albeit with the caveat that the calculations of the excitation energies are highly complex [37,38,44].

Regarding the issue of streamer velocity acceleration thresholds, the following conclusions can be made. Whilst there is insufficient data from this investigation to pinpoint the exact acceleration voltages for each concentration, there is evidence that the addition of 0.33% DEA gives slow streamers at 280 kV. A concentration of 0.22% DEA already leads to fast streamers at 236 kV. Therefore, the minimum sufficient concentration lies somewhere between 0.22% and 0.33%. Data in [9] suggests that the exact acceleration voltage does shift

with the concentration of the additive. Even though in the present study the voltage is not increased until the point of acceleration, there is clear evidence of streamer slowing in the voltage range 220–280 kV for the DEA concentration range 0.33% to 1% (related to the specific test setup described in Section 2.1).

Toluene, on the other hand, does not lead to a statistically significant increase of acceleration voltage compared to pure cyclohexane (1% toluene accelerates in the of range 209 kV and 216 kV, and cyclohexane accelerates in the range of 192 kV and 213 kV, approximately). In the voltage range 220–280 kV, both 1% and 10% toluene in cyclohexane are ineffective at slowing streamers. This may arise since the IP of toluene is not sufficiently low enough to promote side branching early in the streamer propagation process.

Comparing the data for pure cyclohexane and that of 2% toluene in Figure A5 and Table A1 with the main data shown in Figure 4, it shows that indeed both cases of pure cyclohexane and cyclohexane with toluene additive have effectively the same acceleration voltage range that is approximately between 180 kV and 210 kV (for the test conditions used herein).

The study of the excitation energies of DEA and toluene and corresponding modelling of the streamer branching is recommended as future work. A further understanding of the relationship between the degree of streamer branching and the corresponding threshold voltage for transition into slower streamers is necessary. Furthermore, the present work could be extended to test the different concentrations of DEA and their acceleration voltages for better understanding of the optimal DEA concentration effects. Future work could also include testing the negative polarity case.

5. Conclusions

The concept of altering streamer propagation velocity in liquid insulation has been explored. It has been demonstrated that a relatively simple experimental screening method can indicate whether an additive (such as DEA or toluene) will be effective at slowing streamers within a given voltage range in cyclohexane (for the point-plane, 70 mm, 1.2/50 μ s lightning impulse, positive polarity case). DEA was effective at increasing the TTB (and thus slowing the streamer velocity) for concentrations of 0.33% and above, in the voltage range \approx 200–280 kV. Toluene was not effective in the same voltage range, at both 1% and 10% concentrations. The time-to-breakdown measurement method employed was validated using high speed imaging, and consequently, it was possible to discriminate between fast (likely 4th mode) and slow (2nd mode) streamers.

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Conflicts of Interest: The authors declare no conflict of interest.

Appendix A

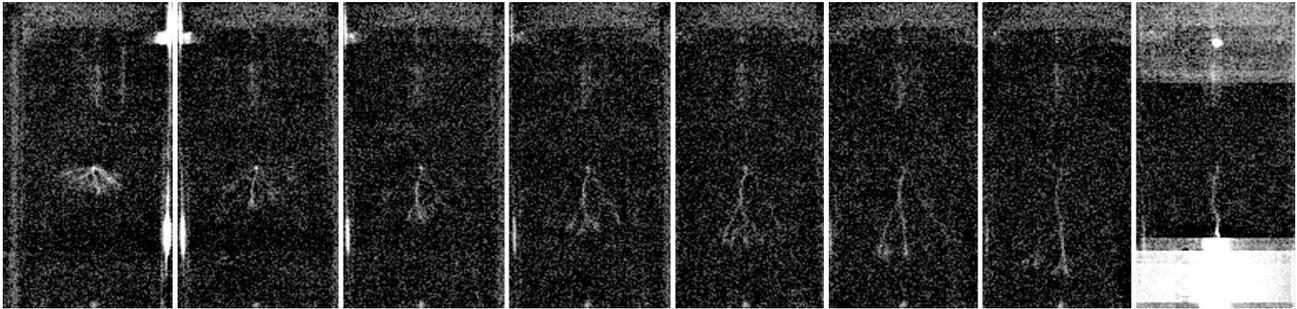


Figure A1. 1% DEA in cyclohexane, positive polarity, 228 kV. 4.16 μ s between frames, Time-To-Breakdown (TTB) \approx 40 μ s.

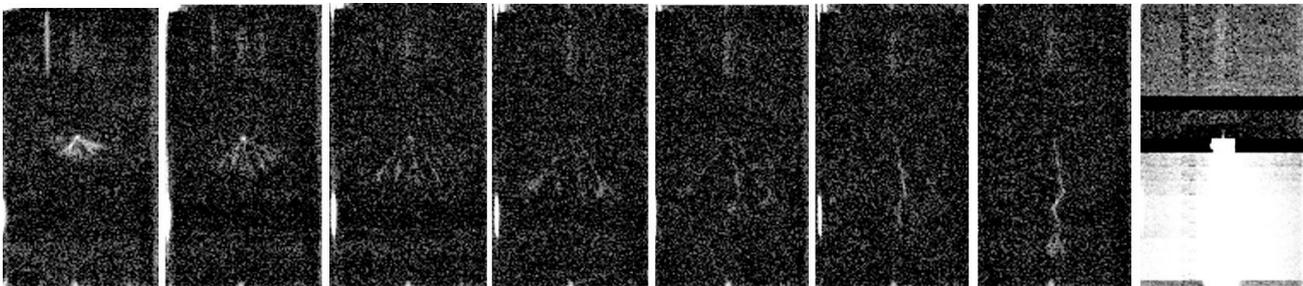


Figure A2. 1% DEA in cyclohexane, positive polarity, 227 kV. 4.16 μ s between frames, Time-To-Breakdown (TTB) \approx 40 μ s.

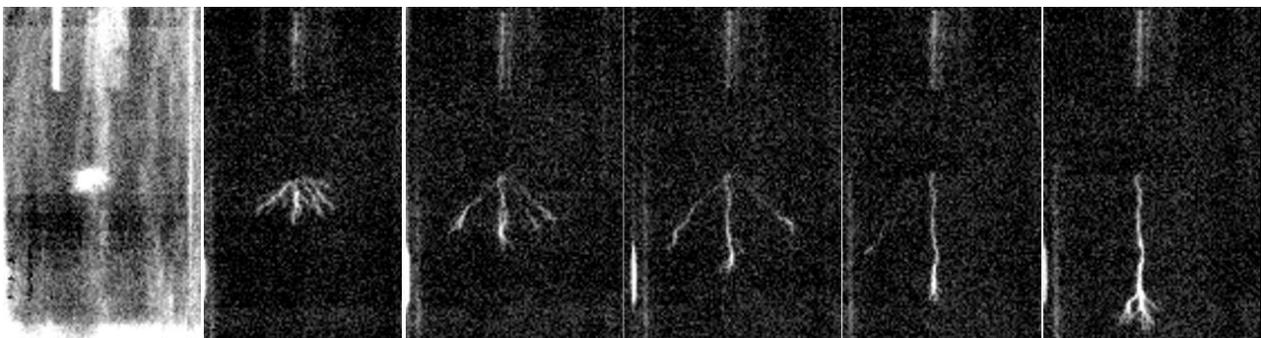


Figure A3. 0.35% DEA in cyclohexane, positive polarity, 239 kV. 5 μ s between frames, Time-To-Breakdown (TTB) \approx 40 μ s.

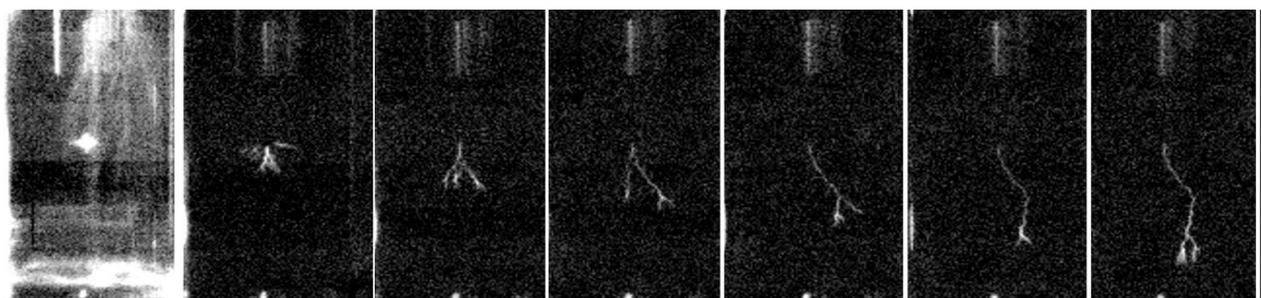


Figure A4. 0.35% DEA in cyclohexane, positive polarity, 238 kV. 5 μ s between frames, Time-To-Breakdown (TTB) \approx 40 μ s.

Appendix B

Table A1. The time to breakdown and inferred overall streamer velocity for pure cyclohexane and mixtures of 2% toluene and 0.33% DEA of in cyclohexane. Applied voltage was a positive polarity lightning impulse (1.2/50 μ s) with a 70 mm gap (point-plane arrangement). This data serves as a check of the repeatability of the results presented in Section 3.4, as a new needle and new batches of liquids were used.

Liquid	Peak Voltage [kV]	Time to Breakdown [μ s]	Approximate Streamer Velocity [mm/ μ s]
Cyclohexane	205	32	2.2
	227	0.75	93.3
	233	0.75	93.3
	246	0.75	93.3
	251	0.75	93.3
	252	0.75	93.3
Cyclohexane + 2% Toluene	192	32	2.2
	205	0.75	93.3
	224	0.75	93.3
	236	0.75	93.3
	239	0.75	93.3
	240	0.75	93.3
	246	0.75	93.3
	280	0.75	93.3
	205	0.75	93.3
Cyclohexane + 0.33% DEA	233	32	2.2
	236	34	2.1
	239	32	2.2
	252	40	1.8
	258	34	2.1
	265	40	1.8
	272	34	2.1

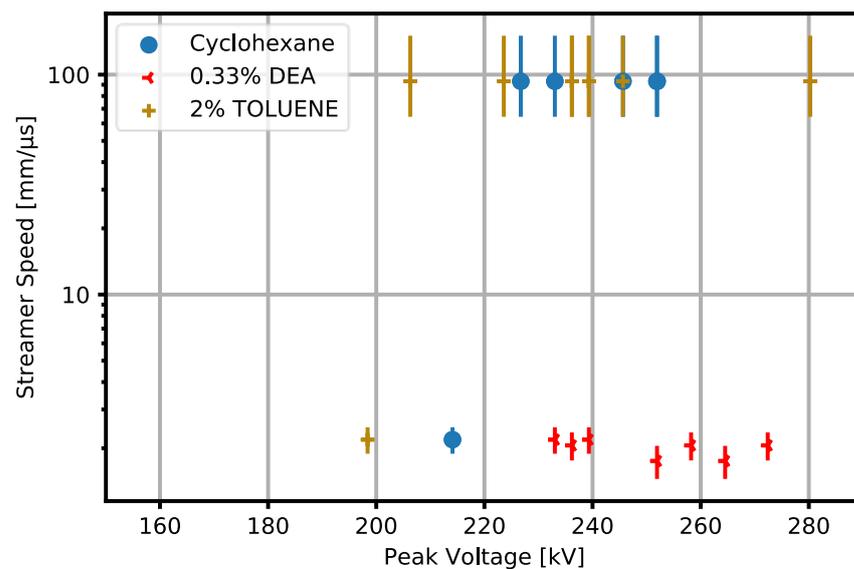


Figure A5. The approximate overall streamer velocity of streamers for cyclohexane and mixtures of cyclohexane with 0.33% DEA and 2% toluene. Applied voltage was a positive polarity lightning impulse (1.2/50 μ s) with a 70 mm gap (point-plane arrangement). This data serves as a check of the repeatability of the results presented in Section 3.4, as a new needle and new batches of liquids were used.

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