Observation of Prebreakdown and Breakdown Phenomena in Liquid Hydrocarbons II. Non-Uniform Field Conditions

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ABSTRACT

The prebreakdown and breakdown processes have been recorded in n-hexane toluene and Marcol 70, both in a pure state and with selected impurities. The study was carried out using a point-plane geometry. A low ionization potential additive had only a small effect on the breakdown voltage or the streamer propagation speed but did significantly alter the shape of the prebreakdown streamer when the needle was positive, an anode. For a negative needle, a cathode, chemical impurities affected the breakdown voltage. The significance of these findings is discussed in detail.

INTRODUCTION

In a preceding paper [1] results were presented of prebreakdown and breakdown phenomena occurring in liquid hydrocarbons under approximately uniform field conditions using high-speed photographic techniques. These studies were carried out with highly purified hydrocarbons and the results indicated streamer initiation occurring predominantly at the cathode (cathode event). A second mechanism was observed to occur less frequently near th anode (anode event). To elucidate these mechanisms, further studies were undertaken under non-uniform field conditions using these same highly purified liquids [2]. While the electrode separation (the gap) was 2 mm for the uniform field condition, it was 3 to 5 mm in the point-plane study. Again, the same high-speed photographic techniques were utilized. The latter results confirmed the appearance of streamers at the negative needle (cathode), but also showed streamer growth from the positive needle (anode) in conformity with the findings of other researchers [3]. To examine the possibility of generalizing these observations and to gain a basic understanding of the mechanism responsible for the initiation as well as the propagation of these streamers, further studies were undertaken. In one phase of this study well-defined impurities were added intentionally and in a second phase the influence of gap size was investigated. The results of these investigations are summarized in this report. In the next section the experimental conditions are described, then the results obtained are summarized. The significance of the findings is discussed in the last section.

EXPERIMENTAL

The equipment used in this study is described in detail in [1,2]. The test fluids used here were purified n-hexane and toluene, chemical grades of these two fluids (containing both oxygen and water) as well as Marcol 70 [4], a naphthenic white oil. The impurities included chemical grade N,N^1-Dimethylaniline (DMA), Antistatic Additive #3 (ASA 3) which is a mixture of 1/3 chromium salicilate, 1/3 calcium sulfosuccinate and 1/3 vinyl copolymer in kerosene (50/50 mixture by volume), water and air. The concentration of the ASA3 impurity is referred to on the basis of active ingredients. The impurities were injected with the help of syringes into the cell system without exposing the fluid to the ambient atmosphere.

The cell design used in this study was, in principle, the same as that used in earlier studies [1,2] except for the cell's overall dimensions which were increased 2.5 fold. The stainless steel body was replaced by a structure machined from polytetrafluroethylene (PTFE). All tubing was also made from PTFE. The electrode system consisted of a highly polished steel disk of 8.9 cm diameter with well-rounded edges and a phonograph needle. The voltage pulse was of trapezoidal shape rising in aobut 4 μs to crest voltage, remaining at or near this level for 3 μs, and decaying to zero in another 4 μs with occasional overshoot to voltages of opposite polarity. The electrode gap was illuminated with a Xenon flash lamp or an Argon laser. The shadowgraphs produced by the events in the gap were recorded with an image converter camera capable of framing speeds up to 2×10^7 frames/s. The results obtained with this set-up are presented in the next section.

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RESULTS

In this section the observations and measurements made when the needle electrode was a cathode will be presented first, and those relating to a needle anode will be summarized subsequently.

Needle Cathode (negative)

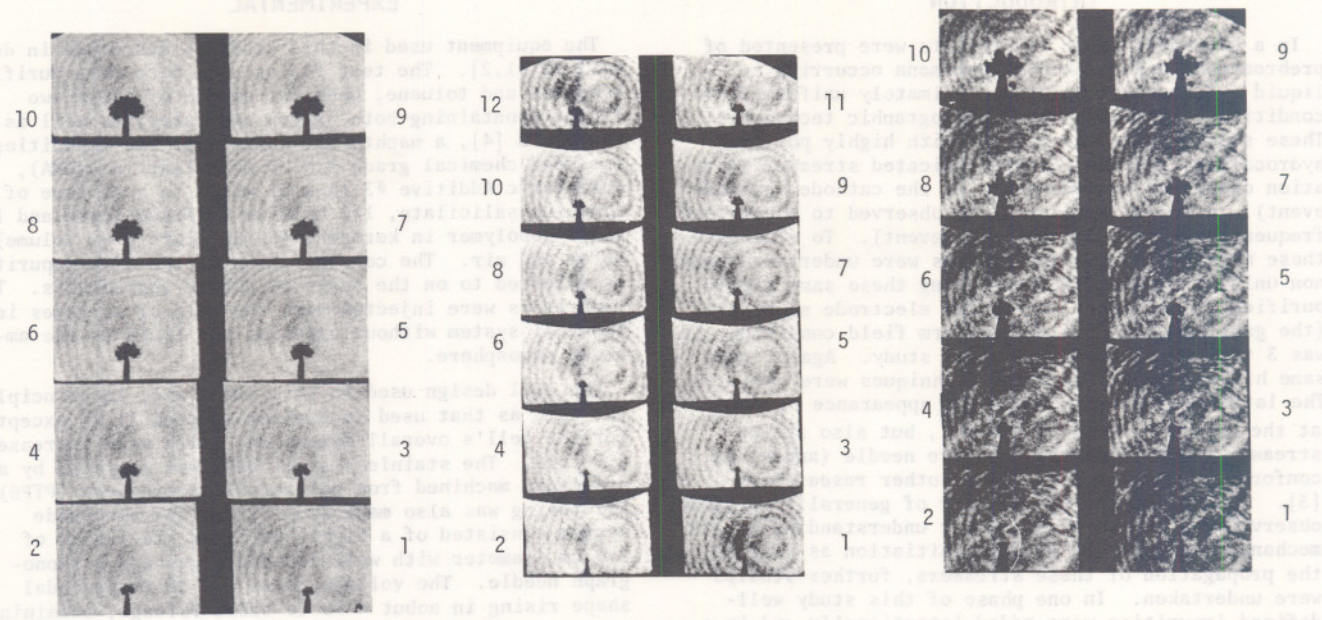
Examination of the numerous photographs of streamer development and breakdown produce the following general observations. Upon application of an external potential, streamers appear in the form of dense bushes near the tip of the needle. Their growth is not very regular, particularly in the later stages when their lateral growth exceeds their vertical one (Fig. 1). Their average growth rate is of the order of 10^4 cm/s. With highly purified, paraffinic hydrocarbons such as hexane, cyclohexane, or isooctane and the available power supply, it was impossible to cause breakdown when the gap was 0.3 cm or larger. Under these conditions one was able to observe occasional breakdown during a polarity reversal of the voltage pulse (Fig. 2). In toluene and contaminated liquids, it was possible to observe breakdown regularly in 0.3 cm gaps (Fig. 3).

These bush-like streamers do not, in principle, traverse the gap to cause breakdown. At a certain stage in their growth, which appears to depend on the purity of the liquid, secondary streamers develop from one or several sites on the periphery of the bush (Fig. 4). These secondary streamers grow at an average rate approaching 10^7 cm/s and they are very difficult to observe even at the highest framing speeds available. They appear more readily in contaminated liquids than in pure ones which may perhaps explain the fact that the breakdown voltage of pure liquids is higher than that of contaminated ones. A different picture prevails when the needle is the anode as described in the following paragraphs.

Needle Anode (positive)

The streamer appears to grow from the anode into the liquid and there is no visual indication of charge carrier injection at the plane cathode (Fig. 5). Similar to the observation made for the case when the needle is a cathode, there seem to occur streamer developments at two significantly different rates. These differences are not readily noticeable when studying gaps of 0.64 cm or less. They can be resolved, however, at large spacings such as 1.27 and 2.54 cm (Fig. 6.).

As shown in Table 1, toluene breaks down at a higher applied voltage than does n-hexane. For the purpose of this study the breakdown voltage V_{BD} is defined as the voltage at which the secondary streamer reaches the anode and establishes a conductive path. Since the voltage wave is trapezoidal, this V_{BD} can occur at times when the voltage is decaying from the crest voltage V_{CR} to zero. This situation holds for all electrode gaps studied. The reason for the lower breakdown voltage of n-hexane when compared to toluene may be found in the neraly three times faster growth of the streamers in n-hexane. Marcol 70, which is in composition intermediate between linear aliphatic n-hexane and aromatic toluene contains both linear and branched aliphatic as well as cycloaliphatic molecules. It has a higher viscosity than n-hexane. Its streamer growth rate is intermediate between the two pure hydrocarbons. At the larger gap spacings of 1.27 and 2.54 cm the viscosity of the medium appears to influence the overall growth process. This aspect will be discussed later on.

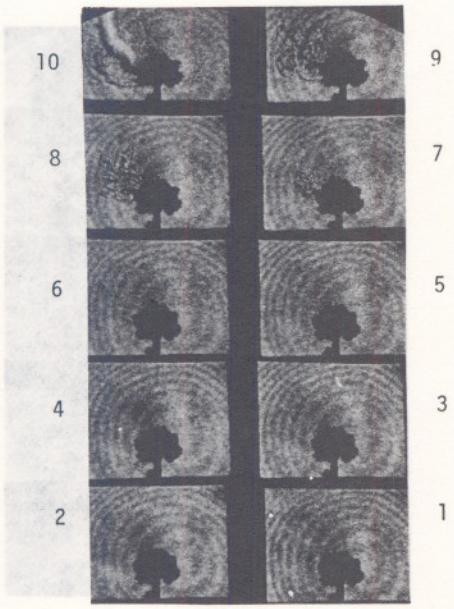


n-hexane
0.64 cm gap
156 kV Crest Voltage

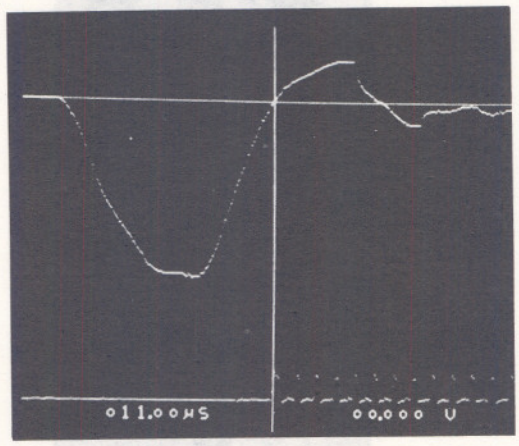
cyclohexane
0.5 cm gap
120 kV Crest Voltage

toluene
0.3 cm gap
168 kV Crest Voltage

Fig. 1: Streamer growth in pure hydrocarbons (needle cathode, 2×10^6 frames/s, trapezoidal pulse)

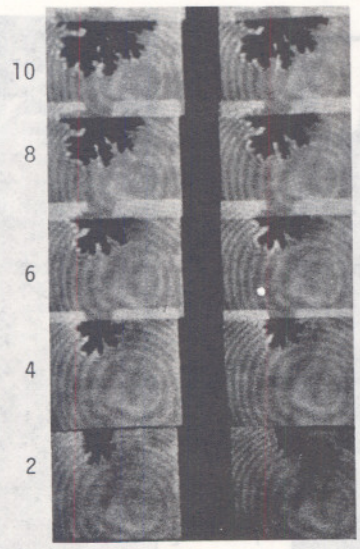


Crest Voltage - 180 kV
Camera fired 11 μs after pulse starts



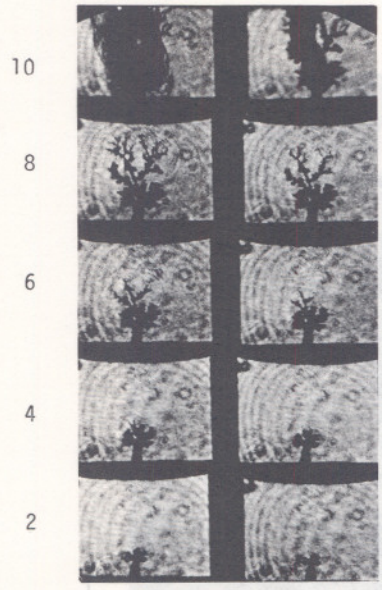
Voltage trace of pulse
Camera Signal

Fig. 2: Breakdown on polarity reversal of applied voltage pulse (n-hexane, needle initially cathode, 0.64 cm gap, 2×10^6 frames/s, trapezoidal pulse)



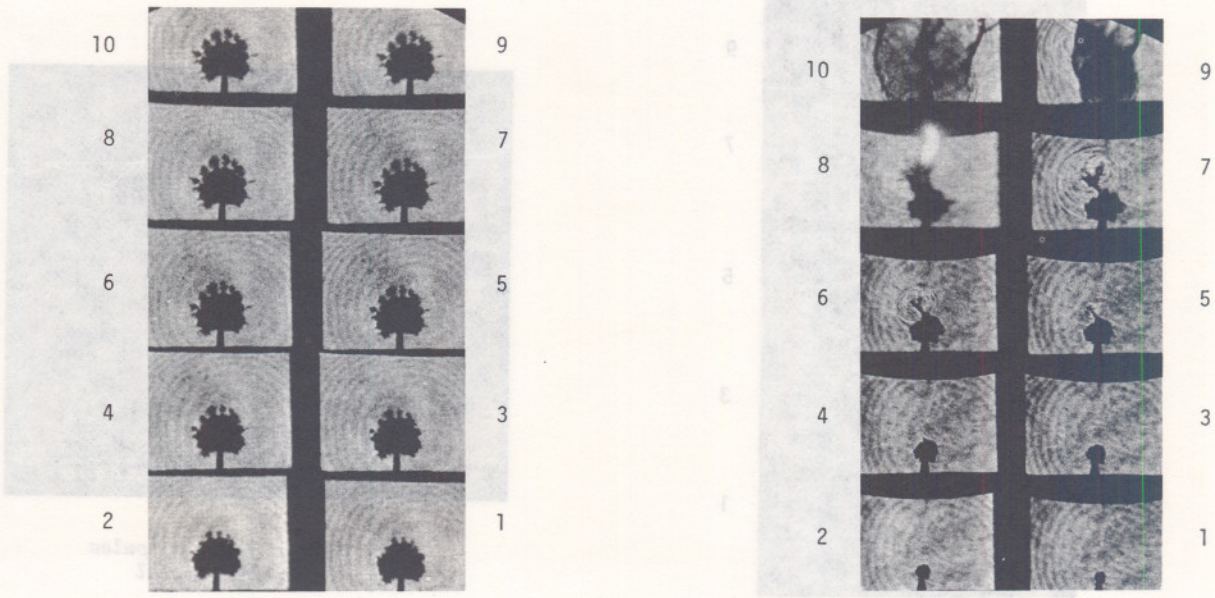
n-hexane + 20 ppm ASA 3
156 kV
5×10^6 f/s
17×

Crest Voltage
Framing Speed
Magnification



Toluene + 50 ppm ASA 3
72 kV
2×10^6 f/s
17×

Fig. 3: Effect of contaminents on streamer growth and breakdown (needle cathode, 0.3 cm gap, trapezoidal pulse)

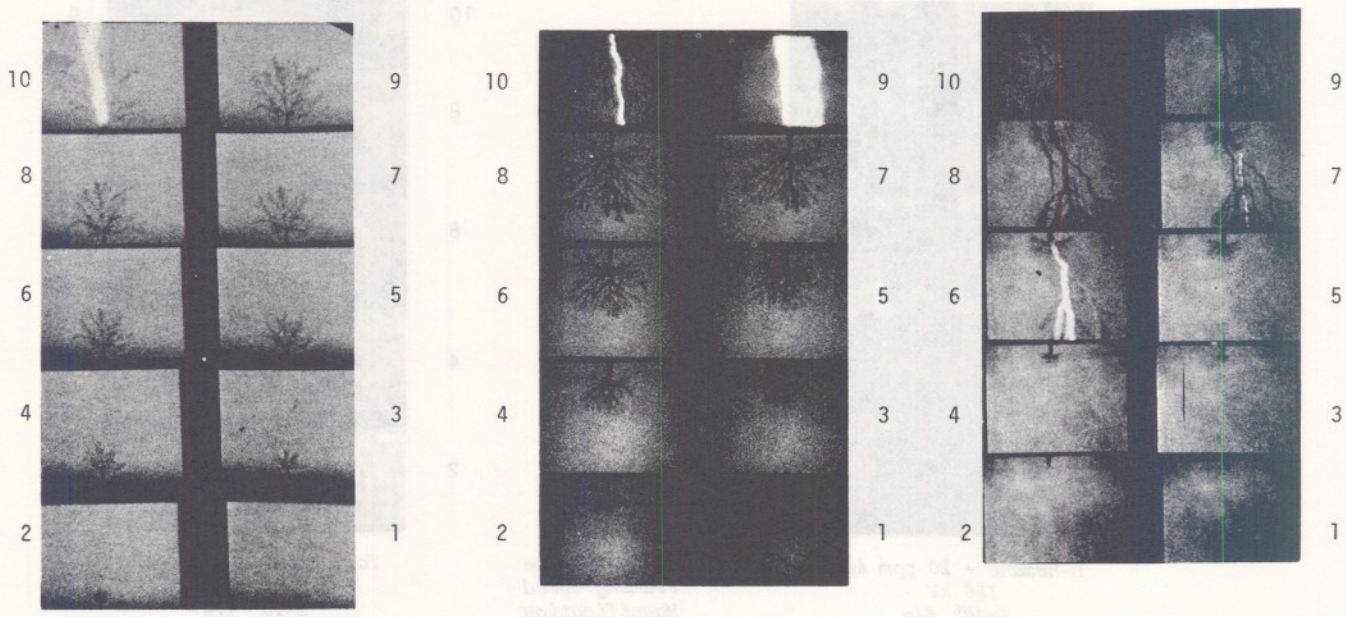


n-hexane
204 kV

fluid
Crest Voltage

Toluene
120 kV

Fig. 4: Secondary Streamer Development (0.3 cm gap, needle cathode 2×10^6 frames/s, trapezoidal pulse)

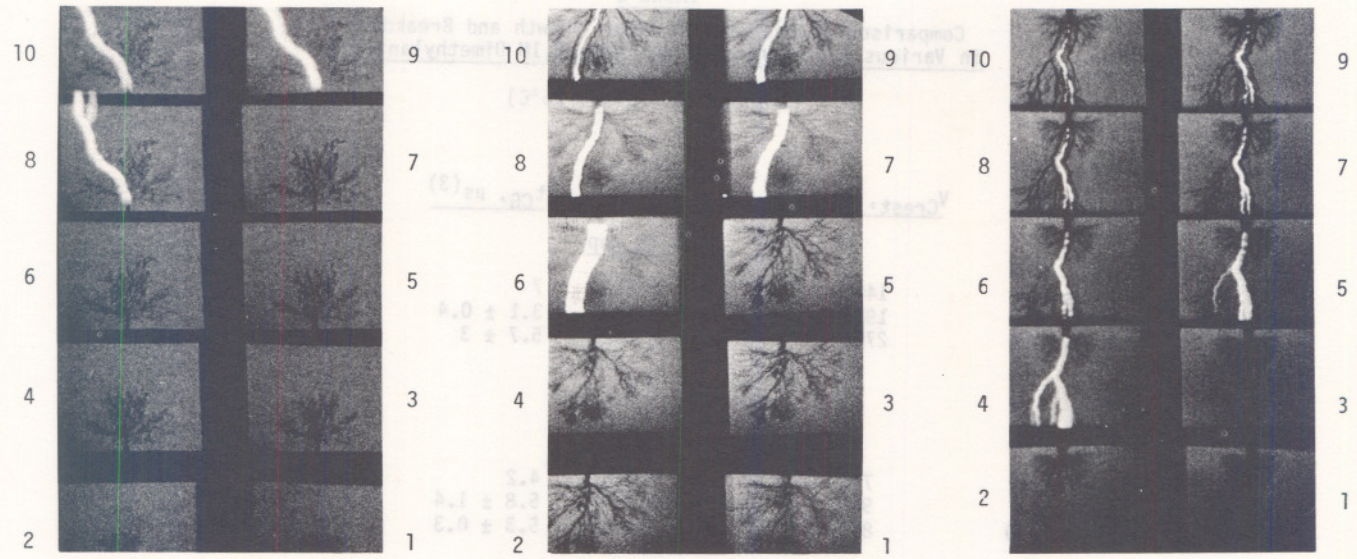


n-hexane
5×10^6 f/s

Marcol 70
2×10^6 f/s

Toluene
2×10^6 f/s

Fig. 5: Positive streamer growth in liquid hydrocarbons (1.27 cm gap, needle anode, crest voltage 96 kV, trapezoidal pulse)



n-hexane
1.27 cm gap
96 kV crest voltage

Marcol 70
0.64 cm gap
60 kV crest voltage

Toluene
0.6 cm gap
96 kV crest voltage

Fig. 6: Secondary streamer development in liquid hydrocarbons (needle anode, 2×10^7 frames/s, trapezoidal pulse)

TABLE 1

Comparison of Positive Streamer Growth and Breakdown in Various Hydrocarbons

(Trapezoidal Pulse, 25°C)

Liquid	V_{crest}, kV(1)	V_{BD}, kV(2)	t_{CG}, μs(3)	Growth Rate in cm/s x 10^{-5}
		2.54 cm gap		
n-hexane	122	117.6 ± 2.6	3.0 ± 0.5	3.4 ± 0.2
toluene	152	148.6 ± 3.5	3.1 ± 0.4	1.2 ± 0.1
Marcol 70	228	227.7 ± 1.1	5.1 ± 0.7	2.1 ± 0.3
		1.27 cm gap		
n-hexane	72	69.9 ± 1.6	2.8 ± 0.7	3.0 ± 0.5
toluene	96	92.7 ± 3.1	2.8 ± 0.9	1.1 ± 0.5
Marcol 70	96	96.4 ± 1.7	4.7 ± 0.3	1.8 ± 0.1
		0.63 cm gap		
n-hexane	60	54.2 ± 5.9	1.2 ± 0.4	2.9 ± 0.3
toluene	72	70.9 ± 1.5	3.1 ± 0.6	1.1 ± 0.1
Marcol 70	48	48.5 ± 0.8	3.3 ± 0.2	1.6 ± 0.1

(1) Nominal crest voltage
(2) Voltage at which breakdown occurred, average of seven consecutive determinations
(3) Time to cross gap

TABLE 2

Comparison of Positive Streamer Growth and Breakdown in Various Hydrocarbons Containing 0.1M Dimethylaniline

(Trapezoidal Pulse, 25°C)

Liquid	V_{Crest}, kV(1)	V_{BD}, kV(2)	t_{CG}, μs(3)	Growth Rate in cm/s x 10^{-5}
		2.54 cm gap		
n-hexane	148	84.4 ± 21.3	7	3.1 ± 0.2
toluene	192	179.4 ± 15.2	3.1 ± 0.4	1.6 ± 0.3
Marcol 70	276	260.7 ± 18.7	5.7 ± 3	2.8 ± 0.4
		1.27 cm gap		
n-hexane	72	71.2 ± 2.5	4.2	3.1 ± 0.1
toluene	96	79.9 ± 7.5	5.8 ± 1.4	1.6 ± 0.1
Marcol 70	84	84.7 ± 1.8	5.3 ± 0.3	1.9 ± 0.1
		0.64 cm gap		
n-hexane	60	55.5 ± 2.9	1.9 ± 0.2	3.4 ± 0.2
toluene	60	59.7 ± 1.4	3.3 ± 0.6	1.4 ± 0.1
Marcol 70	54	54.4 ± 0.5	2.9 ± 0.2	1.9 ± 0.1

(1) Nominal crest voltage
(2) Voltage at which breakdown occurred, average of seven consecutive determinatons
(3) Time to cross gap

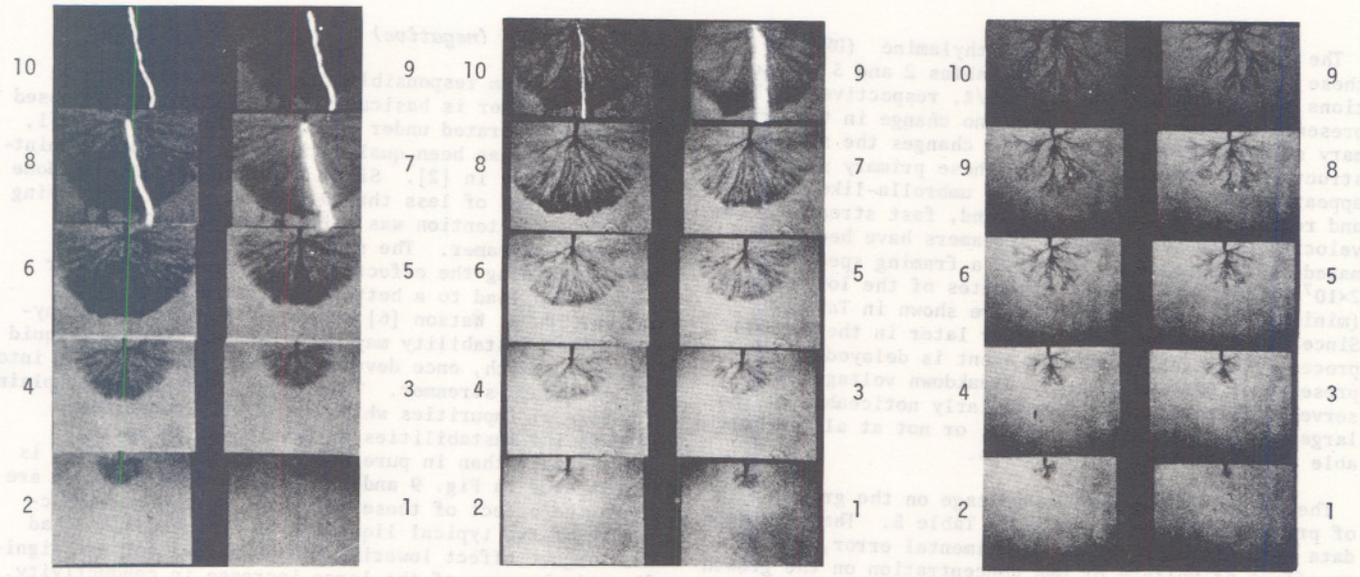
TABLE 3

Comparison of Positive Streamer Growth and Breakdown in Various Hydrocarbons Containing 0.2M Dimethylaniline

(Trapezoidal Pulse, 25°C)

Liquid	V_{Crest}, kV(1)	V_{BD}, kV(2)	t_{CG}, μs(3)	Growth Rate in cm/s x 10^{-5}
		2.54 cm gap		
n-hexane	174	141.4 ± 2.6	7.3 ± 0.6	3.2 ± 0.1
toluene	192	189.5 ± 2.3	4.0 ± 0.6	1.9 ± 0.3
Marcol 70	264	246. ± 16	6.0 ± 0.5	2.7 ± 0.9
		1.27 cm gap		
n-hexane	72	66.4 ± 11.2	4.2	3.0 ± 0.1
toluene	108	98 ± 18	5.6 ± 0.9	1.5 ± 0.2
Marcol 70	96	89 ± 8	5.0 ± 0.3	2.1 ± 0.1
		0.64 cm gap		
n-hexane	60	55.8 ± 3.0	2.0 ± 0.3	2.9 ± 0.3
toluene	96	88.2 ± 2.1	2.8 ± 0.3	1.5 ± 0.3
Marcol 70	84	81.2 ± 2.2	2.3 ± 0.3	1.9 ± 0.1

(1) Nominal crest voltage
(2) Voltage at which breakdown occurred, average of seven consecutive determinations
(3) Time to cross gap

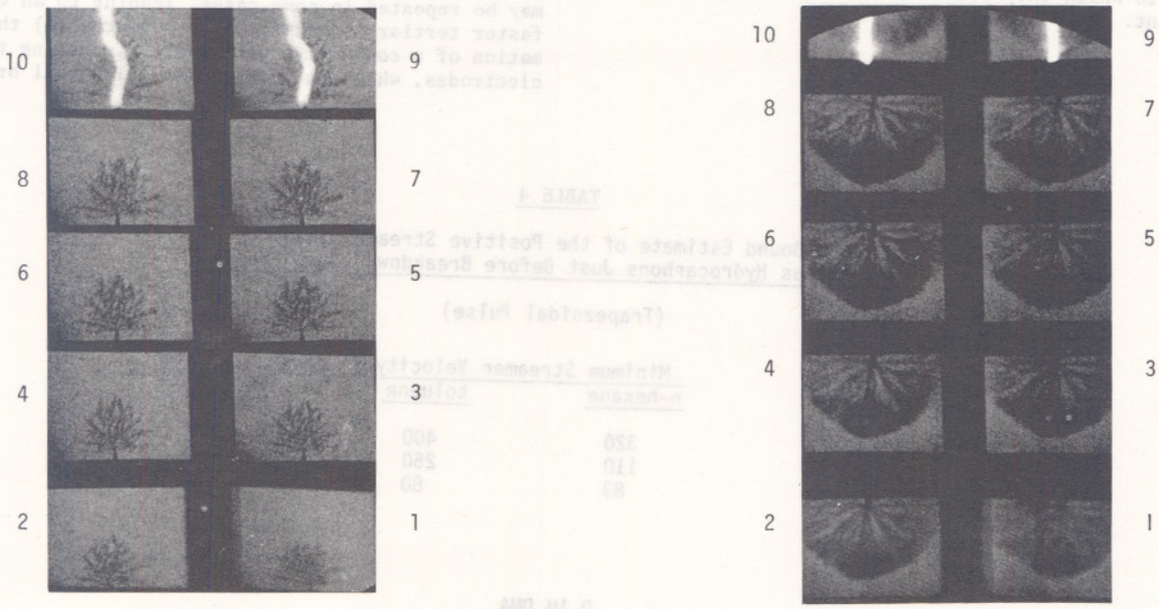


n-hexane
120 kV crest voltage

Marcol 70
96 kV

Toluene
96 kV

Fig. 7: Effect of 0.2 M dimethyl anilin on positive streamer structure (1.27 cm gap, needle anode, 2×10^6 frames/s, trapezoidal pulse)



pure n-hexane

n-hexane plus 0.1 M DMA

Fig. 8: Effect of dimethylaniline on secondary streamer growth (1.27 cm gap, needle anode, 96 kV crest voltage, 2×10^7 frames/s trapezoidal pulse)

The effect of adding N,N^1-dimethylamine (DMA) to these three fluids is shown in Tables 2 and 3 for solutions containing 0.1 and 0.2 mol/ℓ, respectively. The presence of this species causes no change in the primary streamer growth rate but it changes the streamer structure as shown in Fig. 7. These primary streamers appear to propagate in a radial, umbrella-like fashion and retard the onset of the second, fast streamer. The velocity of these secondary streamers have been estimated from photographs taken at a framing speed of 2×10^7 frames/s (Fig. 8). Estimates of the lower bound (minimum) streamer velocities are shown in Table 4. Since these fast streamers occur later in the overall process, the final breakdown event is delayed in the presence of DMA and a higher breakdown voltage is observed. This effect is particularly noticeable at the largest gap used. It is barely, or not at all, resolvable at the smaller gap spacings.

The effect of the applied voltage on the growth rate of primary streamer is shown in Table 5. The available data suggest that, within experimental error, there is no effect of voltage or DMA concentration on the growth rate. The significance of these findings is discussed in the next section.

DISCUSSION AND CONCLUSIONS

The observation and data reported in the preceding Section will be discussed in the same order since the events to which they relate are believed to be quite different.

Needle Cathode (negative)

The mechanism responsible for the formation of the primary streamer is basically the same as that proposed for thsoe generated under uniform field conditions [1, 5,6], and it has been qualitatively discussed for point-plane geometry in [2]. Since the earlier work was done at narrow gaps of less than 0.5 cm it is not surprising that little attention was paid to the advent of the secondary streamer. The studies of larger gaps and those examining the effect of impurities on streamer propagation lead to a better understanding of their origin. P. K. Watson [6] suggested that an electrohydrodynamic instability may occur at the streamer/liquid interface which, once developed, may grow very fast into the secondary streamer. This concept has helped explain the role of impurities which apparently cause these interfacial instabilities to develop at an earlier growth state than in pure dielectric liquids. This is illustrated in Fig. 9 and in Table 6a. In Table 6b are shown the effect of these additives on the ac conductivity of two typical liquids. As expected ASA 3 had the largest effect lowering the breakdown voltage significantly because of the large increase in conductivity. In summary, it appears that the development of breakdown when the needle is negative can be explained in terms of five basic steps: (1) charge carrier injection, which is still poorly understood; (2) formation and growth of the primary streamer; (3) development of an interfacial instability leading to a very rapidly growing secondary streamer that bridges the remainder of the gap. There is evidence [8] that this process may be repeated in some cases, leading to an even faster tertiary streamer which produces (4) the formation of a conductive path short circuiting the electrodes, which in turn causes (5) actual breakdown.

TABLE 4

Lower Bound Estimate of the Positive Streamer Velocity in Various Hydrocarbons Just Before Breakdown (Fast Event)

(Trapezoidal Pulse)

Gap/Liquid	Minimum Streamer Velocity in cm/s x 10^{-5}(1) n-hexane	toluene	Marcol 70
2.54 cm	320	400	300
1.27 cm	110	250	200
0.64 cm	83	80	200
	0.1M DMA		
2.54 cm	82	200	200
1.27 cm	23	120	33
0.64 cm	9	46	15

(1) Determined at lowest crest voltage at which 100% breakdown was observed.

TABLE 5

Effect of Applied Voltage and Impurities on Positive Streamer Growth Rate

(Needle Anode, 2.54 cm gap, Trapezoidal Pulse)

V_{crest}	Growth Rate in cm/s x 10^{-5} n-hexane	toluene	Marcol 70
108	3.6 ± 0.4		
120	3.4 ±0.2*		
144		1 ± 0.1	
168		1.2 ± 0.1*	1.8 ± 0.2
192		1.3 ± 0.2	1.9 ± 0.1
216		1.1 ± 0.3	2.4 ± 0.3
		0.1M DMA	
122	2.5 ± 0.1		
148	3.1 ± 0.2*		
192		1.6 ± 0.3*	2 ± 0.1
216			2.6 ± 0.4
240			2.7 ± 0.4
264			2.7 ± 0.4
276			2.8 ± 0.2*
		0.2M DMA	
148	3.1 ± 0.2		
174	3.2 ± 0.1*		
192		1.9 ± 0.3*	
240			2.3 ± 0.2
264			2.7 ± 0.2*

*First voltage at which 100% breakdown was observed.

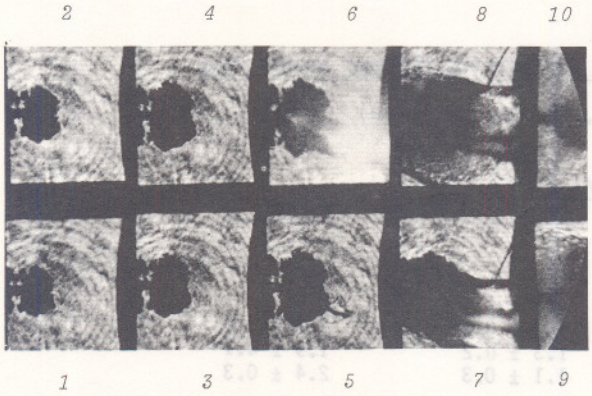
Needle Anode (positive)

The breakdown voltage observed when the needle is positive have been found to be consistently lower than those observed when the needle is negative. The observations reported here have further shown that the change in conductivity or impurity concentration has, within experimental error, no effect on the breakdown voltage or the streamer growth rate. This drastically different behavior is believed to indicate that another mechanism is involved in streamer growth rate and propagation. As suggested in the literature [2,5, 6,9,10] the convergent field literally extracts charge carriers from the bulk of the liquid towards the needle into a more intense field which causes local breakdown resulting in the formation of a conductive channel leading to the needle surface. This channel is believed to act as an extension of the needle anode [11] and the process repeats itself. This process presumably depends on both the local availability of charge carriers and the field at the tip of the channel. A comparison of streamers generated in pure n-hexane with and without DMA illustrates this point (Fig. 10). In the absence of DMA a few streamers are formed and they grow randomly into the liquid. In the presence of DMA many more thin streamers grow in a radial fashion as mentioned earlier. This pattern can be attributed to both the low ionization potential and the high electron affinity of DMA in comparison to n-hexane, Marcol and even toluene. The umbrella-like structure of conductivty streamers produces a geometry approaching that of a sphere-plane, i.e. a nearly uniform field. As pointed out earlier, the breakdown voltage of such a gap is higher than that of a sharp point plane of the same separation. This distinction then explains the difference in the observed breakdown voltage in the presence and absence of DMA.

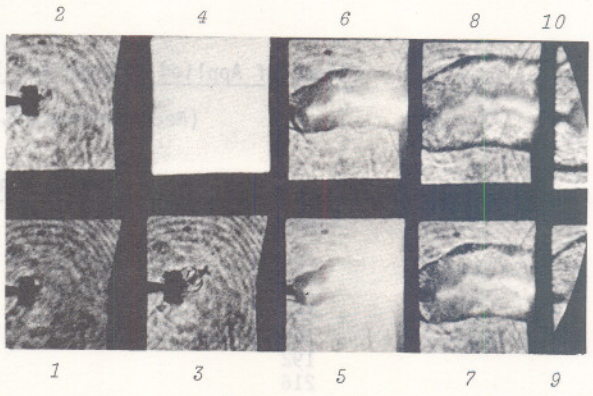
Although no visual evidence exists of the injection of charge carriers at the plane cathode, it has to be assumed that this process occurs continuously. The reason it cannot be made visible is probably due to the existence of numerous injection sites on the cathode surface each involving only small amounts of electrons which, once in the liquid, will move towards the anode needle. The fact that the presence of DMA and other impurities does not alter the rate of growth of these streamers is believed to be consistent with the above explanation. All the impurities do is to provide more charge carriers, hence more channels are formed that propagate at the same speed.

Again the final step, the fast event, is beyond the temporal resolution of the electro-optical equipment when gaps of less than 1.27 cm are used. With 2.54 cm gaps it was possible to study this event in some detail as shown in Fig. 11. The many branches forming near the cathode surface during this fast event seem to support the above argument.

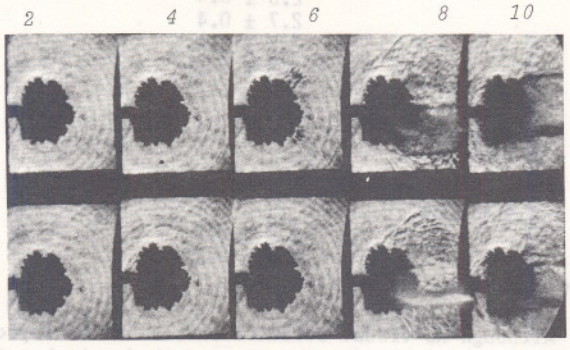
Finally it is appropriate to call attention to the fact that under the conditions discussed here, the light emission after the gap is short circuited, is much weaker, and lasts a shorter time than in the case of the breakdown when the needle is negative. It appears that less energy is stored in the gap in this case than in the latter presumably because of the difference in breakdown voltage. This conclusion is consistent with the proposed concept of thermalized electrons being "removed" from the liquid in this case while energetic electrons are "pushed" into the liquid when the needle is negative and eventually they thermalize, releasing energy to the liquid. This picture also accounts for the lower voltage at which breakdown occurs when the needle is an anode than when it is a cathode.



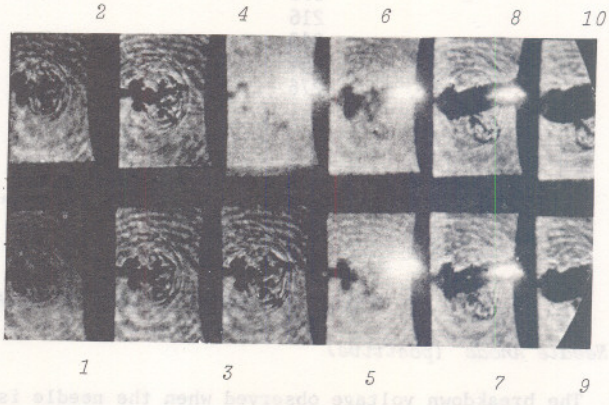
pure n-hexane
192 kV crest voltage
2×10^6 f/s



chem. grade hexane
(saturated with O_2 + H_2O
5×10^6 f/s



n-hexane + 0.15 M DMA
180 kV crest voltage
2×10^6 f/s



toluene + 20 ppm ASA 3
108 kV crest voltage
2×10^7 f/s

Fig. 9: Effect of contaminent on secondary streamer development (needle cathode, 0.3 cm gap, trapezoidal pulse)

In conclusion, it should be noted that sufficient data have been taken in this investigation and in related work by others that a consistent picture is emerging of the breakdown process. There is general agreement on the shape and velocity of the streamers, the effect of impurities, and the effect of voltage variations. The processes are complex with electron emission from the electrode, the local electrical field distribution, and the availability of sufficiently energetic electrons in the host liquid each possibly being the dominant influence on the streamer propagation at different times. Much of the data necessary to develop a quantitative, predictive model of the process now appear to be available, so it is anticipated that the next significant advances will be in process modeling.

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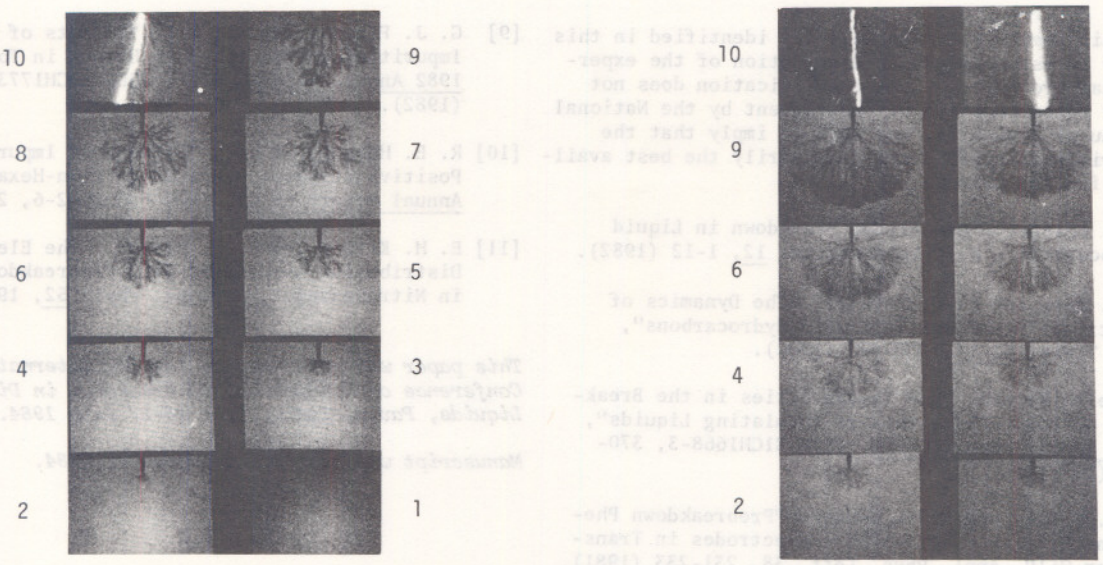
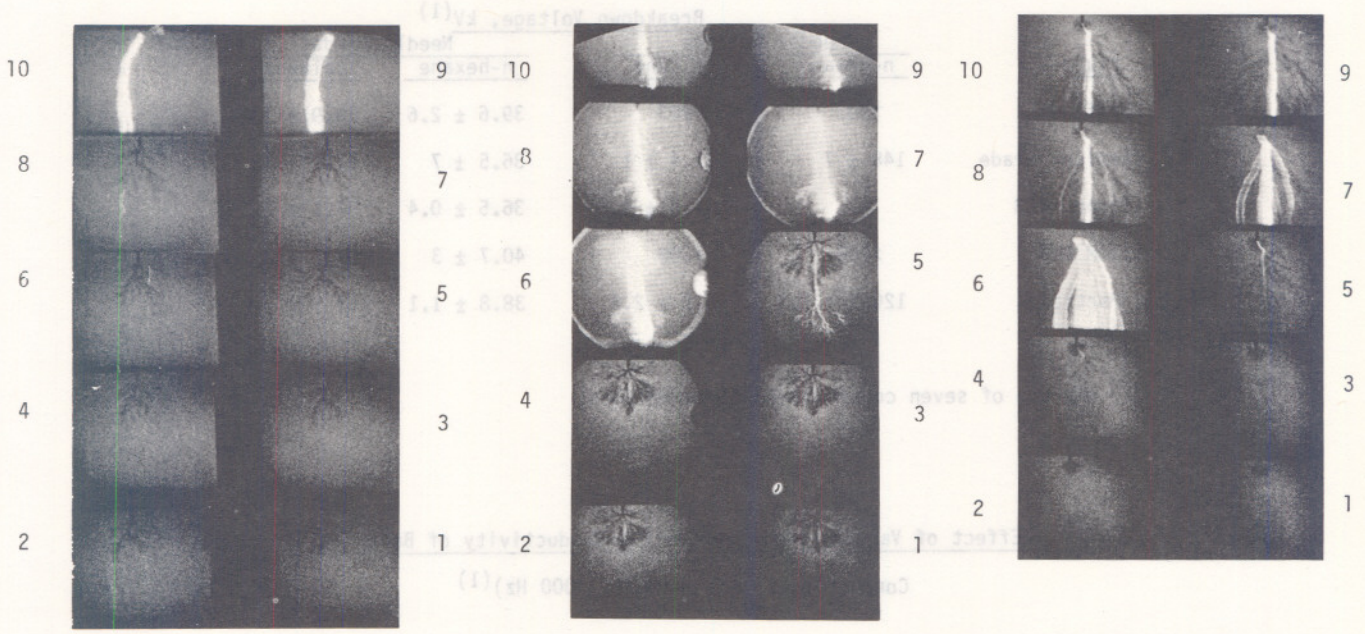


Marcol 70
108 kV

crest voltage

0.1 M DMA in Marcol 70
96 kV

Fig. 10: Comparison of primary streamer growth in pure and DMA containing Marcol 70 (needle anode, 1.27 cm gap, 2×10^6 frames/s, trapezoidal pulse)



pure n-hexane
120 kV crest voltage
2×10^7 frames/s

Marcol 70
240 kV crest voltage
5×10^6 frames/s

Toluene
216 kV crest voltage
2×10^7 frames/s

Fig. 11: Secondary streamer development in liquid hydrocarbons (needle anode, 2.54 cm gap, trapezoidal pulse)

[4] Certain commercial materials are identified in this paper to assure adequate description of the experimental procedure. Such identification does not imply recommendation or endorsement by the National Bureau of Standards, nor does it imply that the materials identified are necessarily the best available for the purpose.

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TABLE 6

A. Effect of Additives on Electrical Breakdown Voltage of Base Fluids

(25°C, 0.3 cm Gap, Trapezoidal Pulse)

	Breakdown Voltage, kV(1)			
	Needle Cathode		Needle Anode	
Fluid	n-hexane	toluene	n-hexane	toluene
Purified	>203	>203	39.6 ± 2.6	58.9 ± 1.1
Chemical Grade	148 ± 7	121.4 ± 1	36.5 ± 7	57.1 ± 18
10 ppm ASA3	-	84.5 ± 0.8	36.5 ± 0.4	57.1 ± 2
0.10M DMA	>200	>200	40.7 ± 3	57.2 ± 2
C Particles	120.7 ± 5.5	95.9 ± 2.5	38.8 ± 1.1	57 ± 2

(1) Average of seven consecutive determinations.

B. Effect of Various Additives on the Conductivity of Base Fluid

Fluid	Conductivity, Scm^{-1} (25°C, 1000 Hz)(1)	
	n-hexane	toluene
Purified	$<1 \times 10^{-16}$	6.25×10^{-14}
Chemical Grade	1.6×10^{-15}	1.1×10^{-13}
10 ppm ASA3	1.9×10^{-10}	5×10^{-10}
0.10M DMA	1.5×10^{-15}	6.3×10^{-13}

(1) Experimental error is 10%