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Time to Breakdown Studies for Liquids of Different Physico-Chemical Nature

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***Abstract*—An automated setup for testing dielectric performance of liquids in a small point-to-plane gap is presented. It provides step voltages with rise times around 30 ns, with amplitudes up to 90 kV. Breakdown studies of thirteen different liquids were conducted with a gap distance of 4 mm in a test cell containing 20 ml liquid. The liquids were circulated and filtered after each breakdown to remove carbonized particles and relax space charges. Time to breakdown versus applied voltage was recorded, and for streamers that stopped mid gap, shadow graphic images of streamer structures were taken. Multivariate correlation studies between streamer properties and physio-chemical parameters of liquids were made, indicating relevance of liquid evaporation and electronic states of molecules.**

Keywords—dielectric liquid; point to plane; impulse; breakdown voltage; acceleration voltage, time to breakdown, multivariate analysis, electronic properties

I. INTRODUCTION AND BACKGROUND

Dielectric liquids are used as insulation in transformers, capacitors, cables and other power and high voltage apparatus. In transformers, having large open volumes, mineral oils have been dominating, whereas in capacitors a variety of synthetic liquids has been used. New liquids, such as synthetic and vegetable esters and gas to liquid, are currently introduced to transformers to replace the mineral oils at all voltage levels.

IEC standards for liquid testing are written with transformer insulation in mind, but does not reveal any differences in functional properties of liquids. IEC 60156, measuring average ac breakdown voltage in a small gap with uniform field, is sensitive to particle content in a liquid and to the relative humidity, but not to liquid composition, whereas IEC 60897, measuring lighting impulse breakdown for negative lightning impulse (LI) in a point to sphere gap mainly reports differences between breakdown voltages [1].

Breakdown in a liquid occurs after a pre-breakdown plasma filled channel, called streamers, which has bridged the insulation gap. For mineral oils it is reported that the time to breakdown vary a lot depending on applied voltage level [2, 3]. Streamers at lower voltages move slowly and a propagating streamer may be quenched during the falling tail of an LI. In mineral oils, typically at twice the breakdown voltage for a step pulse, the streamer velocities will increase by one to two orders of magnitude. Then streamers may then cross a longer gap and result in breakdown during a short LI [4]. It is now well documented that the impulse withstand voltages for larger centimeter gaps, in particular for positive LI, vary depending on the liquids and their electronic properties [5-7]. These streamer velocities and the voltages where acceleration occurs are considered being functional properties of a dielectric liquid. Documentation of streamer properties in larger gaps are time consuming and expensive.

This paper describes an experimental investigation of these functional properties for a small millimeter gap under positive impulse voltages. It is a feasibility study of possibilities for ranking dielectric liquids using a millimeter sized gap for studying streamer velocities, breakdown and acceleration voltages.

Correlations between observed functional properties (breakdown, acceleration voltage and velocity) and a group of physicochemical parameters (e.g. ionization potential, first excitation level, molar mass, density, permittivity, viscosity and boiling temperature) were studied using multivariate analysis.

II. EXPERIMENTAL SETUP AND TEST PROCEDURE

The experiments were performed in a computer-controlled setup in a small point to plane gap in a 20 ml Teflon cell at room temperature using a fast rise step voltages. The point electrode - a tungsten needle - was typically etched to 5 µm point radius and the gap was set to 4 mm unless otherwise stated. A 90 kV impulse generator was used. Rise times of the positive voltage step (referred to the needle polarity) was typically 20 ns, but could increase to 40 ns at higher voltages. Step voltage length was set to 50 µs, long enough for slower streamers to cross. The liquid was circulated via two filters (a 1.2 µm prefilter and a 0.2 µm membrane filter) and a 100 ml liquid reservoir. A camera, either a fast shutter time camera (Proxitronic) or a high-resolution CCD camera with a fast diode flash (Pixelink), were installed to take shadow graphic images of the streamer structure.

The impulses were applied automatically starting at 15 kV and increased stepwise in 5 kV steps up to 80 kV, with 15 impulses per step and a waiting time of 6 minutes between each impulse. The liquid was circulated and filtered between each impulse for 3 minutes. An image was taken for each impulse, and the voltage was recorded. From the voltage shape, the time to breakdown was recorded and the average streamer velocity calculated assuming streamer initiation at 90% peak voltage. This was done automatically, typically with 180 impulses per liquid during an overnight test sequence.

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III. Tested Liquids

The liquids were used as they arrived, directly from bottles or drums. The following liquids were tested (split in three groups). The seven hydrocarbon liquids comprised mineral oils (Marcol 52, Nytro 10XN and Primol 352), neat hydrocarbons (Cyclohexane, n-heptane, Benzene) and capacitor liquids (Jarylec C101). The halogenated liquids were trichloroethylene and three perfluorpolyethers (Galden HT135, HT200 and HT 230). Finally, two esters were tested, being methyl butyrate and Midel 7131. In one case the liquid (Galden HT230) were degassed without making any difference in the results.

IV. Experimental Results

Fig. 1 shows results from the testing of n-Heptane. In this case, there was a clear difference between the breakdown voltage and the acceleration voltage. For some liquids, fast streamers (4th mode) occurred at breakdown voltage, resulting in no recording of 2nd mode streamers, but shadow graphic pictures revealed that streamers were initiated below breakdown voltage, except for Midel 7131. In other cases even 90 kV was not enough to produce the faster mode streamers. Tip erosion was observed during the test sequences. In some cases, with a thinly etched needle, the length of the gap could extended from e.g. 4 mm to 5 mm in the end of the test sequence. n-heptane showed a strong tip erosion, so gap length ended longer than in the start, resulting in a too low velocity for the fast mode streamer. Fig. **2** shows the average velocities of all the tested liquids.

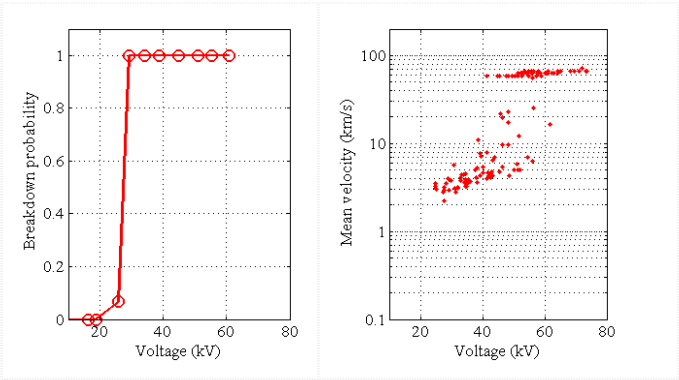

Fig. 1: Breakdown probability and calculated velocity versus step voltage for streamers in n-Heptane

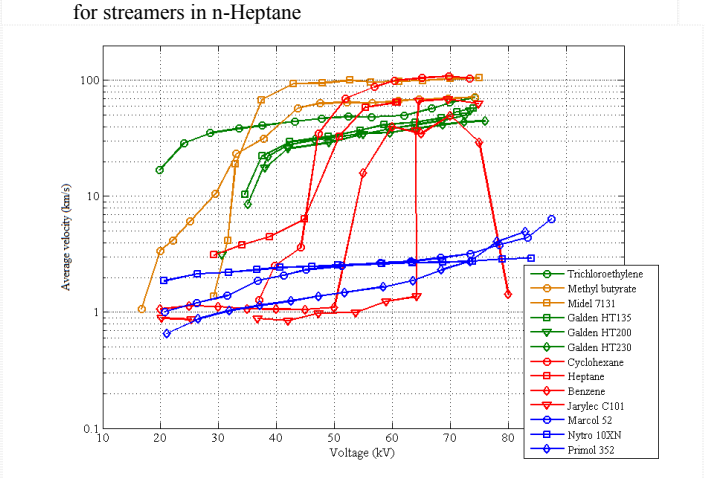

Fig. 2: Average streamer velocities for all tested liquids versus applied voltage, velocities are not corrected for tip erosion.

Shadow graphic images taken with 60 µs exposure time and 3 µs flash is shown in Fig. 3. These pictures are taken from streamers not going to breakdown. When breakdown occurred also light emission from the breakdown would lead to a "double exposure". Branching varied from liquid to liquid. Those showing higher degree of branching would normally show a high acceleration voltage.

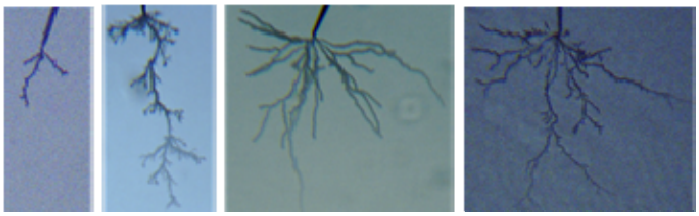

Fig. 3: Images of 2nd mode streamers from different liquids. From left to right: Trichloroethylene, Galden HT230, Jarylec C101 and Nytro 10XN.

The breakdown voltage is defined as the voltage where it is 50 % probability for a breakdown to occur. The acceleration voltage is defined as at the voltage in which the average velocity increases significantly for a slightly higher voltage. For the mineral oils, acceleration could not be reached with the available voltage and acceleration voltages were therefore estimated for these liquids. One can also see from Fig. 2 that for some liquids it is difficult to find a difference between breakdown and acceleration voltages. With these assumptions and limitations the ratio between acceleration voltages and breakdown voltages were calculated and plotted as seen in Fig. **4**.

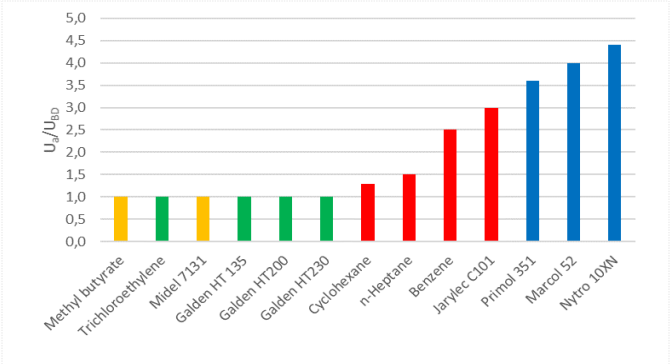

Fig. 4: Ratio between acceleration voltage U_a and breakdown voltage U_{BD} for the different liquids

V. Correlations between results and physio-chemical parameters

No fixed model based on physiochemical parameters (i.e. breakdown and acceleration voltages and streamer velocities) exists that can be used for estimating functional properties of a liquid. One expect that electron avalanches plays a role in joule heating of the liquid needed to form extensions of the plasma channels during propagation. Correlation analysis between measured properties and physiochemical parameters of the liquids we consider relevant, were attempted. These parameters and properties are shown in Table 1.

TABLE 1: PROPERTIES AND PARAMETERS USED IN THE STATISTICAL ANALYSIS

Observed Property	Molecular parameter	Physical parameter
Breakdown voltage	Ionization potential for the base liquid	Density of base liquid
Acceleration voltage	First excitation level for the base liquid	Relative permittivity
Velocity at breakdown	Molar mass	Thermal conductivity
Velocity at acceleration	Cut-off for UV absorption	Dynamic viscosity
Velocity for 4th mode		Heat capacity
		Boiling temperature
		Evaporation enthalpy

A full set of properties was only available for trichloroethylene, methyl butyrate, cyclohexane and benzene. For these a correlation study was done, with results as seen in Table 2.

TABLE 2: CORRELATION TABLE FOR NEAT LIQUIDS

	Functional properties				
Liquid	U_{BD}	U_a	V_{BD}	V_a	V_{4th}
ionization potential	34	53	30	28	61
1st excitation level	89	36	72	67	**100**
molar mass	33	81	95	95	49
liquid density	43	55	**100**	**100**	68
cut-off for UV abs	54	1	75	73	83
permittivity	51	88	19	15	24
thermal cond.	19	34	85	88	16
dynamic viscosity	97	59	52	47	90
heat capacity	2	43	50	51	40
boiling temperature	41	86	11	9	12
evaporation enth.	59	81	12	8	29

The correlation coefficients in Table 2 gives an indication on how well the observed properties correlates with changes in a parameter. The functional properties are breakdown (U_{BD}) and acceleration (U_a) voltages, velocity at breakdown voltage (V_{BD}), acceleration (V_a) and 4th mode (V_{4th}) streamer velocity. A value of 100 indicate a perfect correlation. A scatter plot, as shown in Fig. 5, will elucidate this correlation, which in this case was very clear. On the other hand the apparent correlations between breakdown and acceleration voltages and liquid density showed a horizontal line i.e. it had no influence at all.

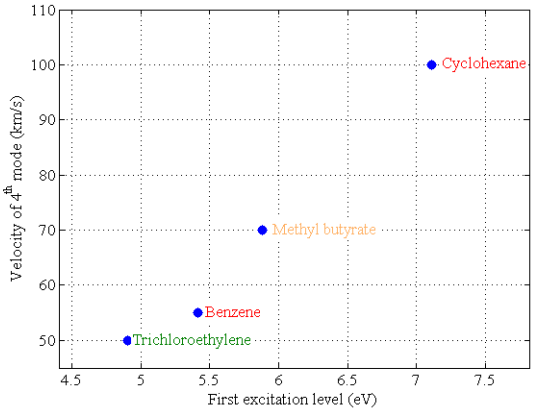

Fig. 5: Correlation between 4th mode streamer velocity and 1st excitation level. The velocities are adjusted for tip erosion.

One problem with this approach occurs for complex liquids such as mineral oils for which well-defined singular molecular parameters does not exist. When increasing the data set to comprise also n-Heptane, Jarylec and the perfluorpolyethers the data on ionization potential and 1st excitation level, UV absorption, viscosity and evaporation enthalpy was incomplete. However, some correlations among the other data were suggested in the analysis:

- Good correlations of breakdown voltage to permittivity.
- Good correlation between 4th mode velocity and molecular mass, density, thermal conductivity, heat capacity and boiling temperature.

When finally comprising all liquids in the correlation study we only had data for density and permittivity for all the liquids. High correlation coefficients could then be found for streamer velocity at breakdown and at 4th mode propagation versus density.

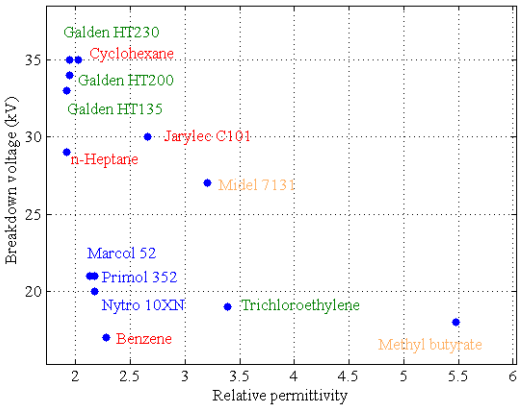

Fig. 6: Correlation between breakdown voltage and permittivity

VI. DISCUSSION

A. Test method

The method looks to have a potential to differentiate between liquids and their functional properties in a small gap, particularly the streamer velocities for different modes. However looking at Fig. 2, we see that for some liquids, the velocities at lower voltages are not documented, and for other liquids (i.e. mineral oils) the acceleration voltage and 4th mode velocity could not be documented.

The major challenges for these experiments were on one hand the complications arising from using a short gap, stressing the needs for accuracy in adjusting distances and for accurate timing of streamer inception: With a rise time of 20-40 ns and the crossing time for a fast mode streamer in the 4 mm gap becomes some 40 ns, which is comparable to the rise time. A large part of the propagation may occur at the rising flank making calculation of velocities inaccurate. However, one will reveal the acceleration voltage. On the other hand, the gap was too long to acquire fast mode breakdown for the maximum available voltage. We attempted to reduce the gap to 3 mm and further down to 2 mm and could then see acceleration for Nytro 10XN at 55 kV at 2 mm. Reducing the gap will result in

problems with determining crossing time for the faster streamers, so a better solution would be to increase the test voltages, which was impossible in this setup.

Tip wear was a problem in the experiments. This could be improved by reducing the energy released during breakdown by reducing the capacitances in the impulse source and by introducing a series resistor. However, a series resistor will increase rise time of the voltage making timing even more inaccurate. Effects on streamer initiation will not be critical. As long as the tip radii is above the critical radius for a liquid [8], in the range 5 - 10 µm one will not see 1st mode, only 2nd mode at initiation and rise time is seen to have little importance on inception for the 2nd mode streamers. One should anyway avoid too sharp tips as they wear quickly. It is therefore advisable to have radii above the critical radius. We found tips in the 5 - 10 µm range to be quite stable.

Velocities were now calculated based on time delay from voltage application (50 %) to breakdown. The camera with a long exposure time (60 µs) gives images (see Fig. 3) that can be used to detect streamer length for non-breakdown streamers so better ratios between fast and slow mode can be determined. In both these approaches, we assume that streamers start without any delay at the rising flank of the voltage, as the tips are sharp.

B. Analysis of liquid behavior

The set-up was designed for quick and low-cost data harvesting based on an idea that streamer parameters like velocities, breakdown and acceleration voltages determined from small scale has a relevance for streamer propagation in larger gaps. More research remain before this can be claimed. On one hand, it is known that the breakdown voltage, and the gap dependence of the ratio between acceleration and breakdown voltage is liquid dependent [9, 10], on the other hand the large ratio seen for mineral oils in larger gaps is also found for these short gap experiments.

The imaging gives information about branching that seem to have a correlation to acceleration. To get a quantitative measure of the branching one could calculate the fractal dimension of the 2nd mode. The images can, as discussed above, also give information in addition to that shown in Fig. 2.

In lack of good models for streamer behavior, multivariate analysis offers a way of elucidating which parameters that have an influence the liquids properties. It is well known that the streamer channels are established through evaporation of base liquids, and one hypothesis is that electronic avalanches that occur around the streamer head is the motor in the energy conversion from electrostatic potential energy to heat.

The documentation of thermodynamic parameters of liquids are quite complete, and we see that parameters such as molecular mass, density, thermal conductivity, heat capacity and boiling temperature has a good correlation to the velocity of the fast propagation mode velocity as was expected.

However, when it comes to molecular electronic parameters data is much scarcer. For complex liquids, like mineral oils, it is impossible to come up with singular values. For neat liquids and for additives some data can be found in literature, or can be found from modeling [11]. As shown in Table 2, 1st excitation level show very good correlation to 4th mode velocity, indication that light emission and absorbance has an influence on the heat production at the streamer tips. The scatter plot gives more confidence in this hypothesis. This may of course be a singularity for these four liquids, not being generally valid. However, such analysis gives a good idea on where to direct further work to establish good physical models.

VII. Conclusion

A set-up and method for investigation small-scale dielectric properties for liquids has been built. Further work remain to show how these properties can be representative for larger scale streamer propagation.

Statistical analysis indicate that thermodynamic and electronic properties of liquids influence streamer propagation velocities.

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