Field dependent conductivity of n-pentane

Hestad, Ø; Linhjell, D; Lundgaard, L ; Samseth, J Ø & Unge, M

Citation for the published version (APA 6th)
Hestad, Ø. L. G., Linhjell, D., Lundgaard, L. E., Samseth, J. Ø., & Unge, M. (2017). Field dependent conductivity of n-pentane 2017 IEEE 19th International Conference on Dielectric Liquids - ICDL (pp. 4): IEEE conference proceedings.
DOI: 10.1109/ICDL.2017.8124606

Field dependent conductivity of n-Pentane

Oystein. L. Hestad, Dag Linhjell, and Lars E. Lundgaard
SINTEF Energy Research
Trondheim, Norway
oystein.hestad@sintef.no

Jarl Ø. Samseth
Department of physics
Norwegian University of Science and Technology (NTNU)
Trondheim, Norway

Mikael Unge
ABB Corporate Research
Västerås, Sweden

***Abstract*— A sensitive differential charge measurement technique was used to measure the conduction current from a sharp needle in a needle plane geometry in n-pentane. The current was measured at voltages up to the threshold for streamer inception in n-pentane with a positive needle polarity. The measured current was compared to finite element models to link the recorded charge with conductivity models for the liquid. A good fit was obtained using the classical Poole-Frenkel conductivity model.**

Keywords—Pre-inception current; Field-dependent conductivity; Finite Element Modeling; Streamers

I. Introduction

The streamer phenomenon in dielectric liquids is the precursor for dielectric breakdown in liquid filled high voltage apparatuses due to high overvoltage [1]. Determining the factors responsible for inception and propagation of this phenomenon is therefore important.

Experimental investigations of the phenomenon show that it consists of a low-density region propagating in the the liquid from a high field region [1], [2]. The low-density region must be at least partly conductive to move the potential from the high field region where the streamer is incepted (i.e. needle electrode or protrusion/contaminant in the insulation material). Both inception and propagation thus require a phase transition from the liquid phase to a conducting phase (plasma), as well as charge buildup at the tip of the streamer of sufficient magnitude to move the electric field into the liquid and facilitate further charge generation in front of the streamer [1], [3], [4].

The electric field distribution in the liquid is clearly important. Estimating the field distribution is typically done by using analytical approximations based on prolate spheroidal or other coordinate systems, or numerical approximations using finite element solvers [5]–[8]. In either case, the approximation is typically based on solving the Laplace equation, without considering charge redistribution caused by nonlinear conduction of the liquid. For most dielectrics the conductivity increases exponentially with the field [9] which can be approximated by

$$\sigma(E)= \sigma_0 \exp(k\, E^{1/n}), \qquad (1)$$

where σ_0 is in [S/m], and k and n are constants. Note that this equation is greatly simplified, as in reality the conductivity will be highly temperature dependent. The majority of field dependent conductivity models can be well fitted to a simplified formula like (1), among these are hopping conduction (n=1), Pole-Frenkel and 1D-Onsager (n=2). n=3 is included because it provides the best fit to some polymers, and n-tridecane. The effect of field dependence on the excitation energy and the ionization potential on conductivity is a possible explanation for such field dependence [10]. Field dependent conductivity reduces the time constant of the dielectric, causing charge redistribution in the liquid on much shorter timescales. Thus for most dielectrics, field-dependent conductivity of the liquid must be included when calculating the field distribution in the dielectric even for very fast transients [9].

This work has been supported by The Norwegian Research Council under the contract 228850/E20, ABB and Statnett.

II. Materials and Methods

A. N-pentane

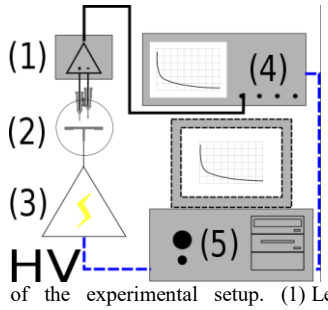
Experiments were performed in spectroscopic grade n-pentane. N-pentane was selected based on the large number of streamer experiments performed in this liquid [11], [12]. No preconditioning of the liquid was performed. Prior to the experiments, the low-field conductivity and permittivity of the n-pentane was measured using an IRLab measurement cell. The relative permittivity was 1.82, while the low field conductivity at room temperature was ~13 pS/m.

B. Experimental setup

A detailed description of the experimental setup can be found in [13]. A sketch of the setup is shown in Fig, 1.

The experimental setup consisted of a test cell with a sharp (~2 µm radius) and a blunt (~1 mm radius) needle over a plane electrode with a needle-plane gap of 5 mm. The sharp needle electrode was made by electrochemical etching of a 100 µm diameter tungsten wire. Both the needle and probe electrode were examined under a SEM microscope before and after the experiments.

A high voltage pulse was applied to the plane electrode. Capacitors were used to integrate the current from the needle and the probe, as described in [13], [14]. A differential amplifier subtracted the signal from the probe to remove the displacement current, which is typically several orders of magnitude larger than the conduction current. The setup was calibrated at low voltages. The sensitivity of the charge measurement using this setup is better than 0.2 pC.



Fig, 1. Drawing of the experimental setup. (1) Lecroy DA 1855A Differential amplifier, (2) Test cell, (3) High voltage pulse battery, (4) Tektronix TDS540 Oscilloscope, (5) Computer.

C. *Finite Element Analysis*

Finite element analysis (FEA) were performed using COMSOL Multiphysics with the AC/DC module (electrical currents interface). A 2D axial-symmetrical model based on the geometry of the test-cell was made. The charge buildup on the needle during and after the voltage rise was found by integrating the displacement field over the surface of the needle. This was performed with a constant conductivity (1e-12 S/m) and with a field dependent conductivity. The results obtained with a constant conductivity was then subtracted from the results obtained with a field dependent conductivity, at the same applied voltage, in order to subtract the displacement current. This is the same principle as performed experimentally where the displacement current is subtracted by using a blunt probe (low field) and a differential amplifier, and the resulting charge versus time obtained by FEA can thus be compared with the experimental results. The model only include charge transport through the field dependent conductivity of the liquid, electrohydrodynamic (EHD) motion is not included in this model.

A parametric sweep where the tree parameters in (1) was varied was run to form a basis for comparison between the measured current and the calculated current for three conductivity models (n=1, n=2 and n=3). All post processing was performed in Matlab where unconstrained nonlinear optimization method was used to fit the three conductivity models to the experimental results.

III. Results and Discussion

A. *Polymeric growth on needle electrode*

SEM images of the needle and the probe before experiments, and the needle after a series had been performed, are shown in Fig. 2. Note the growth of a polymeric substance on the needle over time. This is in line with what has been observed previously in n-Tridecane [15]. As a clear image of the substance growing on the needle was obtained using SEM, the substance must be at least partly conducting. The effect of the polymeric layer is evident in Fig. 3, where three measurement series using the same needle are shown. At a given voltage, the recorded current is higher for the first series than the later series. A small shift towards lower currents is also seen in going from the 2nd to the 3rd series. Previous measurements in n-tridecane indicate that the growth of a polymeric substance on the needles is linked with inception of streamers, no such layer has been observed prior to inception of streamers. The polymerization is thus probably due to formation of radical species during the inception and growth of streamers. The first streamers occured at 9 kV (20 % probability) for the first series (Fig. 4). Thus as the voltage was increased in steps for each series the first results up to 9 kV in series one should not be influenced by the polymeric layer.

In the following, the results obtained in the first series have been used, as this series has the least amount of polymer on the needle electrode.

B. *Measured conduction currents*

The average measured current over the first 50 µs after the end of the rise of the pulse has been plotted in Fig. **5**. These average currents were used for the initial fitting of the results to conductivity formulas. The resulting currents based on FEA using the following formulas for the field dependent conductivity are shown in the figure:

$$\sigma(E)=1.86\cdot10^{-10}\exp(0.0245\,E^{1/3}) \quad (2)$$

$$\sigma(E)=10^{-9}\exp(0.0011\,E^{1/2}) \quad (3)$$

After minimizing the error the resulting charge on the needle in the simulation and from the experiments were compared, the results are shown in Fig. 6. Clearly, it is difficult based on these measurements to distinguish between the two formulas

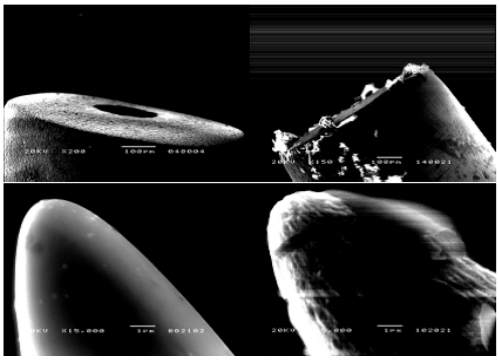


Fig. 2. SEM pictures of the needle and the probe before (left column) and after (right column) current measurements. The effective curvature of the needle increased from 1.9 to ~3 µm during the experiments.

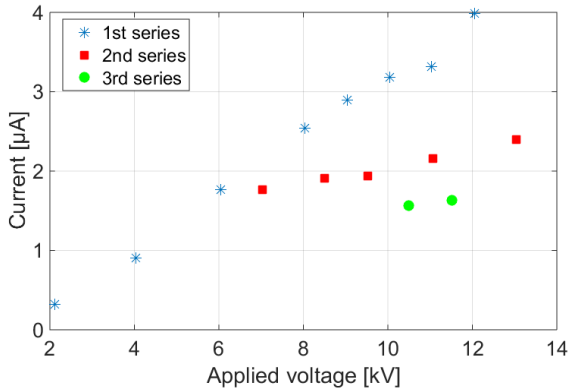


Fig. 3 Average pre-inception currents during the first 50 µs following the voltage rise. Note the decrease in current for group 2 and 3 compared to group 1. Within each group, the experiments were conducted from low to high voltage.

given above. Both provide reasonably good fits to the average current as well as the overall shape of the charge vs. time. Note that prefactor in the exponential in eq. (3) can be calculated directly from classical Poole-Frenkel conductivity, which provides a theoretical foundation for the measured current [16].

C. *Space charge limited fields in n-pentane*

Based on the field dependent conductivity of n-pentane, the space charge limited field (SCLF) as a function of frequency of the applied voltage can be calculated using [9]:

$$E_{lim}=(k^{-1}\ln(\varepsilon_r\varepsilon_0\omega/\sigma_0))^n \quad (4)$$

where E_{lim} is the SCLF, ω is the angular frequency of the applied field (approximately given by the inverse of the rise time for a step voltage), and the rest are constants from (1). The SCLF can also be found graphically by comparing the time constants for the dielectric with the frequency of the applied voltage, $1/\omega= (\varepsilon_r\varepsilon_0)/\sigma(E_{lim}) \rightarrow \sigma(E_{lim})= \varepsilon_r\varepsilon_0\omega$. The result for three dielectric liquids, for which the field-dependent conductivity has been calculated based on the experimental and computational procedure presented in this paper, is shown in Fig. 7. Of the three liquids, n-pentane has the lowest SCLF of approximately 120 MV/m and 170 MV/m for (3) and (2) respectively. Note that for n-pentane, the time constant based on the low field conductivity (13 pS/m) is ~1.3 s, while taking the field dependent conductivity into account the conductivity may reach 10^{-4} S/m or higher at fields above 100 MV/m giving a dielectric time constants of ~160 ns or less.

Fitting the average current (Fig. 5) and the shape of the charge recordings (Fig. 6) provides a basis to determine the formula for field-dependent conductivity. Measurements on n-tridecane indicate that for this liquid a conductivity formula similar to (2) gives the best fit. An argument for this based on

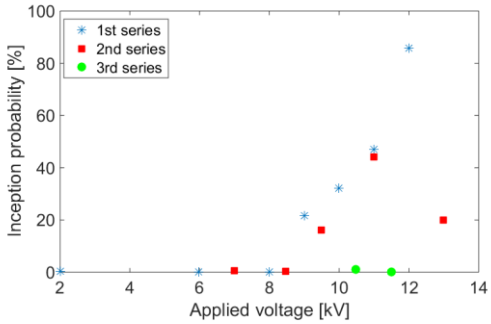


Fig. 4. Inception probability in n-pentane versus applied voltage.

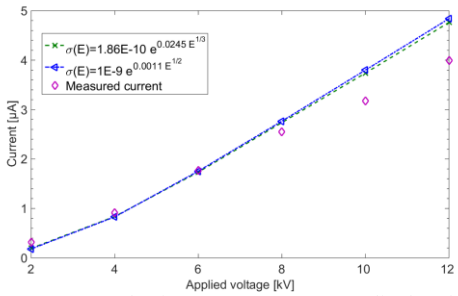


Fig. 5. Average conduction current versus amplitude of applied voltage compared with FEA based on two formulas for field-dependent conductivity.

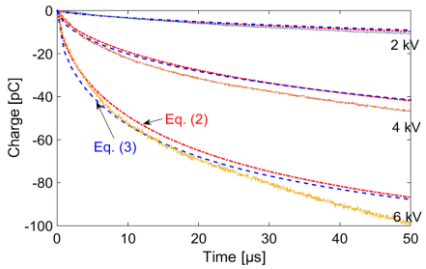


Fig. 6 Comparison between the two conductivity models and the recorded charge. Yellow solid lines represents experimental data, dotted lines are calculated based on (2) and the dashed lines are calculated based on (3).

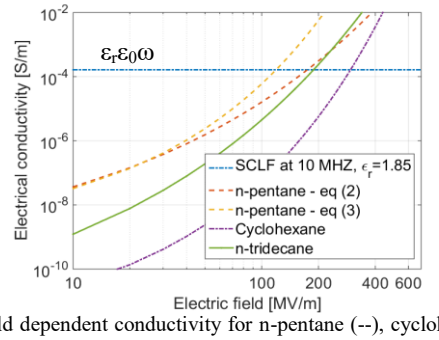


Fig. 7. Field dependent conductivity for n-pentane (--), cyclohexane (-.) and n-tridecane (-) versus field. The intersection with the horizontal line gives an approximate value for the SCLF at 10 MHz.

the field dependence of the 1st excitation energy and the ionization potential was given in [17]. The similarity between the two molecules (n-pentane and n-tridecane) suggests that a similar conduction mechanism may be relevant for n-pentane. Still the experimental results for n-pentane and n-tridecane differ somewhat. While n-tridecane gave a reasonable fit to an equation similar to (3), the prefactor in the exponential (k) could not be related to the standard Poole-Frenkel conductivity model [15]. However, for n-pentane, the calculated prefactor based on the permittivity of n-pentane is is a near-perfect match to that obtained through fitting the measured currents with the the numerical model. Based on this, we believe (3) to be the best model for the high field conductivity of n-pentane. The reasonable fit to two differing conductivity models, does, however highlight the uncertainty in these measurements. As the SCLF is frequency dependent one possible way to distinguish between conductivity models would be to change the rise times of the applied voltages. This frequency dependence of the field distribution is also likely to be cause of the rise time dependence of the inception probability in n-pentane (and other dielectric liquids). The experiments performed under positive polarity should be repeated with negative polarity in n-pentane, to check if (2) and (3) still holds. If the current is caused by high-field conductivity in the bulk this should be the case. For n-tridecane similar current magnitude were measured under positive and negative polarity [15]. For cyclohexane, larger currents were measured for positive than for negative polarity above a threshold voltage, but similar magnitude were measured below this threshold regardless of polarity [15].

As stated above, the SCLF is important to estimate the field distribution in the dielectric before and during propagation of a streamer. The combination of a high electric

field and high conductivity results in a region of high power dissipation near the point electrode, which causes a rapid increase in the temperature in the liquid and may be the reason for inception of slow streamers. The temperature increase in n-pentane with a voltage rise time of 40 ns and amplitude of 9 kV is shown in Fig 8. Note that the calculation did not take any convection in the liquid into account, and the maximum value may thus be somewhat high. The effect of convection on the ns scale should, however, be limited. The temperature increase near the point electrode may be responsible for bubble formation, in which subsequent discharges occur, leading to a 1st mode streamers.

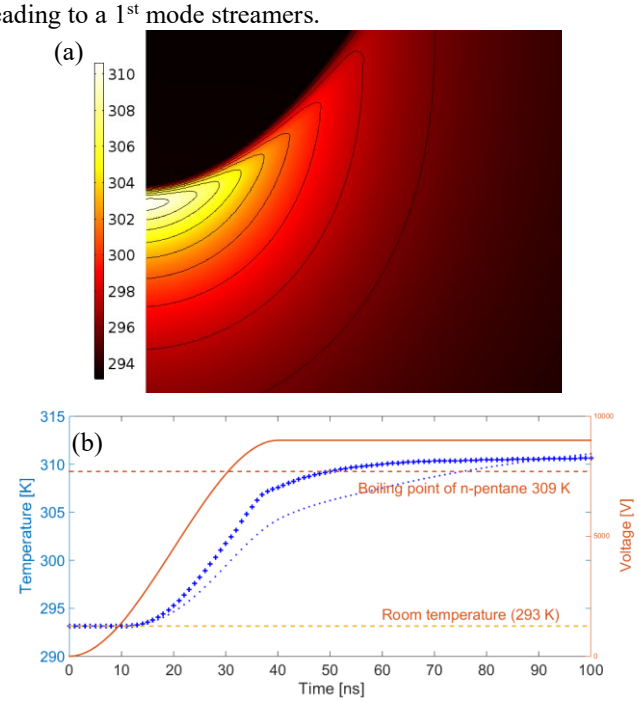


Fig 8. (a) Temperature distribution with contour lines near point electrode at t=100 ns with conductivity given by (3). (b) Maximum temperature along symmetry axis with conductivity given by (2) (+) and (3) (·), and applied voltage over point plane gap (-).

IV. Conclusion

Pre-inception currents in n-pentane were studied using a sensitive differential charge measurement technique. When comparing the measured currents with those computed using Finite Element simulations based on established models for field dependent conductivities, the measured currents fit well to standard Poole-Frenkel conduction. Based on the high field conductivity formula (3), a SCLF of 120-170 kV/mm for n-pentane can be deduced for fast rise times. This indicates that the streamer inception fields reported in literature in liquids never occur due to the formation of a SCLF in the high field region. The energy dissipation in the high field region, given such a field dependent conductivity, is sufficient to explain bubble formation in n-pentane.

Acknowledgment

The authors would like to thank Torstein Grav Aakre for performing the IRLAB conductivity measurements.

References

[1] O. Lesaint, "Prebreakdown phenomena in liquids: propagation 'modes' and basic physical properties," *J. Phys. D. Appl. Phys.*, vol. 49, no. 14, p. 144001, 2016.

[2] A. Denat, "High field conduction and prebreakdown phenomena in dielectric liquids," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 13, no. 3, pp. 518–525, 2006.

[3] O. L. Hestad, A. Vuong, I. Madshaven, and P.-O. Astrand, "Thermal decomposition of cyclohexane by Kinetic Monte Carlo simulations and its relevance to streamer formation," in *CEIDP*, 2016, pp. 404–407.

[4] P. Gournay and O. Lesaint, "On the gaseous nature of positive filamentary streamers in hydrocarbon liquids. II: Propagation, growth and collapse of gaseous filaments in pentane," *J. Phys. D. Appl. Phys.*, vol. 27, no. 10, pp. 2117–2127, Oct. 1994.

[5] R. Coelho and J. Debeau, "Properties of the tip-plane configuration," *J. Phys. D. Appl. Phys.*, vol. 4, no. 9, p. 305, Sep. 1971.

[6] J.-W. G. Hwang, "Elucidating the mechanisms behind pre-breakdown phenomena in transformer oil systems," *Thesis (Ph. D.)--Massachusetts Inst. Technol. Dept. Electr. Eng. Comput. Sci.*, 2010.

[7] H. Yamashita, K. Yamazawa, and Y. S. Wang, "The effect of tip curvature on the prebreakdown streamer structure in cyclohexane," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 5, no. 3, pp. 396–401, 1998.

[8] N. V. Dung, H. K. Høidalen, D. Linhjell, L. E. Lundgaard, and M. Unge, "Effects of reduced pressure and additives on streamers in white oil in long point-plane gap," *J. Phys. D. Appl. Phys.*, vol. 46, no. 25, p. 255501, Jun. 2013.

[9] S. Boggs, "Very high field phenomena in dielectrics," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 12, no. 5, pp. 929–938, 2005.

[10] I. Madshaven, H. S. Smalo, M. Unge, and O. L. Hestad, "Photoionization model for the transition to fast mode streamers in dielectric liquids," in *CEIDP*, 2016, pp. 400–403.

[11] P. Gournay and O. Lesaint, "A study of the inception of positive streamers in cyclohexane and pentane," *J. Phys. D. Appl. Phys.*, vol. 26, no. 11, pp. 1966–1974, 1993.

[12] O. Lesaint and P. Gournay, "Initiation and propagation thresholds of positive prebreakdown phenomena in hydrocarbon liquids," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 1, no. 4, pp. 702–708, 1994.

[13] O. Hestad, L. Lundgaard, and D. Linhjell, "New experimental system for the study of the effect of temperature and liquid to solid transition on streamers in dielectric liquids: application to cyclohexane," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 17, no. 3, pp. 764–774, Jun. 2010.

[14] L. Dumitrescu, O. Lesaint, N. Bonifaci, A. Denat, and P. Notingher, "Study of streamer inception in cyclohexane with a sensitive charge measurement technique under impulse voltage," *J. Electrostat.*, vol. 53, no. 2, pp. 135–146, 2001.

[15] O. L. Hestad, P.-O. Astrand, and L. Lundgaard, "N-tridecane as a model system for polyethylene: comparison of pre-breakdown phenomena in liquid and solid phase stressed by a fast transient," *IEEE Trans. Dielectr. Electr. Insul.*, vol. 18, no. 6, pp. 1929–1946, Dec. 2011.

[16] L. A. Dissado and J. C. Fothergill, *Electrical Degradation and Breakdown in Polymers*. IET, 1992.

[17] H. S. Smalø, Ø. Hestad, S. Ingebrigtsen, and P.-O. Åstrand, "Field dependence on the molecular ionization potential and excitation energies compared to conductivity models for insulation materials at high electric fields," *J. Appl. Phys.*, vol. 109, no. 7, p. 73306, Apr. 2011.