# Features of Current Passage Processes in Liquid Dielectrics at the Injection and Dissociation Mechanisms of Charge Formation

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Abstract—The paper presents the results of the experimental research into the effect of charge formation mechanism (CFM) on the features of current passage processes in liquid dielectrics. The current-voltage characteristics obtained with the sawtooth voltage modulation (the dynamical current-voltage characteristics) underlie the investigation. The special method for the experimental revealing the criteria of the prevalence of the injection or dissociation CFMs was developed and described in detail. Two limiting cases when only one CFM prevailed were realized and investigated. A number of features of current passage processes revealed in the numerical research earlier were experimentally examined and confirmed.

Keywords—Dynamical current-voltage characteristic, hysteresis, field-enhanced dissociation, conductivity

### I. INTRODUCTION

Liquid dielectrics are of a wide use in high-voltage equipment both as insulation (e.g., in transformers) and as the working liquid (e.g., in electrohydrodynamic devices) with the electric conductivity of liquid being one of the most important parameters in both cases. The conductivity is non-linearly dependent on the voltage [1], determined by the aggregate of ion transport mechanisms and charge formation ones, affected by the external conditions [2] and varies when introducing various particles (e.g., seeding ones) into the bulk [3], [4]. The non-linearity of the conductivity in high-voltage range is caused by both the injection (e.g., [5], [6]) and field-enhanced dissociation (or the Wien effect) [7], with it being unknown a priori what is more important. However, the computeraided design of electrohydrodynamic (EHD) devices calls for the answers on the following questions: which charge formation mechanism (CFM) is a prevalent one?, how do correlate their roles?, or which one can be neglected? To answer the questions, it is necessary to develop criteria for revealing emergence of one or another CFM and propose the experimental method, letting one examine the developed criteria. Moreover, the criteria that undemand complicated experimental equipment are the foreground ones since they can be implemented by a wide circle of researches.

Presently, the mentioned questions are topical ones since ascertaining the prevailing CFM is necessary for improving performance of EHD devices and, especially, EHD pumps [8]–[10] and atomizers [11], [12]. However, revealing the prevailing CFM presents a complicated problem in practice, and, when designing an EHD device, it is often believed that the latter operates owing to the one mechanism while it actually works due to another one. Besides, the field-enhanced dissociation is used to withdraw from consideration when designing EHD devices (e.g., [13]).

The answers on the designated questions have not been obtained yet despite their high theoretical and practical value with the corresponding issue being insufficiently discussed in the literature. However, prospects for the development of the subject area have appeared recently due to the improvement of the technique for solving the set of EHD equations in the most complete approach [14]-[17], i.e., with concurrent consideration of the injection and dissociation mechanisms of charge formation. Some features of EHD flow structure at different CFMs have been already revealed basing on the computer simulation [17]. The present research is of a particular interest since the features of current passage processes, which were revealed in the computer simulation earlier, are verified experimentally, and the current-voltage characteristic (CVC) obtained with the voltage sawtooth modulation was chosen to be the main data to compare.

## II. EXPERIMENTAL TECHNIQUE

The application of the CVC obtained with the voltage sawtooth modulation for the investigation of high-voltage conductivity of dielectric liquids was suggested in the experimental work [18], where the corresponding sets of data were called the dynamical current-voltage characteristics or DCVCs. Such approach to the conductivity measurement has a number of advantages over obtaining classical CVC. First and foremost, the main advantage is the rapidity of the experimental data acquisition since there is no need to wait for obtaining steadystate electric-current values, which can take hours or even days [19], [20]. As consequence, the whole measurement process goes on under constant external conditions and, in particular, at constant temperature, which variation strongly affects the electric conductivity of liquid [2], [21]. Besides, the rapidity of the experimental data acquisition makes it possible to verify the reproducibility of the results, which is of great significance on account of complications and instabilities of current passage processes in low-conducting liquids. Further, the rapidity of data acquisition reduces the probability of accidental current fluctuations caused by mechanical impurities (dust), since the latter usually fail to get in the near-electrode area during a

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Fig. 1. Schematic diagram of the experimental set-up.

short voltage modulation period. In addition to everything else, dynamical CVCs (as opposed to classical ones) contain complementary information that is what is used in the present work to ascertain the prevailing CFM.

Fig. 1 presents the schematic diagram of the experimental set-up. It consists of high-voltage source (VIDN-30, OST), high-frequency generator (3409/5, AKIP) for voltage modulation, 2 analog-to-digital converters (ADC) (E14-140, L-Card), CCD camera (746-USB, EVS), and transparent test cell that is filled up with the investigated liquid. A needleplane or wire-plane electrode system with highly non-uniform electric field distribution is used, which provides the conditions for activation of high-voltage CFM without obtaining the electrical breakdown of liquid. The electrode type (radius of curvature, metal, etc.) is chosen depending on the objectives of the experiment. In the present study, a sharp needle (with  $15\pm1 \ \mu m$  curvature radius) and wire (with  $50\pm2 \ \mu m$  radius and 6 cm length) were utilized as high-voltage electrode, whereas transformer oil (with and without butyl alcohol) and liquid petrolatum were used as working liquids. The voltage and current are measured by virtue of 2 separate high-speed ADCs. A CCD camera records the interelectrode gap illuminated by laser to check the absence of mechanical impurities in the vicinity of the needle tip during the measurement.

The important feature of the proposed experimental technique is the necessity to separate two current components capacitive  $(I_C)$  and ionic  $(I_{ion})$  ones:

$$I = I_C + I_{ion} \tag{1}$$

The first component is caused by application of changing voltage to the electrode system, which leads to the emergence of capacitive current:

$$I_C = C \frac{\mathrm{d}V}{\mathrm{d}t} \tag{2}$$

where C is the capacitance of the electrode system, V is the voltage. The component is unconnected with the ion motion in the interelectrode gap while it is dependent on the geometry of the electrode system. The current will flow even if there is no liquid in the cell though the presence of the latter affects the  $I_C$  due to the increase in the electric permittivity, which leads to the capacitance growth. In turn, the second component is exactly the current caused by the ion motion

through the interelectrode gap and therefore it does provide the high-voltage conductivity of a liquid. Thus, to investigate the features of current passage processes in liquid dielectrics one has to examine just the ionic component that can be obtained subtracting the capacitive current from the total one.

Prior to passing to the analysis of the results, one can estimate whether the capacitive current is to be taken into account. The characteristic value of the capacitance of the laboratory electrode system amounts to 1 pF, which provides 10 nA current at 10 kV/s voltage modulation rate. Thus, taking into account the component is requisite when ionic current level is up to 1  $\mu$ A. To obtain exact values of the capacitive current, one has to measure the voltage with further calculation of its time derivative and test the electrode capacitance. The latter can be estimated numerically on the base of computer simulation if the electric permittivity and exact geometry parameters are known. However, it is more convenient to measure the capacitance experimentally, applying to the electrodes short voltage pulse with sufficiently low magnitude (to avoid the activation of high-voltage CFMs) and steep rising edge. Then, the current passing in the first instant of time will consist of the only capacitive component. Consequently, the capacitance will equal to the ratio of the measured current to the voltage time derivative. To provide more precise capacitance measurement, the corresponding calculations are to be performed for a time span with further calculation of the average value, since noises distort the pickup signal. The approach described above let one estimate the capacitance of the system within the same experimental set-up, which makes the method of DCVC investigation more convenient.

Fig. 2 shows an example of the characteristic experimental data measured in the wire-plane electrode system in purified transformer oil. Curves 1 and 2 are the data recorded by ADC and the curve 3 and 4 were calculated as described above. In the considered case, the voltage was modulated from 0 up to 20 kV and back during 3 s, which corresponds to approximately 15 kV/s voltage modulation rate. The total current (Fig. 2a, curve 2) shows a steep change at the first instant after the voltage application, which is provided by the capacitive current presented in Fig. 2b (curve 3). Then, a smooth growth of the current takes place with further increase in voltage due to emergence of the ionic current (Fig. 2b, curve 4). And the total current changes steeply at the instant when the voltage begins decrease (at t = 2 s) since the capacitive current reverses sign.

## III. RESULTS AND DISCUSSION

The DCVC itself (Fig. 3a, curve 1) can be plotted basing on the obtained oscillograms of the voltage and the ionic current (curves 1 and 4 in Fig. 2). Fig. 3a also displays another DCVC (curve 2) observed at one-order-less voltage modulation rate (1.5 kV/s). The characteristics for the purified transformer oil have the following shape. At sufficiently low voltage (up to approximately 5-7 kV), the dynamical CVC shows linear current growth, which corresponds to the range of the Ohm's law applicability. In the voltage range, the dissociation of the impurity molecules is actually the sole CFM while the



Fig. 2. Smoothed pickup signal of voltage (curve 1) and current (curve 2) obtained for the transformer oil and wire-plane electrode system (a) and the corresponding capacitive (curve 3) and ionic (curve 4) current components (b); all curves are normalized to the maximum values of voltage and current, respectively.

migration is the main mechanism of ion transport. The section of more rapid current increase is seen at higher voltages, which corresponds to the activation of high-voltage CFMs (i.e., the injection or field enhanced dissociation) and emergence of EHD flows. The difference between the increasing voltage section and the decreasing voltage one (i.e., so-called hysteresis) is the feature of the presented DCVCs. The similar data were obtained in the experimental research earlier [18] as well as calculated numerically on the base of the complete set of EHD equations [22], [23]. The occurrences of hysteresis and the effect of voltage modulation rate on the current level indicate that the typical time span for steadying of current passage processes is more than that of the voltage change, and the system fails to adjust to the changing conditions. According to the simulation results [22], [23], the both features are the specific ones for the case of the prevalence of the injection CFM. In addition, the observed DCVCs show current fluctuations at voltages in the range from 15 to 20 kV, which is also specific for the injection charge formation. However, it is worth noting that the current surge at 15 kV voltage (curve 2) is unconnected with CFM but caused by accidental appearance of mechanical impurities in the vicinity of the needle tip.

The computer simulation shows that there are two specific time spans for current passage processes in liquid dielectrics. The first one is that of charged-jet crossing the interelectrode gap and the second one is that of space charge accumulation inside the entire bulk. The first one causes the emergence of hysteresis, while the second provides the decrease of current



Fig. 3. Dynamic current-voltage characteristics observed for purified transformer oil at different voltage modulation rates: 1-15 kV/s and 2-1.5 kV/s (a) and for transformer oil with 5% impurity of butanol at 15 kV/s (b).

with lessening the voltage modulation rate. The latter agrees with the fact that curve 2 is located below the curve 1. Besides, the papers [22], [23] show the hysteresis to appear only in the case when the Maxwell relaxation time

$$\tau = \frac{\varepsilon}{\sigma} \tag{3}$$

is greater than the modulation period;  $\varepsilon$  is the electric permittivity,  $\sigma$  is the conductivity. The analogous regularity is observed in the experiment: the hysteresis disappears (Fig. 3b) with the increase in conductivity of the working liquid by introduction of an electron-acceptor impurity (5% of butanol) into the transformer oil. The corresponding Maxwell relaxation time is about 0.1 s (according to the conductivity  $2 \times 10^{-10}$  S/m and electric permittivity  $2 \times 10^{-11}$  F/m) while the voltage changes by 1 kV in 0.2 s. Thus, the current steadying goes faster than voltage changing. Among other things, it is worth noting that the DCVC (Fig. 3b) shows similar current fluctuations in high-voltage range as the previous ones (Fig. 3a).

DCVCs obtained for the needle-plane electrode system with liquid petrolatum being used as the working liquid are presented in Figs. 4 and 5. Since the conductivity of liquid petrolatum is awfully low (less than  $10^{-13}$  S/m) the ohmic current is negligible. The latter makes it possible to study exactly high-voltage conductivity provided by the activation of the injection charge formation or field-enhanced dissociation. Besides, utilizing very sharp needle (with curvature diameter 15  $\mu$ m) as the high-voltage electrode provides an opportunity to raise the maximum value of the local electric





Fig. 4. DCVCs observed for purified liquid petrolatum at positive (a) and negative (b) polarities of the needle electrode at different modulation rates: 1-20 kV/s, 2-6 kV/s, 3-2 kV/s, and 4-0.6 kV/s.

Fig. 5. DCVCs observed for purified liquid petrolatum at positive (a) and negative (b) polarities at different modulation rates (1-20 kV/s, 2-6 kV/s) when the lateral needle surface was covered by solid electrical insulation.

field strength above  $10^8$  V/m avoiding electric breakdown of the interelectrode gap. Since no electron-acceptor or electrondonor impurity was introduced inside the liquid, then the fieldenhanced dissociation CFM is expected to be the prevalent one, at least, at positive polarity when the electron emission is impossible. Among other things, it is worth noting that high voltage was applied to the electrode system during about ten minutes prior to carrying out every set of measurements to obtain stable and reproducible data.

Fig. 4a shows DCVCs observed at positive polarity at two different voltage modulation rates-2 and 20 kV/s. Both characteristics have very similar shape and features. Thus, there is no linear section in the low-voltage range, whereas the observable current emerges only at 15 kV and steeply rises with further increase in voltage. As opposed to the case considered above (Fig. 3), the DCVCs show no hysteresis and are almost independent of the modulation rate. However, a polarity reversal leads to marked changes of DCVCs (Fig. 4b). Though they still show no ohmic section, the current appears at comparably lower voltage (about 8-10 kV), the slope of the characteristics (the angle of curve inclination) slightly lessens, the hysteresis emerges, and, at last, the DCVCs depend on the voltage modulation rate. Besides, the current level increases almost by an order of magnitude in compare with the case of positive polarity.

All the features for both cases are in a quite good agreement with the data obtained in the computer simulation in two limiting cases when only field-enhanced dissociation (the Wien effect) or, on the contrary, only injection is taken into account [22]–[24]. In the former case, the major part of ions emerges in the immediate vicinity to the needle tip, space charge propagates to the counter electrode in the form of very thin jet and is removed from the bulk with very high efficiency. Due to that the corresponding DCVCs show no hysteresis and the current value is independent of the voltage modulation rate. However in the latter case, ions are injected from the major part of the needle surface, charged area is comparably wide and part of ions fails to neutralize at the counter electrode since they are taken away with fluid flow. Besides, the voltage increase at the injection CFM leads to fluctuations of the lateral boundary of the charged area [25], which, in turn, provides instabilities of the total current passing through the external electric circuit [15].

Thus, basing on the conducted comparison between the experimental data and simulation ones, the following conclusions can be drawn: DCVCs obtained at the positive polarity (Fig. 4a) correspond to the case of the Wien effect occurrence whereas the injection CFM is the underlying one at the negative polarity (Fig. 4b). The second part of the conclusion is also confirmed by the fact that the voltage corresponding to the current onset depends on the polarity, which fails to be explained within the scope of the field-enhanced-dissociation model since the Wien effect is independent of the voltage reversal.

As it was written above, ions emerge at the vicinity of the needle tip as well as along the needle base at the injection CFM as opposed to the case of the Wien effect occurrence. This means that the total current has to considerably reduce at the negative polarity while remain almost unchanged at the positive one if the needle base is covered with solid insulation. The supposition is justified by the DCVCs presented in Fig. 5, which were obtained in the case of utilizing the needle electrode (with the same tip radius as before) with whole lateral surface (excluding the needle tip) being covered by ethylene vinyl acetate. The shape of DCVCs at positive polarity actually remains unchanged and the total current goes just 20% down from the original value as opposed to the negative polarity, at which the total current decreases more than twice and the hysteresis is disappeared. This all means that a major part of ions emerges at the lateral needle surface, which confirms the suggestion about prevalence of the injection charge formation at negative polarity.

## IV. CONCLUSION

The paper presents the results of the experimental research into the effect of charge formation mechanism on the features of current passage processes in liquid dielectrics. The special method basing on the so-called dynamical current-voltage characteristics was developed for experimental revealing the criteria of the prevalence of the injection or dissociation charge formation mechanisms. The DCVC features predicted on the base of the computer simulation were revealed and confirmed experimentally. Among them are the follows. DCVC shows hysteresis and the voltage modulation rate affects the current values at the injection mechanism of charge formation whereas the situation is vice versa in the case of the Wien effect occurrence. Moreover, the features agree with theoretical suppositions and numerical computations singly as well as in the aggregate, which corroborates the reliability of the conclusions. In addition, the following conclusion can be made basing on the conducted research. The injection CFM is the underlying one at the negative polarity while the fieldenhanced dissociation prevails at the positive one in the liquid petrolatum when a sharp needle is utilized as the high-voltage electrode.

The research mostly concerns the case of very low electric conductivity of liquid when the ohmic current plays a minor role. The further development of the investigation consists in transition to the liquids with higher conductivity level and carrying out experimental investigation of the effect of CFM on the EHD flow structure.

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