

TRANSIENT CURRENTS IN ORGANIC DIELECTRIC LIQUIDS, POLYMERS, AND LIQUID CRYSTALS

R. ROMANETS, K. YOSHINO¹

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Pridniprovs'ka State Academy of Civil Engineering and Architecture
(24a, Chernyshevsky Str., Dnipropetrovsk, 49600, Ukraine),

¹Osaka University, Department of Electronic Engineering,
Graduate School of Engineering
(2-1, Yamada-Oka, Suita, Osaka 565-0871, Japan)

The phenomena and mechanisms of transient currents in organic dielectric liquids, polymers, and liquid crystals are reviewed.

Introduction

Organic dielectric liquids, polymers, and liquid crystals are very widely used in the insulation of electrical apparatus, transformers, instruments, devices, etc., and their electrical conductivity properties have thus been extensively investigated. Although it is well known that the transient currents flow upon voltage application in these dielectrics, the current transients are complex phenomena depending on materials and conditions. Therefore, there are still many problems yet unsolved.

In this work, the phenomena and mechanisms of the transient currents in organic dielectric liquids, polymers, and liquid crystals are reviewed. There are many publications on this problem, and it is not possible to consider all of them. Therefore, we have chosen only some typical works for consideration. The transient currents in organic dielectrics can be classified as charging, discharging, and reversal characteristics. The main idea of this work is to show the basic features of these transient currents in organic dielectrics. Although the structures of dielectric liquids, polymers, and liquid crystals are very different, it is possible to find some common properties in transient characteristics.

1. Transient Currents in Dielectric Liquids

It is well known that, after application of a step dc voltage to the cell with a dielectric liquid, the transient current appears. The transient current-time characteristic depends on the nature and purity of a liquid, on the range of the electric field, on the electrode material, on temperature, etc. [1, 2]. This

current has been termed as charging (or polarization), and it cannot decay, does not change or increase with time. In dielectric liquids with bulk dissociation, the charging current is known to decay with time in most cases from the initial value until a quasi-steady-state current is achieved. In some cases, the current decreases monotonically and sometimes it has been observed to produce a peak value before decay [3, 4]. The decay of a charging current on time may be described by the exponential law

$$I = I_0 \exp(-\lambda^* t), \quad (1)$$

where I_0 is the initial current and λ is a constant depending on the nature of liquid (Fig. 1, curve 1). The charging current in silicone liquid [5] has been found to vary with time as

$$I = At^{-m}, \quad (2)$$

where A is a constant, and the exponent m ($0 < m < 1$) decreases with increase in temperature. There is a case where the charging current decreases linearly very quickly in the first time interval, and then it drops very slowly to a constant value (Fig. 1, curve 2) [6]. The transient current observed in dielectric liquids is often determined by the injection of charge carriers by electrodes [7, 8]. The transient current in the case of pure benzene has the shape shown in Fig. 1, curve 3 [7], and, in the case of nonpolar liquids doped with a neutral product, it can be represented by curve 4 (Fig.1). For technical oil with conducting particles, it is possible to observe curve 5 (Fig.1) [9], where the charging current is not changed with time.

If the steady-state charging current in a dielectric liquid is reached, the applied voltage is suddenly removed, and two electrodes are short circuited, the so-called discharging (or depolarization) current is observed [9]. Usually, the discharging current (Fig. 2, area A, curve 2) flows in the direction opposite to that of the charging current (Fig. 2, area A, curve 1). Sometimes,

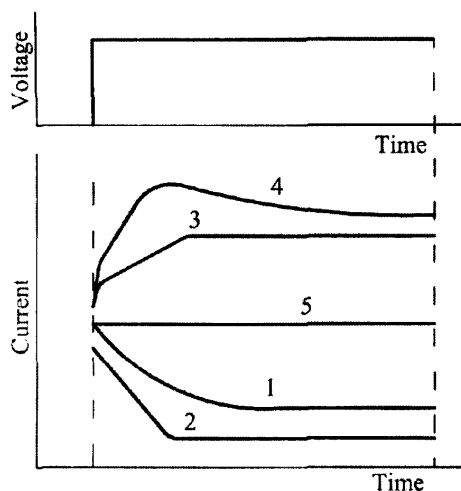


Fig. 1. Typical examples of current dependence on time when the step voltage is applied to the cell with a dielectric liquid

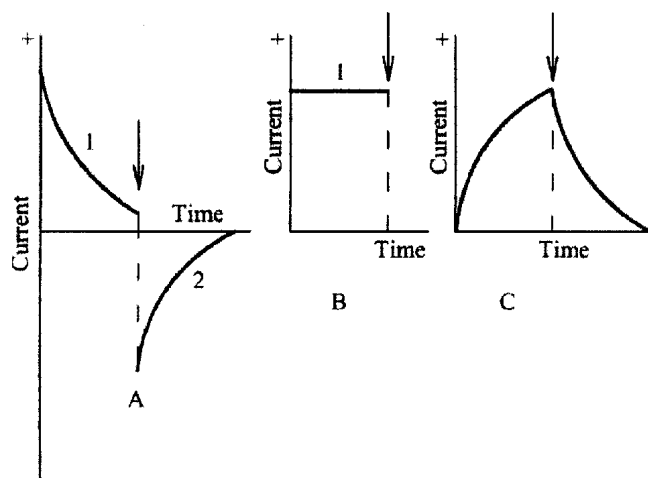


Fig. 2. Typical characteristics of charging (1) and discharging (2) currents in dielectric liquids

the former is equal in magnitude to the latter. The arrows in each area of Fig. 2 correspond to the switch-off of the voltage and the short circuiting.

In liquids containing the conducting particles of impurity, the charging current practically does not change with time (Fig. 2, area B, curve 1). In liquids where the charging current increases with time (Fig. 2, area C, curve 1), the discharging current can flow in the same direction as the charging current (Fig. 2, area C, curve 2).

The measurement of charging and discharging currents was performed with liquid aromatic hydrocarbons (benzene, toluene, etc.) [10, 11]. When the charging current flows, the space charge storages in the immediate vicinity of the electrodes in a test cell with dielectric liquid occurs. This space charge Q was found by investigation of the discharging current, and the value of Q was estimated using the area under the curve of the discharging current as a function of time. At first, this charge increases rapidly enough, showing then the tendency towards saturation [11]. The distribution of an electric field in benzene and toluene at low stress was determined by the probe method and by Kerr's method at a high stress [12, 13]. The formation of heterocharges near electrodes was revealed: a negative space charge near the anode and a positive space charge near the cathode. The accumulation of charge causes a redistribution of the electric field in the electrode gap and explains a decrease of the charging current with time. In investigations of the transient currents in several engine oils, after using them in special machines, the charging current was not changed with time, and

the discharging currents were not practically observed [14]. Such a behavior of the technical oils can be explained by the existence of conducting particles in these dielectric liquids. An unusual behavior of the discharging current is observed in dimethylsiloxane [15], where the discharging current remains negative for low fields, and the discharging current becomes positive for higher fields after an initial time interval. The transition of a discharging current from negative to positive is known as the transition from a heteropolar to homopolar current.

The typical traces of charging and discharging currents in silicone oil AP 100 (Wacker Chemie GmbH) at various temperatures are shown in Fig. 3. In these experiments, the test cell with plane parallel stainless steel electrodes, a guard electrode, and the electrode spacing of 0.2 cm was used (applied voltage of 1000 V). In each part of Fig. 3: (a) for $T = 293$ K, (b) for $T = 313$ K and (c) for $T = 333$ K, curves 1 of charging currents correspond to the time interval of applied voltage of 5 s, curves 2 – 50 s, and curves 3 – 500 s, respectively. The number of discharging currents corresponds to that of charging ones, and the recording rate for all discharging currents is the same. It can be seen that the waveform of the charging and discharging currents depends on the time interval of an applied field and temperature.

At $T = 293$ K (Fig. 3,a), the charging currents flow in the forward direction as the charging ones. At $T = 313$ K (Fig. 3,b), there is the inversion in a direction of the discharging flow. At $T = 333$ K (Fig. 3,c), the discharging currents flow in the direction opposite to

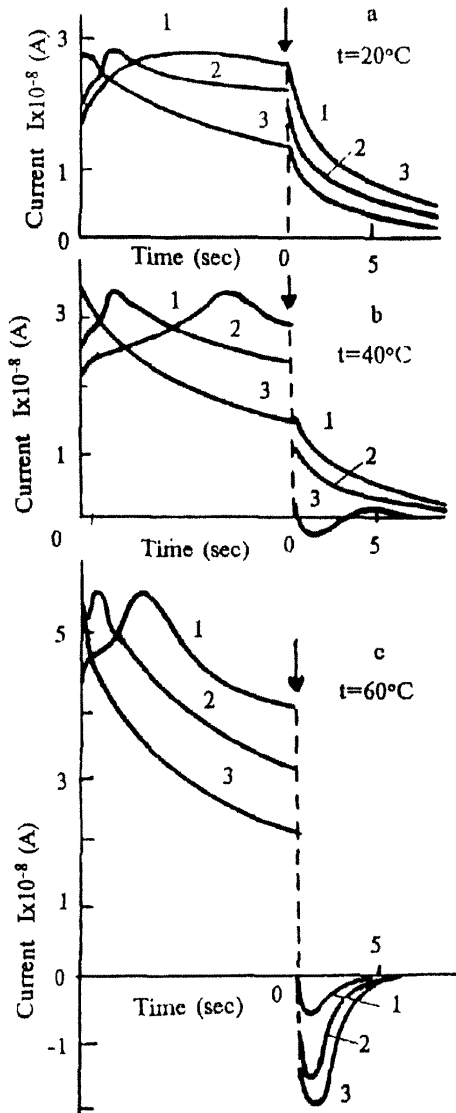


Fig. 3. Typical traces of charging and discharging currents in silicone oil AP 100

that of the charging currents. This behavior of the discharging currents can be explained by the presence of homocharges and heterocharges. The accumulation of homocharges related to the injection conduction at a lower temperature ($T = 293$ K). The superposition the injection and volume conduction takes a place at an intermediate temperature ($T = 313$ K). For a higher temperature ($T = 333$ K), the volume conduction and the accumulation of heterocharges are dominated, and we have the typical discharging current in a dielectric liquid [2].

When the polarity of the applied voltage is reversed, the reversal current flows, and the peak appears in the

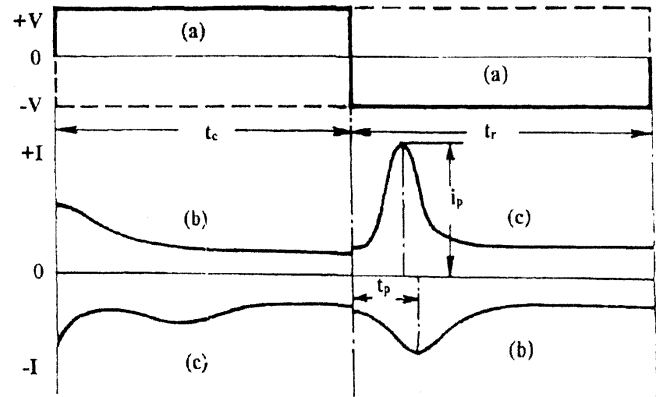


Fig. 4. Typical experimental voltage waveform, charging and reversal currents in dielectric liquids

current waveform. The typical experimental voltage wave-forms (curves *a*), experimental turn-on and field reversal currents (curves *b* and *c*, respectively) are shown schematically in Fig. 4. Curves *b* conform to the decreasing charging and corresponding reversal currents, and curves *c* conform to the increasing charging and corresponding reversal currents. The time interval for the charging and reversal currents are t_c and t_r , respectively. The time at which the current reached its maximum after the voltage reversal is t_p , and the maximum of the reversal current is I_p .

In general, under the first application of a voltage to the cell with dielectric liquids, the charge carriers drift to the oppositely charged electrodes, and a space charge can originate near the electrodes. On reversing the electric field, neutral molecules near the electrode can accept some injected charge. These ions will drift through the liquid as a charged layer, and the reversal current is generated. The time interval for reaching a peak after exchanging the polarity was used in the measurements of the apparent mobility of charge carriers in dielectric liquids under a low field strength, if the time of the voltage-reversal peak is inversely proportional to the electric field [16]. The appearance of the reversal current peak depends on the duration of the charging current t_c . For time interval $t_c \leq 0.5$ s in carbone tetrachloride, the reversal current peak was not observed [17]. One reversal peak appears for the charging time interval $t_c = 2$ s, and there are two reversal peaks for time interval $t_c = 4.5$ s. These two peaks were concluded to be due to positive and negative ions. In some cases, the characteristic curves of reversal current versus time in a dielectric liquid look symmetrically with respect to the polarity of the electric field [3].

On this base, it was assumed that the mobility $\mu = \mu_+ = \mu_-$, where μ_+ is the mobility of a positive charge carrier, and μ_- is the mobility of a negative charge. A repetition of the polarity reversal method was used under various electric fields, gap lengths, and temperatures in order to characterize the relation between the reversal current peak and the mobility of ions in dimethylsiloxanes [18]. It was shown that the reversal current peak seems to approach a certain value after several time repetitions. The calculated value of the apparent mobility of charge carriers depends on many parameters and cannot be represented as the intrinsic mobility of a charge carrier. The investigation of reversal currents in dielectric liquids shows that the shape of the reversal characteristic depends on the nature of a liquid and impurities in it, field strength, time interval, time repetition, electrode gap, temperature, etc. In general, the reversal currents show that the charged layers accumulated near electrodes can act as the reservoirs of positive and negative charge carriers. So, the current wave form under the reverse dc polarity in dielectric liquids can be explained in terms of the monopolar or bipolar injection from these charged layers.

2. Transient Currents in Polymers

The investigations of current-time ($I - t$) characteristics and current-voltage ($I - V$) characteristics were indicated in [19] as simple experimental methods to identify the nature of the traps, their distribution in space and energy depth, and the location of the space charge in polymers. As usual, when a dc field is applied to a polymer sample sandwiched between two plane parallel electrodes, there appear the rapidly changing charging current and a long-term slowly current decaying with time until a steady-state current is achieved. This steady-state current may be by many orders of magnitude lower than the initial value of a charging current. Upon the removal of the dc voltage and if the electrodes become short-circuited, the discharging current flows very often in the opposite direction [20–21]. In low-density polyethylene (LDPE) [20], the charging and discharging currents were observed to be mirror images of each other at all times within of $5 - 10^5$ s at temperatures below 200 K. The transient current I decayed with time lapsed t after the removal of the external voltage, according to the power law

$$I(t) = A(T)t^{-n}, \quad (3)$$

where $A(T)$ is a temperature-dependent factor and $n \leq 1$. At temperatures above 200 K, the charging and

discharging currents began to depart from being mirror images. The typical time dependences of the charging and discharging currents in polymers at a constant field and at various temperatures are presented in [21].

The observed low-temperature peaks in the isochronal discharging currents (the currents at fixed times) at 138 and 203 K in LDPE were explained by the dipolar relaxations accompanying molecular motions with a wide distribution of relaxation times. Above 273 K, a charge carrier hopping was proposed as the predominant mechanism of a transient current at high fields. A similar behavior of charging and discharging currents was observed in polyethylene terephthalate (PET) and polyvinylidene fluoride (PVF₂) [21]. The mechanisms of the time-dependent polarization effects were studied on the basis of the analysis of transient currents behavior with respect to polarizing fields and temperature. It was supposed that the induced polarization may be explained by a permanent orientation of structural dipoles in the applied field, a field-induced non-uniform distribution of trapped charges, or a combination of the two above-mentioned effects.

There is the term of the anomalous transient current which is involved as an electrical current flowing in the direction opposite to the expected one. The anomalous transient currents in polymers can be classified into three groups [22].

The first group consists of anomalous discharging currents. In this case, if a previously polarized sample is short-circuited, the discharging current does not fall asymptotically to zero, but overshoots and flows in the direction of the original charging current [23–25]. The long-term discharging currents in polymers can be ascribed either to the dipole relaxation or to the release of a trapped charge. It was observed that the sign reversal in a discharging current occurs after the application of relatively high fields for enough a long time. There is the suggestion that a high field injects a homocharge. In general, this charge can be non-uniformly distributed, and, after the short-circuiting of the sample, the associated electric field origins the discharging current. If heterocharges are formed from undischarged bulk carriers, the discharging current flows in the opposite direction.

The second group corresponds to anomalous thermally stimulated currents (TSC) [26, 27]. If a previously polarized sample is heated in the short-circuit state, the discharging current typically shows a peak as the temperature rises. The simultaneous observation of TSC and TL (thermoluminescence) of LDPE has been

reported [28]. This seems to be an effective method to distinguish the trap and dipole mechanisms. The “low temperature injection” and “high temperature injection” were used for experiments. The observed TSC peaks are due to the release of trapped carriers.

The third general group of anomalous transient currents is based on various observations of charging currents that increase with time. The appearance of the charging current peaks is associated with the transient time of the space-charge-limited current (SCLC) [28]. In many cases, the charge mobility can be estimated from similar experiments [29].

The method of polarity reversal of the applied voltage was used in the investigation of anomalous transient currents in plasticized polyvinylchloride (PVC) [30]. In the experiment, the series of polarity reversal of the applied voltage was performed. The nonsymmetric polarity effect was observed between the positive and negative currents, and the double current peak appeared in the reversal current-time characteristic. Similar dependences were observed in chlorinated polyethylene (CPE) [31]. While the charging current decreases monotonically, the voltage-reversal current exhibits two peaks. If the second voltage reversal was executed after three hours, the two peaks appeared again. The two peaks are concluded to be due to positive and negative ions. It was found that the time for each peak t_p is inversely proportional to the electric field E , and the values of t_p can be related to the transient time of ions between the electrodes. The carrier mobility μ was obtained from t_p for both peaks by using the equation

$$\mu = d/(t_p \cdot E_r), \quad (4)$$

where d is the thickness of the sample and E_r is the applied field after the voltage reversal.

On the basis of the investigations of transient currents in polymers, various mechanisms have been proposed to interpret the conduction current including the bulk controlled transportation, electrode polarization, dipole orientation, charge injection leading to space charge effects, hopping of charge carriers between trapping sites, and tunneling of charges from an electrode to traps.

3. Transient currents in liquid crystals

Systematic studies of transient currents induced by the switching or the polarity reversal of an applied dc voltage in liquid crystals are of a practical and fundamental interest. The transient charging currents induced by the

application of a step dc voltage have been studied in nematic liquid crystals (NLCs) [32, 33]. Usually, a peak in the transient charging current is observed, and it has been explained as originating in SCLC. This peak is observed in the nematic phase, and it is due to the alignment of the director of liquid crystal molecules in the direction of the applied electric field for materials with a positive dielectric anisotropy. Typical transient charging currents in liquid crystals generated by the step voltage application were represented by transients in 4-cyano-4'-5-alkyl-biphenyl (5CB; BDH Chemical Ltd. K15) at various applied voltages ($T = 296$ K). A prominent peak in the transient current can be seen above the critical voltage, but cannot be observed below the critical voltage and in the isotropic phase. From these experimental results, it was concluded that the transit charging currents are due to the director orientation with the applied electric field. The time at which the current peak has occurred (t_{peak}) was shortened with increasing applied voltages. It was found that the relationship between the peak time and the voltage is superlinear and is expressed as $t_{\text{peak}} \propto V^n$, where $n \approx 2$. The transient charging current was also measured in a double voltage pulse experiment [3.2]. In this experiment after the first application of a voltage pulse (prepulse) and the short-circuit of the LC cell during time interval t_d , the second voltage pulse for the measurement of current transients was applied. The prepulse had influenced the measuring transient current, and two peaks appeared at the short intervals in the shape of this current. The first peak is shifted to a longer time range with increasing t_d , whereas the second peak gradually disappears. Such changes in charging currents cannot be observed in the isotropic phase. This supports the idea of that the alignment of the director of LC molecules in the direction of the electric field plays an important role in the transients. It was concluded that the changes in the charging current shapes were attributed to a prepulse-induced nonuniform distribution of impurity ions which deviates from their thermal equilibrium distribution.

Transient currents induced by the polarity reversal of a dc voltage applied to a LC cell have been studied in [32]. The current peaks in the transient currents induced by this method in 5CB for various applied voltages less than characteristic voltage V_c and also for 3 V which is greater than V_c were observed. For the applied voltages above V_c , the time of the appearance of a peak current decreases with increase in the applied voltage, because the higher drift velocities are observed at higher fields. For the applied voltages less than V_c , an unusual

phenomena appears as the time occurrence of the peak current decreases with the applied voltage. This unusual behavior has been explained by a simple model which is based on the time variation of an external applied voltage after the polarity reversal and the internal electric field which is induced by the absorbed electric charges on the cell substrate.

Conclusions

After the application of a step dc voltage to the cell with a dielectric liquid, polymer, or a liquid crystal, the similar shapes of charging and discharging transient currents can be observed. The comparison of the charging and discharging currents in dielectric liquids and polymers shows that a similar power law dependence of the current as a function of time can be used. In the reversal current characteristics of dielectric liquids and polymers, two peaks can be observed, which reflects the transport of two types of charge carriers (positive and negative). There are many another common features in the transient behavior of organic dielectrics in electric fields. These data show that, for the study of organic dielectrics, the universal methods can be used. Moreover, on the basis of the unification of investigations of the transient (charging, discharging and reversal) currents and current-voltage characteristics, it is possible to get a more information about the nature of conduction processes in organic dielectric materials.

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ПЕРЕХІДНІ СТРУМИ В ОРГАНІЧНИХ РІДКИХ ДІЕЛЕКТРИКАХ, ПОЛІМЕРАХ ТА РІДКИХ КРИСТАЛАХ

Р. Романець, К. Йошино

Резюме

Зроблено огляд явищ та механізмів перехідних струмів в органічних рідких діелектриках, полімерах та рідких кристалах.