

DETERMINATION OF FIELD DISTRIBUTION IN DIELECTRIC LIQUIDS BY THE "DRIFTING LAYER" METHOD

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The motion of a thin layer of ions produced by ionizing radiation is used for determining the distribution of field in dielectric liquids. The field distribution was determined in this way when the measuring chamber was filled with paraffin hydrocarbons, cyclohexane, silicones and carbon tetrachloride.

Introduction

Knowledge of electric field distribution makes it possible to describe the phenomena which occur when current flows through dielectric liquids [1]. For most dielectric liquids the liquid-electrode contact is not ohmic; only a part of the current carriers reaching the electrodes become immediately neutralized. In some aromatic hydrocarbons the charge injection from the electrodes into the liquid also takes place [2, 3]. These processes bring about an accumulation of space charge in the liquid, what deforms the field distribution. Field distribution measurements provide information on the phenomena occurring on the electrodes and also on the quantities which characterize the transfer of charge in the liquid. For example, Silver [4] calculated the mobility of ions of both signs, the recombination and diffusion coefficients, and the efficiency of ion production in a liquid by using the potential distribution measured by Forster [2] in benzene.

The diffusion of charge carriers, because of their nonuniform distribution in a liquid, gives rise to polarization effects described in the papers by Jaffe and Le May [5] and Zaky and House [6]. For high charge densities the decisive role is played by the drift of charge carriers in the electric field generated by space charge. The dependence of current intensity upon applied field and time for the analogous case of current flow in solid insulators was examined, among others, by Mott and Gurney [7], Rose [8], and Many and Rakavy [9].

Hitherto, two methods of examining field distributions in dielectric liquids were in actual use. One of them is a measurement of field distribution with a charge probe [2, 10,

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11, 12], and the other utilizes the Kerr effect [3, 13, 14, 15, 16, 26]. Both methods have limited ranges of application, however. The probe method may be used only in the case of liquids with conductivities not lower than $10^{-15} \Omega^{-1} \text{ cm}^{-1}$; hence, it cannot be used, for instance, for highly purified paraffin hydrocarbons of electric conductivities between 10^{-18} to $10^{-20} \Omega^{-1} \text{ cm}^{-1}$. The method utilizing the electro-optical effect is restricted to liquids with large values of Kerr constant, and was primarily used in the case of nitrobenzene. A method of measuring field distributions without these restrictions, based on the determination of electrostriction pressure, was given by Stuetzer [17, 18], but it did not become a very widely used tool. This vacancy regarding liquids with the lowest values of conductivity is filled by the "drifting layer" method described below. An attempt to apply this method for measuring the field distribution in hexane, was made by Bloor and Morant [10].

Measuring procedure

The operating principle of the measuring arrangement is shown in Fig. 1. At one of the electrodes the ionizing radiation forms a thin layer of ions. Ion motion in the electric field induces a current in the circuit, the intensity of which depends on the rate of drift. The potential drop on the high ohmic resistance U_R or on the capacitance U_C connected with the chamber

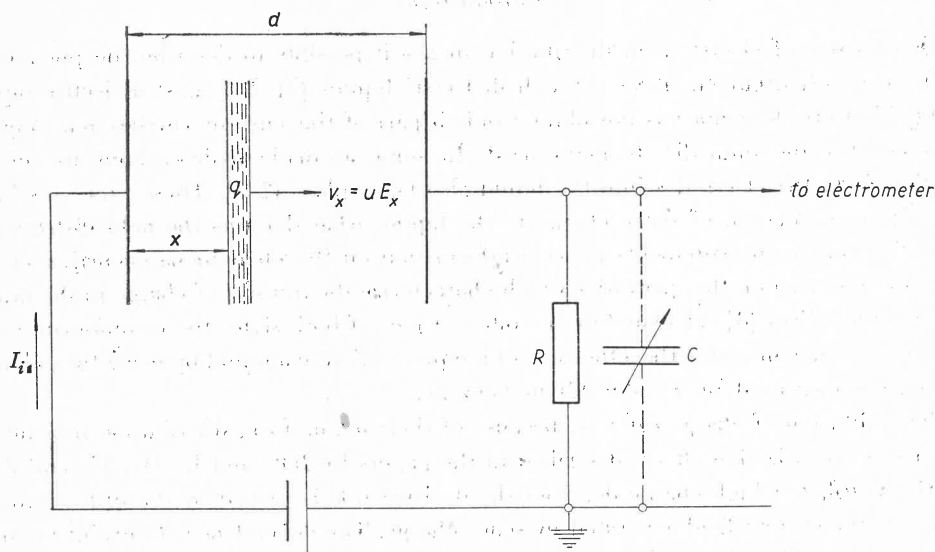


Fig. 1. Principle of field distribution measurement by "drifting layer" method

is recorded on the oscillograph after being appropriately amplified. This method was used by Gzowski and Terlecki [19], Le Blanc [20], Terlecki [21], Allen and Hummel [22], Jachym [23] and Schmidt and Allen [24] for determining the mobility of ions by measuring the time in which the ion layer passed between the electrodes. Improvements of the measuring technique now enables the application of this method in measurements of the distribution

of electric field strength between the chamber electrodes. The relation of current intensity I_i in the external circuit with the field distribution in the liquid is calculated in the following manner. The work which the electric field must perform to displace an ion layer of charge q in a field of strength E_x by the distance dx is equal to the work of a source of current at voltage V during time dt ;

$$I_i V dt = q E_x dx. \quad (1)$$

Taking into account that $dx/dt = u E_x$ (u ion mobility) yield the formula

$$I_i = \frac{qu E_x^2}{V}. \quad (2)$$

This formula says that current intensity is proportional to the square of the field strength at a given point. When the ion layer moves in a homogeneous field of strength $E_0 = V/d$ (d is electrode spacing) the current intensity in the external circuit is constant and is expressed by

$$I_i = \frac{qu E_0}{d} = \frac{qv_0}{d} \quad (3)$$

(v_0 is the velocity of the layer), while the charge collected on the capacitor is proportional to time (Fig. 2a, b, c). If the current intensity in the external circuit alters with time, this is an indication that the field distribution is non-uniform.

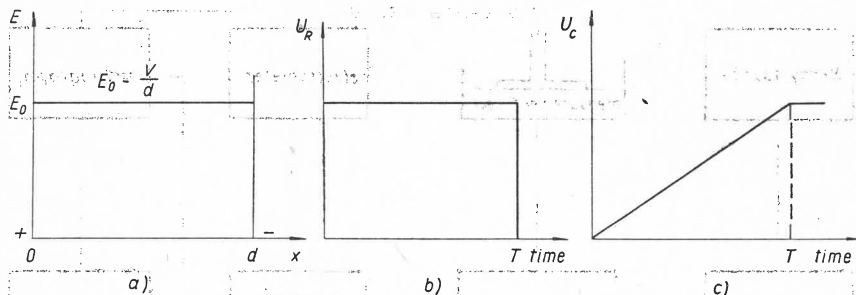


Fig. 2. a) uniform field strength distribution, b) drop of potential U_R on high ohmic resistor during ion migration, c) rise of potential U_C on capacitor during ion layer migration.

For an inhomogeneous field the distance between the ion layer and electrode is a non-linear function of time $x(t)$. From the run of oscillograms it is possible to calculate the electric field strength $E_{x(t)} = E_t$ as a function of time on the basis of Eq. (2). The function $x(t)$ is found from the formula

$$x(t) = \int_0^t u E_t dt \quad (4)$$

which enables calculation of field strength E_x versus distance. The absolute values of field strength and distance are found from the conditions

$$V = \int_0^d E_x dx \quad (5)$$

and

$$d = \int_0^T u E_i dt \quad (6)$$

(T is the time of the ion layer's transition between the electrodes.) If the time dependence of the charge Q_i collected on the capacitor is measured, the current intensity is

$$I_i = \frac{dQ_i}{dt}. \quad (7)$$

The accuracy of this method depends on the fulfilment of the following requirements. The chamber design should guarantee geometrical uniformity of field. The ion layer thickness must be small compared with the regions of field inhomogeneity. The layer charge density should be low enough to make possible to neglect diffusion and Coulomb repulsion of the ions in the layer, and its electric field should not deform the charge distribution in the liquid.

Measuring arrangement

The measuring arrangement is similar to that used in the work by Gzowski and Terlecki [19]. Figure 3 shows the block diagram of the set-up. A major role is played by the electronic key which controls the operation of the entire system. During ionization the key, *via* a switch,

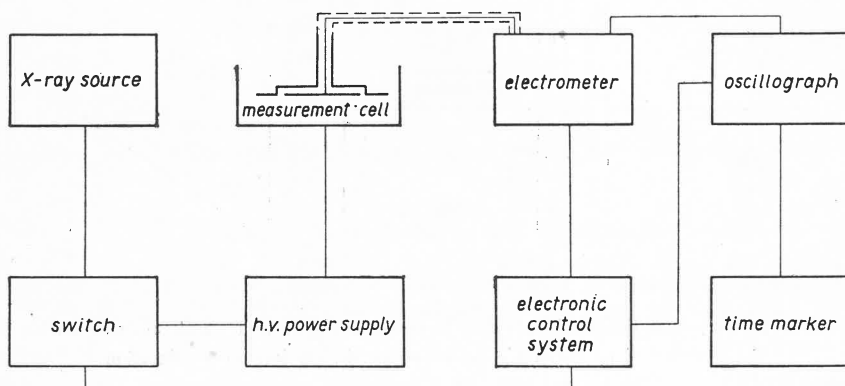


Fig. 3. Block diagram of measuring arrangement

cuts off the high voltage and grounds the electrometer and oscillograph connected to it. Once the ionization is concluded, a voltage is applied to the chamber and then, after a short delay, the electrometer and oscillograph are switched on. The key also enables ionization when voltage is applied to the measuring chamber. The runs of voltage on the high ohmic resistance and on the capacitance, visible on the oscillograph screen, were photographed.

The measuring chamber shown in Fig. 4, filled with the examined liquid, could be shifted along the vertical axis. In this way the ion layer could be formed at any point between the electrodes. The design of the handle allowed precise setting of the beam parallel to the electrodes and collimation of a radiation beam of any required thickness.

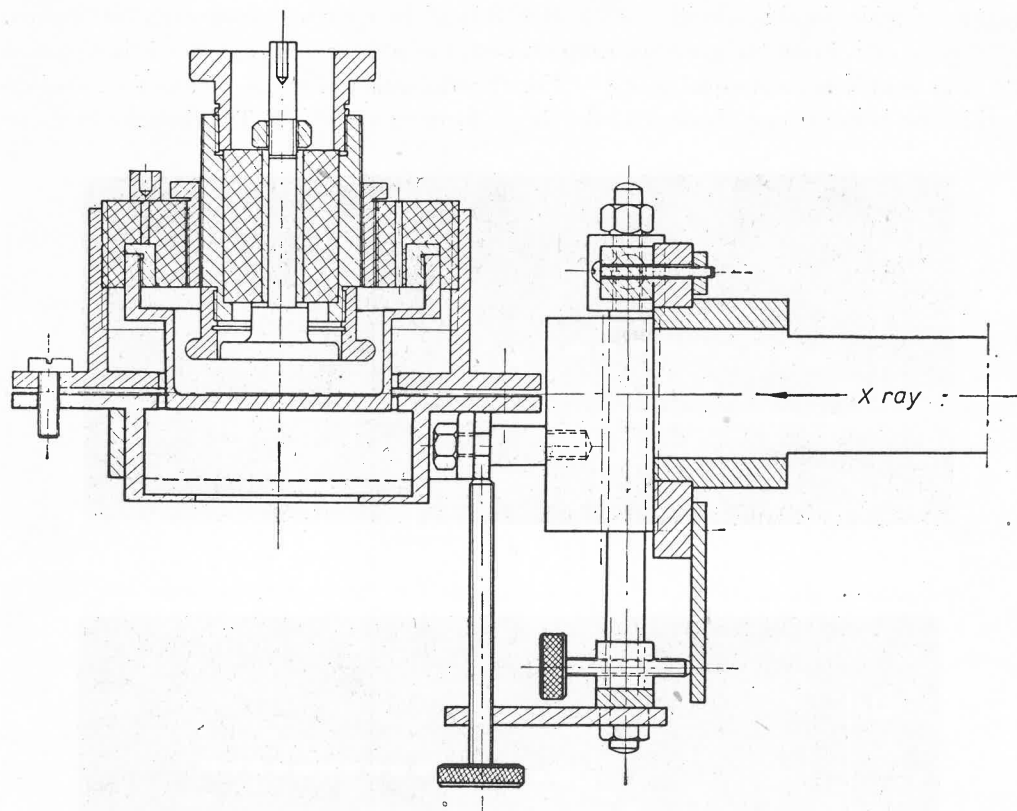
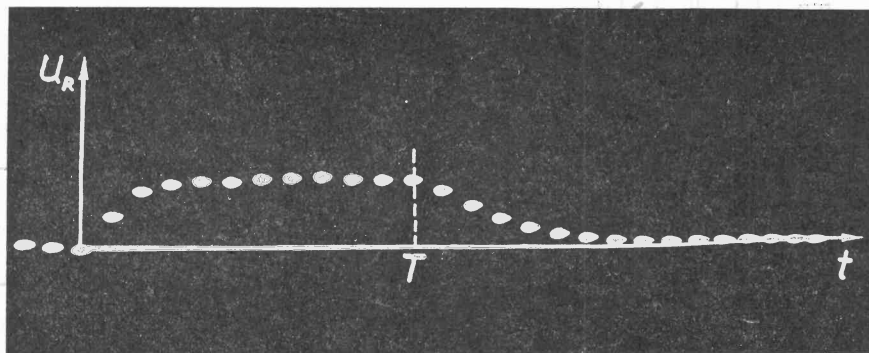


Fig. 4. Diagram of measuring chamber with handle for position control and radiation beam collimator

Results of measurements and discussion

With the method described here the distribution of the electric field in paraffin hydrocarbons, cyclohexane, silicones and carbon tetrachloride were measured. The chamber electrode spacing was from 0.5 cm to 1.4 cm. The applied voltage from a battery was variable within 0.5 kV to 1.5 kV. An ion layer from 0.3 mm to 1 mm thick was formed at one of the electrodes. After a lapse of time of the order of several tenths of a second the carriers of one sign were neutralized on the electrode, and the ion layer of the opposite sign moved between the electrodes. Assuming an approximate equality of the mobilities both positive and negative ions (what is true in liquids insufficiently purified, having a conductivity of order 10^{-14} to $10^{-16} \Omega^{-1} \text{ cm}^{-1}$), the distance of the ion layer front from the electrode at the instance when all the ions of opposite sign are neutralized can be estimated to be nearly 2 mm. The current intensity in the external circuit was less than 10^{-12} A; this corresponds to a layer charge of 10^{-11} coul. In this situation the applicability conditions for the method are satisfied [25]. Measurements were performed for charge carriers of both signs by measuring the voltage on both the capacitor U_C and on the resistor U_R . The oscillograms shown in Figs 5a and b are examples of the voltage curves observed in carefully

purified liquids, in this specific case $\text{Si}_3\text{O}_2(\text{CH}_3)_8$ of an electric conductivity of approx. $10^{-19} \Omega^{-1} \text{cm}^{-1}$. From the potential drop on both the resistor and capacitor it is seen that the field is uniform and equal to $E_0 = V/d$. Determination of the mobility of excess ions in this case is rather easy, because of the simple formula $u = d^2/VT$. The time the ions take



a

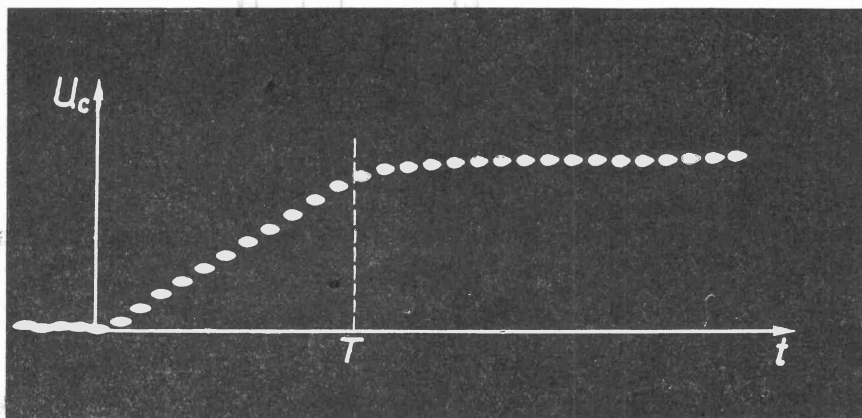
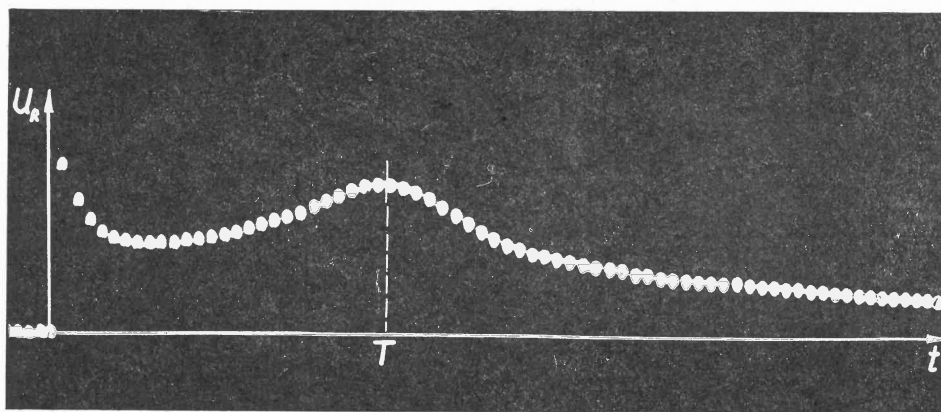


Fig. 5. Time dependence of a) potential U_R on resistor, b) potential U_C on capacitor for $\text{Si}_3\text{O}_2(\text{CH}_3)_8$. Time interval between steps is 0.16 sec. Electrode spacing $d = 1$ cm. Applied voltage $V = 790$ V. Negative carriers: Mobilities: $u_- = 6.6 \times 10^{-4} \text{cm}^2 \text{V}^{-1} \text{sec}^{-1}$, $u_+ = 2.5 \times 10^{-4} \text{cm}^2 \text{V}^{-1} \text{sec}^{-1}$, $\sigma \sim 10^{-19} \Omega^{-1} \text{cm}^{-1}$.

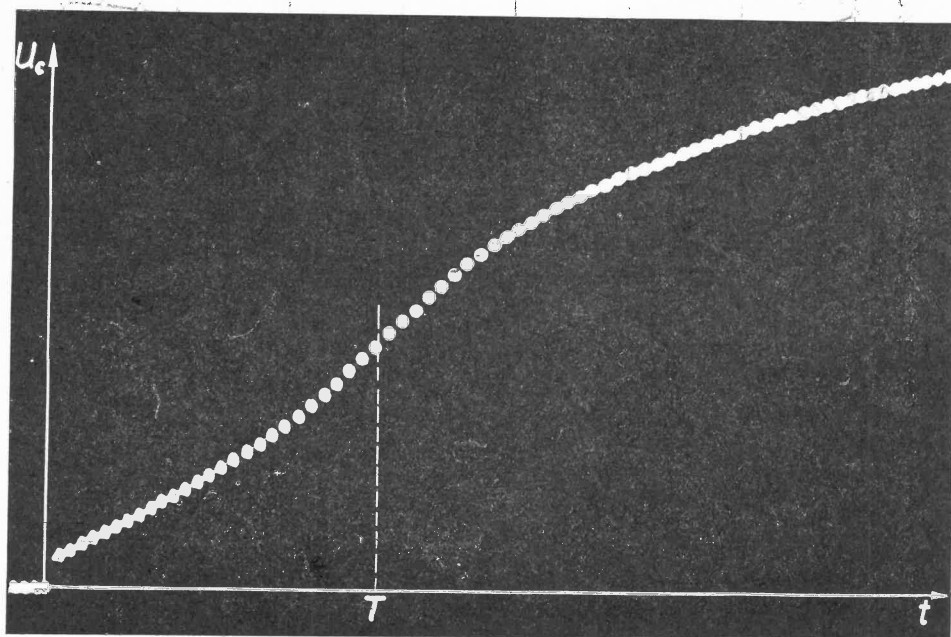
to pass between the interelectrode distance d is strictly defined and easy to read. Different voltage runs are obtained for liquids purposely not purified of a conductivity of the order $10^{-14} \Omega^{-1} \text{cm}^{-1}$, or for a liquid in freshly manufactured measuring chambers.

Results received in such cases are illustrated by the oscillograms in Figs 6a and b. The oscillograms are more complicated than former ones. In accord with what was said above, it should be attributed primarily to the inhomogeneous field distribution between the chamber electrodes. The initial jump of potential in the curve presented in Fig. 6a is associated with the near electrode drop in field strength at the place where the ion layer is formed and with

the temporary presence of the other layer of opposite sign, which had not yet been able to reach the electrode; Next, the migration of the ion layer leads it into a region of lower strength, what causes drop in current intensity in the circuit. A subsequent rise in potential (Fig. 6a) is seen when the ion layer enters the region of stronger field in the vicinity of the



a



b

Fig. 6. Time dependence of a) voltage U_R on resistor, b) voltage U_C on capacitor in the case of an inhomogeneous field distribution (decane: $\sigma \sim 10^{-15} \Omega^{-1} \text{cm}^{-1}$). Electrode spacing $d = 1.4 \text{ cm}$. Applied voltage $V = 902 \text{ V}$. Positive carriers. Interval between steps is 0.16 sec.

other electrode. Attention is attracted by the lack of a sharp boundary at the time when the layer of ions reaches this electrode. This is also seen when either a high ohmic resistor (Fig. 6a) or a capacitor (Fig. 6b) is included in the circuit. It should be remembered, however, that inhomogeneous field distributions are observed in liquids which do not have the lowest values of conductivity. As is known [22, 23], in such liquids ions of much lower

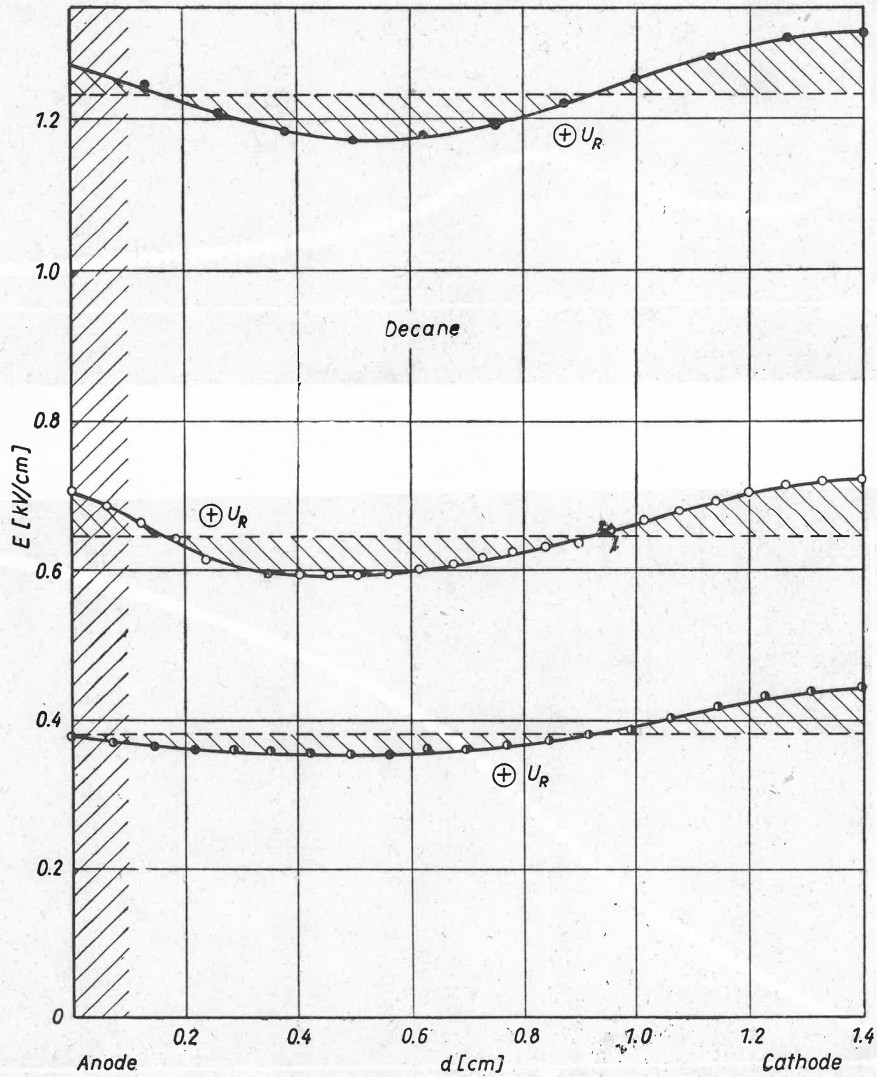


Fig. 7a

mobilities are created during ionization and ion migration, and this gives a diffused picture of the arrival of ions to the electrode. This arrival of slow ions takes some time. In our case we assumed that the arrival time for the layer of excess ions to the electrode is that instant

at which the potential U_R measured on the resistor reaches its second maximum. This time has been compared with the time at which there is the inflexion in the curves of the potential U_c on the capacitor as a function of time. In all cases these times are consistent.

In order to express the field distribution as a function of interelectrode distance from the obtained oscillograms, which give the field distribution as a function of time, calcula-

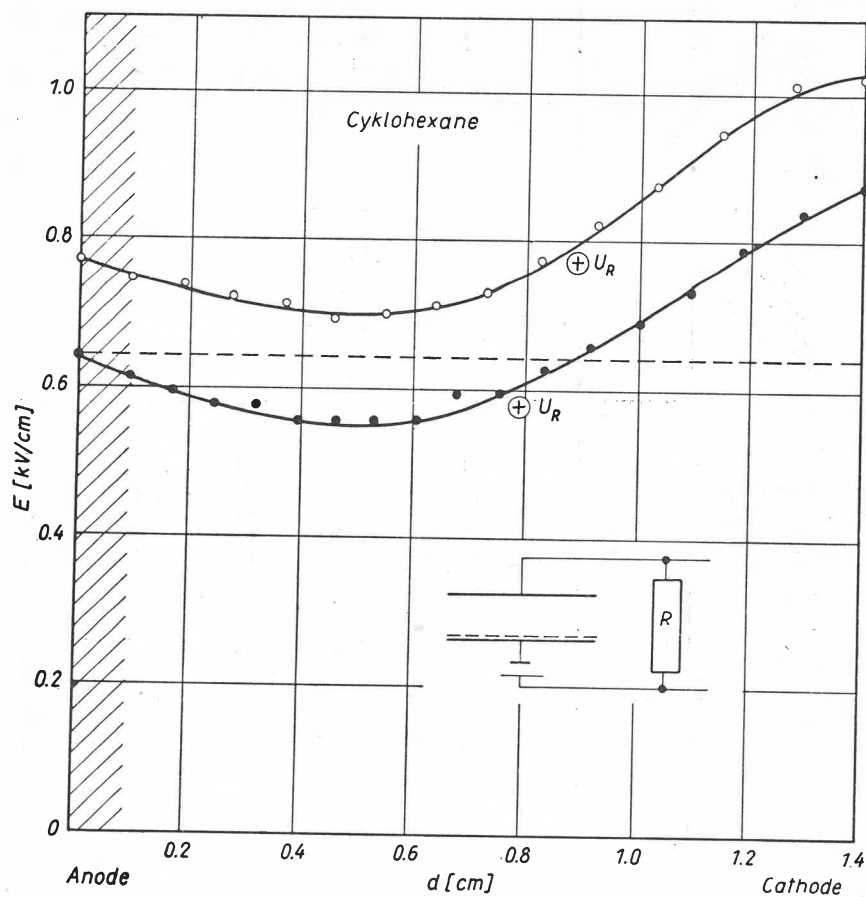


Fig. 7b

tions were made accordingly with the formula (2) and (4)–(7). Field distributions calculated in this way for decane, cyclohexane and carbon tetrachloride are shown in Figs 7a, b and c. In accord with formula (5), the mean electric field strengths are proportional to the areas under the curves in these figures. The dashed lines illustrate the mean values of applied field $E_0 = V/d$, and they run in such a way that the areas above and below the line are equal.

Determination of field analytically, in the sense that a general dependence $E(x, V)$ be given only on the basis of the obtained oscillograms, is difficult. This is because the shape

of the field distribution changes when there is a change in the voltage V applied to the electrodes or the electrode spacing d . This was expounded explicitly in Ref. [3]. Therefore, only the graphical technique was used for giving the field distribution for each value of applied voltage, hence, for each specific oscillogram.

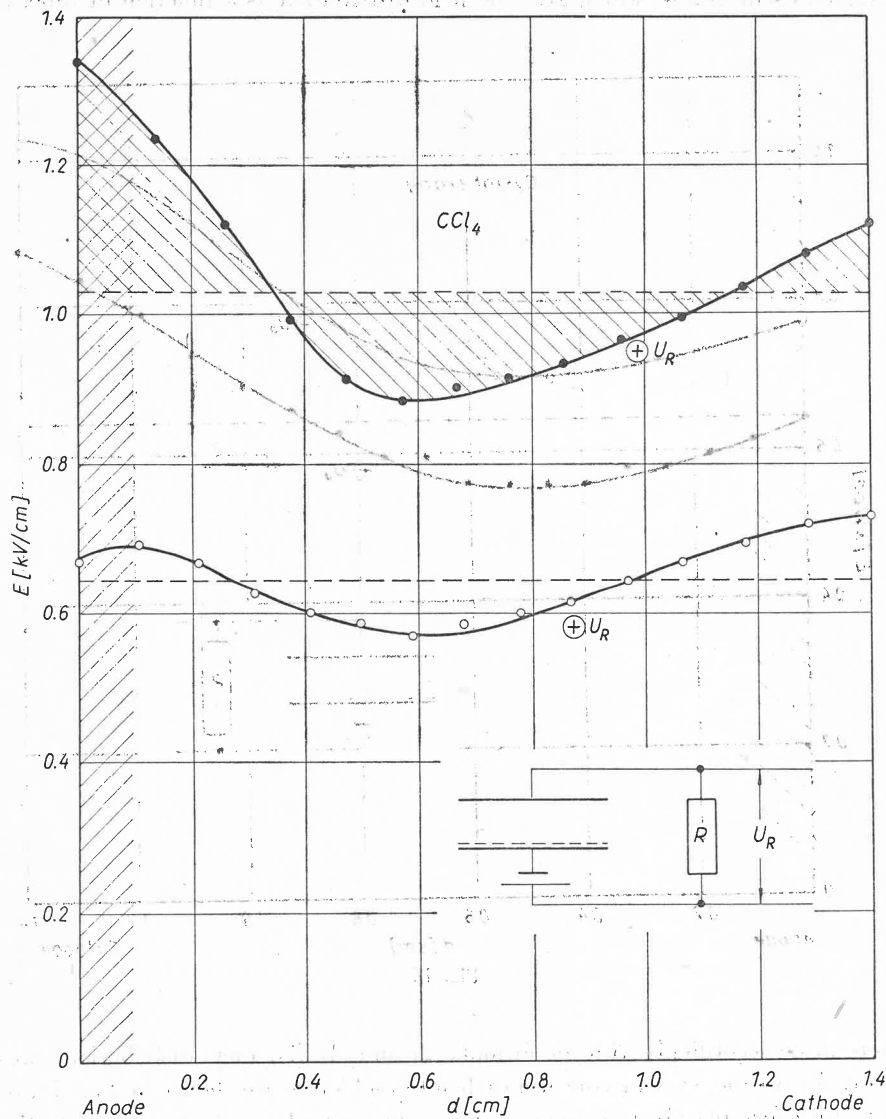


Fig. 7c

Fig. 7. Calculated field distributions between chamber electrodes for a) decane, b) cyclohexane, c) carbon tetrachloride, as a function of applied voltage. $\oplus U_R$ denote the curves obtained from potential drop measurements on resistor during motion of positive ions. The hatched region at the anode represents the ionized volume of liquid. The dashed lines parallel to axis of abscissae represent the mean values of applied field.

Summary

The results of measurements and the discussion in the foregoing indicate a real possibility of applying the described method for field estimate, especially in the events when the conductivity of the liquid ranges from 10^{-18} to $10^{-20} \Omega^{-1} \text{cm}^{-1}$. The electric field distribution considerably affects the shape of the current-voltage characteristics for both natural and ionization conduction. Attention was also turned to the eventuality that an inhomogeneous distribution of field may bear an essential effect on the values of measured mobilities of excess ions in liquids. These values may be systematically undervalued or overvalued, depending on the place where the ion layer originates in the interelectrode space. Extremely thin ion layers, required when electrode spacings are small, can be achieved by photoemission of electrons from the cathode induced by ultraviolet radiation [20, 21]. A drawback of the method described here is first, that it cannot be used for liquids of high natural conductivity when the ionization effect is imperceptible. and, secondly, that at a rather complicated electronic system for detecting the migrating ion layer is required.

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