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KINETICS OF FORMATION OF CHARGE CLUSTERS IN LOW-CONDUCTIVITY LIQUIDS

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Results of an experimental investigation of the space-time characteristics of charge clusters, obtained by the method of light scattering in low-conductivity liquids, are presented.

Introductory Remarks. It is known [1, 2] that electrical conductivity in liquid dielectrics is produced by locally ordered charged microregions (charge clusters) by controlling which (using an electric field) one can regulate the electrophysical characteristics of liquids with the aim of using them in technology (hydrodynamic-electrical transducers). To realize this control over charge clusters, it is necessary to know the kinetics of their formation. Since the theory of the problem is still not developed, experimental methods of investigation are brought to the fore. As one of them, recording of Rayleigh scattering of light transnitted by the investigated low-conductivity liquid can be proposed. The possibility of employing this method stems from the fact that low-conductivity liquids, because of the existence of charge clusters in them, exhibit properties of finely disperse media characterized by:

1) a large difference in conductivity (2–3 orders) between the dispersion inclusions (clusters) and the basic medium (the low-conductivity liquid);

2) fulfillment of the condition $d << \lambda$, where d is the characteristic dimension of a cluster [3, 4]; λ is the wavelength of the probing radiation in the low-conductivity medium.

In this case, according to [3], the following relation can be used:

$$\frac{Q_{\text{scat}}}{Q_{\text{abs}}} = Q = \frac{\pi^3}{9}\left(\frac{d}{\lambda}\right)^3 \frac{(n^2-\chi^2-1)^2+4n^2\chi^2}{n\chi}, \tag{1}$$

and hence

$$d = \frac{3\lambda}{\pi}\sqrt[3]{\left(\frac{n\chi Q}{(n^2-\chi^2-1)^2+4n^2\chi^2}\right)}. \tag{2}$$

Taking into account that $n \approx \sqrt{\varepsilon'}$ and $\chi = \dfrac{\sigma}{2\omega\varepsilon_0\sqrt{\varepsilon'}}$, we can use expression (2) to determine the dimension of the charge clusters d from the experimentally measured Q.

Measurement Procedure. In the experiment (Fig. 1) we simultaneously recorded [4] the power of the incident, transmitted, and scattered radiation of an He–Ne laser.

The linearly polarized light of an He–Ne laser (LGN-222) (λ = 0.63 μm) is divided by a beam-splitting cube (SC) into two beams: beam 1 (B_{inc}) and beam 2 (B_{obj}). To eliminate the influence of the parameters of the beam-splitting cube on the linearity of the polarization of the object beam, a polarizer (P) is used. The instantaneous output power of the laser is controlled by a photodetector (PhD1) using beam 1 (U_{inc}). A quarter-wave plate ($\lambda/4$) converts the linearly polarized light of the second beam into light with circular polarization. A special telescopic system (TS) with small aberrations forms a beam 0.1 mm in diameter. The collimated beam passes parallel to the plane copper electrodes (ELs) (9 × 16 mm) of a measuring cell (MC)

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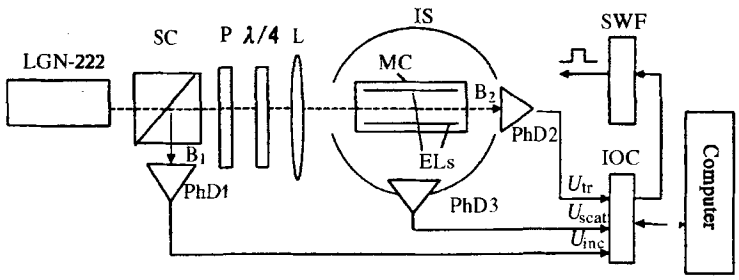


Fig. 1. Schematic diagram of the experimental setup for investigation of the space-time characteristics of charge clusters.

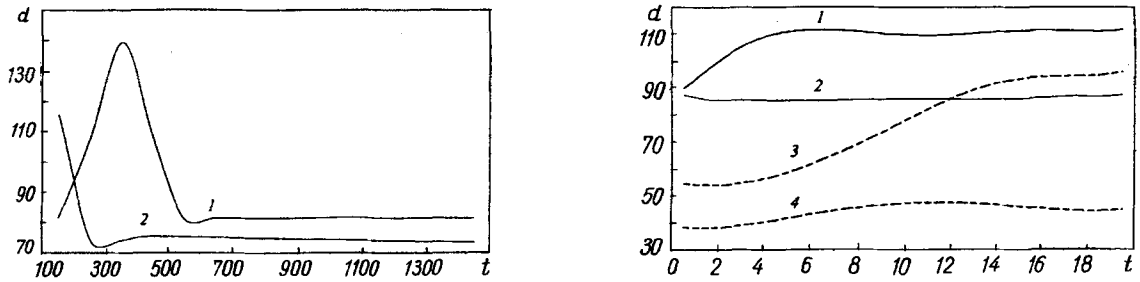


Fig. 2. Results of experimental measurements of the time characteristics of transient processes at the positive electrode (0.1 mm) obtained for a 7% solution of glycerin in diethylsiloxane (1) and a 10% solution of ethylene glycol in heptane (2) at E_0 = 400 V/cm. d, Å; t, μsec.

Fig. 3. Results of experimental measurements of the dimensions of charge clusters d at the positive and negative electrodes for different liquids: 1) diethylsiloxane (+0.3 mm); 2) heptane (+0.3 mm); 3) diethylsiloxane (–0.3 mm); 4) heptane (–0.3 mm). t, min.

placed in an integrating sphere (IS). The measuring cell is moved perpendicularly to the incident light by a micrometric screw; the accuracy of the positioning is ~10 μm. The power of the radiation transmitted by the investigated medium and the power of the radiation scattered by the charge clusters are recorded by photodetectors PhD2 (U_{tr}) and PhD3 (U_{scat}), respectively. The analog signals from the photodetectors are converted into a digital code by an input/output measuring card (IOC) installed in a Pentium 150 computer. The measurement interval (t_{min} = 20 μsec) and the number of measurements are set by programs in accordance with the experimental conditions. The moment of voltage supply to the cell electrodes is taken as the origin of the time scale. The data obtained are processed in the Exel program with an intrinsic set of functions for statistical and mathematical data processing.

The voltage pulses across the electrodes of the measuring cell are formed by a square-wave generator (SWG) (U_{out} = 0–1 kV, τ_r = 100 nsec). The duration of the generated pulses $\tau_p = 10^{-3}$–10^4 is determined by programs in accordance with the experimental conditions. Synchronization of the generated pulses is also realized by programs.

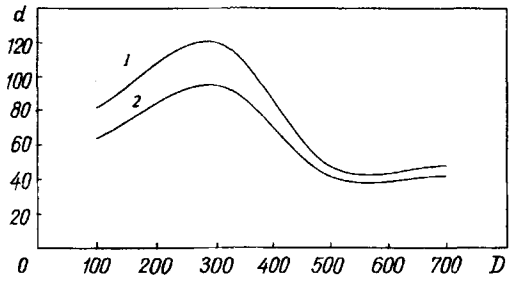
Use of a fast input/output device makes it possible to record the occurring processes in real time.

Obviously, the power of the probing He–Ne laser radiation can be determined as

$$P_{inc}(t) = P_{tr}(t) + P_{scat}(t) + P_{abs}(t). \quad (3)$$

Normalizing to unity, we obtain

$$K_{tr}(t) + K_{scat}(t) + K_{abs}(t) = 1, \quad (4)$$



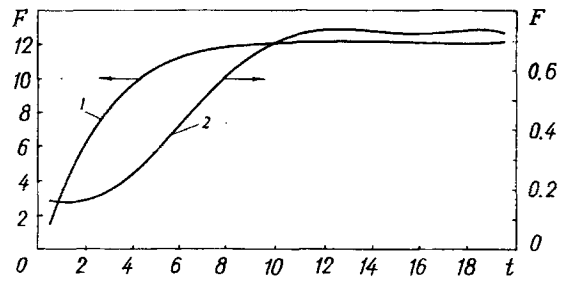


Fig.4. Dependences of the characteristic dimensions of charge clusters d on the near-electrode distance D for different liquids: 1) diethylsiloxane; 2) heptane. D, μm.

Fig. 5. Results of experimental measurements of the rotation of the plane of polarization (F) near the positive (+0.3 mm) electrode, obtained for a 7% solution of glycerol in diethylsiloxane (1) and a 10% solution of ethylene glycol in heptane (2) at E_0 = 400 V/cm. F, deg; t, min.

where

$$K_{tr}(t) = \frac{P_{tr}(t)}{P_{inc}(t)}; \quad K_{scat}(t) = \frac{P_{scat}(t)}{P_{inc}(t)}; \quad K_{abs}(t) = \frac{P_{abs}(t)}{P_{inc}(t)}.$$

Thus, measuring $P_{inc}(t)$, $P_{tr}(t)$, and $P_{scat}(t)$, we can find the transmission and scattering coefficients of the probing radiation, calculate the absorption coefficient from (4), and then determine the characteristic dimension of the charge clusters d from (2). The error in measuring d is determined by the accuracy of recording Q and does not exceed 5%.

Results of Measurements. The experiments performed showed that the dimensions of the clusters change with time most markedly when they are found in the near-electrode layers of the liquid. Here, two transient processes developing at different distances from the electrode were recorded:

1) a rapid process (~100 μsec) occurring at distances of up to 50–100 μm (Fig. 2);

2) a slow process (~5–10 min) occurring at distances of 200–300 μm (Fig. 3).

The dependence of the maximum established dimension of the cluster d on the distance D to the electrode that corresponds to these processes is presented in Fig. 4.

The results obtained should be complemented by the results obtained in [6], where it was established that the slow process correlated with the rotation of the plane of polarization of the light F (Fig. 5).

CONCLUSIONS

We make the following conclusions.

1. The rapid transient process is due to the formation of a dense (predominantly ionic) portion of the near-electrode layer, which correlates with results of [5] obtained by the method of pulsed volt-ampere characteristics.

2. The slow process is due to the formation of a loose (cluster) portion, which correlates with results of [5] obtained by the method of microwave probing and results of [6] obtained by the method of measuring the rotation of the plane of polarization of light transmitted by the investigated low-conductivity medium.

NOTATION

Q_{scat}, Q_{abs}, dimensionless coefficients of scattering and absorption referred to the particle cross-sectional area; n, refractive index of the additive; χ, absorption coefficient of the additive; D, distance from the positive

electrode; d, characteristic dimension of a cluster; λ, wavelength of the probing radiation in the medium of propagation; ε_0, dielectric permittivity; ε, dielectric permittivity of the additive; ε', real part of the dielectric permittivity; σ, conductivity of the additive; ω, angular frequency of the probing radiation; $P_{inc}(t)$, $P_{tr}(t)$, $P_{scat}(t)$, and $P_{abs}(t)$, power of the incident and transmitted radiation and the radiation scattered and absorbed by the clusters, respectively; $K_{tr}(t)$, $K_{scat}(t)$, and $K_{abs}(t)$, normalized coefficients of the transmitted scattered, and absorbed power; E_0, strength of the electric field in the interelectrode spacing; PhD1-PhD3, photodetectors for measuring the incident, transmitted, and scattered powers, respectively; U_{inc}, U_{tr}, and U_{scat}, voltage at the output of photodetectors PhD1, PhD2, and PhD3, respectively; P, polarizer; $\lambda/4$, quarter-wave plate; SWG, programmed square-wave generator; IOD, device for input/output the analog signals into (out of) the computer; F, angle of rotation of the plane of polarization; t_{min}, minimum interval of reading of data into the computer; U_{out}, amplitude of the output radiation of the square-wave generator; τ_r, rise time of the leading edge of a high-voltage pulse; τ_p, duration of a high voltage pulse; t, time. B_1, beam used for control of the laser output power; B_2, object beam. Subscripts: scat, scattering; abs, absorption and absorbed; inc, incident; tr, transmitted; out, output; min, minimum; r, rise; p, pulse.

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