

EFFECT OF TEMPERATURE AND ELECTRIC FIELD STRENGTH ON THE IONIZATION CONDUCTIVITY OF LIQUID AND SOLID CYCLOHEXANE

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The ionization currents induced by ^{137}Cs gamma-rays were measured in a plane-parallel ionization chamber. Measurements were carried out in the -30°C to 40°C temperature range. The build-up time of the current, from the instant ionization is switched on to achievement of a fixed value, is about 10^3 times longer in solid cyclohexane than in the liquid form. The shapes of the current-voltage characteristics are similar for both the solid and liquid cyclohexane forms, but the ionization currents measured in the liquid are some two times higher than the corresponding currents in the solid. In both the liquid and the solid the ionization current increases with increasing temperature. Calculated values of thermal activation energies are lower in the liquid, $W_L \sim 0.05$ eV, than in the solid state, $W_S \sim 0.3$ eV. With increasing electric field strength the activation energy decreases in both solid and liquid cyclohexane.

Introduction

Experimental research on the electric conductivity of dielectric liquids induced by ionizing radiation is well developed [1, 2]. There have also been quite a few researches of this kind dealing with solid dielectrics [3, 4, 5]. Much less work has been carried out, however, on the conductivity induced by ionizing radiation in dielectric substances in both the liquid and solid states with particular consideration of the region near the freezing point [6, 7].

Measuring technique

The principle of the measurement is depicted in the drawings of Fig. 1 (a detailed description of the measuring arrangement will be given elsewhere [8]). The dielectric (of a natural conductivity of $\sigma < 2 \times 10^{-18}$ ohm $^{-1}$ cm $^{-1}$) is ionized by ^{137}Cs gamma-rays (source activity 20 mCi) when the lead shield uncovers the radioactive isotope. The ionization current flowing in the chamber was measured by the electrometer E1 included in the circuit.

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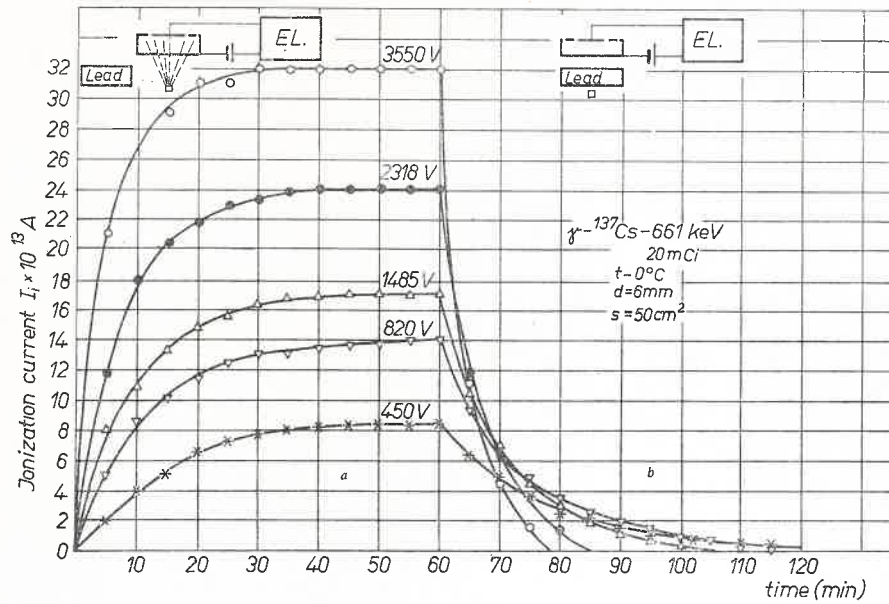


Fig. 1a) Build-up of ionization current from instant of ionization initiation in solid cyclohexane. b) Drop of current after discontinuation of ionization

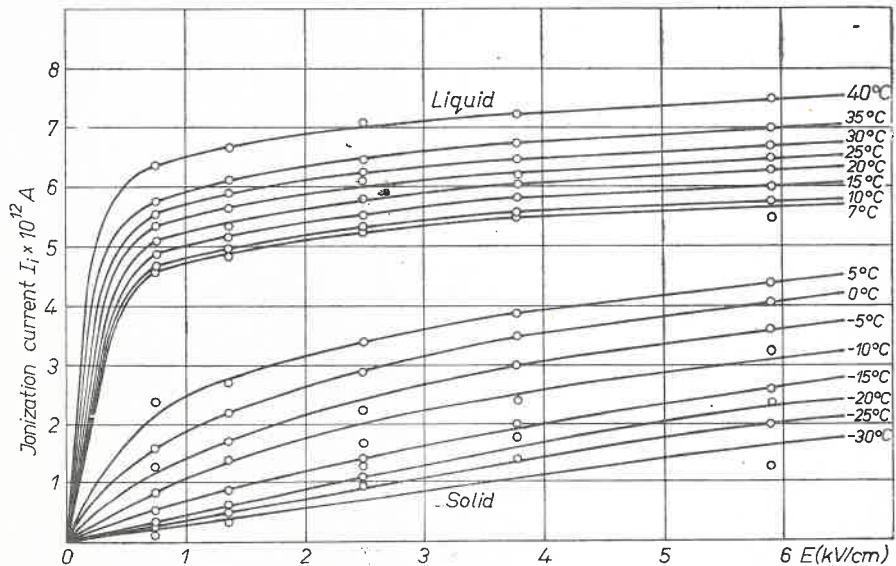


Fig. 2. Current-voltage characteristics in solid and liquid cyclohexane at various temperatures. Electrode spacing $d = 6$ mm, area of collecting electrode $S = 50$ cm²

Results of measurements

In the liquid the ionization current achieves a fixed value several seconds after ionization is initiated. When the liquid is solidified the rise of ionization current lasts much longer, and it reaches a stable value about an hour after ionization is started. The time build-up of the ionization current in solid cyclohexane, and its drop after ionization is discontinued, are illustrated in Fig. 1.

The current-voltage characteristics were determined for the values of ionization currents established thus. The curves pertain to the range of temperatures from -30°C to $+40^{\circ}\text{C}$. As is seen in Fig. 2, the ionization currents in the liquid are higher by several times than in the solid cyclohexane. With an increase of the temperature there is an increase of the ionization current in both the liquid and the solid.

If it is assumed that these are thermally activated processes in both cases [2, 4], it is possible to calculate the activation energy W for them. To this aim, the found values of

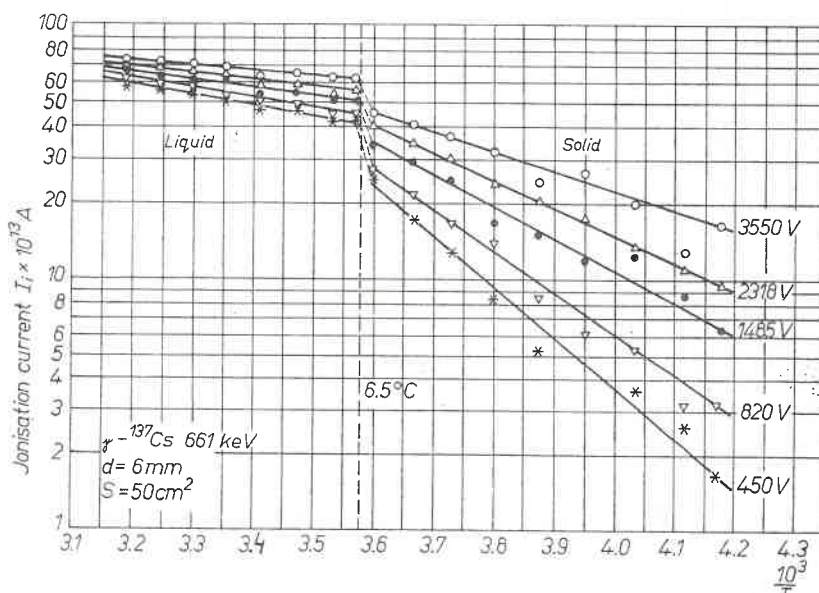


Fig. 3. Dependence of ionization current on reciprocal temperature ($10^3/T$) in semilogarithmic scale

ionization current as a function of temperature are presented in the semilogarithmic system ($\ln I_i, 1/T$). The activation energies W were calculated from the slopes of the curves shown in Fig. 3 in accord with the formulae

$$I_i = I_0 \exp(-W/kT) \text{ and } W = -k \frac{\partial(\ln I_i)}{\partial(1/T)} \quad (1)$$

where T is temperature, k is Boltzmann's constant, and I_0 is a constant independent of temperature. The activation energies for liquid and solid cyclohexane calculated in this

way are presented as functions of electric field strength in Fig. 4. With increasing electric field strength the activation energy decreases in both the liquid and the solid. The drop in activation energy of liquid cyclohexane with increasing field strength had been already ascertained earlier [9, 6].

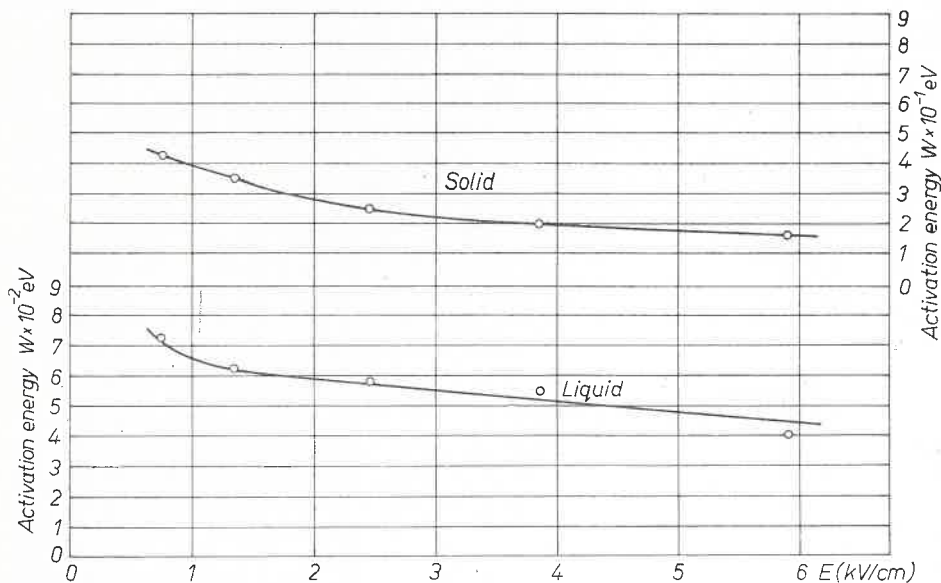


Fig. 4. Dependence of activation energy on electric field strength

Discussion and conclusions

The time curves of the ionization currents in liquid and solid cyclohexane are essentially different. Whereas in the liquid the time within which the current became established at a given voltage did not exceed several seconds, in solid cyclohexane this time equalled about an hour. An analysis of the time dependences of the ionization currents will be made in closer detail later [8]. For the present, we are primarily interested in the problem of the dependence of ionization current upon temperature. The ionization current increases with increasing temperature in both liquid and solid cyclohexane. If it is assumed that in both cases these are thermally activated processes, then the activation energy W will be a measure of the rate of conductivity changes with changes in temperature. The activation energies calculated according to Eq. (1) for liquid and solid cyclohexane for various field strengths, shown in Fig. 4, exhibit both some similarity and some difference for the two states. In both cases the activation energy slightly decreases with increasing electric field strength. It is only in the solid state that the drop in activation energy with increasing electric field strength is somewhat larger. The corresponding values of activation energy for other thermally activated process in liquid cyclohexane are: $W_D = 0.2$ eV for diffusion; $W_\eta = 0.129$ eV for viscosity; $W_n = 0.41$ eV for natural conductivity; and $W_- = 0.131$ eV and $W_+ = 0.165$ for ion mobility [2a].

Comparing the quoted data and Fig. 4, it follows that the value of activation energy for ionization conductivity W_i is the lowest of all. This low value of activation energy W_i in liquid cyclohexane, as compared with other activation energy values, is above all due to the conditions under which it is measured [10]. Contrary to other measurements, the measurement of W_i takes place when the liquid is ionized and at the same time an electric field is acting. This circumstance may also be why the activation energy W_i depends on electric field strength [9].

It seems that the microscopic mechanism of the thermal activation in ionized liquids can be explained in accordance with the theory of Mozumder and Magee [11]. In their model [11] it is assumed that electrons released during ionization remain in a quasi-free state until their kinetic energy becomes lowered to the value of the thermal energy of intermolecular vibrations, $\sim kT$. In the thermal activation process during measurement of the dependence of ionization currents on temperature there occurs a process in which such localized electrons are freed to quasi-free states. Tewari and Freeman [12] interpret thermal activation processes somewhat differently. In their model [12] it is assumed that the excess electrons go over to localized states before their kinetic energy becomes reduced to a value below that of the vibrational energy levels of intramolecular bonds of the molecules (~ 0.2 eV). Experiments show [2] that in the case of the liquid the model of Mozumder and Magee [11] is rather the true one. On the other hand, in the case of solid cyclohexane, when the activation energies W_i are much higher than the corresponding ones for the liquid, the hypothesis of Tewari and Freeman [12] may be correct. It is plausible that in solid cyclohexane the probability of excess electrons being localized on the energy levels of intramolecular bond vibrations is greater than that of their being localized by the levels of intermolecular vibrations. Activation energy values similar to those obtained here have been obtained by Fowler [4] for conductivity induced in solid insulators.

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