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DIELECTRIC PROPERTIES OF POLYETHYLENE

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ABSTRACT

The temperature dependence of dielectric properties in polyethylene was measured in the frequency range from 10 to 10^5 Hz. The frequency dependence of the complex impedance in the complex plane could be fitted by semicircles. The system could be represented by an equivalent circuit of a bulk resistance in series with parallel surface resistance-capacitance combination. The relaxation time, τ, has been evaluated from experimental results. Results reveal that the temperature dependence of τ, is a thermally activated process.

INTRODUCTION

The electrical properties of polymeric materials have attracted interest during the last few decades [1,2]. The reason for this interest is the fact that an appreciable increase in the knowledge of electrical conduction theory as well as structure characteristics have been accomplished from such measurments.

The ac conductivity of these materials shows a frequency-dependent conductivity over a wide range of frequencies [3]. The region of low frequency dispersion approximates to a frequency independent plateau which when tends to obtain extrapolated gives the dc conductivity. In some other conductors, the presence of low frequency dispersion cannont be neglected while determining the effective dc conductivity. Apart from data on ac conductivity, most significant results which may be obtained from dielectric measurements are those on dielectric relaxation times, because observed time constants can be related to molecular absorption mechanism. When a substance exhibits several relaxation times, the average of them can be obtained by a colecole plot of the real versus the imaginary part of the complex dielectric constant [4]. The dielectric measurements have been made of different polymer materials [5,6] and other organic systems [7,8]. Analysis of the ac conductivity for all these substances revealed semiconducting features based predominantly on the hopping mechanism.

The aim of this investigation is to study the dielectric properties of polyethylene as a function of both temperature and frequency.

EXPERIMENTAL

Low density polyethylene (LDPE) films were obtained from plastic company (Alexandria, Egypt). The material is linear and semi - crystalline. Silver paste was painted on the major faces of each specimen as electrodes to form sandwich structures of area ≅0.87 cm^2. The thicknesses of the samples were determined using a micrometer with mean values ranged from 1.0 to 2.5 mm. The linear polyethylene used in the present investigation has the following properties : melt index 0.24 - 0.30 gm, (23°C), density 0.921 - 0.923 gm/Cm^3; crystalline melting point 115°C and softening point 97°C. The dielectric measurements were made as a function of temperature in the frequency range 10 to 10^5 Hz using lock-in amplifier (Stanford type SR 510). Measurements were made while the sample was contained in a cell specially designed to minimize the effect of a copper constantan thermocouple mounted in close proximity to the specimen of interest.

RESULTS AND DISCUSSION

The Cole-Cole diagrams $\varepsilon''(\varepsilon')$ in polyethylene are shown for various temperatures in Fig. (1). A semicircle is obtained for each temperature. The centres of the semicircles at the temperature lie below the real axis. This confirms the idea that there exists a distribution of relaxation times in polyethylene. The data shown in Fig. (1) are used to determine several interesting parameters such as distribution parameter, α, macroscopic relaxation time, τ_o, molecular relaxation time, τ , and its activation energy E_o [9,10]. Knowing α, one can determine τ_o from the relation (10) :-

$$\frac{U}{V} = (\omega \tau_o)^{1-\alpha} \qquad (1)$$

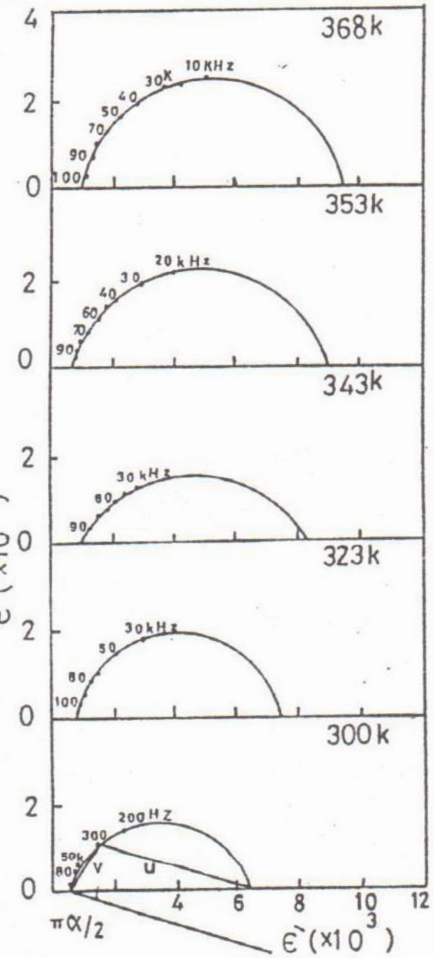

Fig. (1) :

The Cole - Cole digrams in polyethlene.
The frequencies indicated are in Hertz.

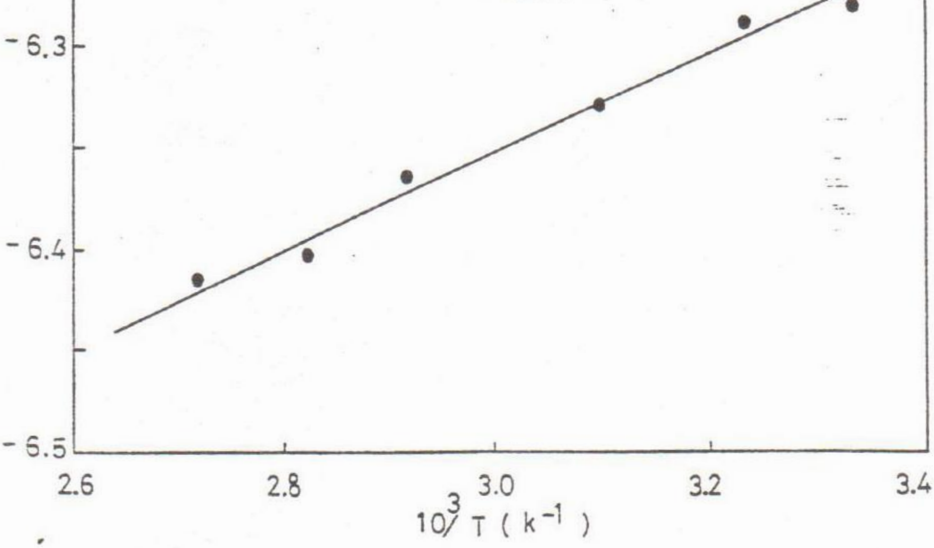

Fig. (2) : Temperature dependence of molecular relaxation time, τ.

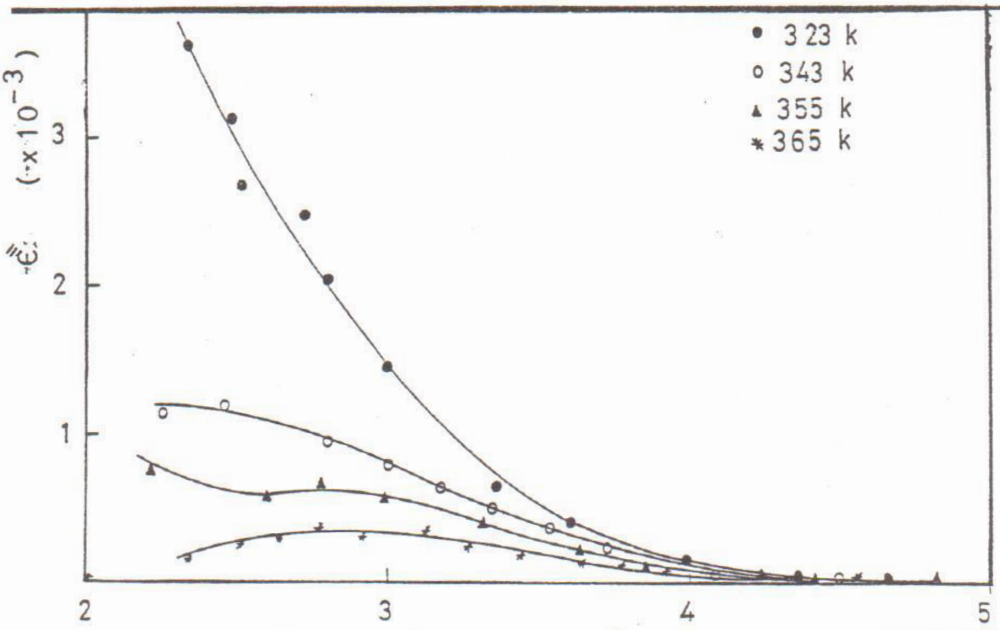

Fig. (3) :

Loss factor ε" vs. log ν for polyethylene at different temperatures.

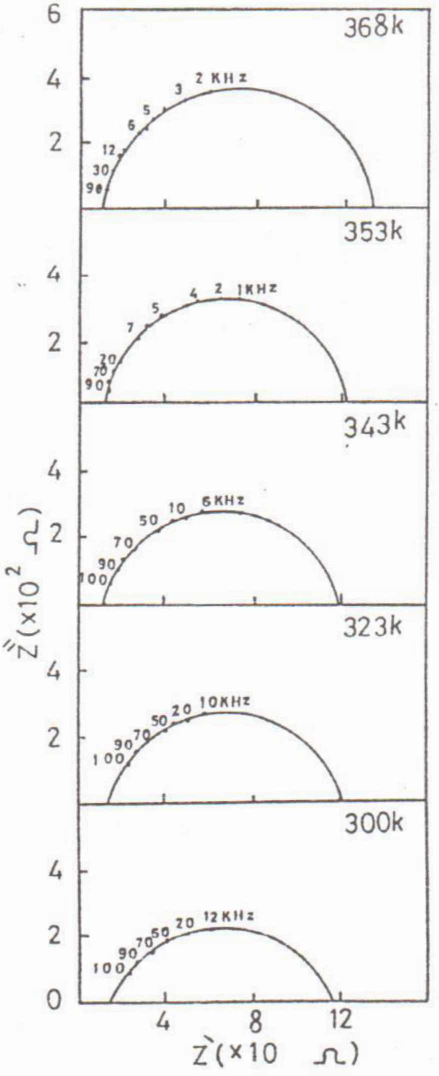

Fig. (4) :

Complex impedance spectra of polyethylene at different temperatures. The frequencies indicated are in Hertz.

Wherer U is the distance on the Cole-Cole diagram between the static dielectric constant ε_o and the experimental point, V the distance between that point and the dielectric constant at the high frequency limit ε_∞ and ω the angular frequency, $2\pi\nu$. Some of these symbols are shown in Fig. (1). The parameter α is equal to zero when the dielectric has only one relaxation time. Whereas, for a distribution of relaxation times it varies between 0 and 1. The extent of the distribution of relaxation times increases with increasing α.

The value of τ_o was found to decrease with increasing temperature. The molecular relaxation time τ, could be evaluated [9] from the formula :

$$\tau = \frac{2\,\varepsilon_o + \varepsilon_\infty}{3\,\varepsilon_o}\,\tau_o \tag{2}$$

The temperature dependence of τ can be described by a thermally activated process [11] of the type :

$$\tau\,(T) = \tau_o \exp\,(E_o/kT) \tag{3}$$

Where τ_o is a constant characterises relaxation time.

Figure (2) shows the plot of the relaxation time as a function of 1/T. By using eqn. (3), The values of τ_o and E_o were found to be $(3.55 \pm 0.02)\ \times 10^{-7}$s and (0.047 ± 0.001) eV, respectively.

The loss factor, ε'', versus log ν for LDPE film is shown in Fig. (3). Similar results have been obtained for linear polyethylene [12,13].

The measured impedance $Z\,(\nu) = Z'\,(\nu) + iz''\,(\nu)$ were plotted in the complex plane for various temperatures. Figure (4) shows typical spectra in relation to the frequency, ν, of the applied sinusoidal voltage. This spectrum configuration leads to an equivalent circuit for polyethylene system comprised of bulk resistance, R_b, in series with parallel R_s - C_s circuit [14,15]; this leads to the established relations :

$$Z' = R_b - R_s / [1 + \omega C_s R_s)^2] \qquad (4)$$

$$Z'' = WC_S R_s^2 / [1 + \omega C_s R_s)^2] \qquad (5)$$

Where R_s is the grain surface resistance, C_s the capacitance of the measured sample and $W = 2\pi\nu$.

Z" vanishes upon approaching both zero and high frequencies, where Z' equals (R_b+R_s) at zero frequency (dc) and only R_b at high frequencies. The values of R_s and R_b for various temperatures were obtained from Fig. (4). The semicircles Z" (Z') dependence would suggest conduction mechanism proceeding in the bulk material with contribution from the surface. The activation energies for the bulk and surface conduction were found to be (0.039 ± 0.002) eV and (0.013 ± 0.001) eV, respectively. The calculated value of ≈0.039 eV for the bulk conduction was estimated from the plot of log σ versus 1/T, Fig. (5).

The frequency dependence of the conductivity σ' versus frequency in logarithmic representation is shown in Fig. (6) for different temperatures. It can be oberved that it is similar to that expressed by a function of frequency [16] :

$$\sigma' \alpha \omega^s$$

The frequency , S, is O < S < 1 . In the case of the transport processes in disordered media [11], the low - frequency conductivity has the same frequency dependence as already observed in the polyethylene material, which characterizes a process of hopping conduction [17].

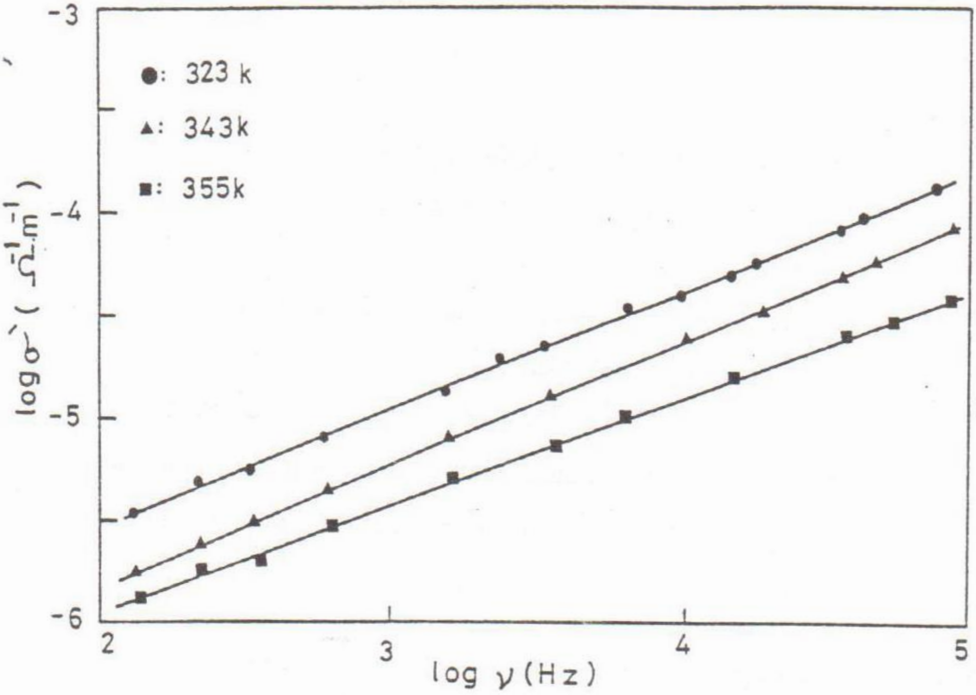

Fig. (5) : Temperature dependence of dc conductivity of polythylene.

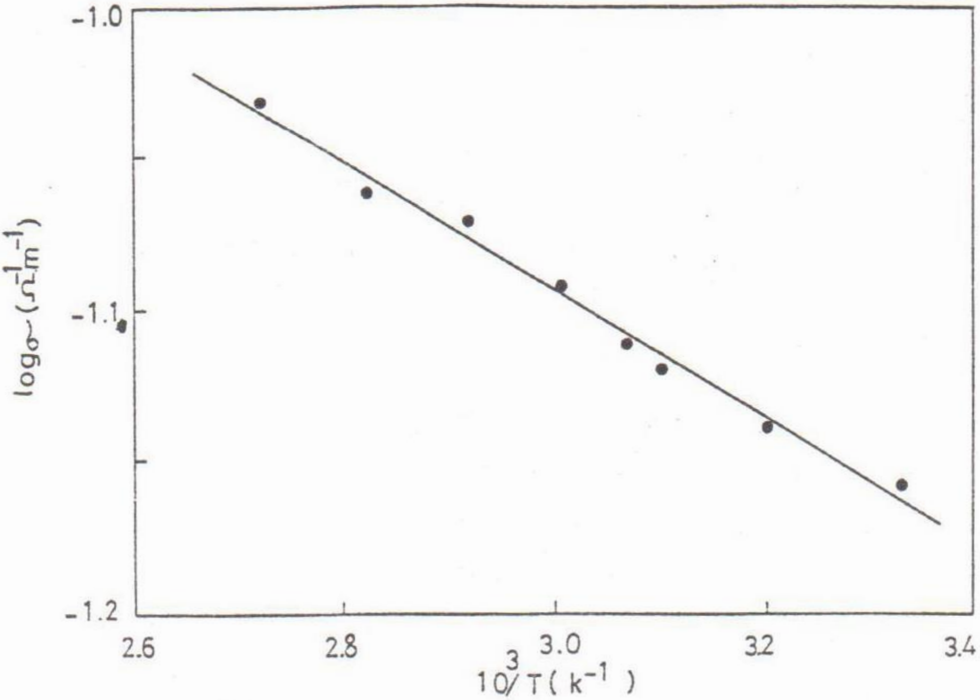

Fig. (6) The frequency dependence of the conductivity of polyethylene at different temperatures.

CONCLUSTION :

From the measurement and analysis of the dielectric properties of polyethylene in the frequency range from 10 to 10^5 Hz at temperature from 300 to 368 K, the following points can be concluded :

1 - The ac conductivity in polyethylene reveals semiconducting features based predominantly on the hopping mechanism.
2 - The complex impedance plots exhibit arcs of circle characteristic of bulk resistance in series with parallel surface resistance - capacitance combination.
3 - The temperatures dependence of molecular relaxation time can be expressed by thermally activated process.

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