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THE PROPAGATION OF ULTRASONIC WAVES IN CONDENSER OIL AT PRESENCE OF DC ELECTRIC FIELD

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1. Introduction

A DC electric field externally applied to a dielectric liquid modifies the acoustic properties of the liquid.

Our earlier studies [1, 9] on the influence of an electric field on the propagation velocity of ultrasonic waves were based on the assumptions that the variations observed were due to changes in the free energy. Theoretical assessments for typical dielectric liquids and nearbreakdown field strengths show that the change in ultrasonic wave velocity should not exceed several mm/s and should occur in times comparable with the time determined by orientational relaxation of the polarization vector related with rotation of the dipolar molecules in the field.

Thus, the change in ultrasonic velocity observed on applying a DC electric field to an oil as a function of its viscosity and the structure and size of its molecules should occur in times of the order of microseconds. However, experiment fails to confirm the above assumptions [1, 9]. In fact, the change in ultrasonic velocity and the times involved are considerably in excess of those calculated theoretically. Hence, in the first part of our present paper, we invoke other theoretically admissible mechanisms to explain the changes in ultrasonic wave velocity in condenser oil in an external DC electric field.

In part 2 we report the influence of an electric field on the amplitude of the absorption coefficient of the wave in mineral non-polar and synthetic dipolar oils versus the electric field strength and temperature. We also propose different explanations for the variations observed. One of the causes of the experimentally observed behaviour of the dielectric liquids subjected to the action of a DC electric field due to the presence of gas bubbles.

2. The electric field surrounding a gas bubble

It will be remembered [2] that a dielectric liquid sample, even if purified repeatedly by physico-chemical methods, still contains an amount of small gas bubbles. The gaseous cavity the bubble is surrounded by a region of strong inhomogeneity of the electric field [6] causing the free electric charges present in a real medium to be sucked away towards the surface of the sphere. If the resultant charge thus attached to the bubble is non-zero, the bubble will drift in the electric field in the direction of the electrode of opposite sign. The electric field distribution inside the bubble and in its neighbourhood is shown in Fig. 1. The region between the two planar parallel electrodes is filled with a liquid dielectric of relative electric permittivity ε_r containing a spherical bubble with an absolute electric permittivity approximately equal to that of vacuum.

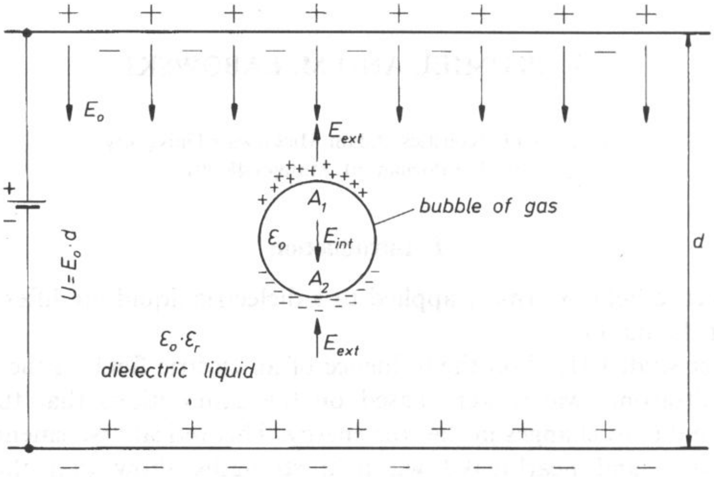

Fig. 1. Electric field distribution within and outside gas bubble in a dielectric liquid.

As the dielectric becomes polarized, electric charge is induced on the surface of the spherical cavity with the surface density distribution D_R given by

$$D_R = D_{\max} \cdot \cos\phi . \tag{2.1}$$

These polarization charges give rise to an electric field of their own independent of the initial field E_0.

The electric field strength E_{int} induced inside the bubble by the charges on its surface is in this case directed parallel to the original external field E_0.

As a result, the real field $E_{\mathrm{int(re)}}$ inside the bubble will be

$$E_{\mathrm{int(re)}} = E_0 + E_{\mathrm{int}} , \tag{2.2}$$

whereas the resultant electric field strength $E_{\mathrm{ext(re)}}$ outside the sphere in its neighbourhood will be given by

$$E_{\mathrm{ext(re)}} = E_0 - E_{\mathrm{ext}} . \tag{2.3}$$

The electric field strength induced within the sphere by the electric charges cosine-distributed on its surface can be shown to amount to

$$E_{\mathrm{int}} = \frac{D_{\max}}{3\varepsilon_0}, \tag{2.4}$$

whence

$$D_{\max} = 3\varepsilon_0 E_{\mathrm{int}}. \tag{2.5}$$

The electric flux $\Delta\Phi$ produced by the charge ΔQ accumulated on the spherical surface element ΔS in the neighbourhood of a point A_1 or A_2 is

$$\Delta\phi = \Delta Q = D_{\max} \cdot \Delta S = 3\varepsilon_0 \cdot E_{\mathrm{int}} \Delta S. \tag{2.6}$$

The flux splits into two parts, the one inside the sphere, $\Delta\Phi_{\mathrm{int}}$, and the other outside the sphere, $\Delta\Phi_{\mathrm{ext}}$. The two can be defined as follows:

$$\Delta\Phi_{\mathrm{int}} = \varepsilon_0 E_{\mathrm{int}} \cdot \Delta S, \quad \Delta\Phi_{\mathrm{ext}} = \varepsilon_0 E_{\mathrm{ext}} \cdot \Delta S. \tag{2.7}$$

This enables us to determine the strength of the electric field E_{ext} induced outside the sphere in the point A_1 or A_2 by the polarization charges:

$$3\varepsilon_0 E_{\mathrm{int}} \cdot \Delta S = \varepsilon_0 E_{\mathrm{int}} \cdot \Delta S + \varepsilon_0 E_{\mathrm{ext}} \cdot \Delta S, \tag{2.8}$$

i.e.,

$$E_{\mathrm{ext}} = 2E_{\mathrm{int}}. \tag{2.9}$$

Hence, we get from (2.3)

$$E_{\mathrm{ext(re)}} = E_0 - 2\mathrm{E}_{\mathrm{int}}. \tag{2.10}$$

As we know [6], the normal components of the resultant field strength at the boundary between two dielectrics have to be inversely proportional to the electric permittivities:

$$\frac{E_{\mathrm{ext(re)}}}{E_{\mathrm{int(re)}}} = \frac{E_0 - 2E_{\mathrm{int}}}{E_0 + E_{\mathrm{int}}} = \frac{\varepsilon_0 \cdot 1}{\varepsilon_0 \varepsilon}, \tag{2.11}$$

whence

$$E_{\mathrm{int}} = E_0 \frac{\varepsilon_r - 1}{1 - 2\varepsilon_r}. \tag{2.12}$$

With regard to the initial component of the field, E_0, we finally obtain the following expressions fot the real electric field existing inside and outisde the bubble:

$$E_{\mathrm{int(re)}} = \frac{3\varepsilon_r}{1 + 2\varepsilon_r} E_0, \quad E_{\mathrm{ext(re)}} = \frac{3}{1 + 2\varepsilon_r} E_0. \tag{2.13}$$

In the neighbourhood of the air bubble, the local electric field is weakened because the coefficient of relative permittivity of the liquid is $\varepsilon_r > 1$, whereas within the bubble it increases compared to the initial field.

For example, in the case of synthetic oil for which $\varepsilon_r = 4$ the field at the surface of the bubble falls to one third, and a considerable local field inhomogeneity arises in its neighbourhood. The respective field gradient attaches the free charge to the bubble and causes the latter to wander towards the neutralizing electrode.

The resultant electric field strength inside the bubble — which is much in excess of its macroscopic value — lowers the electric breakdown field value of the oil, an effect undesirable in the case of insulating systems.

In strong electric fields the breakdown value of the gas in the bubble obeys Paschen's law according to which the pre-breakdown voltage U_p is dependent on the diameter of the bubble d and the pressure p.

With a view to assess the electric breakdown value, the voltage across the diameter of the bubble along the lines of force of the field can be assumed to be given by $U \approx E_0 \cdot \varepsilon_r \cdot d$, where E_0 is the field strength in the oil, ε_r its relative electric permittivity, and d the diameter of the bubble. If U attains the pre-breakdown value U_p, electric discharge takes place inside the bubble. For an air bubble with $d = 0.1 - 1$ mm at 25°C and a pressure of $p = 1000$ hPa, the discharge voltage $U_p = 4.3 \cdot d^{0.63}$ [10].

Hence, the electric field strength E_p [kV/mm], at which ignition of incomplete discharges in an air bubble of diameter d [mm] takes place is found to be

$$E_p = \frac{4.3 \cdot d^{0.63}}{d \cdot \varepsilon_r}. \tag{2.14}$$

By Eq. (2.14) ignition in bubbles of diameter $d = 1$ mm immersed in mineral oil with $\varepsilon_r = 2.2$ should occur at $E_p = 2$ kV/mm, whereas in bubbles of diameter $d = 0.1$ mm — at $E_p = 4.6$ kV/mm.

In the mineral oil studied by us, where $d = 2 \cdot R_{ef} = 0.0916$ mm and $\varepsilon_r = 2.2$, the theoretical breakdown value amounted to $U_p = 0.95$ kV, whereas the electric field strength $E_p = 4.7$ kV/mm and was strongly in excess of that applied in our measurements. In practice, the formation and growth or vanishing of bubbles in oil can be due to a number of factors, such as temperature, moistness, the chemical composition of the oil, and the electric field strength.

An intense electric field can moreover destroy the molecular structure of the paraffine hydrocarbons by disrupting the bonds between the carbons and hydrogens, leading to the emergence of hydrogen and methane bubbles. On the other hand, molecules of aromatic hydrocarbons adsorb atoms of free hydrogen thus impeding the formation of hydrogen bubbles.

All this contributes to modify the acoustic properties of the medium under the action of an external electric field, both by affecting the propagation velocity of ultrasonic waves and their absorption coefficient in the medium. Apparently, these factors act in a way to remove the gas bubbles from the liquid [10].

Tests carried out by applying an AC field failed to affect the ultrasonic propagation velocity and signal amplitude.

3. Experimental

3.1. The method

The change in absorption coefficient of the ultrasonic wave on application of the DC field was recorded by the pulse method. A block diagram of our setup is shown in Fig. 2. In the course of our studies we measured the amplitude of the ultrasonic pulse of frequency 6 MHz before and after applying the DC electric field at right angles to the oil sample. The DC electric field ranged from 0 to 300 kV/m and the temperature from 0 to 40°C. The application of analog memory enabled us to record the ultrasonic pulse amplitude which, on traversing an analog-digital converter, was fed into the computer memory. Examples of the time-variations of the ultrasonic pulse amplitude recorded for the two oils investigated are given in Figs. 3a and 3b.

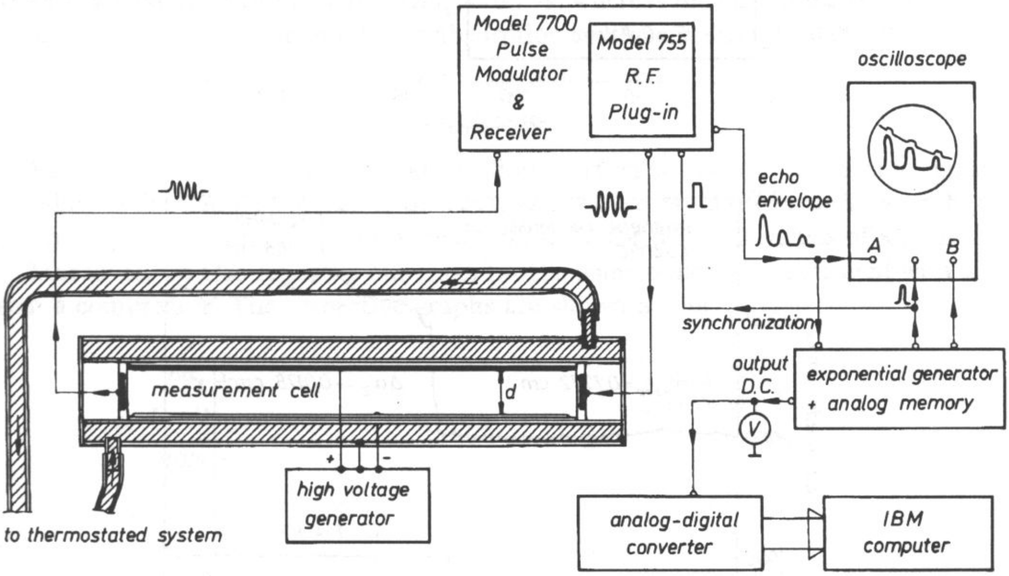

Fig. 2. Block diagram of our setup for determination of the amplitude absorption coefficient of an ultrasonic wave propagating in a dielectric liquid.

The procedure adopted by us involved calculating the mean value $\langle u_0 \rangle$ of the ultrasonic pulse amplitude over a period of 120 s preceding application of the external DC field as the mean value $\langle u_E \rangle$ during 120 s with the field switched on.

Let $\langle u_0 \rangle$ and $\langle u_E \rangle$ correspond, respectively, to the zero-field amplitude absorption coefficient $\langle \alpha_0 \rangle$ of the wave and $\langle u_E \rangle$ to its variation $\langle \Delta\alpha_E \rangle$ after once the electric field E_0 was applied. The field modifies the acoustic properties of the oil. Now $\langle u_0 \rangle$ and $\langle u_E \rangle$ can be expressed as follows:

$$\langle u_0 \rangle = U_A \exp^{-\langle \alpha_0 \rangle L} \quad \text{and} \quad \langle u_E \rangle = U_A \exp^{-\langle \alpha_E \rangle L}, \tag{3.1}$$

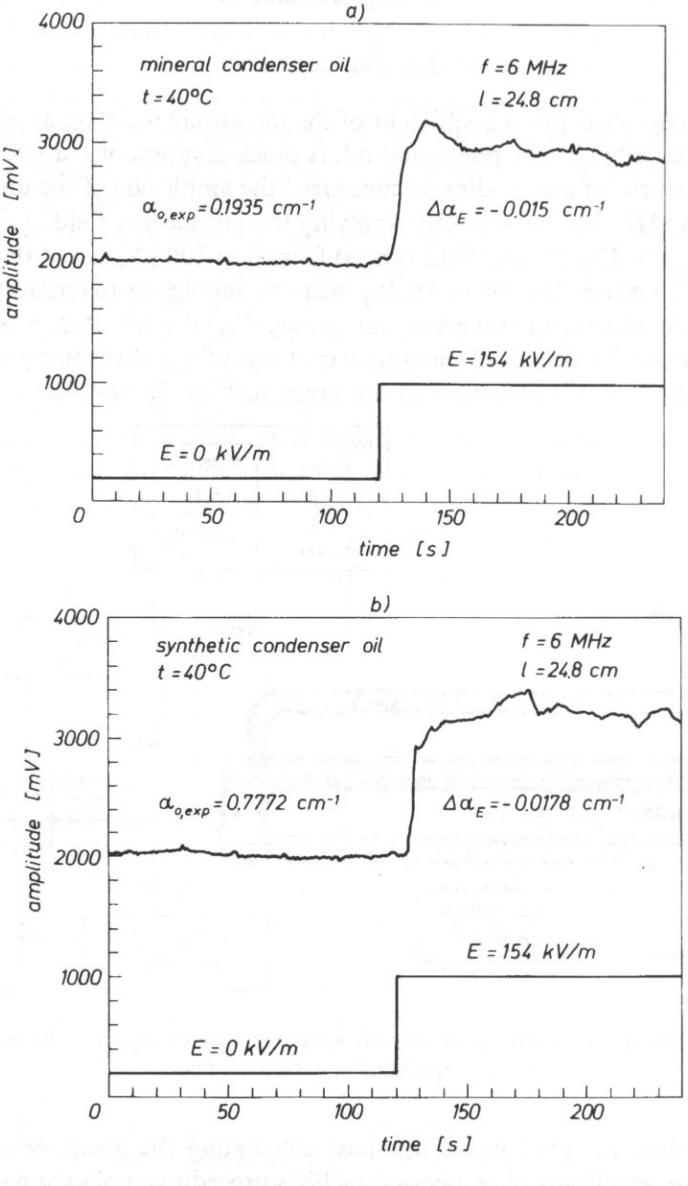

Fig. 3. The changes in ultrasonic pulse amplitude (6 MHz) prior to and after application of an external electric field E=154 kV/m; a) in mineral condenser oil (MCO), b) in synthetic condenser oil (AKB).

where $l = 24.8$ cm is the axial distance between the measuring transducer and U_A the ultrasonic pulse amplitude obtained from the receiver transducer at $l = 0$.

Hence, we derive the following expression for $\Delta\langle\alpha_E\rangle$, the variation of mean amplitude absorption coefficient of the wave due to the external field, as measured directly in our experiment:

$$\Delta\langle\alpha_E\rangle=\langle\alpha_E\rangle-\langle\alpha_0\rangle=\frac{1}{l}\cdot\ln\left(\frac{\langle u_0\rangle}{\langle u_E\rangle}\right). \tag{3.2}$$

Thus, in order to determine $\Delta\langle\alpha_E\rangle$ we only have to measure the length l of the path traversed by the wave in the oil and the ratio of the pulse amplitudes prior to and after applying the field. The relative error incurred when measuring the change in amplitude absorption coefficient, $\Delta(\Delta\langle\alpha_E\rangle)/(\Delta\langle\alpha_E\rangle)$ calculated by the full differential method, is

$$\frac{\Delta(\Delta\langle\alpha_E\rangle)}{\Delta\langle\alpha_E\rangle}=\frac{\Delta u(\langle u_E\rangle+\langle u_0\rangle)}{\langle u_E\rangle^2\ln^2\left(\frac{\langle u_0\rangle}{\langle u_E\rangle}\right)}+\frac{\Delta l}{l}. \tag{3.3}$$

With the values $l=24.8$ cm, $\Delta l=0.01$ cm, $\Delta u=1$ mV, $\langle u_0\rangle=1$ V and $\langle u_E\rangle=2$ V, we arrive at a relative error of $\Delta(\Delta\alpha_E)/(\Delta\alpha_E)=0.2\%$, whereas the absolute error incurred in measuring the change in amplitude absorption coefficient $\Delta(\Delta\alpha_E)=0.000055\ \mathrm{cm}^{-1}$.

4. Results

Variations in the amplitude absorption coefficient of the ultrasonic pulse travelling in the sample exposed to different values of external field are shown in Fig. 4. Similarly Fig. 5 shows variations temperature T for different values of kV/m.

Moreover, we also repeated our experiments, using mineral oil saturated with air from a compressor. The respective graphs are shown in Fig. 6.

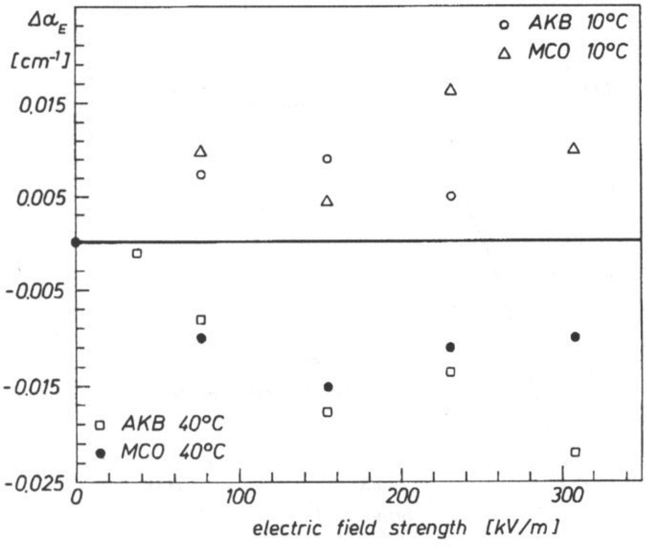

Fig. 4. Variations in amplitude absorption coefficient $\Delta\alpha_E$ of the ultrasonic wave in the oils versus the external electric field strength E.

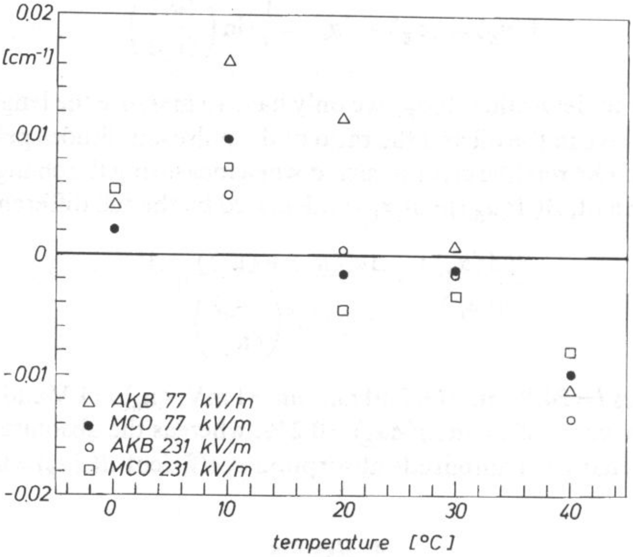

Fig. 5. The same as in Fig. 4 for the temperature of the oils.

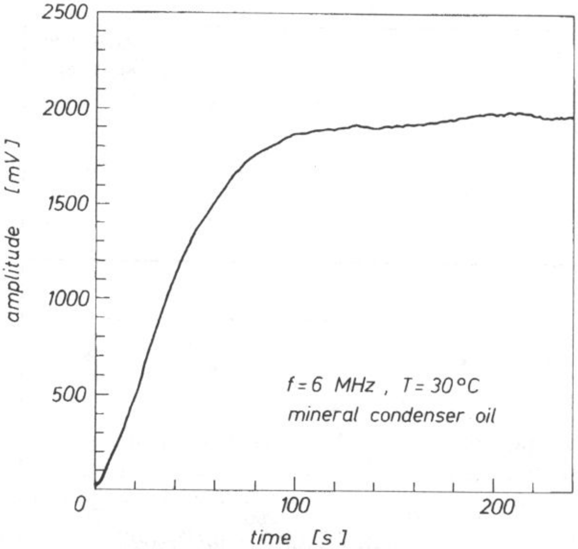

Fig. 6. Time-variation of the ultrasonic pulse amplitude 6 MHz, 30°C on traversal of a given path in the mineral oil after the production therein of a cloud of air bubbles.

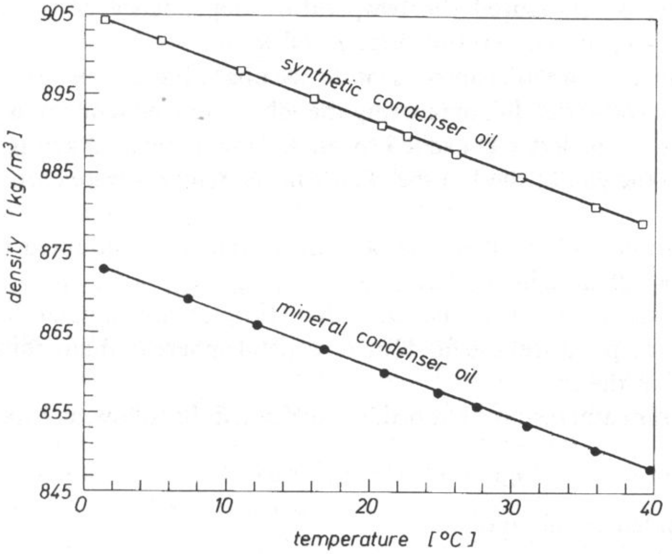

Fig. 7. The density $\rho(T)$ of the oils versus the temperature T.

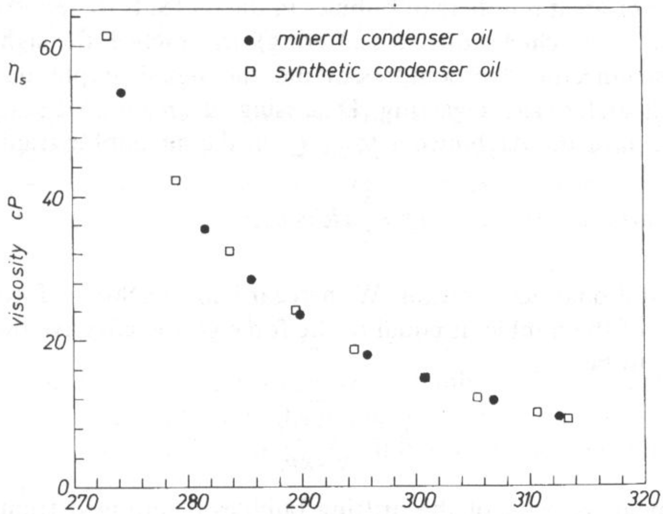

Fig. 8. The laminar viscosity $\eta_s(T)$ of the oils as a function of temperature.

Additionally, we measured the temperature dependence of the density ρ and laminar viscosity η_s on temperature Fig. 7 and 8.

We determined ρ with microprocessor device operating on the basis of the method proposed by KRATKY *et al.* [8], measuring the vibration frequency of a small, narrow tube in the form of the letter U excited by an AC magnetic field acting on a constant magnet fixed to the end of the U-tube. These measurements were carried out with an accuracy of 10^{-4}.

The correlation coefficient of the $\rho(T)$ measurements amounted to -0.9998 in mineral oil and -0.9999 in synthetic oil.

Laminary viscosity η_s was measured with a Höppler viscosimeter determining five times (for each temperature) the fall time of a metal sphere of diameter 15.57 mm and $\rho = 8140$ kg/m^3 in the oil.

To the experimental results obtained, we attributed the following exponential for η_S:

$$\eta_s(T) = 1.335 \cdot 10^7 \cdot \exp^{-0.0455 \cdot T}, \tag{4.1}$$

for mineral condenser oil, and

$$\eta_s(T) = 2.335 \cdot 10^7 \cdot \exp^{-0.0474 \cdot T}, \tag{4.2}$$

for synthetic oil, where the temperature is in Kelvin [K] and the viscosity is expressed in units of [cP].

5. Discussion of the results

The presence of great numbers of bubbles in the oil leads to very strong damping of the ultrasonic wave (chiefly due to scattering). In practice, the signal vanished.

Only on disconnecting the compressor did the signal amplitude begin to rise slowly as the oil underwent degassing. Degassing of an oil of density ρ_0 proceeds through the action of the Archimedes force Q on the air bubble (radius R):

$$Q = \frac{4}{3} \pi R^3 g \rho_0, \tag{5.1}$$

where g is gravitational acceleration. With regard to the Stokes force F which, at uniform motion of the bubble, is equal to the force Q, the effective radius R_{ef} of the bubble is found to be

$$R_{ef} = \sqrt{\frac{9 v \eta_s}{2 g \rho_0}}, \tag{5.2}$$

with v — the mean velocity of the drifting bubbles determined from the recorded variation in ultrasonic pulse amplitude in the course of degassing.

With the values occurring in the preceding example, $\rho_0 = 854.23$ kg/m^3, $\eta_s = 13.33 \cdot 10^{-3}$ Ns/m^2, $v = 294$ μm/s, we obtained for the effective radius of the bubble $R_{ef} = 45.8$ μm.

If incomplete discharge takes place in the bubble oxygen O_2 goes over into ozon O_3. Since ozon combines with the surrounding oil (especially at higher temperatures) the bubbles decrease in size [10]. Consequently, the ultrasonic amplitude absorption coefficient decreases. This process may well be responsible for the characteristic shape of the $\Delta\alpha_E(E, T)$ graphs of Figs. 4 and 5 obtained experimentally. In both cases the ultrasonic absorption specifically decreases at higher temperatures, at which diffusion of the gas out of the bubbles proceeds more intensely.

6. Ultrasonic wave propagation in a liquid with gas bubbles

Bubbles impede the propagation of ultrasonic waves, particularly so if the vibration eigenfrequency of the bubbles lies close to the frequency inducing their vibrations.

The whole process is fundamentally determined by the following factors:

- part of the acoustic energy is dissipated by the oscillating bubble which, as it were, becomes a spherical source emitting outwards in all directions,
- the temperature of the bubble rises as a result of its periodic oscillations induced by the ultrasonic wave and heat is transmitted to the surrounding medium, and
- losses in energy also come from the arisal of hydrodynamic microcurrents in the liquid medium surrounding the oscillating bubble.

The description of absorption and scattering of ultrasonic waves propagation in a liquid medium with gas bubble involves the following concepts:

- the effective active damping cross-section σ_e,
- the effective active absorption cross-section σ_a, and
- the effective active scattering cross-section σ_r.

These quantities are interrelated as follows:

$$\sigma_e = \sigma_a + \sigma_r. \tag{6.1}$$

Thus, the effective damping cross-section σ_e is meant to denote the area of the plane perpendicular to the direction of incidence of the ultrasonic wave for which the transmitted energy is equal to the sum of the energies absorbed and scattered by the bubbles. The active absorption cross-section σ_a and active scattering cross-section σ_r are defined as follows:

$$\sigma_a = \frac{Q_a}{I_p}, \qquad \sigma_r = \frac{Q_r}{I_p}, \tag{6.2}$$

where Q_a — is the energy flux absorbed by the bubbles, Q_r — is that scattered by the bubbles, and I_p — is the intensity of the incident wave.

In determining the values of these quantities use should be made of the following expressions [3]:

$$\sigma_e = \frac{4\pi r^2\left(\frac{\delta}{\eta}\right)}{\left(\frac{f_0^2}{f^2} - 1\right)^2 + \delta^2}, \tag{6.3}$$

$$\sigma_a=\frac{4\pi r^2\left(\frac{\delta}{\eta}-1\right)}{\left(\frac{f_0^2}{f^2}-1\right)^2+\delta^2},\tag{6.4}$$

$$\sigma_r=\frac{4\pi r^2}{\left(\frac{f_0^2}{f^2}-1\right)^2+\delta^2},\tag{6.5}$$

with f_0 — the resonance frequency of the bubble, f — its oscillation frequency, r — its radius, δ — the damping coefficient of its oscillations in the liquid medium, and $\eta=2\pi r/\lambda$ — the ratio of its circumference and ultrasonic wavelength.

For amplitudes small compared with the radius of the bubble, the resonance frequency of its oscillations is equal to

$$f_0=\frac{1}{2\pi r_0}\sqrt{\frac{3\gamma\left(P+\frac{2\sigma_i}{r}\right)}{\rho}},\tag{6.6}$$

with P — the pressure in the medium, $\gamma=C_p/C_v$ — the ratio of the specific heat at constant pressure and constant volume, σ_1 — the coefficient of surface tension on the gas-liquid boundary, and ρ — the density of the liquid medium.

For bubbles with a radius $r>15$ μm the influence of the surface tension on the pressure inside the bubbles can be neglected, and Eq. (27) reduces to

$$f_0=\frac{1}{2\pi r_0}\sqrt{\frac{3\gamma P}{\rho}}.\tag{6.7}$$

7. Acoustic degassing of liquids

A liquid containing gas bubbles becomes degassed under the action of an acoustic wave, especially if the wave is of high intensity.

The process of degassing runs in three stages, consisting of

(i) an increase in size of the small bubbles due to diffusion of the gas present in solution in the liquid;

(ii) coagulation of pairs and groups of small bubbles into bubbles of considerable size; this stage is dominated by the action of acoustic fluxes as well as Bjerkness and Brillouin forces; and, finally,

(iii) rapid flow of the now enlarged gas bubbles towards the surface of the liquid and their removal from the system.

The enhancement of the degassing process by the acoustic wave is due to the specific behaviour of the gas bubble, considered as an oscillator with one degree of freedom.

The analysis of the interaction between the bubble and the wave is greatly simplified if one assumes $r \ll \lambda$ (where λ is the acoustic wavelength in the liquid).

The periodical variations in pressure in the medium induced by the ultrasonic wave force the bubble to oscillate. Its radial oscillations obey the following linear differential equation [4]:

$$m\frac{\partial^2 v}{\partial t^2}+b\frac{\partial v}{\partial t}+kv=-P_A e^{i\omega t}, \tag{7.1}$$

where v denotes the deviation in volume of the bubble from its stationary value v_0, k is its elasticity coefficient, ω the angular frequency, P_A the pressure amplitude, t time, b dissipation coefficient, ant m generalized mass.

The general solution of Eq. (7.1) is

$$v(t)=\frac{-P_A e^{i(\omega t-\varphi)}}{\sqrt{m^2(\omega_0^2-\omega^2)^2+b^2\omega^2}}, \tag{7.2}$$

with φ the shift in phase between the inducing force and the oscillation, and $\omega_0=(k/m)^{0.5}$ the oscillation eigenfrequency of the bubble.

The deviation in volume from its stationary value becomes maximal at excitation by a wave of frequency close to that of the eigenfrequency of the bubble.

Under this condition, the rate at which diffusion of the gas into the bubbles proceeds is particularly high. They coalesce, they grow in size, thus considerably accelerating the process of degassing. In this process an important role belongs to the Bjerkness forces acting between two oscillating spheres (bubbles) acoustically excited to oscillate in a liquid medium.

If the oscillation frequencies of the two spheres are equal the force of interaction between them is [4, 5]:

$$F_B=\frac{4\pi\rho a^2 b^2 u_a u_b}{l^2}\cos\varphi, \tag{7.3}$$

with a, b their radii, l the distance between their centres, u_a and u_b the oscillation velocities of their surfaces, and ρ the density of the liquid.

Above, the shift in phase φ between the two bubbles is

$$\varphi=\operatorname{arctg}\frac{A-B}{1+AB}, \tag{7.4}$$

where the quantities A and B are

$$A=\frac{\delta_1\omega_{01}^2}{\omega_{01}^2-\omega^2}, \quad B=\frac{\delta_2\omega_{02}^2}{\omega_{02}^2-\omega^2}. \tag{7.5}$$

with δ_1 and δ_2 denoting the respective oscillation damping coefficients and ω_{01}, ω_{02} the angular eigenfrequencies.

The oscillation velocities of the surfaces of two bubbles of radii a and b in a liquid of density ρ forced to oscillations under the action of an ultrasonic wave exerting a pressure P_A are, respectively,

$$u_a=\frac{P_A}{a\omega\rho\sqrt{\left(\frac{\omega_{10}^2}{\omega^2}-1\right)^2+\delta_1^2}}, \qquad u_b=\frac{P_A}{b\omega\rho\sqrt{\left(\frac{\omega_{20}^2}{\omega^2}-1\right)^2+\delta_2^2}}. \tag{7.6}$$

Obviously, at resonance i.e. for $\omega=\omega_0$ the pulsation velocity becomes maximal. On insertion of these expressions into (6.7) we arrive at the followig expression for the Bjerkness force acting between the two spheres:

$$F_B=\frac{4\pi ab\,P_A^2\cos\varphi}{\rho\omega^2 l^2\sqrt{\delta_1^2+\left(\frac{\omega_{01}^2}{\omega^2}-1\right)^2}\sqrt{\delta_2^2+\left(\frac{\omega_{02}^2}{\omega^2}-1\right)^2}}. \tag{7.7}$$

In the case of in-phase oscillations ($\varphi=0$) the two spheres experience a force of attraction, whereas if ($\varphi=\pi$) they mutually undergo repulsion.

In practice, in-phase oscillation take place if the oscillating bubbles are more or less of the same size greatly enhancing the process of degassing.

Thus ultrasonic investigation is seen to permit the measurements of the effective size of gas bubbles and the gas content in oils. The method should be of considerable interest with regard to applications in the Oil Industry.

Acknowledgements

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