LOW-FREQUENCY DIELECTRIC PROPERTIES OF ACETONE

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Abstract – Some peculiarities of the dielectric behaviour of the interaction of translation and rotation components of heat motion for acetone molecules were studied in the present research. As a criterion for relative importance of these components, comparison of the activation energy of translation was chosen: electro conductivity, viscosity and dielectric relaxation. Observed results are discussed in the frame of translation-rotation dynamics of the molecular clusters of acetone.

Keywords: dielectric spectroscopy, dielectric properties of liquids, molecular spectroscopy, clusters, acetone.

I. INTRODUCTION

The dielectric spectroscopy of molecular liquids studies the processes of formation of the orientation component of the electric polarization with the aim of obtaining the information about the structure of matter, in which there are rotation motion of the structure unity-carriers of the constant electric dipoles. Traditional interpretation of the relaxation dispersion-absorption spectra is based on two postulates [1, 2]: 1 – unique molecules are only a type of dipoles carriers 2 – orientation mobility of carrier is determined by its structure and nearest neighbour.

At the same time, structuring of amorphous matter only partly coincides with the definition of nearest neighbour. In addition, more different type of local ordering, - intermediate ordering, and reflects existence of many level hierarchy of kinetic stable molecular clusters [3 – 7]. When comparing with accidental and non-structured fluctuations, for each cluster there are finite, fixed number of molecules and fixed number of the physical - chemical characteristics.

Due to relatively high sensitivity the spectral methods are considered as a main source of the experimental information about the clusters when comparing with so-called direct diffraction methods. It is well known that clusterization takes place in more than 90% of molecules in some aliphatic monoalcoholes, amides, acetic acid, and also in pure water, and water solutions of some amino acids   [8 – 13]. Common for all these compounds is that they have strong structural effect of intermolecular hydrogen bonding. Strong anisotropy of angular correlation for clusterized molecules gives good conditions for the experimental detection of the intermediate ordering in such mediums.

Clusterization in so - called «non - associated» liquids, which are built on the base of isotropic and comparatively weak Van der Waals intermolecular forces, is less pronounced. Characteristic for such objects decreasing of the space - time distribution of the intermediate and nearest order result to approaching the dielectric parameters of supposed Van der Waals clusters and unique molecules.

The main aim of the present research is to consider the problem of some dielectric aspects of local structuring in non-associated liquids. We supposed, that under quantitative comparison of rotational dynamics for unique molecules and Van der Waals clusters there should exist noticeable difference of the size of region in the range of which the elemental acts of relaxation is realized, which correspond to these two types of the dielectrically active structural unit. As a criterion for such discrepancy we choose the comparison of the temperature coefficients (activation energy) of the dielectric relaxation time, shearing viscosity and the electrical conductivity. Such an approximation was chosen because all of these three transportation phenomena is considerably different when comparing they translation and vibration input in respective dynamic process.

Acetone was chosen as a typical representative of non - associated liquids. Some information about the dielectric properties of acetone are given in the review [14], about viscosity in Ref. [15]. The method of dielectric spectroscopy allows to study the dielectric properties of object in more details due to high value of the dielectric susceptibility (static) of acetone, and due to high degree of molecules dissociation, respectively. Using high - frequency measurements in the range of $10^5 \div 10^6 \, Hz$ excludes totally many source of mistake, which are characteristic for standard methods of measuring the electroconductivity at zero frequency.

II. THEORY

According to theory of Debay [1] the dispersion of the dielectric susceptibility ε' and related dielectric losses ε'' are described as follows:

$$\varepsilon' = \varepsilon_\infty + \left(\varepsilon_\circ - \varepsilon_\infty\right)\left[1+\left(\frac{f}{f_m}\right)^2\right]^{-1} \qquad (1)$$

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$$\varepsilon'' = \left(f \big/ f_m \right)\left(\varepsilon_{\circ} - \varepsilon_{\infty}\right)\left[1 + \left(\frac{f}{f_m}\right)^2\right]^{-1} \qquad (2)$$

where f - is the frequency of the voltage applied to the sample; f_m – is thye critical frequency, corresponding to the maximal value $\varepsilon'' = (\dfrac{\varepsilon_0 - \varepsilon_{\infty}}{2}$ when $f = f_m$) and determined according to the relation

$$f_m = \left(2\pi\tau\right)^{-1} \qquad (3)$$

where τ - is the dielectric relaxation time, which characterizes the speed of the formation process of the orientation component of the dielectric polarization.

Dielectric susceptibilities ε_0 and ε_{∞} fix the frequency range, corresponding to the amplitudes of the orientation component of the polarization according to the equations

$$\frac{f}{f_m} << 1 \text{ or } f \cong 0{,}01 f_m \text{ and } \varepsilon' = \varepsilon_0 \qquad (4)$$

$$\frac{f}{f_m} >> 1 \text{ or } f \cong 100 f_m \text{ and } \varepsilon' = \varepsilon_{\infty} \qquad (5)$$

In both cases the dielectric losses $\varepsilon'' \to 0$. At the same time, in real dielectrics always $\varepsilon'' \neq 0$, due of the fact that at the high and low wings of the relaxation spectra there are overlapping components of the low-frequency edge of the rotation - vibration spectra and Ohm's losses caused by the electric charge transfer. In the latter case equation (2) is replaced by follows equation

$$\varepsilon'' = \varepsilon''_d + \varepsilon''_{\gamma} \qquad (6)$$

where the dipole input to ε'_d by taking into consideration (4) is given as follows

$$\varepsilon''_d = (\varepsilon_0 - \varepsilon_{\infty})(\frac{f}{f_m}) \qquad (7)$$

and Ohmic one as

$$\varepsilon''_{\gamma} = 1{,}8 \cdot 10^{12} \frac{\gamma}{f}, \qquad (8)$$

where γ – electrical conductivity in the units of Ohm^{-1}cm^{-1}. If (7) and (8) is given as below

$$\frac{\varepsilon''_d}{f} = (\varepsilon_0 - \varepsilon_{\infty}) \cdot f_{m^{-1}}, \qquad (9)$$

$$\varepsilon''_{\gamma} f = 1{,}8 \cdot 10^{12} \gamma, \qquad (10)$$

then the right part of (9) and (10) contains components which are not frequency dependent (at fixed temperature). This peculiarity allows using the values of the dielectric susceptibilities, which have been measured at frequencies, corresponding to condition (4), for the separation of the relaxation and Ohmic input in dependence on the character of frequency dependence of multiply $\varepsilon'' f$ and $\dfrac{\varepsilon''}{f}$.

III. METHOD

To determine the Ohmic branch of the absorption spectra in the frequency range of $10^5 \div 10^6 Hz$ we used the modified differentiation method of the complex conductivity (admittance) $Y^* = G + iB$ sample of dielectric by $\varepsilon^* = (\varepsilon' - i\varepsilon'')$ resonance measurements using Q - meter

These values are given by next relations

$$Y^* = i\omega C_0 \varepsilon^* \text{ or } G = \omega C_0 \varepsilon'$$

$$B = \omega C_0 \varepsilon''$$

where $i = \sqrt{-1}$; ω - angular frequency, C_0 – is the capacity of empty sample holder ($\varepsilon' = 1$; $\varepsilon' = 0$).

By using the effect of amplification by voltage allows to connect the active part of the G admittance with the inverse value of the resonance Q^{-1} value of the resonance quality factor (Q – meter). Complex part B of admittance is compensated by self reactivity of contour at fixed frequency and is determined by the resonance value of the capacity C of calibrated capacity. The formulas for the standard methods are looking like

$$\varepsilon' = (C_{mc} - C)C_0^{-1} \qquad (11)$$

$$\varepsilon'' = (Q_{\varepsilon}^{-1} - C)C_{mc}C_0^{-1} \qquad (12)$$

where subscripts «mc» and «ε» are related to the resonance measurements of the resonance contour of Q meter under free contacts and when the sample is connected to the Q - meter, respectively.

It is worth to mention that (11) and (12) is obtained for the case when the admittance of the sample holder is equal to the admittance of the measurements contour of Q - meter. But, in many real cases measured admittance Y_{ε}^* is always differs from the real admittance $Y^*(\varepsilon)$ to the value of the parasitic admittance Y_{par}^*:

$$Y_{\varepsilon}^* = Y^*(\varepsilon) + Y_{par}^* \qquad (13)$$

At frequencies not higher than about 5 MHz, main input to Y^*_{par} is given by the additional part, which is necessary for coupling the sample holder to the thermostat and, respectively, making longer the connector with Q - meter. In latter case, this results to additional and difficultly understands part to the measurements of capacities and quality factors (Q – meter).

Differential modification of the standard method allows to exclude necessity to take into account this additional part by introduction new adjustment to the resonance, when the sample holder without the sample is connected with the Q-meter. In this case, by analogy with (13):

$$Y^*_1 = Y^*(1) + Y^*_{par} \qquad (14)$$

Where Y_1^* and $Y^*(1)$ measured and real admittance of the sample holder without sample. Extracting (14) from (13) excludes the input of parasitic admittance:

$$Y^*_\varepsilon - Y^*_1 = \left[Y^*(\varepsilon) + Y^*_{par} \right] - \left[Y^*(1) + Y^*_{par} \right] = Y^*(\varepsilon) - Y^*(1) \quad (15)$$

Standard transformations show that there is new modified relations corresponding to equation (15):

$$\varepsilon'' = 1 + (C_1 - C_0^{-1}) C_0^{-1} \qquad (16)$$

$$\varepsilon'' = (Q_\varepsilon^{-1} - Q_1^{-1}) C_{mc} C_0^{-1}$$
or $\qquad\qquad (17)$
$$\varepsilon'' = (Q_\varepsilon^{-1} - Q_1^{-1}) C_1 C_0^{-1}$$

Due to the relation $C_1 = C_{mc}$. Formally, relations (12) and (18) are equal. At the same time, coupling the differences $\Delta C = C_1 - C_\varepsilon$ and $\Delta Q^{-1} = Q_\varepsilon^{-1} - Q_1^{-1}$, to the same holder noticeable decreases the probability of systematic mistake when estimate ε''.

Experiments were performed at frequencies 0,5; 1; 2; 3; 5 MHz and temperatures 20, 25, 30, 35, 40° C.

As a sample holder we used a capacity of cylindrical type. Temperature kept constant. Acetone with trade mark ChC (chemical clean) was used without preliminary purification.

IV. RESULTS

Results of measurements are summarized in Table 1, which consists of two parts, containing values measured with and without sample.

Table 1.Resonance capacities C_ε C_1(pF) and quality factors (Q – meter)Q_ε, Q_1 of acetone at different frequencies F(MHz) and temperatures t°(C).

f	0,5		1,0		2,0		3,0		0,5	
t	C_ε	Q_ε	C_ε	Q_ε	C_ε	Q_ε	C_ε	Q_ε	C_ε	Q_ε
20	298,0	26	175,5	30	144,5	47	187,0	76	146,5	88
25	299,0	29	176,0	33	145,0	49	188,0	74	147,0	86
30	299,0	29	176,5	33	145,5	50	188,5	76	147,5	90
35	301,0	30	178,5	36	146,0	54	190,0	80	148,5	90
40	302,0	32	179,0	39	147,0	51	190,0	76	149,0	83
C_1	309,0		186,5		155,0		197,0		156,0	
Q_1	167		150		162		165		162	

The leveling of the temperature – frequency dependences, $C_\varepsilon, Q_\varepsilon$ and $\Delta C = C_1 - C_\varepsilon$ and $\Delta Q^{-1} = Q_\varepsilon^{-1} - Q_1^{-1}$ were made with the least square coefficients method.

$$\Delta C = a_{\Delta c} + b_{\Delta c} t \qquad (18)$$

$$\Delta Q = a_{\Delta Q} + b_{\Delta Q} t \qquad (19)$$

The values of the empirical relations (18) – (19) which were approximated taking into consideration the temperature dependences ΔC, ΔQ under different frequencies are given in Table 2.

Table 2. Coefficients of the equations (18)–(19) determined by the least square method f (MHz)

f	0,5	1,0	2,0	3,0	5,0
$a_{\Delta C}$	15,38	14,79	13,96	13,25	11,86
$b_{\Delta C}$	-0,200	-0,190	-0,176	-0,163	-0,129
$a_{\Delta Q} \cdot 10^4$	362,78	304,84	169,03	76,99	51,65
$b_{\Delta Q} \cdot 10^6$	-262,8	-237,2	-104,3	-22,0	0,0

Estimated according to the data of Table 2 values of C, and also known from literature values of the dielectric susceptibility ε_0 for acetone in function of temperature is give by:

$$\varepsilon_0 = -8,8718 + \frac{8817,452}{T} \qquad (T = 273,2 + t^0 C), \qquad (20)$$

We used these estimated values for determination of C which is in the formulas (16) and (17), according to relation

$$C_0 = \Delta C (\varepsilon_0 - 1) \qquad (21)$$

Table 3. Parameter C_0, for the dielectric susceptibility ε'' and electrical conductivity γ for acetone at different frequencies and temperatures.

f	0,5		1,0		2,0		3,0		5,0		$\gamma \cdot 10^6$	
t	C_0 (pF)	ε''	C_0	ε''	C_0	ε''	C_0	ε''	C_0	ε''	for (23)	for (27)
20	0,537	17,85	0,519	9,25	0,492	4,67	0,472	3,03	0,438	1,840	5,10	5,06
21	0,530	17,96	0,513	9,27	0,486	4,69	0,466	3,06	0,434	1,857	5,12	5,10
22	0,521	18,02	0,506	9,31	0,480	4,72	0,461	3,08	0,430	1,874	5,15	5,15
23	0,516	18,11	0,500	9,33	0,474	4,74	0,455	3,11	0,425	1,896	5,19	5,19
24	0,509	18,21	0,493	9,38	0,468	4,77	0,449	3,15	0,421	1,914	5,23	5,24
25	0,502	18,28	0,486	9,42	0,462	4,80	0,444	3,17	0,417	1,932	5,26	5,29

26	0,495	18,35	0,480	9,45	0,456	4,82	0,438	3,21	0,413	1,951	5,29	5,33
27	0,488	18,49	0,473	9,49	0,449	4,86	0,432	3,24	0,409	1,970	5,33	5,38
28	0,480	18,60	0,466	9,54	0,443	4,89	0,426	3,28	0,404	1,994	5,38	5,42
29	0,473	18,75	0,458	9,61	0,436	4,93	0,420	3,31	0,402	2,004	5,42	5,47
30	0,465	18,87	0,451	9,66	0,429	4,98	0,414	3,35	0,395	2,040	5,48	5,51
31	0,457	19,00	0,444	9,72	0,423	5,01	0,408	3,39	0,391	2,061	5,52	5,56
32	0,450	19,14	0,437	9,77	0,416	5,05	0,402	3,43	0,386	2,087	5,57	5,61
33	0,442	19,30	0,429	9,85	0,409	5,10	0,396	3,47	0,382	2,109	5,62	5,65
34	0,434	19,47	0,422	9,91	0,402	5,15	0,389	3,52	0,377	2,137	5,68	5,70
35	0,426	19,58	0,414	9,99	0,395	5,20	0,383	3,56	0,372	2,166	5,74	5,75
36	0,417	19,87	0,406	10,08	0,388	5,25	0,376	3,62	0,367	2,195	5,81	5,80
37	0,409	20,06	0,399	10,15	0,381	5,31	0,370	3,67	0,362	2,226	5,88	5,84
38	0,401	20,27	0,391	10,24	0,374	5,36	0,363	3,73	0,357	2,257	5,95	5,89
39	0,393	20,44	0,383	10,34	0,366	5,43	0,356	3,79	0,352	2,289	6,02	5,93
40	0,384	20,80	0,375	10,44	0,359	5,50	0,350	3,84	0,347	2,363	6,12	5,98

The values of C_0 and ε'' presented in Table 3 were calculated by equation (18) by using $\Delta Q^{-1} = Q_\varepsilon^{-1} - Q_1^{-1}$ from the table 2 and C_1 from Table 1. It is seen that in all cases ε'' increases with temperature and this is characteristic for Ohmic input. According to (10) $\varepsilon'' f$ should be constant at different frequencies at fixed temperature. According to data $\varepsilon'' f$ given in Table 4 this criterion obeyed.

Table 4. $\varepsilon'' f$ calculated by using the data of Table 3 at different frequencies f (MHz) and temperatures $t(^0C)$.

$\dfrac{t}{f}$	0,5	1,0	2,0	3,0	5,0
20	8,9	9,3	9,3	9,1	9,2
25	9,1	9,4	9,6	9,5	9,7
30	9,4	9,7	10,0	10,1	10,2
35	9,8	10,0	10,4	10,7	10,8
40	10,4	10,4	11,0	11,5	11,8

Equalization of (10) by logarithmic method

$$\lg \varepsilon'' = \lg \gamma' - \lg f \quad (\gamma' = 1,8 \cdot 10^{12} \gamma) \tag{22}$$

Allows approximate the frequency dependence for the conductivity of acetone by equation:

$$y = a + bx \qquad (b = -1) \tag{23}$$

Values of γ found for 5 different fixed frequencies are presented in Table 3. By the other side, the temperature dependence of the conductivity of acetone is given by:

$$\gamma = \gamma_0 \exp\left(-\frac{E_\gamma}{RT}\right) \tag{24}$$

(E_γ 's the energy of activation of conductivity, R – Boltsman constant), or in linear form:

$$\ln \gamma = \ln \gamma_0 - \frac{E_\gamma}{RT} \tag{25}$$

Estimation of γ_0 and E_γ (equation 26) by using least square method gives:

$$\gamma = 6,945 \cdot 10^{-5} \exp\left(-\frac{1525,35}{RT}\right)$$

and $\tag{26}$

$E_\gamma = 1525,35\ cal/mol$

Values of γ calculated according to (27) are presented in the last column of Table 3. The differences between two set of data for γ do not higher than 1%,. The latter is the evidence of good coincidence of the obtained data in the frequency range $10^5 \div 10^6$ Hz.

In the frame of our task we performed same estimations by suing the data on viscosity [15] and the time of dielectric relaxation [14] for acetone, according to the next relations dew scribing the temperature dependences:

$$\eta(puaz) = 21,225 \cdot 10^{-5} \exp\left(\frac{-1525,35}{RT}\right)$$

and $\tag{27}$

$$E_\eta = 1583,71 \frac{cal}{mol}$$

$$\tau(sec) = 1,94 \cdot 10^{-13} \exp\left(\frac{1650}{RT}\right)$$

and $\tag{28}$

$$E_\eta = 1650 \frac{cal}{mol}$$

Table 5. Experimental [15] and calculated by using (27) viscosities (puaz) for acetone at different temperatures $t(^0C)$

t	-20	-10	0	10	20	30	40	50	60
$10^2\ \eta_{exp}$	0,500	0,442	0,395	0,356	0,322	0,293	0,268	0,246	0,230
$10^2\ \eta_{cal}$	0,495	0,439	0,393	0,355	0,322	0,294	0,271	0,250	0,232
Δ, %	-1,0	-0,7	-0,5	-0,3	0,0	+0,3	+1,1	+1,6	+0,9

Table 6. Experimental [14] and calculated by using (28) time of the dielectric relaxation $\tau(sec)$ for acetone at different temperatures $t(^0C)$

t	1	20	40
$10^{12}\ \tau_{exp}$	3,98	3,34	2,76
$10^{12}\ \tau_{cal}$	4,02	3,30	2,75
Δ, %	+1,0	-1,2	-0,4

The experimental and calculated values of η, τ tau are compared in Table 5,6. The differences also are shown which do not higher than 1% in both cases.

V. DISCUSSION

The results of comparison ob data obtained in the present work with those existing in the literature shows that there is

exist the next relation between the activation energies of the electro-conductivity, viscosity and dielectric relaxation:

$$E_\tau \geq E_\eta \geq E_\gamma \qquad (29)$$

The tendency of decreasing for activation energies is seen for three conduction mechanisms in the above given sequence, respectively. Taking into consideration that the conductivity is a translational process, one may conclude that there is strong correlation between sequence and vibration mobility in acetone.

According to Frenkel model [2], elementary act of relaxation takes place via one step vibration transfer of unique molecule through barrier, created by nearest molecules. In modern consideration the free volume added to this model as a necessary condition for realization of transition. It is supposed that translational transition of molecule is finished when rotational re-orientation to angle $2\pi/3$ radian is achieved [16]. Energy, necessary for creation of vacancy is determined as the difference of the activation energies for translation and vibration diffusion, respectively. Such interpretation is on contradiction with the experimental results which shows equality for these two values for acetone.

Translation - vibration interaction is postula-ted also in one particle model of Debay [1]. According to this model relaxation act is understand as sequence of plural small angle turn of polar molecule in viscous medium. Formula given by Debay connects the time of dielectric relaxation of molecule with its volume and viscosity. It is noticeable that only unique fact, that formula of Debay takes place are such a dissolved solutions for which condition is obey [8,17]:

$$V_{DA} \geq V_{DNA},$$

where V – is the volume of molecule, index «DA» and «DNA» correspond to the dielectrically active and dielectrically not active molecules, respectively. There is similarity on a quantitative level with the cluster model, which takes into consideration next conditions:

1) Dielectric relaxation of cluster is a collective process and includes next stages:
a) collapse of cluster b) reorientation of free clusters b) formation of new cluster with new direction of the effective dipole. Second stage is most long in time, because of its determines with the size and form of the molecule. This fact is a reason for existing two different scales for the process of collapse-reformation of clusters. More fast scale respects to the dielectrically inactive clusters, which can be considered as an analogue above mentioned small molecules of non-polar solvents.

2) In difference to the dielectric relaxation, in other transport phenomena such a many step cluster dynamics is absent. For this reason the sequence $E_\tau \geq E_\eta \geq E_\gamma$ is observed. In case of water all three activations are close to each other. In case of normal butyl alcohol $E_\eta = 4500 \dfrac{cal}{mol}$,

$E_\tau = 7100 \dfrac{cal}{mol}$.

3) Most of the acts of collapse – formation are the dielectrically inactive. Input to the orientation polarization comes only from those processes in which the projection of the effective dipole moment is change for given direction.

4) Energy of interaction of clusterized molecules are higher than the thermal energy of motion. Stability of cluster may be disturbed only in case of optimal close relation of cluster with other molecules. Latter results to breaking the cluster with its further formation as a next step.

VI. CONCLUSION

It may be stated, that acetone is characterized by strong correlation of rotation and translation dynamics, which reveal in the comparatively close values of the activation energies for the transport of the electric charge, mass and electric dipoles. Experimentally proved such correlations allow supposing existence of stable clusterization of acetone molecule. Taking into consideration high electric dipole of acetone, which is considered as a main factor ruling the clusterization one may conclude that the main factor is a dipole - dipole interaction. In this connection one may suppose, that observed unique dispersion in the dielectric spectra of acetone are due of the superposition of individual cluster input. Addition of highly polarized liquids may increase the frequency range of the dielectric spectrum of acetone and make more suitable conditions for its solving.

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