# DIELECTRIC PROPERTIES OF NON-ASSOCIATED LIQUIDS IN LOW-FREQUENCY REGION

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Some peculiarities of dielectric development interaction of translation and rotation components of acetone molecule heat motion are studied.

### Introduction.

The dielectric spectroscopy of molecular liquids studies the formation processes of orientation component of electric polarization with the aim of information obtaining about medium structure in which the rotation (pivoting) motion of structural units-carriers of constant electric dipoles is carried out. The traditional integration of relaxation spectrums of dispersion-absorption is based on two postulates [1]: 1) single molecules are single type of dipole carriers; 2) carrier orientation mobility is defined by its structure and short range ordering.

Besides, the structuring of amorphous mediums only partially coincides with short range ordering conception. One more extensive variety of local ordering is called intermediate order [2] reflects the existence of multilevel hierarchy of kinetically stable molecular clumps-clusters [3-5]. The final strongly fixed number of molecules and concrete set of physicochemical characteristics corresponds to each cluster in contrast to occasional and structureless formations.

The spectroscopic methods are the main source of experimental information about clusters in the result of their more high sensitivity to structuring on the level of intermediate order in the comparison with possibility of so-called "direct" diffraction methods. In particular, it is established in the limits of dielectric spectroscopy method of condensed mediums that clustering captures more than 90% molecules in some aliphatic mono-alcohols, amides and acetic acid and also in pure water and aqueous solutions of some amino-acids. The presence of structure-forming effect of intermolecular hydrogen bond is the general for all these objects. The strongly expressed anisotropy of angular correlation of clustered molecules causes the favorable conditions for experimental revealing of intermediate order in these mediums.

The clusterization of so-called "non-associated" liquids constructed on the base of isotropic and comparably weak Van der Waals intermolecular interactions is less obvious one. The decrease of space-time distribution of intermediate and short range orderings character for these objects leads to approaching of dielectric parameters of supposed Van der Waals clusters and single molecules.

### Aim of work.

The aim of present investigation is in that consideration of sole dielectric agenda aspects of local structurization in "non-associated" liquids. It is supposed that the essential dissimilarity of region dimensions within the limits of which the relaxation elementary acts corresponding to these two types of dielectrically active structural units realize, should exist at quantitatively comparable rotation dynamics of single molecules and Van der Waals clusters. The comparison of time temperature coefficients ("activation energies") of dielectric relaxation, shear viscosity and conductivity on constant current has been chosen in the capacity of criteria of such dissimilarity, i.e. these three transfer phenomena essentially differ on relation of translation and rotation contributions in corresponding dynamic process.

The acetone is chosen as typical representative of "nonassociate liquids". Any information on systematic investigations of acetone conductance in known literature is absent. The existing information is separated and is contradictory. The present information is separate and contradictory. The method of dielectric spectroscopy allows us to safely feel this blank because of big equilibrium ("static") acetone dielectric constant and enough high degree of dissociation of molecules correspondingly. The use of high-frequency measurements in range  $10^5 \div 10^6$  Hz is totally excluded that many difficultly taking into consideration the error sources which are related to standard methods of liquid conductance definition at zero frequency.

The experiment has been carried out on Q-meter Tesla BM-560 at frequencies 0,5; 1; 2; 3; 5 MHz and temperatures 20, 25, 30, 35, 40°C. The condensator of cylindrical type is applied in the capacity of holder. The sample temperature is supported as constant by ultra-thermostat.

#### **Investigation theory**

According to Debye [1] the dispersion of dielectric constant  $\varepsilon'$  and dielectric loss  $\varepsilon''$  connected with dispersion are described by the equation:

$$\varepsilon' = \varepsilon_{\infty} + \left(\varepsilon_{\circ} - \varepsilon_{\infty}\right) \left[ I + \left(\frac{f}{f_m}\right)^2 \right]^T$$
(1)

- - i

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

Here *f* is frequency (Hz) of external electric field applied to the sample;  $f_m$  is cutoff frequency corresponding to maximal value  $\varepsilon_M^{''} \left(=\frac{\varepsilon_0 - \varepsilon_\infty}{2} \text{ at } f = f_m\right)$  and defined by the

relation:

$$f_m = (2\pi\tau)^{-1},$$

where  $\tau$  is dielectric relaxation time characterizing the formation process rate of orientation component of electric polarization.

The dielectric constants  $\varepsilon_0$  and  $\varepsilon_{\infty}$  fix the boundaries of frequency range corresponding to amplitude ( $\varepsilon_0 - \varepsilon_{\infty}$ ) of orientation polarization according to following conditions:

$$f_{f_M} << 1 \text{ or } f \cong 0,01 f_M \text{ and } \varepsilon' \equiv \varepsilon_0$$
 (4)

$$f/f_M >> 1 \text{ or } f \cong 100 f_M \text{ and } \varepsilon' \equiv \varepsilon_{\infty}$$
 (5)

The dielectric loss  $\varepsilon'' \rightarrow 0$  in both cases. However, in real dielectrics always  $\varepsilon'' \neq 0$ , i.e. the low-frequency edge of oscillation-rotation spectrum created by transfer of electric charges

In last case the equation (2) is exchanged by the following"

$$\varepsilon^{''} = \varepsilon^{''}_d + \varepsilon^{''}_{\gamma} \tag{6}$$

where dipole contribution  $\mathcal{E}_{d}^{''}$  is given with taking into consideration (4)

$$\varepsilon_{d}^{"} = \left(\varepsilon_{0} - \varepsilon_{\infty}\right) \left(\frac{f}{f_{M}}\right) \tag{7}$$

And ohmic deposition is given according [15]:

$$\varepsilon_{d}^{''} = \left(\varepsilon_{0} - \varepsilon_{\infty}\right) \begin{pmatrix} f \\ f_{M} \end{pmatrix} \tag{8}$$

[1] P. Debay. Polyarnie molekuli, M-L, 1931. (in Russian)

[2] Proc. 12<sup>th</sup> Int.Conference Amorphous and Liquid Semiconductors. J.Non-Cryst. Solids, 1987, 97-98, pp.39; 155; 203. where  $\gamma$  is conductivity on constant current in units Om<sup>-1</sup>cm<sup>-1</sup> at expression (7-8) in the following form:

$$\frac{\varepsilon_d^{''}}{f} = \frac{\varepsilon_0 - \varepsilon_\infty}{f_M} \tag{9}$$

$$\varepsilon_d^{''} f = 1.8 \cdot 10^{12} \,\gamma \tag{10}$$

The right parts contain the values don't depending on frequency (at fixed temperature). Thus peculiarity allows us to use the dielectric loss value measured on the frequencies satisfying to the condition (4) for division of relaxation and ohmic depositions on character of frequency dependencies of

composition of  $\varepsilon'' f$  and private  $\varepsilon''/_f$ .

#### Conclusion

In summary we can confirm that acetone is characterized by strong correlation of rotational dynamics and translation one that is revealed in the comparison of activation energy for electric charge transfer processes, mass and electric dipoles. The experimentally proved presence of such correlation gives the basis to suppose the existence of enough expressed and stable clustering of acetone molecules. Taking under consideration the acetone electric dipole moment it is necessary to consider the dipole-dipole interaction as the main factor managing by clustering. In this connection there are basis to suppose that the single region of dispersionabsorption observable in acetone dielectric spectrum, is created by superposition of individual cluster contributions. The additions of high-polar liquids are able to essentially extend the frequency extent of acetone dielectric spectrum and thus, to create more favorable conditions of its solution.

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## QEYRİASSOSİATİV MAYELƏRİN AŞAĞI TEZLİK DİAPAZONDAKI DİELEKTRİK XASSƏLƏRİ

Aseton molekullarının istilik hərəkətinin translyasiya və fırlanma təzahürlərinin bəzi dielektrik xüsusiyyətləri öyrənilmişdir.

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## ДИЭЛЕКТРИЧЕСКИЕ СВОЙСТВА НЕАССОЦИИРОВАННЫХ ЖИДКОСТЕЙ В ДИАПАЗОНЕ НИЗКИХ ЧАСТОТ

Изучены некоторые особенности диэлектрических проявлений взаимодействия трансляционной и вращательной составляющих теплового движения молекул ацетона.

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