

doi: 10.15407/ujpe60.09.0854

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KINEMATIC SHEAR VISCOSITY OF WATER, AQUEOUS SOLUTIONS OF ELECTROLYTES, AND ETHANOL

PACS 66.20.Cy, 83.60.Fg

The nature of the kinematic shear viscosity in associated (water and aqueous solutions of electrolytes) and strongly associated (alcohols) liquids has been studied. The behavior of the kinematic shear viscosity is shown to be governed by orientational correlations and the translational motion of molecules, which is characteristic of argon. The former mechanism dominates in the supercooled area and in normal states close to the triple point. The latter one is responsible for the viscosity at higher temperatures. The characteristic temperature t_H separating those areas is found to be close to the triple point in the case of water and electrolyte aqueous solutions, and to the critical point in the case of ethanol. The agreement with experimental data is quite satisfactory.

Keywords: viscosity, water, ethanol.

1. Introduction

The temperature dependence of the shear viscosity in liquids is usually described by an expression that contains a sum of exponential terms [1–3],

$$\eta = \eta_1 \exp(\varepsilon_1/t) + \eta_2 \exp(\varepsilon_2/t) + \dots, \quad (1)$$

where $t = T/T_c$ is the dimensionless temperature, T_c the critical temperature of the analyzed liquid, and the Boltzmann constant is considered to equal unity. Expression (1) is a result of the quasicrystal approach for the description of a molecular thermal motion in liquids [4]. In the framework of this model, liquid molecules vibrate around their temporal equilibrium positions during a certain characteristic time τ_0 before shifting into new temporal equilibrium positions. If the characteristic time of those transitions, τ_1 , is substantially shorter than τ_0 , the applied approach is eligible. Transport processes in such liquids are described in the framework of activation theory [4, 5]. The quantitative agreement with experimental data is reached by introducing the temperature dependence for the parameters η_1 and η_2 (as well as ε_1 and ε_2). As a result, the agreement with experimentally observed viscosity values [6–8] is achieved for ionic liquids and liquids capable of transforming into the glass state. However, the physical sense of introduced parameters remains obscured.

In works [9–11], the idea of the “quasicrystal” character of the molecular thermal motion was demonstrated to be inapplicable to argon, because the corresponding activation energy, ε_{Ar} , is much lower than the energy of thermal motion, i.e. $\varepsilon_{Ar} < t$.

For diluted aqueous solutions of electrolytes, the shear viscosity is higher in comparison with that for pure water. In works [12–14], a hydrodynamic theory was developed for the motion of ions surrounded by hydration shells. The principal idea consists in that the ion locally changes the density of water in its vicinity and, in such a manner, changes the specific volume per water molecule. While calculating the specific volume variation, both the direct interaction of an ion with water molecules and its indirect action through neighbor molecules was taken into account. As a result, an expression for the shear viscosity was obtained. The structure of this expression completely reproduces the Einstein formula, in which the specific volume is the sum of volumes of hydrated ions. The specific features in the motion of cations and anions in electrolyte solutions were also studied in work [15].

2. Kinematic Shear Viscosity of Pure Water

In works [16–18], it was shown that there exists such a temperature t_H that the character of the thermal motion of water molecules is crystal-like at $t < t_H$

and argon-like at $t > t_H$. Accordingly, the behavior of the kinematic shear viscosity of water at $t < t_H$ is described by the activation mechanism and, at $t > t_H$, by the argon-like one.

The same is evidenced by the behavior of the dipole relaxation time τ_d . In the interval $t < t_H$, this parameter demonstrates an exponential dependence [19], and the ratio $R = \eta/\tau_d$ remains almost invariant in a wide temperature interval, although both η and τ_d change considerably [20, 21]. At $t < t_H$, the temperature dependence of the shear viscosity in water can be approximated by an expression of the type (1).

For water, the temperature t_H is close by magnitude to the temperature t_D , which has a sense of the molecular dipole ordering temperature,

$$t_D = \frac{d^2}{3k_B T_c \langle r_{12} \rangle^3} \approx 0.45 \div 0.5,$$

where $t = T/T_c$, T_c is the critical temperature of water, $\langle r_{12} \rangle$ the average distance between water molecules, and d the dipole moment of a water molecule. At $t > t_H$, orientational correlations are insignificant. Therefore, rotations of water molecules result in that the effective interaction potential between the molecules becomes spherically symmetric, and the motion of water molecules is argon-like. At $t < t_H$, the behavior of the kinematic shear viscosity substantially differs from the argon-like one owing to the presence of strong orientational correlations.

The relationship between the reduced quantities – the kinematic shear viscosity $\tilde{\nu} = \nu/\nu_R$, where ν_R is the regularized value of kinematic shear viscosity in a vicinity of the critical point, the normalized temperature $t = T/T_c$, and the normalized specific volume $\tilde{v} = v/v_R$, where v_R is the regularized value of specific volume – will be discussed on the basis of the similarity principle applied to the corresponding states [22]. The regularized value of kinematic shear viscosity ν_R is determined with the help of a parabolic extrapolation of the viscosity values into the critical region [18]. The regularized value of specific volume v_R is proportional to the specific volume at the critical point: $v_R = \lambda v_c$, where the multiplier λ is introduced to exclude the influence of dimers, trimers, and other clusters of higher orders, which are formed near the critical point [23, 24].

Let us consider the behavior of the kinematic shear viscosity in the supercooled states of water. The

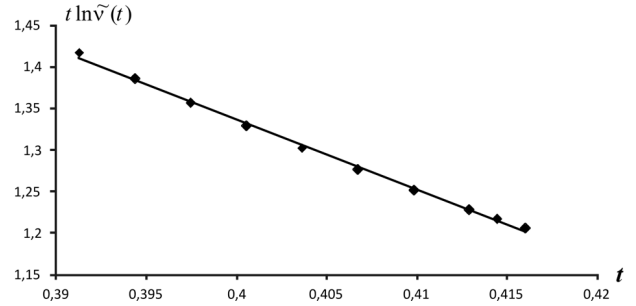


Fig. 1. Temperature dependence of the quantity $t \ln \tilde{\nu}(t)$ along the water–vapor coexistence curve. Points correspond to experimental data [25]; the straight line is their approximation

temperature dependence of the quantity $t \ln \tilde{\nu}(t)$ at the water–vapor coexistence curve is depicted in Fig. 1. One can see that the temperature dependence of the normalized kinematic shear viscosity of water is described by the formula

$$\tilde{\nu}^{(w)}(\tilde{v}, t) = \tilde{\nu}_0 \exp(\varepsilon_H/t), \quad (2)$$

where $\tilde{\nu}_0 = 2.079 \times 10^{-4}$ and $\varepsilon_H = 4.73$. The dimensionless activation energy $\varepsilon_H = E_H/k_B T_c$ practically coincides with the ground-state energy of a water dimer [26, 27]. We would like to emphasize that the ground state of a water dimer is mainly formed by electrostatic forces [28]. The contribution of electrostatic forces exceeds the contribution from the direct electron shell overlapping between the oxygen and hydrogen atoms (in effect, this is an H-bond between two water molecules in the dimer) in a ratio of about 10:2.5. However, it is the total energy (the electrostatic interaction and the hydrogen bond) that is often identified as the H-bond energy and the energy of dimer vacancy capture (see work [29] and references therein).

At $t_H < t < 0.95$, due to the rotation of water molecules, the main water properties are determined by the averaged potential, which looks like the Lennard-Jones one [29, 30]. For this reason, we may assert that the kinematic shear viscosity of water can be described using the argon-like expression

$$\tilde{\nu}^{(w)}(\tilde{v}, t) \approx \frac{\zeta_0(t)}{(\tilde{v} - \tilde{v}_0^{(w)})^{1/3}}. \quad (3)$$

In order to finding the parameters $\zeta_0(t)$ and $\tilde{v}_0^{(w)}(t)$, the dependence of the quantity $\tilde{\nu}^3(t)\tilde{v} = \tilde{\nu}^3\tilde{v}_0 + \zeta$ on $\tilde{\nu}^3$ is used (Fig. 2).

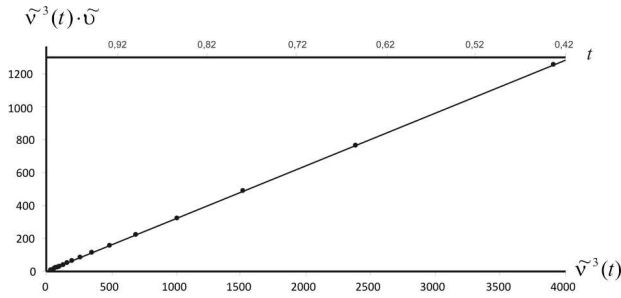


Fig. 2. Dependence $\tilde{\nu}^3(t)\tilde{\nu}$ versus $\tilde{\nu}^{-3}$ and its straight-line approximation

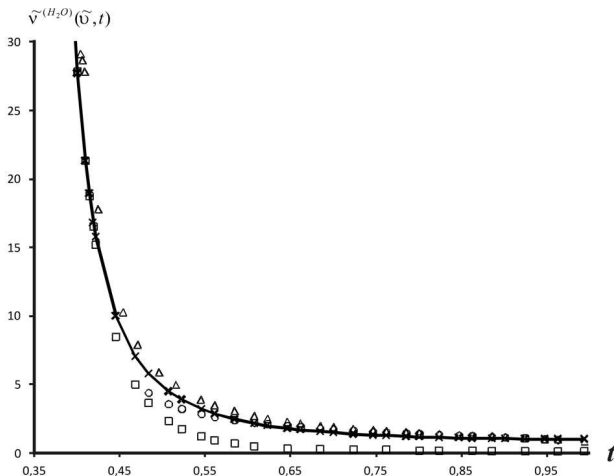


Fig. 3. Comparison of various contributions to $\tilde{\nu}^{(H_2O)}(\tilde{\nu}, t)$: (□) the exponential contribution in Eq. (1), (○) the argon-like contribution, (△) the sum of both contributions in Eq. (1), and (×) experiment [31]

In such a way, we found that $\zeta_0 = 0.643$ and $\tilde{\nu}_0 = 0.358$. These values of the parameters ζ_0 and $\tilde{\nu}_0$ coincide with their counterparts for liquid argon [16]. We would like to emphasize that the argon-like behavior of the kinematic shear viscosity manifests itself at $t > 0.5$. At the same time, in the temperature interval $t < 0.5$ with strong orientational correlations, just the latter govern the behavior of the shear viscosity, so that this parameter is described by formula (2). We may assume that the viscosity of water can be approximated by a superposition of the exponential contribution (2) and the argon-like one (3) in the whole region of liquid state existence:

$$\tilde{\nu}_w(\tilde{\nu}, t) \approx \tilde{\nu}_0 \exp(\varepsilon_H/t) + \frac{\zeta_0}{(\tilde{\nu} - \tilde{\nu}_0^{(w)})^{1/3}}. \quad (4)$$

Note that the excluded volume per molecule, $\tilde{\nu}_0^{(w)}(t)$, depends on the temperature insignificantly.

In Fig. 3, the experimental values of viscosity and various terms in Eq. (4) are compared. One can see that, in the temperature interval $t > 0.55$, the exponential contribution to the shear viscosity is so small in comparison with the argon-like contribution that it can be neglected here.

3. Kinematic Shear Viscosity of NaCl Aqueous Solution

First, let us consider the kinematic shear viscosity of a NaCl aqueous solution with the molar fraction of salt $x = 0.07$. At this concentration, there are approximately 13 water molecules per anion-cation pair, which coincides with the number of water molecules in the hydration spheres of ions [12]. Since ions are spherically symmetric, they practically do not affect the character of orientational correlations. In this connection, a similarity between the behavior of the kinematic shear viscosity for the NaCl aqueous solution and for water should be observed. The analysis of experimental values on shear viscosities [25, 32] completely confirms this supposition. For instance, in the temperature interval $252 \text{ K} < T < 315 \text{ K}$, the experimental data are described by the exponential dependence

$$\tilde{\nu}(\tilde{\nu}, t) = \tilde{\nu}_0 \exp(\varepsilon_0/t),$$

where $\tilde{\nu}_0 = 18.75 \times 10^{-4}$ and $\varepsilon_0 = 4.28$. Here, the normalization is carried out with respect to the regularized shear viscosity value, which was obtained by extrapolating the experimental values for the kinematic shear viscosity of the solution to the critical temperature of water, T_c . The dimensionless activation energy is determined, as was done above, by the formula $\varepsilon_0 = E_0/k_B T_c^{(w)}$. The ε_0 -value for the NaCl solution is a little smaller than for water, which is associated with a partial destruction of tetragonal configurations in the diluted electrolyte solution. The amplitude growth of the shear viscosity in the electrolyte solution by approximately a factor of nine in comparison with water may probably testify that water molecules partially move together with the hydration complex [28].

In the temperature interval $333 \text{ K} \lesssim T \lesssim 423 \text{ K}$, the behavior of the kinematic shear viscosity has the

argon-like character (Eq. (3)) with the parameter values $\zeta_0 = 0.922$ and $\tilde{v}_0 = 0.260$. Note that the value $\tilde{v}_0 = 0.260$ corresponds to the specific volume per molecule at the temperature $T \approx 315$ K. This value is lower than the corresponding value for water by 21%, which is in quite good agreement with the density growth for the electrolyte of a given concentration with respect to water: $\left(\frac{n_{\text{el}}}{n_w} - 1\right) \times 100\% = 20\%$. The substitution of the activation mechanism governing the formation of the kinematic shear viscosity for the argon-like one occurs at $T \approx 315$ K. The difference between the experimental data and the values calculated by formulas (2) and (3) do not exceed 1–5%. A comparison with experimental data is shown in Fig. 4.

In the case of electrolyte solutions, the exponential and argon-like dependences of the shear viscosity match near the crossover temperature ($T = 315$ K), but a little worse than in the case of water. Therefore, it is difficult to single out an interpolation formula which would successfully describe experimental data, as formula (4) does.

4. Kinematic Shear Viscosity of Alcohols

In this section, the kinematic shear viscosity of monoatomic alcohols like methanol and ethanol will be considered. These alcohols are classed to strongly associated liquids. Numerous spectroscopic experiments [33, 34] dealing with infra-red absorption spectra reveal the presence of dimers, trimers, and tetramers practically in the whole region of their liquid state.

A direct evidence for the clustering in liquids is the magnitude and the temperature dependence of the dipole relaxation time τ_D [19, 35]. Experimental data for ethanol in comparison with those for water are tabulated in Table. One can see that, at temperatures $T/T_c < 0.42$, which correspond to supercooled states of water, the dipole relaxation times for water and ethanol considerably exceed the corresponding times of the free rotation for water molecules (0.7 ps) and ethanol (2 ps). As the temperature grows, the dipole relaxation time of water exponentially decreases and, at $T/T_c \approx 0.7$, turns out close to the period of free rotation. In the case of ethanol, the dipole relaxation time also falls down exponentially, but remains much larger than the free rotation period within the whole interval of liquid ethanol existence. This means that

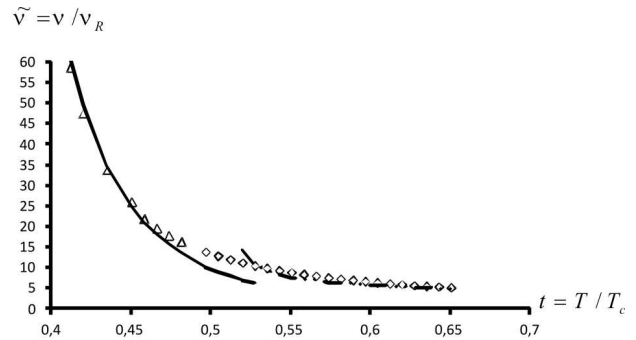


Fig. 4. Dependences of the normalized kinematic shear viscosity in a NaCl aqueous solution on the normalized temperature. Triangles correspond to experimental values in the temperature interval 252–308 K, and diamonds in the temperature interval 333–423 K [25, 32]. The solid curve corresponds to formula (2), and the dashed one to formula (3)

the character of thermal motion in ethanol substantially differs from the argon-like one. In other words, molecular orientations in ethanol change owing to the activation mechanism. This fact is also confirmed by the estimation of the dipole ordering temperature for ethanol molecules,

$$t_D = \frac{d^2}{3k_B T_c \langle r_{12} \rangle^3} \approx 0.85 \div 0.87,$$

which corresponds to $\langle r_{12} \rangle = 2.15$ Å (this is a distance between ethanol molecules in the equilibrium dimer configuration [35]) and to the dipole moment of an ethanol molecule $d = 1.69$ D. Hence, orientational correlations dominate in the temperature interval of existence for liquid ethanol; i.e. at $t < t_D$, ethanol molecules aggregate into clusters of various orders: dimers, trimers, tetramers, and so on. Those facts allow us to draw a conclusion that, within the whole

Experimental values of dipole relaxation time τ_D for pure water and ethanol at various temperatures

$t = T/T_c$	τ_D , ps	
	H ₂ O	C ₂ H ₅ OH
0.4	100	≈500
0.5	14	350
0.58	6	140
0.6	5	130
0.7	2	45

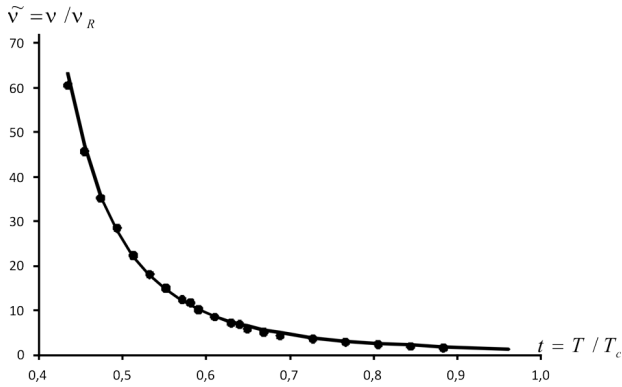


Fig. 5. Kinematic shear viscosity of ethanol as a function of the temperature. Circles correspond to experimental data [37–39], and the solid curve exhibits the values calculated by formula (2)

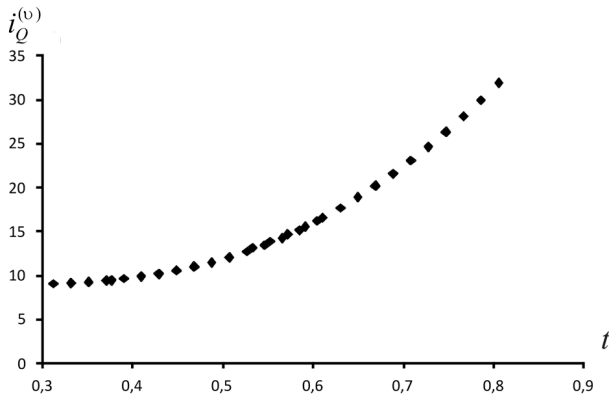


Fig. 6. Temperature dependence of the number of thermal degrees of freedom per molecule, $i_Q^{(v)}$, for liquid ethanol according to experimental data [37]

temperature interval of existence for liquid ethanol, the behavior of its kinematic shear viscosity has the activation character (Fig. 5).

Hence, the experimental data completely confirm our conclusion about the purely activation character of the shear viscosity in liquid ethanol within the whole temperature interval of its existence. In the case of ethanol, the values of the parameters entering formula (2) are $\tilde{\nu}_0 = 423.8 \times 10^{-4}$ and $\varepsilon_0 = 2.98$, i.e. the dimensionless activation energy turns out a little lower and the pre-exponential multiplier much larger than in the case of water. A similar situation took place, when the shear viscosity in electrolyte aqueous solutions was analyzed. Here, again, the considerable growth of the pre-exponential factor is as-

sociated with the relative motion of clusters (dimers, trimers, tetramers, and so forth).

In view of the crucial role played by orientational correlations, let us dwell on another fact that confirms this conclusion. Let us consider the behavior of the heat capacity of liquid ethanol. In order to describe the relative magnitudes of various contributions to the heat capacity, the dimensionless quantity $i_Q = 2C_V/k_B N_A$, where C_V is the heat capacity of the system at a constant volume, and N_A the Avogadro constant, will be used. In works [19, 40], this parameter was called the number of thermal degrees of freedom per molecule. The i_Q -value differs from the standard number of molecular degrees of freedom in that the number of vibrational degrees of freedom in it is twice as large. The maximum value for the number of thermal degrees of freedom for the translational and rotational motions of a molecule equals 12: $i_Q^{(tr)} + i_Q^{(rot)} = 12$. This value corresponds to translational and orientational vibrations in solid ethanol. In liquid ethanol, the sum $i_Q^{(tr)} + i_Q^{(rot)}$ is by 1 or 2 smaller, which is confirmed by estimations made for water [19, 40]. Accordingly, we will assume that the number of vibrational thermal degrees of freedom equals $i_Q^{(v)} \approx i_Q - 12$. The corresponding temperature dependence of $i_Q^{(v)}$ is depicted in Fig. 6.

The vibrational contributions to the heat capacity of liquid ethanol are formed by low-frequency intramolecular vibrations and vibrations of hydrogen bonds between the molecules (see above). In the framework of this model, we can explain the temperature dependence of the number of vibrational thermal degrees of freedom (this will be done in a separate work.). However, even without a detailed analysis, Fig. 6 evidently testifies to a considerable role of clustering effects.

5. Discussion of Obtained Results

In this work, the physical nature of the kinematic shear viscosity in associated liquids – water, aqueous solutions of electrolytes, and ethanol – is discussed. Water and ethanol molecules have dipole moments comparable by magnitude. It is shown that the characteristic temperatures of the dipole ordering in water and aqueous solutions of electrolytes are close to the temperatures of their ternary points. From whence, it follows that the character of thermal mo-

tion in those liquids is argon-like almost in the whole temperature interval of their existence in the liquid state. As a consequence, the behavior of their kinematic shear viscosities is also similar. At the same time, in the interval of supercooled states, as well as in normal states located in a vicinity of the ternary point, the character of molecular thermal motion is mainly driven by strong dipole correlations, which gives rise to the activation mechanism of thermal motion. Therefore, in this interval, the temperature dependence of the kinematic shear viscosity has the exponential character.

In contrast, the characteristic temperature of the dipole ordering in liquid ethanol turns out close to the critical point temperature. In other words, practically in the whole temperature interval, where ethanol is liquid, the thermal motion of its molecules is governed by strong dipole correlations. As a result, dimers, trimers, tetramers, and other higher-order clusters are formed. The domain of existence of argon-like molecular motion is absent. The kinematic shear viscosity is described by the exponential expression typical of the activation mechanism within the whole temperature interval.

The result of our analysis brings us to a conclusion that the conventional ideas used for the description of the kinematic shear viscosity in electrolyte solutions on the basis of hydration effects, as well as the models of molecular motion between the voids in the hydrogen bond network, are essentially simplified.

To summarize, I would like to sincerely thank Academician L.A. Bulavin, the person of jubilee, for stimulating this research. I am also very grateful to Prof. N.P. Malomuzh for his permanent attention to the work.

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Received 12.05.15.

Translated from Ukrainian by O.I. Voitenko

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КІНЕМАТИЧНА ЗСУВНА В'ЯЗКІСТЬ ВОДИ,
ВОДНИХ РОЗЧИНІВ ЕЛЕКТРОЛІТІВ ТА ЕТАНОЛУ

Резюме

У роботі досліджується природа кінематичної зсувної в'язкості асоційованих (вода та водневі розчини електролітів) і сильно асоційованих (спирти) рідин. Показано, що поведінка зсувної в'язкості визначається: 1) орієнтаційними кореляціями і 2) поступальним рухом молекул, характерним для аргону. Перший механізм є визначальним у переохолодженій області та нормальних станах, прилеглих до потрійної точки системи. Другий механізм визначає в'язкість системи при більш високих температурах. Встановлено, що значення характерної температури t_H , що розділяє ці області, є близькою до потрійної точки у воді та водних розчинах електролітів і до критичної точки в етанолі. Узгодження з експериментальними даними у всіх окреслених випадках є цілком задовільним.