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COMPARISON BETWEEN VISCOELASTIC PROPERTIES OF ALIPHATIC ALCOHOLS AND THEIR FLUORO-SUBSTITUTED ANALOGS

The viscoelastic properties of liquid aliphatic alcohols (propanol-1, pentanol-1, and heptanol-1) have been compared with the properties of their fluoro-substituted analogs. Experimental data on the density, kinematic viscosity, and sound velocity of the liquids, as well as their critical and van der Waals parameters simulated by artificial neural networks, are used. The comparison of the properties of indicated alcohols with those of water, where there is a continuous network of hydrogen bonds, and hydrogen peroxide, where there are hydrogen bonds, but not their network, shows that the change in the physical properties after the fluoro-substitution is associated with the density of hydrogen bonds in the liquid.

Keywords: fluoro-substitutional alcohols, water, hydrogen peroxide, shear viscosity, sound speed, artificial neural networks.

1. Introduction

The study of the structure and physical properties of monoatomic aliphatic alcohols and their fluoro-substituted analogs is one of the challenging tasks

of modern physics of fluids. Of considerable interest is a comparison of the physical properties of liquids from the same homologous series and the substitution one obtained by replacing hydrogen atoms with fluorine ones in the molecules of fluoro-substituted liquids. However, information about the supramolecular structure and mechanisms of molecular processes occurring in fluoro-substituted liquids is scarce and sometimes contradictory.

Fluoro-substituted alcohols have found their application in pharmacology and organic synthesis as specific solvents, the components of high-temperature

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coolants, lubricants, and adhesives, as well as in the manufacture of pesticides and polymeric materials [1–3]. They continue to master new areas of application; in particular, they are used in biotechnology for stabilizing peptides [4], biomodifying enzymes [5], changing the structure of protein macromolecules [6], modifying the properties of lipid membranes [7], and so forth. Despite the widespread application of fluoro-substituted alcohols in various domains, their physical properties still remain poorly studied.

The study of viscoelastic properties of the indicated alcohols in the liquid phase makes it possible to trace the features of relevant molecular processes and their dependence on the molecular structure. The obtained results can be used while studying individual molecular liquids [8–10] and solutions of high-molecular compounds and biopolymers [11–13].

At the current stage of forecasting the physical properties of halogen-substituted organic compounds taking their structure into account, the application of artificial neural networks deserves special attention. As is known, neural networks require reliable experimental data, and the quality of their prediction is higher, if more relevant data for the corresponding homologous series are available.

Since the transition from saturated aliphatic alcohols to their fluoro-substituted analogs should affect the character of hydrogen bonds between the molecules, we will compare, in this work, the viscoelastic properties of those alcohols with the corresponding properties of water, in which there exists a continuous network of hydrogen bonds, and the properties of hydrogen peroxide, where the hydrogen bonds exist, but do not form a network. The presence of a hydrogen network in water leads to a number of unique features of the behavior of water as a solvent [14–16].

The aim of the work was to compare the viscoelastic properties of liquid aliphatic alcohols from the same homologous series with the properties of their fluoro-substituted analogs using available experimental data, model equations, and data from artificial neural networks.

2. Experimental Part and Calculation Methods

As the objects to study, we took normal aliphatic alcohols of the analytical grade: propanol-1 (C_3H_7OH ; hereafter, $PrOH$), pentanol-1 ($C_5H_{11}OH$, $PenOH$),

heptanol-1 ($C_7H_{15}OH$, $HepOH$), and their fluoro-substituted analogs 1H,1H,3H-tetrafluoropropanol-1 ($HCF_2CF_2CH_2OH$, $4F-PrOH$), 1H,1H,5H-octafluoropentanol-1 ($H(CF_2)_4CH_2OH$, $8F-PenOH$), 1H,1H,7H-dodecafluoroheptanol-1 ($H(CF_2)_6CH_2OH$, $12F-HepOH$), as well as water, H_2O , and hydrogen peroxide, H_2O_2 . All fluoro-substituted alcohols had a purity of 99.0% and were synthesized at the Department of Chemistry of Fluoroorganic Compounds of the Institute of Organic Chemistry of the National Academy of Sciences of Ukraine.

Experimental measurements of the rheological parameters of the indicated alcohols were carried out in the temperature interval of 293–363 K using standard methods. The density ρ was determined by means of the pycnometric method with an error of 0.05%. The kinematic viscosity ν was studied using the capillary viscometry method with a measurement error not exceeding 1.0%.

The speed of sound propagation C was measured making use of the pulse method with the distance change at a frequency of 27.5 MHz. The total error of measurements did not exceed 0.1%. The technique of measuring the sound speed was described in detail in work [17].

In modern thermodynamic databases Refprop [18], ThermodataEngine [19], WTT [20], and others (in open access, there are Minirefprop [21], SRD69 [22], and Coolprop [23]), the temperature dependences of the main thermodynamic quantities for liquids along their liquid-vapor coexistence curve – in particular, the density ρ and the shear viscosity $\eta = \rho\nu$ – are presented in the form of certain analytic functions that reliably describe physical quantities.

The calculations and the data presentation for the density of liquids ρ can be performed using the Tate [24], Huang–O’Connell [25], and Kesselman [26] equations, as well as the Benedict–Webb–Rubin (BWR) equation [27] and its modifications by Starling (BWRS), Jacobson and Steward (mBWR), and others [28]. For the shear viscosity η , the corresponding equation looks like

$$\ln \frac{\eta}{\eta_0} = \sum_i a_i f(T)^{n_i}, \quad (1)$$

where η_0 , a_i , and n_i are definite constants (the PPDS9, ViscosityL, Yaws.Viscosity, and DIPPR101 equations) [19, 20].

In the absence of raw experimental data, the temperature dependences of the density ρ along the coexistence curve can be predicted using the Rackett, Gunn–Yamada, and Hankinson–Brobst–Thomson methods [29–31]. For the temperature dependences of the shear viscosity η along the coexistence curve, this can be done using the Letsou–Stiel, Joback, and Orrick–Erbar methods [32–34].

In addition, such data can be obtained via simulation using artificial neural networks based on the most reliable experimental data and quantum mechanical calculations of molecular parameters, namely, Mol-instants [35] or ChemRTP [36] simulations. Mol-instants simulations make it possible to obtain the temperature dependences of thermodynamic quantities. At the same time, ChemRTP simulations give reference values of physical quantities, which should be used together with modern thermodynamic similarity methods [37, 38]. It should be noted that, in recent years, simulation methods based on artificial neural networks have achieved substantial progress, which allows the calculations of physical quantities to be carried out with an error close to the determination errors of these quantities in modern experimental studies [39, 40].

According to the principle of corresponding states [37, 38], the dimensionless transformation of the temperature T , the density ρ , and the adiabatic compressibility coefficient $-(\partial V/\partial P)_S$ was performed using the values of their critical parameters. For the speed of sound

$$C = \left(\frac{\partial P}{\partial \rho} \right)_S = V \left(- \frac{\partial P}{\partial V} \right)_S^{0.5}, \quad (2)$$

the dimensionless form of the corresponding expression looks like

$$\begin{aligned} C^* &= \frac{V}{V_C} \left(- \left(\frac{\partial P}{\partial V} \right)_S \frac{V_C}{P_C} \right)^{0.5} = \\ &= \frac{\rho_C}{\rho} \left(- \left(\frac{\partial P}{\partial V} \right)_S \frac{1}{P_C \rho_C} \right)^{0.5} = C \left(\frac{\rho_C}{P_C} \right)^{0.5}. \end{aligned} \quad (3)$$

Here, the specific volume V is measured in m^3/kg units, and the density $\rho = 1/V$ in kg/m^3 units. The values of the critical parameters T_C , P_C , and ρ_C for aliphatic alcohols, water, and hydrogen peroxide were taken from literature sources [22, 39, 40]. For the fluoro-substituted analogs of aliphatic alcohols, due

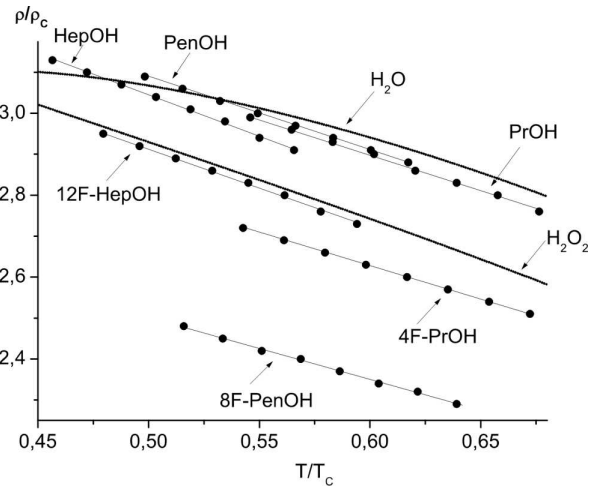


Fig. 1. Temperature dependences of the densities of aliphatic alcohols, their fluoro-substituted analogs, water, and hydrogen peroxide

to the lack of experimental data, the relevant values were obtained by simulation using artificial neural networks [35, 36]. The applied critical parameter values are quoted in Table 1.

3. Comparison of the Temperature Dependences of the Density for Alcohols, Water, and Hydrogen Peroxide

Since the fluoro-substitution in alcohols leads to changes in hydrogen bonds, it is desirable to trace the influence of hydrogen bonds on the properties of alcohols. For this purpose, experimental data on the density of alcohols were obtained using the methods described in Section 2, and data on the density of water and hydrogen peroxide were also calculated.

Table 1. Critical temperatures (T_C), critical pressures (P_C), and critical densities (ρ_C) for aliphatic alcohols, their fluoro-substituted analogs, water, and hydrogen peroxide

Liquid	CAS	T_C , K	P_C , kPa	ρ_C , kg/m^3	Source
PrOH	71-23-8	535.9	5197.3	269	[35, 40]
PenOH	71-41-0	588.0	3839.6	264	[35, 40]
HepOH	111-70-6	641.8	3096.0	262	[35, 40]
4F-PrOH	76-37-9	540.0	4124.6	546	[35]
8F-PenOH	355-89-6	568.0	2773.7	673	[35]
12F-HepOH	335-99-9	611.0	2100.3	597	[35]
H ₂ O	7732-18-5	647.1	22064	32.2	[35, 40]
H ₂ O ₂	7722-84-1	727.98	16202	111	[35, 40]

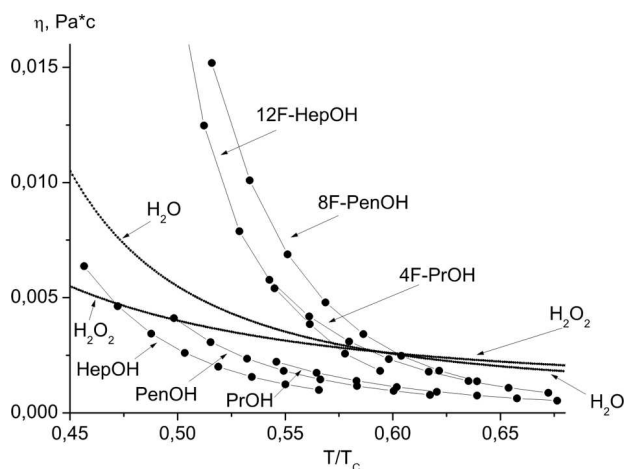


Fig. 2. Temperature dependences of the shear viscosities of aliphatic alcohols, their fluoro-substituted analogs, water, and hydrogen peroxide

In Fig. 1, experimental data [8] are presented for the temperature dependence of the densities of aliphatic alcohols and their fluoro-substituted analogs, as well as the corresponding data for water and hydrogen peroxide obtained from modern physical databases [21–23, 39, 40]. The analysis of this figure shows that, for saturated aliphatic alcohols, provided that the temperature is constant, the reduced density firstly increases with the growth of the number of carbon atoms, and then decreases. At the same time, as one can see, for fluoro-substituted analogs of aliphatic alcohols, on the contrary, the density firstly decreases at a constant temperature as the number of carbon atoms increases, and then increases.

At a certain reduced temperature lower than 0.68, the reduced density of fluoro-substituted aliphatic alcohols, in contrast to the saturated aliphatic alcohols, firstly decreases with the growth in the number of carbon atoms, and then increases. At the same time, for unsubstituted aliphatic alcohols, the reduced density firstly increases with the increase in the number of carbon atoms at the same reduced temperature, and then decreases.

It should be noted that the reduced density of water, where there exists a network of hydrogen bonds, in the range of reduced temperatures above 0.53 is larger than that for saturated aliphatic alcohols. In hydrogen peroxide, where hydrogen bonds exist, but there is no hydrogen bond network, the reduced density at all reduced temperatures is less than that for

the saturated aliphatic alcohols, but larger than for their fluoro-substituted analogs.

4. Comparison of the Temperature Dependences of Viscosity for Alcohols, Water, and Hydrogen Peroxide

We experimentally obtained the kinematic viscosity ν of aliphatic alcohols and their fluoro-substituted analogs. In Fig. 2, the temperature dependences of the shear viscosity η calculated from our experimental data for the kinematic viscosity ν and the density ρ ($\eta = \rho\nu$) are exhibited for aliphatic alcohols and their fluoro-substituted analogs [8–10]. We also plotted the corresponding dependences for water and hydrogen peroxide calculated on the basis of the most reliable literature data for their ν and ρ [21–23, 39, 40]. Figure 2 demonstrates a qualitative difference in the temperature dependences of the shear viscosity for aliphatic alcohols and their fluoro-substituted analogs: they form two separate groups. At the same time, the temperature dependences of shear viscosity for water and hydrogen peroxide intersect each other, being not similar to the corresponding dependences for aliphatic alcohols and their fluoro-substituted analogs.

For a more detailed comparison of the viscosity properties of aliphatic alcohols, their fluoro-substituted analogs, water, and hydrogen peroxide, let us analyze the dependences of the logarithm of the kinematic viscosity of the studied substances on the inverse temperature. According to work [10], the indicated dependences for the examine fluoro-substituted alcohols can be represented by straight lines in the researched temperature interval. Furthermore, according to literature data [22, 23, 39], such dependences for saturated aliphatic alcohols – for example, ethanol – remain also linear in the interval of reduced inverse temperatures $T_C/T = 1.4 \div 2.4$. The results obtained are presented on the same scale in Fig. 3 for saturated alcohols, in Fig. 4 for their fluoro-substituted analogs, and in Fig. 5 for water and hydrogen peroxide.

The analysis of Figs 3 and 4 testifies to a qualitative difference between the two homologous series of alcohols. For aliphatic alcohols from propanol to heptanol at a certain identical temperature, the kinematic viscosity decreases with the growth in the number of carbon atoms within the whole interval of reduced temperatures (see Fig. 3). On the contrary, for fluoro-

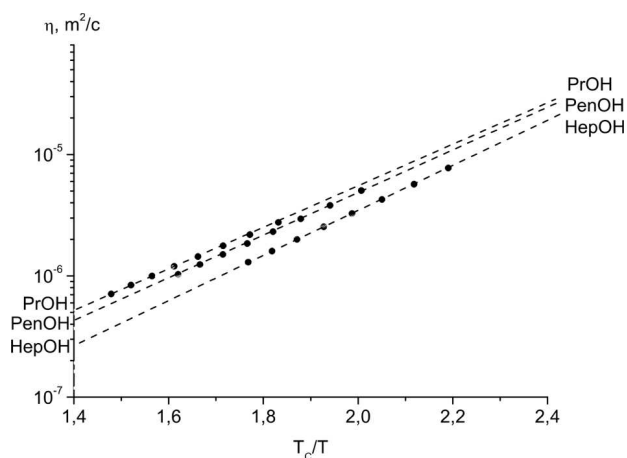


Fig. 3. Temperature dependences of the logarithm of kinematic viscosity for aliphatic alcohols

substituted analogs (see Fig. 4) at a certain identical reduced temperature in the vicinity of the critical point, the kinematic viscosity decreases with the growth in the number of carbon atoms. As the temperature decreases, the character of the corresponding dependences changes to opposite: the kinematic viscosity increases at a certain identical temperature in a vicinity of the freezing temperature with an increase in the number of carbon atoms. For water and hydrogen peroxide, the dependences of the logarithm of the kinematic viscosity on the inverse temperature are not straight lines that intersect each other (see Fig. 5).

5. Temperature Dependences of the Speed of Sound and the Coefficient of Adiabatic Compressibility

We experimentally measured the sound propagation speeds C in aliphatic alcohols and their fluoro-substituted analogs [9] at various temperatures; they are presented in Fig. 6. In addition, Fig. 6 also exhibits data on the sound propagation speed in water and hydrogen peroxide calculated using the methods described in Section 2. The analysis of this figure testifies that, in water, where there is a network of hydrogen bonds, the speed of sound propagation C^* is lower than that in saturated aliphatic alcohols. In hydrogen peroxide, where there are hydrogen bonds but not their network, it is higher than that in saturated aliphatic alcohols, but lower than in their fluoro-substituted analogs. Those facts testify to the influence of hydrogen bonds and, especially, the

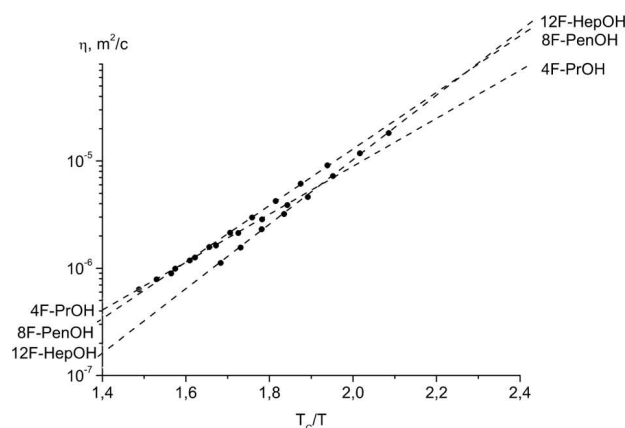


Fig. 4. Temperature dependences of the logarithm of shear viscosity for fluoro-substituted analogs of aliphatic alcohols

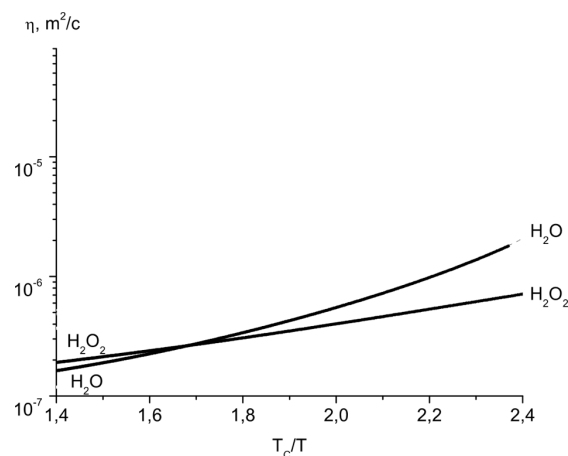


Fig. 5. Temperature dependences of the logarithm of shear viscosity for water and hydrogen peroxide

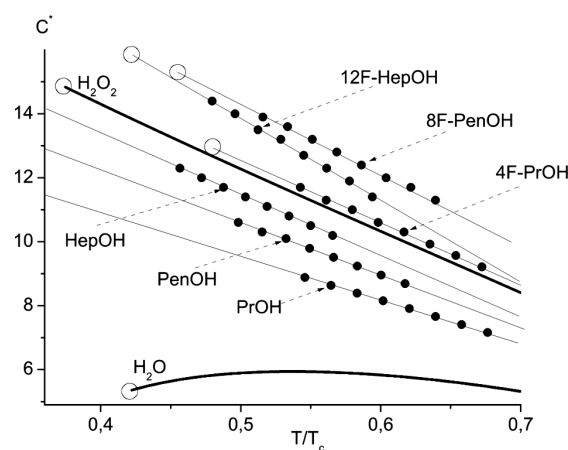


Fig. 6. Temperature dependences of the reduced sound propagation speeds C^* in aliphatic alcohols, their fluoro-substituted analogs, water, and hydrogen peroxide

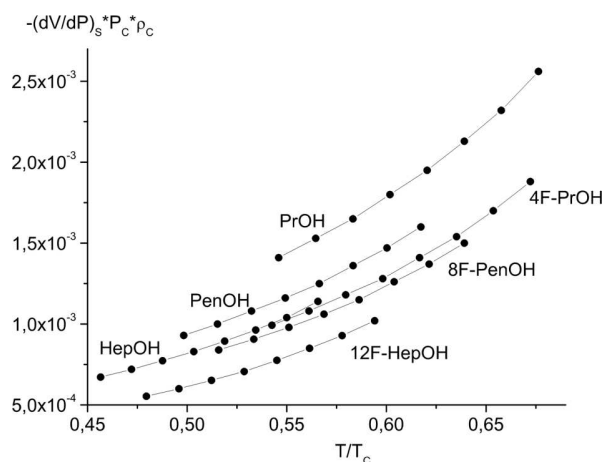


Fig. 7. Temperature dependences of the reduced adiabatic compressibility coefficient for aliphatic alcohols and their fluoro-substituted analogs

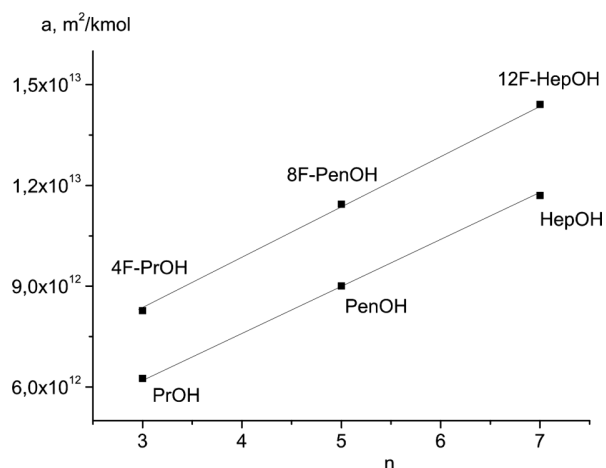


Fig. 8. Dependences of the intermolecular Van der Waals interaction parameter a on the number of carbon atoms n in the molecule for aliphatic alcohols and their fluoro-substituted analogs

availability of their network on the sound propagation speed.

Figure 7 demonstrates the calculated temperature dependences of the reduced adiabatic compressibility coefficient $-(\partial V/\partial P)_s P_c \rho_c$ for aliphatic alcohols and their fluoro-substituted analogs. The analysis of this figure shows that the indicated parameter decreases when transferring from saturated aliphatic alcohols to their fluoro-substituted analogs. As one can see, within the homologous series, for both fluoro-substituted and unsubstituted alcohols, the reduced adiabatic compressibility coefficient decreases with

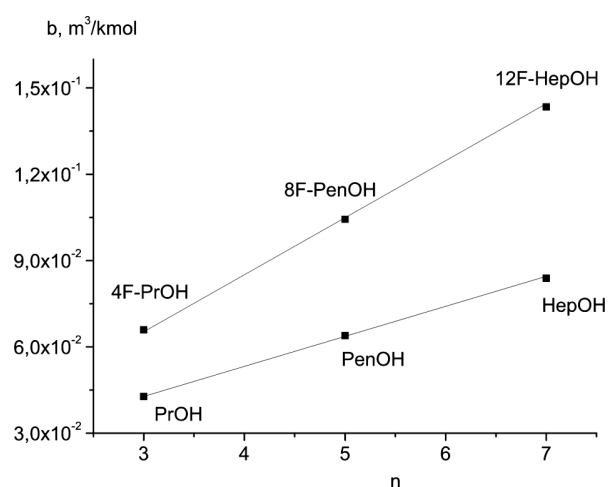


Fig. 9. Dependences of the intermolecular Van der Waals interaction parameter b on the number of carbon atoms n in the molecule for aliphatic alcohols and their fluoro-substituted analogs

Table 2. Molar masses (M_r), Van der Waals parameters (a , b), and molecular non-centricity parameter (w) for aliphatic alcohols, their fluoro-substituted analogs, water, and hydrogen peroxide

Liquid	$M_r \times 10^3$, $\frac{\text{kg}}{\text{mol}}$	$a \times 10^{-12}$, $\frac{\text{m}^2}{\text{kmol}}$	b , $\frac{\text{m}^3}{\text{kmol}}$	w	Source
PrOH	60.095	6.2585	0.042759	0.567822	[35, 40]
PenOH	88.1482	9.0067	0.063943	0.620963	[35, 40]
HepOH	116.2013	11.701	0.083888	0.687522	[35, 40]
4F-PrOH	132.0569	8.274	0.065908	0.662169	[35]
8F-PenOH	232.0719	11.444	0.104402	0.743963	[35]
12F-HepOH	332.0869	14.413	0.143407	0.808113	[35]
H ₂ O	18.0153	1.7296	0.011522	0.345624	[35, 40]
H ₂ O ₂	34.0147	3.0703	0.017895	0.452743	[35, 40]

the growth in the number of carbon atoms. This may be associated with the reduction of the role of hydrogen bonds at fluoro-substituting aliphatic alcohols, as well as the growth of the non-centricity parameter for longer alcohol molecules.

6. Discussion

To find the physical origins of the difference between aliphatic alcohols and their fluoro-substituted analogs, the dependences of the van der Waals parameters a and b , which enter the cubic equations of state (van der Waals, Redlich–Kwong, Soave–

Redlich–Kwong, Peng–Robinson, and their modifications [39]), on the number of carbon atoms are analyzed. The indicated parameters characterize the intensity of intermolecular interaction and the forbidden volume, and, accordingly, they are related to the energy and entropy contributions to the pressure in the system [41].

The values of the van der Waals parameters a and b , as well as the non-centricity parameter of the molecule w , are given in Table 2. The values of these parameters for aliphatic alcohols, water, and hydrogen peroxide were taken from literature sources [21–23, 39, 40]. For fluoro-substituted analogs of aliphatic alcohols, they were obtained by simulation using artificial neural networks [35, 36]. The resulting data are presented in Table 2.

The dependences of the van der Waals parameters a and b on the number of carbon atoms for aliphatic alcohols and their fluoro-substituted analogs are plotted in Figs. 8 and 9. The presented dependences demonstrate the growth of the van der Waals parameters a and b when going from aliphatic alcohols to their fluoro-substituted analogs. Furthermore, although the growth behavior of the parameter a is identical for both homologous series, it is qualitatively different for the parameter b . As the number of carbon atoms increases, the parameter characterizing the forbidden volume in the system grows faster for fluoro-substituted alcohols than for unsubstituted ones. It can be assumed that this occurs due to a denser mutual arrangement of long alcohol molecules because of their twisting, which leads to the corresponding changes in the entropic contribution to the pressure [41]. In our opinion, it is the parameter b that can be considered as a characteristic of the qualitative difference between different homologous series.

7. Conclusions

On the basis of obtained experimental data for the viscoelastic properties for aliphatic alcohols and their fluoro-substituted analogs and using the simulation of the values of critical and van der Waals parameters for fluoro-substituted alcohols with the help of artificial neural networks, we have shown that the homologous series of aliphatic alcohols and their fluoro-substituted analogs have a number of physical differences.

1. At a certain reduced temperature less than 0.68, the reduced density of fluoro-substituted aliphatic al-

cohols, unlike that of saturated aliphatic alcohols, firstly decreases with the increase in the number of carbon atoms and then increases. At the same time, for unsubstituted aliphatic alcohols, the reduced density at the same reduced temperature firstly increases with the growth in the number of carbon atoms and then decreases.

2. For aliphatic alcohols and their fluoro-substituted analogs at a certain identical reduced temperature in a vicinity of the critical point, the kinematic viscosity decreases with the increase in the number of carbon atoms. For aliphatic alcohols at a certain identical reduced temperature in a vicinity of the freezing point, in a vicinity of the freezing point, kinematic viscosity decreases with the increase in the number of carbon atoms, whereas, for their fluoro-substituted analogs, on the contrary, it increases.

3. The reduced sound propagation speed, which is reciprocal to the reduced density, increases due to the fluoro-substitution of aliphatic alcohols. In our opinion, this is associated with a reduction in the concentration of hydrogen bonds in the liquid. Note that, in water, where there is a network of hydrogen bonds, the sound propagation speed is lower than in saturated aliphatic alcohols.

4. The adiabatic compressibility coefficient decreases, when transferring from saturated aliphatic alcohols to their fluoro-substituted analog. It also decreases for fluoro-substituted and unsubstituted alcohols with the increasing number of carbon atoms.

5. A comparison of the properties of alcohols with those of water, which has a continuous network of hydrogen bonds, and with the properties of hydrogen peroxide, where there are hydrogen bonds, but not their network, shows that the change in physical properties after the fluoro-substitution is connected with the change in the number of hydrogen bonds per unit volume.

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ПОРІВНЯННЯ В'ЯЗКОПРУЖНИХ ВЛАСТИВОСТЕЙ АЛІФАТИЧНИХ СПИРТІВ ТА ЇХ ФТОРЗАМІЩЕНИХ АНАЛОГІВ

У роботі проведено порівняння в'язкопружних властивостей рідких аліфатичних спиртів пропанолу-1, пентанолу-1

та гептанолу-1 із властивостями їх фторзаміщених аналогів. Використано експериментальні дані по густині, кінематичній в'язкості та швидкості поширення звуку і симуляції штучними нейронними мережами величин критичних параметрів та параметрів Ван дер Ваальса досліджених рідин. Порівняння властивостей спиртів із властивостями води, в якій є суцільна сітка водневих зв'язків, та властивостями перекису водню, де є водневі зв'язки, але сітка відсутня, показують, що зміна фізичних властивостей при фторзаміщенні пов'язана зі щільністю водневих зв'язків у рідині.

Ключові слова: фторзаміщені спирти, вода, перекис водню, зсувна в'язкість, швидкість звуку, штучні нейронні мережі.