РОЗДІЛ 3 ХОЛОДИЛЬНІ ТА СУПУТНІ ТЕХНОЛОГІЇ

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VISCOSITY OF TERNARY SOLUTIONS COMPOSED OF PROPYLENE GLYCOL, ETHANOL AND WATER

This paper presents the experimental data on the kinematic viscosity of water / ethanol / propylene glycol solutions that are prospective as coolants. An experimental setup with capillary viscometers was used to measure the kinematic viscosity of samples in the temperature range from 243K to 303K and in the wide range of mass fractions of components. Experimental uncertainty of the obtained experimental data does not exceed 0.4 %. Solutions compositions were chosen according to needs of the refrigeration industry in order to provide a possibility of their application at temperatures 233 K and higher. The new model for prediction of the kinematic viscosity of coolants within wide interval of reduced temperatures $0.1 \le t = 1 - T/\overline{T}_C \le 0.6$ *(*$\overline{T}_C$ *is the pseudocritical temperature) based on the limited experimental data both for pure substances and aqueous solutions of mono- and polyalcohols is presented in paper.*
Keywords: *Secondary coolant; Kinematic viscosity; Ternary solutions; Propylene glycol; Water; Ethanol; Experiment; Modelling.*

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I. INTRODUCTION

Aqueous solutions of ethylene glycol and propylene glycol are widely used as secondary coolants and heat-transfer fluids. As coolants, they have following advantages: low crystallization temperature, low corrosivity (compared to brines). Taking into account low toxicity requirements, the-application of propylene glycol in secondary loop refrigeration systems has a significant advantage compared to ethylene glycol. However, propylene glycol has a higher viscosity compared to ethylene glycol, which leads to increase of the power consumption for circulation pumps in secondary coolant circuits. Apart of that, low toxicity aqueous solutions of propylene glycol are widely used in refrigeration plants in the food industry.

Despite of a wide area of application, secondary loop refrigeration systems have certain number of drawbacks. First of all, above-mentioned systems have a high material capacity due to require additional equipments for a circuit of the secondary coolant compared to direct cooling systems. In addition, presence of the additional circuit requires lower temperatures in an evaporator of the refrigeration system compared to direct cooling systems (approximately by 4 - 6 K). Last drawback leads to losses in values of the coefficient of performance. Last but not least, secondary loop refrigeration systems consume an additional power required for circulation pumps.

The impact of the above-mentioned negative factors can be compensated by application of nanotechnologies during the development both of new compressor nanooil and of perspective nanocoolants [1, 2].

Additives of metal and metal oxide nanoparticles into ethylene or propylene glycol based coolants is a very promising way to increase the efficiency of refrigerating plants. An addition of nanoparticles into the coolant leads to an increase both in values of the thermal conductivity of a nanofluid and heat transfer coefficients in heat exchangers [1-4]. All mentioned above will promote to reduce of the material capacity of the equipment. However, presence of nanoparticles in the multicomponent coolant lead to increase of viscosity of the heat transfer fluid [2-4].

The information on thermophysical properties of nanocoolants composed of ethylene glycol or propylene glycol is very rare in the literature. It should be mentioned, that there are few studies related with prospects of the application of nanofluids as coolants in combustion engines [5]. A number of authors showed that the main problem of the application nanoparticles as additives to secondary coolants and heat-transfer fluids is the significant increase of the relative (compared to the base fluid) viscosity at low temperatures [6, 7].

For preliminary assessment of the influence of nanoparticles additives on the properties of the heat transfer fluids the viscosity of two different nanofluids has been measured at low temperatures typical for

refrigeration. The object of the present study is industrial coolant (inhibited aqueous solution of propylene glycol 54 % by mass). Two types of additives were used: nanoparticles of the TiO_2 (produced by Sigma Aldrich, size less than 25 nm in powder) and Al_2O_3 (produced by Wenzhou Jingcheng Chemical Co, α-modification, size 10±5 nm in powder).

Experimental values of relative viscosity for nanofluids prepared from inhibited aqueous solutions of the propylene glycol (54% by mass) and two different types of nanoparticles are presented in Figure 1.

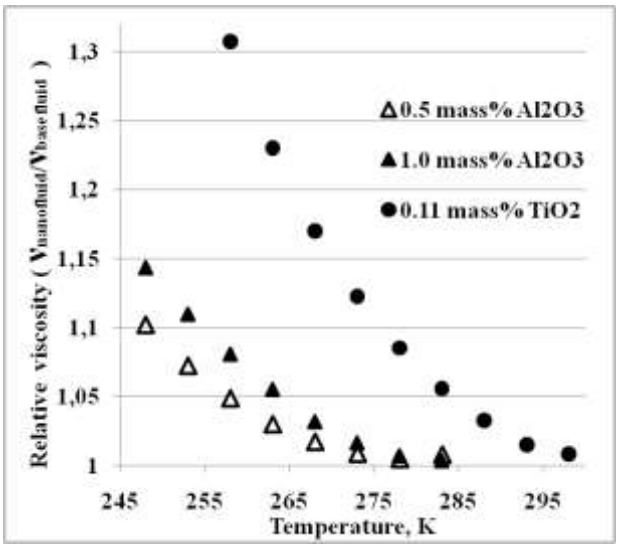

Figure 1 *– Temperature dependencies of relative viscosity for the inhibited aqueous solutions of the propylene glycol (54% by mass) with nanoparticles of TiO_2 and Al_2O_3.*

The information in Figure 1 and literature source [7] allows to make several conclusions. First of all, the significant increase of relative viscosity of nanofluids is observed at low temperatures, where an orientation interaction between polar molecules becomes significant. Secondly, the size and the shape of nanoparticles define the effect of the increase of the relative viscosity.

In order to compensate the negative effect of nanoparticles presence on the viscosity of nanocoolants, an addition of monoalcohols into solutions of water and glycols is proposed by authors of present paper. The addition of monoalcohols into aqueous solutions will promote both decrease of the crystallization temperature and reduce of the viscosity at low temperatures.

Thus, the using of an optimal value of concentrations of nanoparticles and monoalcohols can promote both to an enhancement of the heat transfer coefficients in heat exchangers and reduce of the energy consumption of pumps which provide circulation of secondary coolants. Selection of ethanol as component of coolants can be explained by several factors: low viscosity of ethanol at low temperatures typical for refrigeration, low toxicity, low crystallization temperature (159 K), low price on the market.

The diversity of components of multicomponent coolants (water, ethylene glycol or propylene glycol, ethanol, nanoparticles, corrosion inhibitors, etc) must be taken into account during development of a new generation of nanocoolants. For such reason, a choice of an optimal composition of nanocoolants is a complex task. A solution of this problem significantly depends on the development of methods for prediction of thermophysical properties of multicomponent liquid systems. At the first stage of the development of new nanocoolants, thermophysical properties (especially viscosity) of water/ethanol/propylene glycol solutions should be studied experimentally. The task of the selection of the optimal composition of ternary systems can be simplified if the information on viscosity of such multicomponent systems is available.

The viscosity of aqueous solutions of ethylene glycol and propylene glycol was studied at temperatures above 273 K [8-12]. However, it should be mentioned, that the experimental information on viscosity at low temperatures typical for refrigeration for the above-mentioned solutions is rare in the literature [8, 9]. The data on viscosity of ternary aqueous solutions of mono- and polyalcohols are very limited. For example, thermophysical properties of water/ethanol/ethylene glycol and methanol/ethanol/propylene glycol solutions are presented in [13, 14]. The information on viscosity of solutions composed of propylene glycol, water and ethanol is not presented in the literature. Taking into account all mentioned above, the purpose of presented study was the experimental and modeling study of the concentration dependence of the viscosity for water/ethanol/propylene glycol solutions.

II. EXPERIMENTAL SECTION

Anhydrous grade propylene glycol (1,2-propandiol) with mass fraction purity 99.79 mass% was used to prepare ternary solutions. The water content of propylene glycol was 0.050 % by mass and measured with a Karl Fisher analysis. Ethanol with mass fraction of water 5.80% was used. The water mass fraction was taken into account at calculation the ternary solution concentration. Finally, bidistilled water was used. The mass amounts of each component were determined using high precision balance (± 0.0005 g).

The experimental set up with Ubbelohde capillary viscometers (suspended-level viscometers) were used to measure the kinematic viscosity of samples. Uncertainty in temperature measurements was not exceed 0.2 K, uncertainty in viscosity measurement was not exceed 0.4 %.

Compositions of studied solutions are shown in Figure 2 by points. These ternary compositions cover the region where water/ethanol/propylene glycol as coolant can be used. Concentrations of the solutions components were chosen with respect to the capability of their application in refrigeration systems (values of the melting point and viscosities at low temperatures).

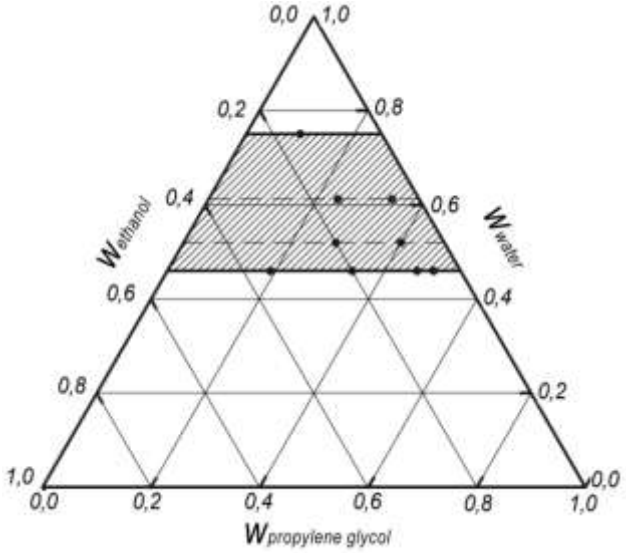

Figure 2 *− Ternary diagram for water / ethanol / propylene glycol solutions (a shaded area shows mixtures compositions which are typical for the application in refrigeration systems): ● − mass fractions in which the values of kinematic viscosities were measured*.

The experimental data on the kinematic viscosity of solutions composed of water, ethanol and propylene glycol are shown in Table 2. In order to evaluate the dynamic viscosity of mixtures samples, the density was measured. Experimental data on density of the considered ternary systems were published previously in the temperature range 248-303 K [16].

III. MODELING SECTION

The most of information on viscosity of coolants is presented in the literature in a form experimental data obtained within long-term or expensive measurements with further extrapolation in the low temperature range. Different approaches are used to evaluate the viscosity of multicomponent liquids [17, 18]. For example, mixing rules, the self-referencing method and Lohrenz–Bray–Clark model based on data on the density and viscosity of the pure components are widely used by many authors in calculations of the viscosity of multicomponent mixtures [19]. It worth to mention several theoretical models: the rough hardsphere model, the friction theory model, the free volume model and the Vesovic–Wakeham method [19].

Table 1 − Kinematic viscosities of the ternary solutions (x_1 water + x_2 ethanol + (1 - x_1 - x_2) propylene glycol) as a function of mole fractions x_1 and x_2 obtained in the range of temperatures T = (245.1 to 303.2) K.

x_1/w_1	x_2/w_2	*Experimental values*							
0/0	*0/0*	*T, K*	245.2	253.1	263.1	278.2	293.2		
		$v \cdot 10^6$, m^2/c	4289	1644	586.2	168.5	59.65		
0.7722/ 0.4598	*0.0329/ 0.0501*	*T, K*		245.6	253.1	273.1	283.2	293.2	
		$v \cdot 10^6$, m^2/c		160.4	78.53	17.49	10.04	6.295	
0.7658/ 0.4592	*0.0524/ 0.0804*	*T, K*	248.0	258.2	267.8	273.0	281.4	291.7	
		$v \cdot 10^6$, m^2/c	113.5	47.02	21.55	15.59	9.764	5.977	
0.7434/ 0.4599	*0.1263/ 0.1998*	*T, K*		253.1	263.2	273.3	283.2		
		$v \cdot 10^6$, m^2/c		41.18	19.55	13.19	7.945		
0.7160/ 0.4590	*0.2135/ 0.3500*	*T, K*		248.8	258.2	273.2	283.2	293.2	303.2
		$v \cdot 10^6$, m^2/c		38.36	20.89	8.627	5.537	3.921	2.772
0.8008/ 0.5195	*0.0604/ 0.1002*	*T, K*	245.1	253.1	266.2	273.2	283.2	292.2	
		$v \cdot 10^6$, m^2/c	115.30	55.35	19.34	12.41	7.315	4.874	
0.7821/ 0.5194	*0.1179/ 0.2002*	*T, K*	245.9	253.2	266.1	273.1	283.2	293.1	303.1
		$v \cdot 10^6$, m^2/c	82.82	44.16	17.31	11.37	6.791	4.329	2.981
0.8591/ 0.6100	*0.0274/ 0.0498*	*T, K*		253.1	263.2	273.1	283.2	293.2	303.2
		$v \cdot 10^6$, m^2/c		41.05	19.14	10.15	6.064	3.879	2.688
0.8408/ 0.6101	*0.0809/ 0.1501*	*T, K*	248.4		263.1	273.2	283.2	293.2	303.2
		$v \cdot 10^6$, m^2/c	57.82		17.86	9.569	5.686	3.673	2.549
0.9006/ 0.7488	*0.0707/ 0.1502*	*T, K*			263.1	273.2	283.2	293.2	303.2
		$v \cdot 10^6$, m^2/c			11.08	6.156	3.774	2.529	1.801

Remarks:
x_1 and x_2 – mole fraction of water and ethanol, consequently; w_1 и w_2 – mass fraction of water and ethanol, consequently.

The analysis of inaccuracy in the prediction of solution viscosities by different models allows to make following conclusions:

- the existing in the literature models do not provide an acceptable quality of data on solutions viscosity;
- it is necessary to develop new models which require a minimum quantity of initial empirical information.

The analysis of the above-mentioned prediction methods shows that in conditions, when experimental data

on the viscosity of multicomponent coolants are limited, the method based on "structure-property" could provide an acceptable accuracy of calculations [19, 20]. The viscosity of multicomponent mixtures can be calculated by following equation:

$$\frac{1}{\eta_{mix}} = a_\eta \left(V_{mix} - Or_{mix}\right)^{b_\eta}, \qquad (1)$$

where η_{mix} is the dynamic viscosity of n-component solution, Pa·s; a_η и b_η are empirical coefficients; V_{mix} is the molar volume of a liquid at the boiling line, $m^3 \cdot mol^{-1}$; Or_{mix} is the orthochor of solution, $m^3 \cdot mol^{-1}$.

The values of orthochor for individual substances can be calculated by relationships proposed in [20]. The initial information necessary for orthochor calculation is following: density or surface tension or molar volume at boiling point. For the multicomponent refrigerants, orthochor can be calculated by following additivity rule:

$$Or_{mix} = \sum_i x_i \cdot Or_i, \qquad (2)$$

where x_i is the molar fraction of i-th component, $mol \cdot mol^{-1}$; Or_i is the orthochor of i-th component, $m^3 \cdot mol^{-1}$.

According to [20], the equation (1) describe experimental values of viscosity for non-associated substances and solutions within a wide range of reduced temperatures $0.005 \leq t \leq 0.45$. Values of reduced temperatures for pure substances can be calculated by Eq. (3), and for solutions by Eq. (3*):

$$t = 1 - T/T_C, \qquad (3)$$

$$t = 1 - T/\bar{T}_C, \qquad (3^*)$$

where T_C is the critical temperature of a substance, K; $\bar{T}_C$ is the pseudocritical temperature of solution, which can be obtained from the solution density data [16, 20].

Some important factors should be emphasized for application of the correlation (1) when predicting viscosity of coolants at low temperatures. First of all, the temperature interval that typical for application of coolant in refrigeration generally corresponds to the range $t > 0.45$, that is unsuitable to area of self-similarity for proposed in literature correlations. Secondly, significantly increasing of the viscosity in polar and associated liquids at low temperatures ($t > 0.45$) related with an existence of orientation effects during intermolecular interactions. In low temperature range (coolants vapor pressure insignificant), the traditional thermodynamic criteria such as acentric factor or Riedel's criterion, unable to expand the area of self-similarity of Eq. (1).

The authors research shows that for extension of the range of application of the correlation (1), it's favorable to use ψ-factor (the factor of complexity of intermolecular interaction [21]). It is necessary to emphasize an important advantage of ψ-factor in studies of the viscosity. The ψ values can be calculated for various substances and solutions from the available in literature information by the correlation proposed in [21]:

$$\psi = 0.1 \cdot \ln(T_{nb}) - 0.122 \cdot \ln(V_{nb}) + 0.006, \qquad (4)$$

where T_{nb} is boiling temperature, K; V_{nb} is the molar volume of liquid at boiling point, $cm^3 mol^{-1}$.

By the authors opinion, the ψ-factor characterizes the deviation of the values of intermolecular interactions that typical for various polar substances from these values that typical for the non-polar spherically symmetric molecules. Thus, ψ-factor, as opposed to the different thermodynamic similarity criteria, is most adapted to taking into account the orientation effects in polar and associated substances.

Our analysis showed that deviations between calculated viscosities Eq. (1) and experimental data in the range of reduced temperatures $0.45 \leq t \leq 0.6$ depend on both ψ-factor and reduced temperature.

Taking into account above-mentioned facts, following correlation for viscosity of pure refrigerants and their solutions at low temperatures typical for refrigeration is proposed:

$$\frac{1}{\eta_{mix}} = a_\eta \left(V_{mix} - Or_{mix}\right)^{b_\eta} \cdot \left(1 + \psi \cdot t^8\right), \qquad (5)$$

Verification of the proposed correlation was carried out for the individual substances listed in Table 2. In Table 2 the initial information for calculation and characteristic parameters for these substances are also listed.

Table 2 – The initial information necessary in viscosity calculations and characteristic parameters for different pure substances

Substance	M, kg/kmol	T_C, K	T_{nb}, K	Electric dipole moment, D	ψ
Water	18.015	647.25 [22]	373.12 [22]	1.855 [22]	0.2403
Ethanol	46.068	513.9 [22]	351.39 [22]	1.6909 [22]	0.0876
Methanol	32.042	512.6 [22]	337.63 [22]	1.7 [22]	0.130
Ethylene glycol	62.07	720±10 [23]	470.45 [10]	2.20±0.02 [9]	0.112
Propylene glycol	76.09	676±1 [23]	461.35 [10]	3.63 [9]	0.075
Ethane	30.069	305.32 [22]	184.57 [22]	0 [22]	~ 0.0
Isobutan	58.122	407.81 [22]	261.4 [22]	0.132 [22]	~ 0.0
Butan	58.122	425.13 [22]	272.66 [22]	0.05 [22]	~ 0.0
Nonane	128.26	594.55 [22]	423.91 [22]	0.07 [22]	~ 0.0
Dodecane	170.33	658.1 [22]	489.3 [22]	0 [22]	~ 0.0

Average absolute percent deviation as integral criterion of an error in calculations of viscosities was used:

$$AAPD = \frac{100\%}{N} \cdot \sum_{i=1}^{N} \left| \left(\eta^{[\,]} - \eta^{calc} \right) / \eta^{[\,]} \right|, \qquad (6)$$

where N is the number of points in the temperature range where the initial viscosity data were taken; $\eta^{[\,]}$ is the viscosity value from literature source; η^{calc} is the viscosity value calculated by Eq. (1) or Eq. (5).

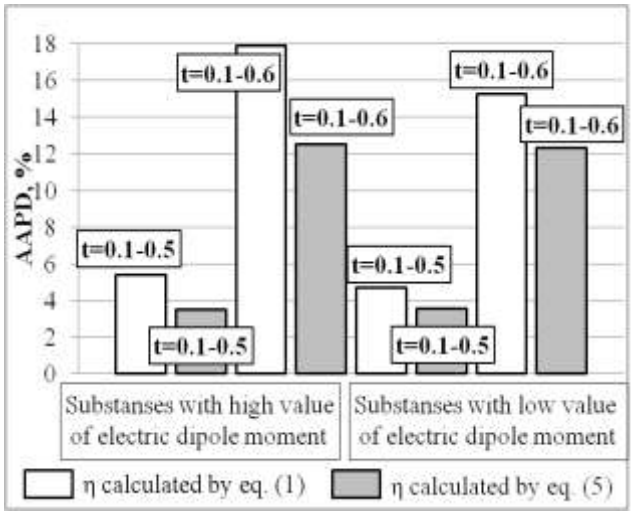

Figure 3 *− Average absolute percent deviation between the experimental and predicted by the correlations (1) and (5) viscosity values for two groups of individual substances listed in Table 2. Deviations were obtained with help of the initial information in the ranges of reduced temperatures t = 0.1 - 0.5 and t = 0.1 - 0.6*

Figure 3 shows that including in Eq. (1) ψ-factors allows to increase the prediction quality for viscosity of the different pure substances in the wide range of temperatures.

In order to adapt the correlation (5) to prediction of viscosity of multicomponent coolants authors proposed to use following additivity rule at evaluation of ψ-factors:

$$\psi_{mix} = \sum_{i} x_i \cdot \psi_i \,, \qquad (7)$$

where ψ_i is the factor of complexity of intermolecular interaction of i-th component of the solution.

The verification of the proposed model for ternary solutions water/ethanol/propylene glycol was done by calculation of relative deviation:

$$dev = \left(\left(\eta^{calc} - \eta^{exp} \right) / \eta^{exp} \right) 100\% \,, \qquad (8)$$

where η^{exp} is the experimental viscosity.

Deviations between viscosity values calculated by the Eq. (5) and experimental data presented in Table 1 are shown in Figure 4.

Figure 4 shows that the proposed model (see Eq. (2) - (5)) can provide the sufficient accuracy for prediction of viscosity of multicomponent coolants. Maximum deviations are indicated at low temperatures for solutions with high values of mass fraction of water. At the same time, acceptable prediction capabilities of the model (Eq. (2) - (5)) for aqueous solutions allows to make a preselection the optimal composition of new multicomponent coolants. Estimated by the above-mentioned model composition values can be used as a basis for the development of a new generation of nanocoolants based on solutions water, ethanol and propylene glycol.

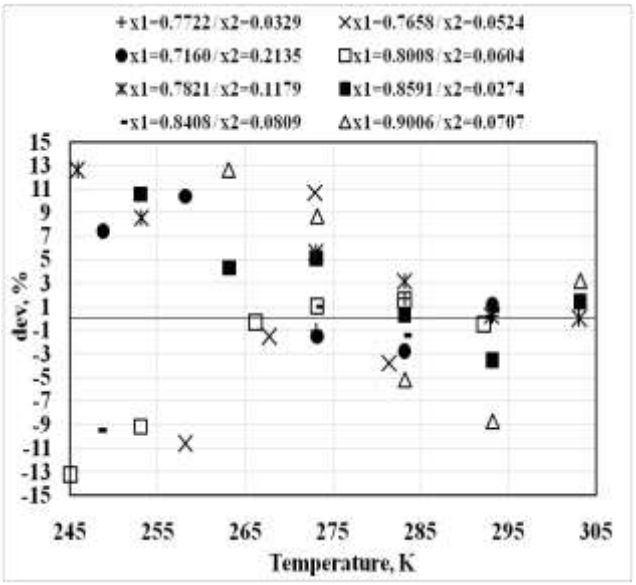

Figure 4 *− The deviations of viscosity values calculated with using Eq. (5) from experimental data listed in Table 1 for solutions water (x_1) / ethanol (x_2) / propylene glycol.*

IV. CONCLUSION

The availability of the accurate information on thermophysical properties of aqueous solutions of mono- and polyalcohols is necessary condition to choose the optimal composition of multicomponent coolants. However, the information on viscosity of prospect coolants may be absent in the low-temperature range typical for refrigeration. In such complicated conditions, a combination of the precision of the experimental information on viscosity in the limited temperature range and organizational principles of predictive models can be considered as a promising approach in studies of viscosity of multicomponent coolants and heat-transfer fluids.

The viscosity prediction methods for solutions of associated liquids which have high values of the dipole moments are insufficiently developed in the literature.

Proposed by authors' model for prediction of viscosity of multicomponent coolants composed by water, mono- and polyalcohols can be recommended for practical application and for further usage in development of new models for coolants thermal conductivity and heat capacity prediction at low temperatures. The authors' further research will be dedicated to solving of these problems.

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ДОСЛІДЖЕННЯ В'ЯЗКОСТІ ТРИКОМПОНЕНТНИХ ВОДНИХ РОЗЧИНІВ ПРОПІЛЕНГЛІКОЛЮ І ЕТАНОЛУ

У статті наведено експериментальні дані з кінематичної в'язкості розчинів вода / етанол / пропіленгліколь, перспективних як холодоносії. Вимірювання виконано з використанням капілярних віскозиметрів в інтервалі температур 243-303 К та у широкому діапазоні концентрацій компонентів. Похибка отриманих експериментальних даних не перевищує 0,4 %. Концентрації компонентів розчинів варіювали, виходячи з вимог, що висуваються до холодоносіїв, які можуть застосовуватися в холодильному обладнанні при температурах від 233 К та вище. На основі отриманих експериментальних даних розроблено нову методику прогнозування в'язкості як чистих речовин, так і водних розчинів одноатомних і багатоатомних спиртів у інтервалі зведених температур $0{,}1 \le \bar{t} = 1 - T/\bar{T}_C \le 0{,}6$, *де* $\bar{T}_C$ *– псевдокрітична температура розчину.*

Ключові слова: *Проміжний холодоносій; В'язкість; Розчини; Пропіленгліколь; Вода; Етанол; Експеримент; Методи розрахунку.*