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EXPERIMENTAL STUDY OF THE EFFECT OF NANOPARTICLES OF TiO_2 ON THE THERMOPHYSICAL PROPERTIES OF THE REFRIGERANT R141b

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Наведено результати експериментального дослідження теплофізичних властивостей холодоагенту R141b, розчину R141b/поверхнево-активна речовина (ПАР) Span-80 і нанофлюїду R141b/Span-80/наночастинки TiO_2. Вміст як ПАР, так і наночастинок TiO_2 у об'єктах дослідження складав 0,1 мас. %.

Виміри проведено на лінії кипіння в інтервалах температур (273…293) К для густини, (293…343) К для поверхневого натягу, (300…335) К для динамічної в'язкості, (293…348) К для теплопровідності, (261…334) К для ізобарної теплоємності.

Показано, що вплив ПАР та наночастинок TiO_2 на густину холодоагенту R141b був незначним і сумірним із невизначеністю експериментальних даних (до 0,08 %). Добавки сумісно ПАР та наночастинок TiO_2 сприяли зниженню поверхневого натягу R141b на величину до 0,3 % у порівнянні з чистим R141b. Добавки сумісно ПАР та наночастинок TiO_2 в R141b сприяли збільшенню в'язкості на (0,8…1,0) %, а добавки ПАР призводилили до суттєвого зниження в'язкості – на (3,5…5,0) % у порівнянні з в'язкістю чистого R141b. Показано, що добавки ПАР в R141b суттєво не впливають на теплопровідність (ефект не перевищував 0,25 %), а добавки сумісно ПАР та наночастинок TiO_2 призводять до збільшення теплопровідності холодоагенту (0,3…1) %. Було зафіксовано зниження на (1,5…2,0) % питомої ізобарної теплоємності при введенні у R141b сумісно ПАР та наночастинок TiO_2 та незначне збільшення теплоємності при додаванні ПАР (до 1,0 %).

Зроблено висновок, що вплив добавок наночастинок та ПАР на теплофізичні властивості холодоагенту R141b є неоднозначним та непрогнозованим. Результати експериментальних досліджень впливу наночастинок на теплофізичні властивості холодоагенту підтверджують необхідність розробки методів моделювання цих властивостей на основі урахування наявності на поверхні наночастинок структурованої фази базової рідини або ПАР.

Ключові слова: холодоагент R141b, нанофлюїд R141b/наночастинкиTiO_2, густина, поверхневий натяг, питома ізобарна теплоємність, теплопровідність, в'язкість

1. Introduction

Application of nanofluids as working fluids is one of the methods to improve the vapor compression refrigeration systems performance, and consequently, reduce the costs of cold production.

Nanofluids are colloidal systems containing the nanoparticles of solid metals, metal oxides, or carbon allotropic modifications. The nanofluids are named "nanorefrigerant" in a case the hydrocarbons or halogenated hydrocarbons (the working fluids of vapor compression refrigeration systems) are used as the base fluid.

Performed in the past two decades studies have shown that the addition of small amounts of nanoparticles can significantly change the thermophysical properties of the base fluid [1–10]. The application of nanofluids as coolants and working fluids in thermal power systems (including refrigeration) can contribute to the intensification of heat transfer processes in various heat exchangers [11–13].

Therefore, the study of the influence of nanoparticle additives on various thermophysical properties of refrigerants is of current importance. New empirical information on the thermophysical properties of nanorefrigerants is needed both for the development of calculation models for predicting the nanofluids thermophysical properties and for the modeling of heat transfer processes.

2. Literature review and problem statement

Some papers can be mentioned among the studies devoted to the investigation of the nanorefrigerants thermophysical properties [1–11].

In paper [1], the data on thermal conductivity and viscosity for nanofluids R134a/Al_2O_3 at the volume fraction of nanoparticles from 1 to 5 % is presented. The increase in thermal conductivity by (4...7) % and viscosity by 3 % at the volume fraction of nanoparticles of 1 % in the temperature range of (300...325) K has been shown. However, such properties as density, heat capacity and surface tension for the considered nanofluid have not been investigated. That fact limits the possibility of calculating the characteristics of the heat transfer process.

In paper [2], the data on the thermal conductivity and viscosity of the nanorefrigerants R134a/CuO at the nanoparticle concentrations of 1 to 5 vol. % at temperatures of 300 to 320 K are presented. The obtained results show that the thermal conductivity of CuO/R134a nanorefrigerant increased (by 11.5 % for a nanofluid containing 5 vol % of nanoparticles compared with a nanofluid containing 1 vol % of nanoparticles) and specific heat capacity decreases with the augmentation of particle concentrations (by 12.5 % for a nanofluid containing 5 vol % of nanoparticles compared with a nanofluid containing 1 vol % of nanoparticles). The results of measurements of the viscosity and density of the nanorefrigerant showed a significant increase with the increase of nanoparticles volume fractions: more than 2-time viscosity increase for a nanofluid containing 5 vol % of nanoparticles compared with a nanofluid containing 1 vol % of nanoparticles. The obtained results [2] indicate the unpromising nature of the studied nanofluids. Since the negative effect of increasing viscosity at high concentrations of nanoparticles eliminated the positive effect of thermal conductivity enhancement. It would be much more interesting to study these effects at low (up to 1 vol. %) fractions of nanoparticles, as well as to study the possibility of using surfactants to control the viscosity of the nanofluid.

In paper [3], thermal conductivity and viscosity of the Al_2O_3/R141b nanorefrigerant for nanoparticles concentrations of 0.5 to 2 vol. % at temperatures of 5 to 20 °C have been investigated. The highest observed thermal conductivity and viscosity were 1.626 and 1.79 times greater for a nanofluid containing 2 vol % of particles compared with the properties of the base fluid. As in [1], such important properties for the analysis of heat transfer processes as density, heat capacity and surface tension for the considered nanofluid [3] have not been investigated.

The results of a detailed experimental investigation on thermophysical properties of the nanofluid isopropanol/Al_2O_3 have been presented in [4]. But the shortcoming of this study is the lack of data on thermal conductivity of the investigated system.

The reviews of experimental investigations of nanofluids thermophysical properties performed by a number of researchers are presented in [5–7]. But the analysis of information of the reviews [5–7] did not allow us to draw a conclusion about the influence of the adding of nanoparticles on the properties of base fluids. The main reason for this fact is the absence of agreement in the results of various authors.

It has been found in [8] that the nanorefrigerants have a much higher and strongly temperature-dependent thermal conductivity at very low particle concentrations than pure refrigerants. This can be considered as one of the key parameters for enhanced performance of refrigeration and air conditioning systems.

There are papers dedicated to investigating the influence of not only concentration but also the nanoparticles material and shape [9] and nanoparticles size on the nanorefrigerants thermophysical properties. The detailed review of performed studies of nanorefrigerant thermophysical properties is presented in [11]. But the analysis of the information presented in [9–11] did not allow us to draw a conclusion about the influence of the considered characteristics on thermophysical properties of nanofluids.

It has been shown within the performed analysis that the published experimental studies dedicated to the thermophysical properties of nanofluids do not contain the complete information necessary to predict the characteristics of the heat transfer process by using the existing models.

The expediency of nanorefrigerants practical application is analyzed in [12, 13], where the results of the study of heat transfer during nanorefrigerants boiling in evaporators are presented.

The authors of the paper [12] presented the results of the study of the flow boiling heat transfer characteristics of four nanorefrigerants (R141b/Cu, R141b/Al, R141b/Al_2O_3 and R141b/CuO) in an internal thread copper tube. They found that the maximum heat transfer coefficient of the four kinds of nanorefrigerants increased by 17–25 %, the average heat transfer coefficient increased by 3–20 %, and the maximum heat transfer coefficient of the R141b/Cu nanorefrigerant increased by 25 %. But the obtained effects [12] have not been compared with a variation in thermophysical properties of nanorefrigerants, which does not allow understanding the reason for the heat transfer enhancement during boiling.

The authors of the paper [13] investigated the nucleate pool boiling heat transfer of the nanorefrigerant 141b/TiO_2 nanoparticles (at 0.01, 0.03 and 0.05 vol %) at different pressures. Pool boiling experiments of nanofluids were conducted and the results were compared with those of the base refrigerant. The results indicate that the nucleate pool boiling heat transfer deteriorated with increasing particle concentrations, especially at high heat fluxes. At 0.05 vol %, the boiling heat transfer curves were suppressed. But in the paper [13], there is no comparison of the experimental results with the heat transfer coefficients during nanorefrigerant boiling calculated with using the existing correlations with taking into account the thermophysical properties.

The results of experimental investigations on the influence of the nanoparticles adding in refrigerants on the performance of the vapor compression refrigeration system are presented in [4, 11, 14]. The prospects of using the nanorefrigerants for enhancement of the coefficient of performance of vapor compression refrigeration systems are shown. However, the variation in the studied parameters in connection with the influence of nanoparticle additives on the thermophysical properties of nanorefrigerants has not been analyzed in the reviews [4, 11–14]. This fact does not allow analyzing the main factors determining the change in the performance parameters of refrigeration systems that use nanorefrigerants as working fluids.

Thus, at present, there are no experimental studies containing the complex investigation of thermophysical properties of any nanorefrigerant prepared according to a certain technology using identical samples of nanoparticles.

It should be emphasized that the lack of accurate information on the thermophysical properties of nanorefrigerants does not allow us to correctly interpret the data on the effect of nanoparticles on both the heat transfer processes and enhancement of the coefficient of performance of refrigeration systems.

Several serious problems must be taken into account for the successful introduction of nanorefrigerants to the industry. It is worth noting that nanofluids are complex multicomponent thermodynamic systems, which consist of the base fluid, nanoparticles and, sometimes, surfactants.

Due to the thermodynamic instability of colloidal systems, the size of nanoparticles with sorbed on their surface surfactant or base fluid molecules and, consequently, the fraction of this interfacial phase, can change with temperature [15–17]. These changes can lead to variation in the nanorefrigerant thermophysical properties. The additional studies of the temperature dependence of changes in the nanofluids structure are needed for the correct interpretation of the experimental data on the thermophysical properties of nanorefrigerants. According to the mentioned above, the investigations of the colloidal stability of nanorefrigerants and determination of the nanoparticles hydrodynamic diameter and the amount of base fluid or surfactant sorbed on the surface of nanoparticles are needed.

Thus, not only the concentration of the components but also the size and shape of the nanoparticles, the presence of surfactants, the technology of nanofluids preparation, etc. influence on the properties of nanofluids. For this reason, the presented in the mentioned above studies data on the influence of low concentrations of nanoparticles on viscosity, surface tension, thermal conductivity and specific heat capacity are often not in agreement with each other even for systems with nanoparticles with the same chemical nature.

As follows from [7, 18, 19], there are no accurate for practical purposes thermodynamic models for the calculation of such nanofluids properties as thermal conductivity, viscosity and heat capacity. As a rule, the presented in the literature correlations do not take into account the influence of nanoparticle size, the presence of surfactants in nanofluids, the structural changes in the layer of the molecules of base fluids or surfactants sorbed on the nanoparticles surface [7, 18, 19]. At the same time, some researchers state [7] that the developed models for the calculation of thermophysical properties are quite applicable for nanofluids with a low concentration of nanoparticles.

As follows from the above review, the study of the influence of low concentrations of nanoparticles and surfactants on the complex of thermophysical properties of halogenated hydrocarbon refrigerants is required to develop the thermodynamic models for predicting the nanorefrigerants properties and to analyze the heat transfer processes with nanorefrigerants.

3. The aim and objectives of the study

The aim of this work is an experimental investigation of the influence of TiO_2 nanoparticles on the density, surface tension, specific isobaric heat capacity, viscosity and thermal conductivity of the R141b refrigerant.

To achieve the set aim, the following objectives were to be accomplished:

– to justify the choice of a model system based on the halogenated hydrocarbon refrigerant with adding the metal oxide nanoparticles and to prepare the colloidally stable samples of nanofluids for experimental investigation their thermophysical properties;

– to perform the experimental study of the density, viscosity, surface tension, thermal conductivity and specific isobaric heat capacity for samples: R141b, R141b/surfactant and R141b/surfactant/TiO_2 nanoparticles;

– to analyze the variation in thermophysical properties of the base refrigerant in the presence of a surfactant and TiO_2 nanoparticles with using the obtained experimental data.

4. Materials and methods used in the study of thermophysical properties of the R141b refrigerant, R141b/Span-80 surfactant solution and R141b/Span-80 surfactant/TiO_2 nanoparticles nanofluid

4. 1 Justification of the selection of the objects of study and their preparation

Taking into account the variety of refrigerants used in refrigeration systems, it is advisable to perform the tasks of the study for the model system. The R141b refrigerant was chosen as the base fluid for the model system in this study. This refrigerant has been widely used for model studies by many researchers [3, 10, 12, 13, 20, 21]. The nanorefrigerant preparation has not been difficult because R141b has the low vapor pressure at ambient temperatures. In addition, the R141b refrigerant is in thermodynamic similarity with halogenated hydrocarbons refrigerants, which have been widely used in the refrigerant industry.

TiO_2 nanoparticles are the industrial product with low cost and they are often considered by researchers as a promising additive to halogenated hydrocarbons refrigerants [4, 11, 14]. It should be taken into account that the concentration of nanoparticles in the working fluids is recommended to be low (up to 0.5 wt. %) in orders to ensure good colloidal stability and low cost of the nanorefrigerants.

In addition, information on the thermophysical properties of the R141b/TiO_2 nanorefrigerant is necessary for the thermodynamic interpretation of previously obtained data on the boiling processes of this nanofluid [20, 21]. The effects obtained in the study of the thermophysical properties of the model system R141b/nanoparticles can be extended to other halogenated hydrocarbons refrigerants, which are widely used in the refrigeration industry and in air conditioning systems.

According to the aforementioned, the R141b refrigerant, CAS No. 1717-00-6 (manufactured by Zhejiang MR Refrigerant Co. Ltd, China) was selected as the base fluid for nanofluid preparation in the presented study. TiO_2 nanoparticles were used as additives. According to the manufacturer's information, the size of the nanoparticles in the powder does not exceed 25 nm, CAS No.1317-70-0 (Sigma-Aldrich).

The technology of preparation and results of the investigation of the colloidal stability of the R141b/TiO_2 nanofluid containing various types of surfactants have been presented in detail in [21]. The selection of the surfactants type and their concentrations and the stability of the R141b/TiO_2

nanofluids containing anionic, cationic or nonionic surfactants have been studied in [21]. Based on the obtained results [21], it was concluded that the best colloidal stability among the considered surfactants is provided by the Span-80 nonionic surfactant.

Additional studies conducted as part of this work confirmed the sufficient colloidal stability of the R141b/TiO_2 nanoparticles system in the presence of Span-80 CAS No. 1338-43-8 (Sigma-Aldrich).

It should be taken into account that not only nanoparticles but also surfactant additives will contribute to the change in thermophysical properties of the base fluid. Therefore, the research objects of this work are:

R141b refrigerant without nanoparticles and surfactants additives – R141b;

– solution of the R141b refrigerant and Span-80 surfactant (0.1 wt.%) – R141b/Surf.;

– nanofluid consisting of the R141b refrigerant, Span-80 surfactant (0.1 wt.%) and TiO_2 nanoparticles (0.1 wt.%) – R141b/Surf./TiO_2.

4. 2. Methods and equipment used in the experimental study of the thermophysical properties

In the samples preparation, the amount of the components has been determined by the gravimetric method. The GR-300 precision balance with an uncertainty of 0.5 mg has been used.

The measurements of the density for the object of the study were performed by the pycnometric method using the experimental setup described previously in [22, 23]. The glass cell containing the testing substance (pycnometer) was placed in the thermostat equipped with an automatic temperature control. The thermostat temperature stability has been estimated to be within 0.02 K. For measuring the level of the liquid phase of the sample in the pycnometer, the KM 8 cathetometer with an uncertainty of 0.015 mm was used.

To investigate the surface tension, the modified differential method of capillary rise on the experimental setup presented in [22, 23] has been applied. The method's advantage is the use of more than one capillary pairs for measuring the height differences of the liquid rising. The capillary constant a^2 is calculated as the weighted average value. The use of more than one capillary pairs significantly increases the accuracy of the experimental data.

The following relationship was used for the calculation of the surface tension for the object of the study:

$$\sigma = 0.5 \cdot g \cdot a^2 (\rho' - \rho''), \qquad (1)$$

where σ is the surface tension; ρ' and ρ'' are the density of liquid and vapor phases, g is acceleration of gravity; a^2 is the capillary constant.

Data for the vapor density of R141b were taken from the REFPROP database [24].

Experimental study of the dynamic viscosity for the samples of the object of the study has been performed on the Heppler viscosimeter (based on the rolling ball method). The Heppler viscosimeter instrument uncertainty was 1.0 % in the whole range of measurement. The measured dynamic viscosity and density data have been used to calculate the kinematic viscosity.

The specific isobaric heat capacity measurements were performed using an adiabatic calorimeter. The detailed description of the experimental setup was reported in [25]. The combined standard uncertainty of measuring the absolute temperature of the container with the sample did not exceed 0.03 K. The combined standard uncertainties of determining the heat input did not exceed $2.5 \cdot 10^{-5}$ J and the temperature difference – 0.0045 K.

The thermal conductivity measurements of the objects of the study were performed using two independent methods: stationary hot-wire method and transient hot-wire method. The diameter of the hot-wire was 0.1 mm, the thickness of the sample of the studied substance was 0.55 mm. Method of thermal conductivity measurement is presented in [26]. As was shown by the performed analysis, stationary hot-wire method allows obtaining reliable data for colloidal time-stable nanofluids samples.

It should be noted that the key problem when studying the thermophysical properties of nanofluids is the stability of nanoparticles to agglomeration and sedimentation in the range of experimental parameters. The nanoparticles in nanofluids can coalesce and precipitate. This leads to a variation in the nanoparticles concentration in a colloidal solution and a variation in the thermophysical properties of nanofluids with time. In this regard, most of the measurements of thermal conductivity have been obtained using the transient hot-wire method, which has sufficient accuracy and quickness (the measurement process takes no more than a few seconds).

5. Experimental results of the investigation of the thermophysical properties of the R141b refrigerant, R141b/Span-80 solution and R141b/Span-80/TiO_2 nanoparticles nanofluid

The density of the R141b refrigerant, solution of R141b/Span-80 and nanofluid of R141b/Span-80/TiO_2 has been measured in the temperature range from 273 to 298 K. The temperature range for density measurement was taken to be narrower than in the study of the other properties because the effect of nanoparticle additives on the density of the base fluid was insignificant.

The experimental data of the density for the object of the study are shown in Table 1.

Table 1

Experimental values of the density ρ for the objects of the study along the liquid-saturation line*

R141b		R141b/Surf.		R141b/Surf./TiO_2	
T (K)	ρ (kg·m^{-3})	T (K)	ρ (kg·m^{-3})	T (K)	ρ (kg·m^{-3})
273.22	1270.7	273.55	1270.1	273.21	1271.3
273.24	1270.4	280.17	1258.4	280.08	1259.1
278.20	1261.3	287.29	1245.9	287.68	1245.6
279.96	1258.8	293.63	1234.5	292.38	1237.3
283.70	1251.7	297.33	1228.1	298.51	1226.3
287.40	1245.4	–	–	–	–
292.87	1235.5	–	–	–	–
298.02	1226.4	–	–	–	–

*Note: * – standard uncertainty of temperature mesurement u(T)=0.1 K. Expanded uncertainty of density mesurement U(ρ)=(7.2–8.8) kg·m^{-3} (0.95 level of confidence).*

Temperature dependences of the density in the temperature range from 273 to 293 K for the objects of the study were fitted by the equation:

$$\rho = a + b \cdot T + c \cdot T^2. \qquad (2)$$

The experimental data on the density and relative deviations for the density of R141b/Surf. and R141b/Surf./TiO_2 from the density of pure R141b are shown in Fig. 1.

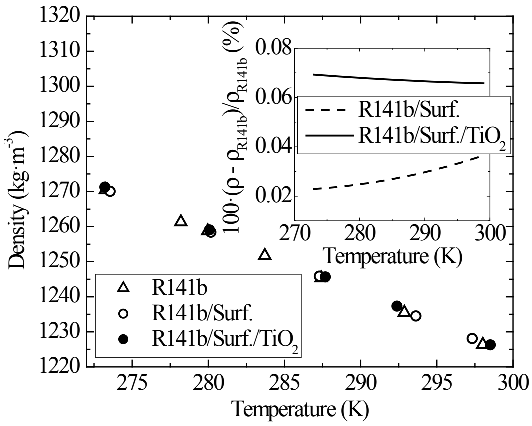

Fig. 1. Temperature dependences of the density of the objects of the study and deviations $100 \cdot (\rho - \rho_{R141b})/\rho_{R141b}$ of the density of R141b/Surf. or R141b/Surf./TiO_2 from the density of pure R141b

The experimental data of the capillary constant a^2 and data on the surface tension σ of the object of the study obtained from the experiment and Eq. (1) are shown in Table 2.

Obtained data on the surface tension of the object of the study in the temperature range from 293 to 343 K were fitted by the equation:

$$\sigma = a + b \cdot T. \qquad (3)$$

The experimental data on the surface tension and relative deviations for the surface tension of R141b/Surf. and R141b/Surf./TiO_2 from the surface tension of pure R141b are shown in Fig. 2.

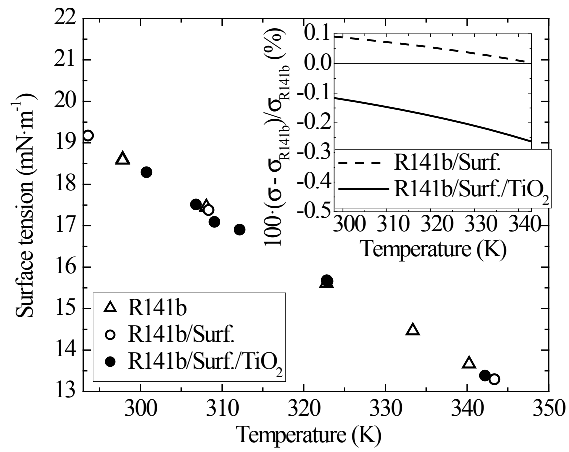

Fig. 2. Temperature dependences of the surface tension of the objects of the study and deviations $100 \cdot (\sigma - \sigma_{R141b})/\sigma_{R141b}$ of the surface tension of R141b/Surf. or R141b/Surf./TiO_2 from the surface tension of pure R141b

The experimental data of the dynamic viscosity η and kinematic viscosity ν of the object of the study are shown in Table 3.

Table 2

Experimental values of the capillary constant a^2 and surface tension σ for the object of the study along the liquid-saturation line*

R141b			R141b/Surf.			R141b/Surf./TiO_2		
T (K)	a^2 (mm²)	σ (mN·m⁻¹)	T (K)	a^2 (mm²)	σ(mN·m⁻¹)	T (K)	a^2 (mm²)	σ (mN·m⁻¹)
297.83	3.082	18.60	293.60	3.155	19.17	300.76	3.046	18.29
308.07	2.943	17.45	308.35	2.932	17.38	306.82	2.947	17.51
322.79	2.706	15.61	322.83	2.718	15.68	309.07	2.932	17.09
333.35	2.562	14.47	343.36	2.407	13.30	312.15	2.872	16.90
340.26	2.456	13.66	–	–	–	322.93	2.715	15.66
–	–	–	–		–	342.19	2.416	13.38

*Note: * – Standard uncertainty of temperature mesurement u(T)=0.1 K. Expanded uncertainty of capillary constant mesurement $U(a^2)$=(0.014–0.022) mm², expanded uncertainty of surface tension determinationt U(σ)=(0.11–0.24) mN·m⁻¹ (0.95 level of confidence)*

Table 3

Experimental values of the dynamic viscosity η and kinematic viscosity ν for the object of the study along the liquid-saturation line*

R141b			R141b/Surf.			R141b/Surf./TiO_2		
T (K)	η (mPa·s)	ν (mm²·s⁻¹)	T (K)	η (mPa·s)	ν (mm²·s⁻¹)	T (K)	η (mPa·s)	ν (mm²·s⁻¹)
300.55	0.4021	0.3271	299.81	0.3849	0.3127	306.16	0.3839	0.3152
302.99	0.3912	0.3195	301.51	0.3807	0.3102	313.00	0.3622	0.3006
315.18	0.3505	0.2920	313.24	0.3438	0.2855	324.03	0.3294	0.2786
318.58	0.3425	0.2870	314.53	0.3339	0.2779	333.15	0.3079	0.2647
322.89	0.3310	0.2794	323.91	0.3136	0.2653	–	–	–
331.80	0.3083	0.2644	324.12	0.3164	0.2677	–	–	–
335.23	0.3003	0.2592	333.18	0.2966	0.2550	–	–	–
–	–	–	333.47	0.2931	0.2521	–	–	–

*Note: * – standard uncertainty of temperature mesurement u(T)=0.1 K. Expanded uncertainty of dynamic viscosity mesurement U(η)=(0.0032–0.0043) mPa·s, expanded uncertainty of kinematic viscosity determinationt U(ν)=(0.0036–0.0049) mm²·s⁻¹ (0.95 level of confidence)*

Obtained data on the kinematic viscosity of the object of the study in the temperature range from 300 to 335 K were fitted by the equation:

$$\lg(\lg(\nu+a))=b-c\cdot\lg(T). \quad (4)$$

The experimental data on the kinematic viscosity and relative deviations for the kinematic viscosity of R141b/Surf. and R141b/Surf./TiO_2 from the kinematic viscosity of pure R141b are shown in Fig. 3.

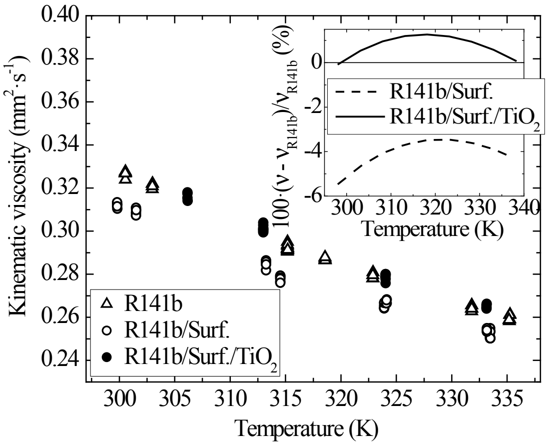

Fig. 3. Temperature dependences of the kinematic viscosity of the objects of the study and deviations $100\cdot(\nu-\nu_{R141b})/\nu_{R141b}$ of the kinematic viscosity of R141b/Surf. or R141b/Surf./TiO_2 from the kinematic viscosity of pure R141b

The experimental data of the thermal conductivity λ of the object of the study are shown in Table 4.

Table 4

Experimental values of the thermal conductivity λ for the objects of the study along the liquid-saturation line*

R141b		R141b/Surf.		R141b/Surf./TiO_2	
T (K)	λ ($W\cdot m^{-1}\cdot K^{-1}$)	T (K)	λ ($W\cdot m^{-1}\cdot K^{-1}$)	T (K)	λ ($W\cdot m^{-1}\cdot K^{-1}$)
294.19	0.09511	293.81	0.09538	293.77	0.09642
315.16	0.08906	304.83	0.09151	314.93	0.08964
324.76	0.08617	327.71	0.08498	334.48	0.08365
347.22	0.07936	347.05	0.07950	348.26	0.07993

*Note: * – standard uncertainty of temperature mesurement u(T)=0.1 K. Expanded uncertainty of thermal conductivity mesurement U(λ)= =(0.0011 – 0.0014) $W\cdot m^{-1}\cdot K^{-1}$ (0.95 level of confidence)*

Obtained data on the thermal conductivity of the object of the study in the temperature range from 293 to 348 K were fitted by the equation:

$$\lambda=a+b\cdot T. \quad (5)$$

The experimental data on the thermal conductivity and relative deviations for the thermal conductivity of R141b/Surf. and R141b/Surf./TiO_2 from the thermal conductivity of pure R141b are shown in Fig. 4.

The experimental data of the specific isobaric heat capacity C_p of the objects of the study are shown in Table 5.

Obtained data on the specific isobaric heat capacity of the object of the study in the temperature range from 261 to 334 K were fitted by the equation:

$$C_P=a+b\cdot T^2+c\cdot T^4. \quad (6)$$

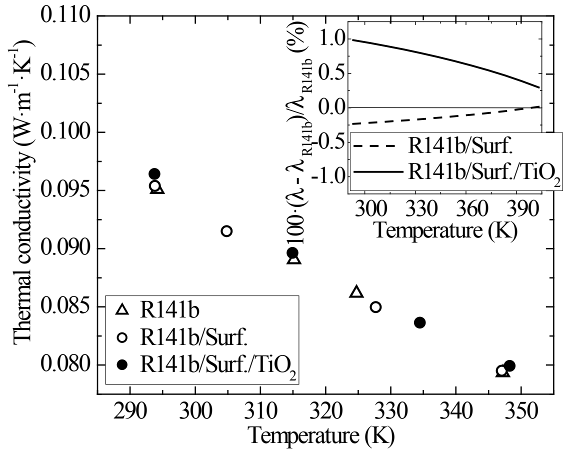

Fig. 4. Temperature dependences of the thermal conductivity of the objects of the study and deviations $100\cdot(\lambda-\lambda_{R141b})/\lambda_{R141b}$ of the thermal conductivity of R141b/Surf. or R141b/Surf./TiO_2 from the thermal conductivity of pure R141b

Table 5

Experimental values of the specific isobaric heat capacity C_p for the objects of the study*

R141b		R141b/Surf.		R141b/Surf./TiO_2	
T (K)	C_p ($J\cdot kg^{-1}\cdot K^{-1}$)	T (K)	C_p ($J\cdot kg^{-1}\cdot K^{-1}$)	T (K)	C_p ($J\cdot kg^{-1}\cdot K^{-1}$)
261.38	1116.4	280.34	1128.0	277.44	1107.0
265.46	1119.5	284.40	1132.1	281.31	1108.8
269.30	1121.4	288.45	1143.6	285.41	1113.3
273.35	1121.8	292.34	1146.4	289.47	1118.4
277.40	1123.0	296.45	1151.6	293.29	1120.6
281.43	1127.0	300.40	1167.2	297.32	1128.9
285.44	1131.3	304.44	1173.9	301.31	1135.8
289.43	1135.8	308.34	1182.5	305.40	1144.0
293.40	1140.0	312.43	1188.5	309.45	1154.4
296.37	1147.7	316.39	1191.3	313.36	1162.3
300.40	1154.6	320.33	1197.5	317.35	1166.1
304.35	1163.2	324.35	1203.5	321.32	1177.9
308.38	1169.4	327.70	1219.0	325.38	1185.1
312.39	1177.7	–	–	329.40	1191.0
316.38	1185.9	–	–	332.85	1193.8
320.39	1191.3	–	–	–	–
324.38	1196.7	–	–	–	–
328.33	1206.1	–	–	–	–
332.36	1213.0	–	–	–	–
334.42	1217.7	–	–	–	–

*Note: * – Standard uncertainty of temperature measurement u(T)=0.1 K. Expanded uncertainty of specific isobaric heat capacity measurement $U(C_p)$=(8.7–9.8) $J\cdot kg^{-1}\cdot K^{-1}$ (0.95 level of confidence).*

The experimental data on the specific isobaric heat capacity and relative deviations for the specific isobaric heat capacity of R141b/Surf. and R141b/Surf./TiO_2 from the specific isobaric heat capacity of pure R141b are shown in Fig. 5.

The coefficients of equations (2)–(6) and fitting standard errors of experimental results are presented in Table 6.

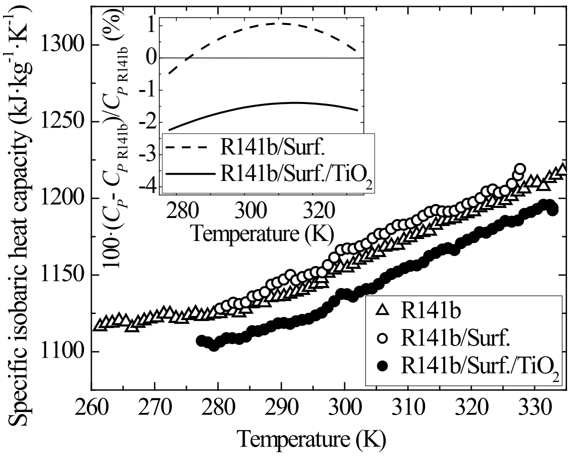

Fig. 5. Temperature dependences of the specific isobaric heat capacity of the objects of the study and deviations $100\cdot\left(C_p - C_{p\,R141b}\right)/C_{p\,R141b}$ of the specific isobaric heat capacity of R141b/Surf. or R141b/Surf./TiO_2 from the specific isobaric heat capacity of pure R141b

Table 6

Coefficients of Eqs. (2)–(6) for the calculation of the thermophysical properties of the R141b refrigerant, R141b/Span-80 solution and R141b/Span-80/TiO_2 nanoparticles nanofluid along the liquid-saturation line

Object of study	R141b	R141b/Surf.	R141b/Surf./TiO_2
Density ρ ($kg\cdot m^{-3}$)			
a	1726.2	1737.8	1730.6
b	–1.5710	–1.6560	–1.5931
c	$-35.606\cdot10^{-5}$	$-19.700\cdot10^{-5}$	$-32.242\cdot10^{-5}$
Fitting standard error ($kg\cdot m^{-3}$)	0.2403	0.0976	0.0415
Surface tension σ ($mN\cdot m^{-1}$)			
a	53.450	53.775	53.288
b	–0.11698	–0.11795	–0.11663
Fitting standard error ($mN\cdot m^{-1}$)	0.0455	0.0356	0.0876
Kinematic viscosity ν ($mm^2\cdot s^{-1}$)			
a	0.83490	1.12876	1.02431
b	10.2758	2.0751	3.2907
c	4.62609	1.16083	1.68443
Fitting standard error ($mm^2\cdot s^{-1}$)	0.00189	0.00301	0.00211
Thermal conductivity λ ($W\cdot m^{-1}\cdot K^{-1}$)			
a	0.18264	0.18180	0.18558
b	$-29.724\cdot10^{-5}$	$-29.514\cdot10^{-5}$	$-30.408\cdot10^{-5}$
Fitting standard error ($W\cdot m^{-1}\cdot K^{-1}$)	0.000114	0.000330	0.000292
Specific heat capacity C_p ($J\cdot kg^{-1}\cdot K^{-1}$)			
a	1167.80	731.53	956.22
b	$-28.064\cdot10^{-4}$	$64.973\cdot10^{-4}$	$11.741\cdot10^{-4}$
c	$29.421\cdot10^{-9}$	$-18.806\cdot10^{-9}$	$9.1252\cdot10^{-9}$
Fitting standard error ($J\cdot kg^{-1}\cdot K^{-1}$)	3.35	2.53	2.52

Comparison of the obtained values of fitting standard errors with the uncertainties of the experimental data confirms the adequacy of the proposed fitted dependencies.

6. Discussion of the results of studies on the influence of the additives of TiO_2 nanoparticles and Span-80 surfactant on the thermophysical properties of the R141b refrigerant

As it was shown in the performed experimental study, the effect of the Span-80 surfactant and TiO_2 nanoparticles on the density of R141b was within the measurement uncertainties. It is necessary to state a common for all samples tendency to increase the density of the liquid with the addition of even small amounts of surfactants and nanoparticles. The obtained results are reliable because density measurements were performed on exactly the same experimental setup for all objects of the study. In this case, the deviations of the experimental data and the data obtained by fitting equations (fitting standard errors in Table 6) are quite low.

As we can see from Fig. 2, the additives of the Span-80 surfactant to the R141b refrigerant practically had not provided the variation in surface tension (the effect does not exceed the measurement uncertainty). The addition of TiO_2 nanoparticles with the Span-80 surfactant to R141b had provided a slight decrease in the surface tension in the entire temperature range of the experiment (up to 0.3 % at high temperatures). These results are in conformity with the results of the study of the nanoparticles influence on the surface tension of refrigerants and solutions with oils presented in [23]. The obtained effect can be explained by an increase in the concentration of nanoparticles in the surface layer of the liquid phase. This fact contributes to a decrease in the surface energy and, consequently, to a decrease in the surface tension. It should be emphasized that the decrease in the working fluid surface tension can contribute to the enhancement of the heat transfer process during boiling.

As we can see from the results of the study (Fig. 3), the Span-80 surfactant additives in the R141b refrigerant contribute to the major decrease in viscosity – (3.5…5) %. This effect is very significant since the surfactant concentration in the R141b refrigerant is low. But the presence of both the Span-80 surfactant and TiO_2 nanoparticles in the refrigerant leads to an increase in viscosity – up to 1 %. The increase in viscosity is one of the negative effects that limit the nanofluids introduction into the industry. The presence of the surfactant contributes to providing only a slight increase in viscosity for the studied system (R141b/surfactant Span-80/TiO_2). In this case, the increase in energy consumption for the circulation of the working fluid will be insignificant.

As we can see from the experimental results presented in Fig. 4, the additives of the Span-80 surfactant in the R141b refrigerant do not contribute to the significant influence on the thermal conductivity (effect does not exceed 0.25 %). However, the addition of TiO_2 nanoparticles contributes to the increase (from 0.3 to 1 %) in the thermal conductivity of the R141b refrigerant. As is well known, the thermal conductivity enhanced is the main factor that contributed to the growth of interest in the application of nanofluids as coolants and working fluids of refrigeration systems. Recently, the observed in a number of studies effects of enhancement in the thermal conductivity of base fluids containing nanoparticle additives [5, 7, 27] have been doubted. The obtained effects were attributed to the incorrect implementation of the experimental measurements [27]. However, it should be emphasized that the experimental data (Table 4) were obtained with the correct estimation of the noise effects and with variations in temperature differences in the studied sample with using exactly the same measuring cell for all objects of the study. The obtained by adding a small amount of nanoparticles (0.1 wt. %) effect of the thermal conductivity increase can be considered as significant. The observed increase in the refrigerant thermal conductivity will contribute to the intensification of heat transfer processes in the equipment of refrigeration systems.

As follows from the results of the study (Fig. 5), the addition of the Span-80 surfactant to the R141b refrigerant does not have a significant effect on the specific isobaric heat capacity. The addition of TiO_2 nanoparticles leads to a significant decrease in the specific isobaric heat capacity of the refrigerant – by (1.5 ... 2) %. The obtained result cannot be explained only by the lower value of the specific heat capacity of the nanoparticles material. It should be noted that the additivity rule is often used by various authors [7] when determining the heat capacity of nanofluids. The specific heat capacity for the studied thermodynamic system calculated by the additive rule had the values exceeding the obtained experimental data for the nanofluid.

The presence of a structured layer of molecules of the base fluid (or surfactant molecules) on the surface of nanoparticles can explain the significant decrease in specific heat capacity and increase in thermal conductivity of nanofluids [15–17]. The results of the experimental studies confirm the requirement to develop the methods for predicting the thermophysical properties of nanofluids. These methods can be based on taking into account the presence of a structured phase consisting of the base fluid or surfactant molecules on the surface of nanoparticles. This phase has properties different from the properties of the base fluid or surfactant in the bulk fluid. To solve this task, additional measurements in a wider range of temperatures and concentrations of nanoparticles (including finding the optimal concentration of nanoparticles) should be performed.

The obtained data on the thermophysical properties of the studied system can also be used for developing models of predicting the heat transfer coefficients during nanofluids boiling.

It should be noted that both the nanoparticles concentration and the technology for preparing nanofluids are significantly influenced by the thermophysical properties of nanofluids and heat transfer processes. Therefore, the results obtained in this work can be extended to similar systems only if the identical preparing technology will be used.

7. Conclusions

1. The choice of a model system based on the R141b halogenated hydrocarbon refrigerant with adding TiO_2 metal oxide nanoparticles has been justified.

The nanorefrigerant preparation has not been difficult because R141b has the low vapor pressure at ambient temperatures. In addition, the R141b refrigerant is in thermodynamic similarity with halogenated hydrocarbons refrigerants, which have been widely used in the refrigerant industry. TiO_2 nanoparticles are the industrial product with low cost and they are often considered by researchers as a promising additive to halogenated hydrocarbons refrigerants.

It was found that the R141b/TiO_2 nanoparticles system remains colloidally stable only in the presence of a surfactant. Among surfactants of various nature, the best stability of this system has been provided by the Span-80 non-ionic surfactant. The satisfactory stability of the R141b/TiO_2 nanoparticles system in the presence of Span-80 CAS No.1338-43-8 (Sigma-Aldrich) has been confirmed ~~at~~ in the studies within this work. Thus, as a surfactant for the R141b/TiO_2 nanoparticles nanofluid, the Span-80 surfactant at a concentration 0.1 wt. % in the solution can be recommended to use.

2. The experimental study of density, viscosity, surface tension, thermal conductivity and specific isobaric heat capacity for samples: R141b, R141b/Span-80 surfactant and R141b/Span-80 surfactant/TiO_2 nanoparticles has been performed.

The thermophysical properties of the objects of the study have been performed along the liquid-saturation line in the t~~h~~emperature ranges of (273...293) K for the density, (293...343) K for the surface tension, (300...335) K for the kinematic viscosity, (293...348) K for the thermal conductivity and (261...334) K for the specific isobaric heat capacity.

3. The analysis of the influence of the Span-80 surfactant and TiO_2 nanoparticles on the density, viscosity, surface tension, thermal conductivity and specific isobaric heat capacity of the R141b refrigerant has been performed with using the obtained experimental data.

It was shown that the effect of surfactants and TiO_2 nanoparticles on the density of the R141b refrigerant was insignificant and within the uncertainty of the experimental data (up to 0.08 %). Additions of both the surfactants and TiO_2 nanoparticles contributed to a decrease in the surface tension of R141b by up to 0.3 % in comparison with pure R141b. Additives of both the surfactants and TiO_2 nanoparticles in R141b contributed to an increase in viscosity of (0.8...1.0) %, and additives of surfactants led to a significant decrease in viscosity – by (3.5...5.0) % compared to the viscosity of pure R141b. It was shown that surfactant additives in R141b did not significantly influence the thermal conductivity (the effect did not exceed 0.25 %), and additions of both the surfactants and TiO_2 nanoparticles lead to an increase in the thermal conductivity of the refrigerant by (0.3...1) %. A decrease of the specific isobaric heat capacity by (1.5...2.0) % was observed by adding the surfactants and TiO_2 nanoparticles to R141b. The slight increase in the specific isobaric heat capacity by adding the surfactants to R141b was observed (up to 1.0 %).

It was concluded that the influence of the addition of nanoparticles and surfactants on the thermophysical properties of the R141b refrigerant is ambiguous and unpredictable. The results of experimental studies on the effect of nanoparticles on the thermophysical properties of a refrigerant confirm the importance of developing methods for predicting these properties. This method can be based on taking into account the presence of a structured phase of the base fluid or surfactant molecules on the surface of nanoparticles.

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METHOD FOR DETERMINING THE THICKNESS OF A BINDER LAYER AT ITS NON-UNIFORM MASS TRANSFER INSIDE THE CHANNEL OF A HONEYCOMB FILLER MADE FROM POLYMERIC PAPER

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Сендвічеві панелі зі стільниковим заповнювачем на основі полімерного паперу «Nomex» широко використовуються у відповідальних конструкціях різноманітного призначення. В процесі виробництва таких панелей деякі чинники технологічного процесу, такі як нанос зв'язуючого на стільниковий заповнювач, температурні режими сушки і полімеризації нанесеного шару, найбільш істотно впливають на фізико-механічні характеристики готових виробів. Специфічним фактором виробництва стільникового заповнювача з полімерного паперу є його багатостадійне просочення складом апретуючого, а потім зв'язуючого на заключних операціях з наступним сушінням і термообробкою стільникових блоків. В результаті цих операцій має місце нерівномірний тепло- і масоперенос (міграція) зв'язуючого від центральної площини панелі до периферійних торцевих її зон. Досліджено закономірності цього нерівномірного тепло- і масопереносу зв'язуючого уздовж довжини стільникового каналу. Показано, що ці явища обумовлені гідродинамічним рухом зв'язуючого, що викликані градієнтами температури, його густини та коефіцієнта поверхневого натяжіння. На основі цього розроблено метод визначення товщини шару зв'язуючого уздовж каналів стільників при відомих (заданих) законах зміни густини та поверхневого натяжіння уздовж довжини чарунки стільникового заповнювача. Метод дозволяє технологічними засобами знизити нерівномірність масопереносу, забезпечивши потрібний допуск на фізико-механічні характеристики стільникових заповнювачів з полімерного паперу. Розглянута задача масопереносу дозволила глибше розкрити механізми формування нерівномірного шару просочення на етапах процесу сушіння при виробництві стільникового заповнювача з полімерного паперу. Використовуючи отримані механізми і технологічні можливості регулювання характеристик зв'язуючих, можливо поліпшити рівномірність товщини шару уздовж каналів стільників до значень, які забезпечать потрібний допуск на фізико-механічні характеристики стільникового заповнювача

Ключові слова: стільниковий заповнювач, фізико-механічні характеристики, полімерний папір, нерівномірний тепло- і масоперенос, зв'язуюче, гідродинамічний рух, рівномірність просочення

1. Introduction

Extension of the scope of application of sandwich structures with a honeycomb filler in various areas of technology in some cases became possible solely using the honeycombs based on polymer paper "Nomex". Such structures have several unique features: lightness at a high level of mechanical characteristics, good heat and sound insulating characteristics, high fatigue resistance and shock suppression [1, 2].

These properties led to their widespread use in important structures for aircraft interiors, for heat and noise insulation of the underbody space of carrier rockets, as well as in building structures [3, 4].

In the process of production of such structures, some factors of the technological process significantly affect phys-