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Thermodynamic and Transport Properties of Some Alternative Ozone-Safe Refrigerants for Industrial Refrigeration Equipment: Study in Belarus and Ukraine[1]

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The study of several hydrofluorocarbons (HFC) and fluorocarbons (FC) and their binary mixtures that have no ozone-depleting ability is being carried out in the framework of Belarus National Program. The fluids include HFCs R134a, R152a, R125, and R32, and FC R218. The following properties are being investigated: (1) phase equilibrium parameters including the boiling and condensing curve and critical point, thermophysical properties at these parameters, and heat of evaporation; (2) isobaric and isochoric heat capacity, ethalpy, and entropy in the gas and liquid state; (3) speed of sound, thermal conductivity, viscosity, and density in the gas and liquid state; (4) dielectric properties and surface tension; (5) behavior of combined construction materials inside the refrigerant medium; and (6) solubility in compressor oils and other technological characteristics. The series of results obtained by authors during the period 1990–1993 is presented.

KEY WORDS: phase transition; refrigerants; transport properties; thermodynamic properties.

1. INTRODUCTION

In different countries including the Commonwealth of Independent States (CIS), an active search for substitutes to the CFCs is being conducted. The

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technologies for their synthesis are also being developed. The authors of this paper represent research institutes where several hydrofluorocarbons (HFCs) and fluorocarbons (FCs) that do not deplete ozone are under study. These include HFCs R134a (CH_2FCF_3), R152a (CH_3CHF_2), R125 (CHF_2CF_3), and R32 (CH_2F_2), FC R218 ($CF_3CF_2CF_3$), and their mixtures. The results obtained by the authors during 1990–1993 are discussed.

2. METHOD AND APPARATUS

2.1. Phase Equilibrium Parameters and Density

A variable-volume apparatus was used to measure phase equilibrium parameters [1]. The experimental cell consists of a cylindrical tube of 300-mm length made of molybdenum glass. The internal diameter of the upper part of the cell (one-third of its length) is 3.4 mm, and that of the lower part 9.6 mm. Mercury is used as the medium to change the volume of cell and to transmit the pressure from the investigated fluid to a piston manometer with oil as a working liquid. To reduce the time needed to reach thermodynamic equilibrium the cell is supplied with a magnetic mixter. The height of mercury is measured visually with a cathetometer to determine the volume (V) of investigated fluid. The relation between the mercury meniscus location and the volume of fluid is determined by calibration. Corrections to this dependence take into account meniscus curvature, capillary depression, thermal extension, and deformtion of the cell volume with pressure.

The critical state is determined by observing the appearance and disappearance of the meniscus of the fluid contained in the cell.

The experimental cell is placed in an isolated ethanol bath controlled with a platinum resistance thermometer. Temperature variations within the cell do not exceed 0.005 K during the measurement period.

The uncertainty of phase equilibrium parameters measured is estimated to be 0.02 K for temperature (T) and 0.15% for most of the pressure values (P) obtained. The apparatus described above was also used to measure the density (ρ) of refrigerants in the gas and liquid phases with errors of 0.35 and 0.3%, respectively.

2.2. Thermal Conductivity

The static coaxial cylinder method for measuring the thermal conductivity (λ) of refrigerants was implemented. The apparatus was operated in a relative mode. Standard samples of helium, argon, and xenon were used for the gas calibration, and carbon dioxide and toluene were used for the liquid calibration to determine the device constants. The apparatus is presented in detail in Ref. 2.

The experimental cell consists of two concentric cylinders of about 300-mm length, separated by a gap of 0.17 mm. A calorimetric heater of 4 W is placed inside the smaller cylinder of about 11.7 mm in diameter. The temperature difference between cylinders is measured with a six-junction differential thermocouple. Pressure is measured with a standard Bourdon tube gauge with an accuracy of 0.4%. The cell is placed in a massive copper thermostat with a four-sectional heater to provide isothermal conditions along the cell. The temperature variation while measuring does not exceed 0.01 K. We estimate the uncertainty of thermal conductivity measurements to be 3.5% as a maximum.

2.3. Viscosity

To measure the viscosity (μ) of refrigerants we applied the falling-cylinder mthod. The method and apparatus have been described in detail in previous publications [2, 3]. A hollow stainless-steel cylinder of 105-mm length falls through the fluid investigated contained within a calibrated tube about 1 m in height and about 30 min in diameter. The cylinder is turned inside the tube around its axis by means of tangential holes made in its body. The cavity of the tube is connected to a standard Bourdon tube gauge for pressure measurement with an uncertainty of 0.4%. The apparatus is placed in a liquid bath with a temperature variation of less than 0.005 K. The temperature is measured with a platinum resistance thermometer with a standard error of 0.02 K. The device constants were determined by calibration with *n*-hexane as a standard sample. It was estimated that the uncertainty in measurement of viscosity in the liquid phase was less than 2.8%.

2.4. Speed of Sound

Speed of sound (ω) in the liquid phase of refrigerants is measured by means of the impulse method at a frequency of 2.1 MHz. The device, which consists of an acoustic cell placed inside a liquid bath and which we recently described in detail in Ref. 4, was examined and tested using water and toluene as the standard samples. The dispersion of measured data around those of the standard did not exceed 0.07%. The pressure and temperature in the acoustic cell were measured with a tense-resistance detector and platinum resistance thermometer, and uncertainty did not exceed 0.1%

and 0.02 K, respectively. The values of speed of sound in the liquid refrigerants can be measured with a standard deviation of not more than 0.2%.

3. EXPERIMENTAL DATA

3.1. Samples of Refrigerants Used

The samples of refrigerants studied were synthesized by the Research & Production Association, State Institute of Applied Chemistry, St. Petersburg, Russia, and kindly provided to the authors. As certified by the supplier, the refrigerants have purities as given in Table I. Right before the experiments the refrigerants are degassed by means of freezing and pumping. The samples of refrigerant binary solutions were prepared in the laboratory by weighing. The composition of each solution is determined with a standard error of about 0.001 weight fraction.

3.2. Phase Equilibrium Diagram and *PVT* Data

The pressure–composition and density–composition relations of investigated fluids at dew and bubble points along the saturated vapor/liquid curve were measured in a temperature range from 270 K to the critical point. Three solutions, i.e., R152a + R134a, R218 + R134a, and R152a + R218, and their components were studied. The experiment was carried out as a constant temperature, while the pressure in the cell was measured as a function of the cell volume. The P–ρ parameters were fixed and isotherms $P(\rho)_{T=\text{const}}$ were plotted for each fluid.

The values of pressure P'' and density ρ'' at the dew point, and P' and ρ' at the bubble point were located by the change in the slope of isotherms while crossing the phase transitions. About 100 experimental points in the P–ρ-diagram were determined [5] for the above solutions. The difficulty of

Table I. Purity of Samples (Weight %)

Component	Refrigerant				
	R32	R134a	R152a	R218	R125
Main fluid	99.99	99.91	99.98	99.997	99.90
Water	0.0005		0.006	0.003	
HCFCs other than the main fluid	0.008 (R21, R143a)	0.09 (R133)	0.01		0.1 (R116)
Nonvolatile remainders	0.0023		0.004		

localizing the dew point on the experimental isotherms is related to the rounding of the first-order phase transition [6] due to the effect of physical adsorption of gas inside the surface capillary system on approaching condensation.

The measurements in the vicinity of the critical point were implemented for three compositions of each solution investigated. Parameters of critical-point T_C, P_C, and ρ_C for R152a + R134a and R218 + R134a were determined as a result of measurements (Table II).

In addition to a series of P–ρ measurements close to the phase equilibrium state the study of the *PVT* relation was carried out along isotherms for the azeotropic composition of R218 + R134a ($X_{\mathrm{R134a}} = 0.405$; X_{R} is the molar fraction of refrigerant R in the solution) and for two compositions of R152a + R134a ($X_{\mathrm{R134a}} = 0.353$ and 0.594). More than 300 experimental values were measured [7]. The range of conditions investigated was 0.2–5.6 MPa, 14–1430 $\mathrm{kg \cdot m^{-3}}$, and 270–365 K.

3.3. Thermal Conductivity and Viscosity

Thermal conductivity values were determined along isotherms in the range of 290–405 K and 0.2–20 MPa in the liquid and gas phases. The following refrigerants and mixtures were studied: R32, R125, R134a, R152a, R218, R218 + R152a ($X_{\mathrm{R152a}} = 0.1234$), and R152a + R134a ($X_{\mathrm{R134a}} = 0.7995$). About 540 and 470 experimental values were obtained in the liquid and gas phases, respectively [8].

More than 180 experimental data points on the viscosity of liquid R134a, R152a, and R32 were measured [9, 10]. The pressure ranged from saturated liquid pressure up to 16 MPa. The lowest temperature measured in the liquid phase was about 290 K, and the maximum value was about

Table II. Experimental Values of Critical Point Parameters of Refrigerant Mixtures

Mixture	X_{R134a}	T_c (K)	P_c (MPa)	ρ_c ($\mathrm{kg \cdot m^{-3}}$)
R218 + R134a	0	345.05	2.671	628.0
	0.196	343.1	2.8312	610.0
	0.358	344.07	3.0150	599.6
	0.419	345.43	3.1007	581.6
	1.0	374.25	4.059	509.1
R152a + R134a	0	386.57	4.517	365.0
	0.182	384.75	4.4261	392.9
	0.496	381.79	4.2739	434.8
	0.750	378.06	4.1585	468.9
	1.0	374.25	4.059	509.1

360, 345, and 320 K for R134a, R152a, and R32, respectively. The values of viscosity at the coexistence–curve parameters were masured between 170 and 220 K.

3.4. Speed of Sound

The experimental data on speed of sound in the liquid phase of refrigerants such as R134a, R152a, and R32 were obtained at pressures ranging from the saturated liquid pressure up to 20 MPa About 210 experimental values of speed of sound in liquid R134a and 120 of those in liquid R32 were received at temperatures of 200 to 360 K [11–13]. The range of temperature covered with 54 experimental values of speed of sound in liquid R152a constitutes 285–360 K [2].

3.5. Oil–Refrigerant Solubility Diagram

We are investigating the suitability of synthetic oils of two types for the refrigerant mixture R152a + R134a. One of these oils, XF-22c-16, of mineral type, is available in the CIS and is manufactured on a large scale as an oil adapted to R22. Another tested oil is adapted to R134a, namely, XF-134. We studied the oil–refrigerant solubility diagram for nine compositions of refrigerant solution with the weight fraction of oil ranging from 0.05 to 0.45. We should emphasize that this study is in the beginning stage.

4. RESULT AND DISCUSSION

4.1. Transport Properties

4.1.1. Viscosity

Our experimental data on viscosity of R134a are in good agreement with data obtained in [14] using capillary method. We approximated both sets of data in the temperature range 257–360 K and at a pressure up to 15 MPa by the following model:

$$\ln(\mu/\mu_{0T}) = \sum_{j=0}^{4} K_{1j}\omega t^{-j} + K_{20}\omega^2 \tag{1}$$

where $t = T/T_C$, $\omega = \rho/\rho_C$, and μ_{0T} is the viscosity of low-density gas that is defined as

$$\ln \mu_{0T} = \ln \mu_{T_c} + 0.9\theta(t)$$

$$\theta(t) = \ln t + 0.5 \left(\frac{t-1}{t}\right)^2 \left(\frac{t-1}{t} - \ln t\right)\left[1 - 0.1 \left(\frac{t-1}{t}\right)^4\right]$$

where μ_{0T_C} is calculated according to Ref. 15, and it is the viscosity of fluid in the state of low-density gas at the critical-point temperature T_C; here and below T is in K, P in MPa, and μ in mPa · s. Our 42 values for the viscosity of liquid R152a and 62 values for the viscosity of liquid R32 can be correlated in the range of parameters of 290–345 K and at pressures up to 16 MPa by the same Eq. (1).

For three refrigerants investigated the viscosity on the saturation curve is given by

$$\mu_S = \mu_C \exp\left(K_0\tau^{1/3} + \sum_{i=1}^{4} K_i\tau^i + M\tau^{11}\right) \tag{2}$$

where the regular part of the viscosity μ_C at the critical point is a calculated constant and $\tau = 1 - t$.

Fitting constants of Eqs. (1) and (2) are shown in Table III. A root-mean-square deviation (σ) of 0.9% in experimental and calculated values was obtained in the ranges of approximation using these models. Equation (2) for viscosity on the saturated liquid curve can be used in the region of 250–360 K. Our data are in good agreement with values of viscosity of saturated liquid recently published in Ref. 16. The differences do not exceed 4% (see Fig. 1 for R134a).

Table III. Constants in Eqs. (1) and (2)

Constant	T32	R134a	R152a
K_{10}	1.07709	0.187119	0.108920
K_{11}	−0.54366	0.013503	0.514127
K_{12}	0.42106	0.486772	0.118678
K_{13}	−0.45576	0.290849	−0.31820
K_{14}	0.27990	−0.29070	0.122203
K_{20}	0.09837	0.157395	0.213063
T_C (K)	351.26	374.25	386.44
ρ_C (kg · m^{-3})	434	511	368
K_0	3.4351	2.6313	2.4511
K_1	−8.7262	0.5584	−0.7855
K_2	58.3567	2.780	14.322
K_3	—	3.8379	−26.412
	168.365		
K_4	189.628	0	7.7502
M	0	313.7	0
μ_C (mPa · s)	0.030	0.032	0.028
μ_{0T_C} (mPa · s)	0.0134	0.0146	0.0125

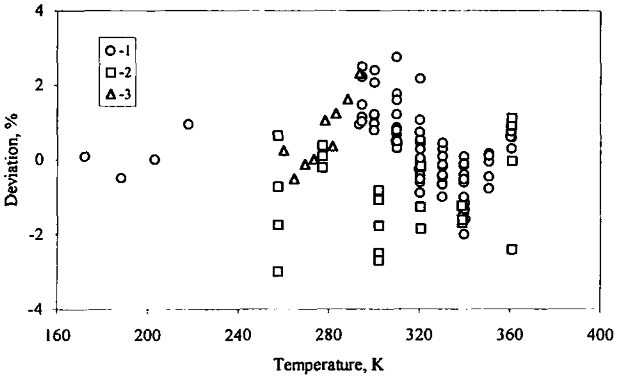

Fig. 1. Deviation of different data on the viscosity of liquid R134a calculated by Eq. (1): (1) results of the present measurements; (2) data of Ref. 14; (3) data of Ref. 16.

4.1.2. Thermal Conductivity

While analyzing the experimental data on thermal conductivity of refrigerant R134a and R152a, one can easily discover that disagreement exists among the data obtained at different spacings. This indicates that (i) refrigerants are only partially transparent to infrared rays and (ii) our data obtained at a clearance of 0.17 mm reflect the molecular mechanism of conductivity rather than other known data [10, 14] where radial clearance amounts to about 1.0 mm. Our data show a good agreement with data [17–19] obtained at an effective gap/radial clearance less than 0.20 mm, i.e., close to ours (see Fig. 2).

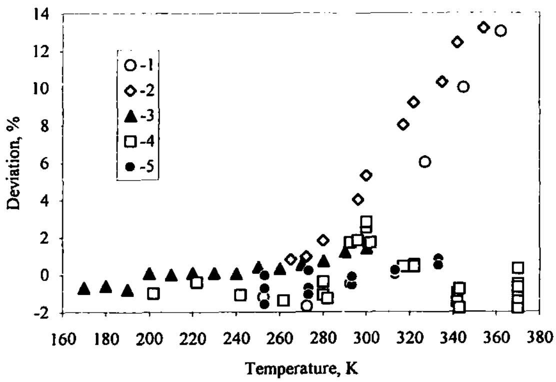

Fig. 2. Deviation of different experimental data on the thermal conductivity of R134a correlated by Eq. (4): (1) data of Ref. 14; (2) data of Ref. 10; (3) data of Ref. 19; (4) data of Ref. 17; (5) data of Ref. 18.

Table IV. Parameters of Eq. (4) for Calculation of Thermal Conductivity of Different Refrigerants in the Liquid Phase

Refrigerant	Coefficients a_{ij}						Temperature range (K)	Pressure range (MPa)	σ (%)
	a_{00}	10^4a_{01}	10^7a_{02}	10^3a_{10}	10^6a_{11}	10^5a_{20}			
R134a	0.24362	−6.3502	2.9621	−0.71998	5.4297	−1.22125	170...400	0.98...19.6	1.3
R152a	0.25443	−5.0073	−0.5627	−1.1949	7.6098	−2.3559	160...400	0.98...20.0	1.6
R218	0.21490	−8.3030	8.5995	−0.80567	6.7479	−3.0353	290...400	1.18...15.0	1.0
R32	0.35789	−4.9894	−10.0173	−9.2273	36.5032	−5.1003	275...405	1.67...12.0	1.1
R125	0.30046	−10.9307	9.2025	−4.3474	18.4259	−2.3503	295...405	1.67...11.0	1.2
R152a + R218	0.22632	−8.4148	8.1065	−2.1712	9.96575	0	290...400	1.08...10.0	0.7
R152a + R134a	0.28794	−8.8357	6.2803	−2.6013	10.9124	0	295...400	0.98...10.0	0.8

Table V. Parameters of Eq. (4) for Calculation of Thermal Conductivity of Different Refrigerants in the Gas Phase

Refrigerant	Coefficients a_{ij}						Temperature range (K)	Pressure range (MPa)	σ (%)
	$10^3 a_{00}$	$10^5 a_{01}$	$10^7 a_{02}$	$10^3 a_{10}$	$10^5 a_{11}$	$10^4 a_{20}$			
R134a	−3.56062	4.0801	0.5105	3.0601	−0.8897	4.9611	230...400	0.20...1.96	1.3
R152a	9.4858	−5.5508	2.2605	13.2186	−3.7082	8.8401	250...400	0.29...4.02	1.2
R218	4.9043	−0.96499	1.1785	7.0464	−1.8655	4.0253	295...400	0.20...3.68	1.2
R32	12.4704	−5.8822	1.9665	5.1568	−1.3051	2.3023	295...405	0.29...2.30	0.6
R125	5.9204	−1.3084	1.3047	4.5364	−1.2126	3.1198	295...405	0.29...2.45	0.5
R152a + R218	−2.7351	4.2226	0.4813	3.1206	−0.6357	0	295...405	0.20...1.05	0.7
R152a + R134a	14.3816	−7.7461	2.4293	10.496	−2.9635	7.15635	295...400	0.29...3.23	0.9

Table VI. Coefficients of Eqs. (5)–(7) for R134a, R152a, R218, and Their Mixtures

	Refrigerant						
					R152a + R134a		
Cofficient	R134a	R142a	R218	R218 + R134a, $X_{\mathrm{R134a}} = 0.405$	θ_0	θ_1	θ_2
α_R	7.4029	7.1920	7.4446	7.2721	7.2159	−0.2989	0.4996
b	6.2822	5.3553	6.8456	6.2812	5.3549	2.5416	−1.6205
B'	1.7198	1.7311	1.6623	1.6942	1.7296	0.02957	−0.0331
β'	0.3282	0.3263	0.3297	0.3234	0.32467	0.03181	−0.0270
B''	2.3256	2.2985	2.2785				
β''	0.3445	0.3273	0.3348				

We used a simple polynomial to correlate the data obtained in both the liquid and the gas phase. The values of thermal conductivity of the refrigerants investigated (R32, R125, R134a, R152a, R218) and of their mixtures (R218 + R152a and R152a + R134a) were described by the expression

$$\lambda = \sum_{i=0}^{2} \sum_{J=0}^{2} a_{ij} P^i T^j \tag{4}$$

where λ is in $\mathrm{W \cdot m^{-1} \cdot K^{-1}}$. The polynomial constants, the standard deviation, and the range of parameters where Eq. (4) is valid are presented in Tables IV and V.

4.2. Thermodynamic Properties

4.2.1. Phase Equilibrium Parameters: Critical Point

As a result of our investigation the parameters of the boiling and condensation curves for some compositions of refrigerants studied were obtained. To analyze these data, we used the following equations for the vapor pressure and coexistence curve of each component [20]:

$$\ln \frac{P_C}{P_S} = \alpha_R \ln \frac{T_C}{T} + b \left(\ln \frac{T_C}{T} \right)^{2.64} \tag{5}$$

$$\ln \frac{\rho'}{\rho_C} = B' \left(\ln \frac{T_C}{T} \right)^{\beta' \left[1 - 1.113 \frac{(\ln(T_C/T))^{0.4}}{\ln(\ln(T_C/T))} \right]} \tag{6}$$

$$\ln \frac{\rho_C}{\rho''} = B'' \left(\ln \frac{T_C}{T} \right)^{\beta'' \left[1 + 4.86 \frac{(\ln(T_C/T))^{0.6}}{\ln(\ln(T_C/T))} \right]} \tag{7}$$

The constants α_R, b, B', B'', β', and β'' in Eqs. (5)–(7) for R134a, R152a, and R218 are given in Table VI. The deviations of the experimental data do not exceed the measurement uncertainty in the temperature interval from boiling point at atmosphere pressure up to the critical point.

Experimental values of the critical parameters of refrigerant binary mixtures investigated (see Table II) are approximated with polynomials:

$$T_C = \sum_{i=0}^{2} t_i X^i_{\mathrm{R134a}}, \qquad P_C = \sum_{i=0}^{2} p_i X^i_{\mathrm{R134a}}, \qquad \rho_C = \sum_{i=0}^{1} r_i X^i_{\mathrm{R134a}} \tag{8}$$

where coefficients t_i, p_i, and r_i are given in Table VII for each refrigerant solution.

While analyzing our results we determined the R134a concentration that related to azeotropic composition of the mixture R218 + R134a: $X_{\mathrm{R134a}} = 0.405$. Within the temperature range of 220 K to critical value T_C, this azeotropic point does not change with temperature. Therefore we used the same model, i.e., Eqs. (5)–(7) presented above, to describe the phase equilibrium diagram of the mixture R218 + R134a. Constants for this mixture are also given in Table VI.

Our results of measurements of phase equilibrium parameters of the R152a + R134a mixture agree with the conclusion of Refs. 21 and 22: within experimental error this fluid should be considered as a quasi-azeotropic solution at temperatures above 240 K. This means that the investigated solution has the same parameters for the boiling and condensation curve. In the region below 240 K this mixture forms a minimum-pressure azeotrope. The phase equilibrium diagram of such a fluid can also be presented by Eqs. (5)–(7) if we propose each coefficient in the model to be presented by a polynomial form like $\Theta = \sum_{k=0}^{2} \theta_k X^k_{\mathrm{R134a}}$, where Θ means any coefficient in Eqs. (5)–(7), and constants θ_k are given in Table VI. The deviations of experimental results and calculated values are less than 0.4% (P') and 0.7% (ρ') in the temperature range investigated (270–353 K).

Table VII. Constants of Eq. (8)

	R218 + R134a			R152a + R134a		
i	t_i	p_i	r_i	t_i	p_i	r_i
0	345.06	2.6675	632.51	386.50	4.5189	364.24
1	−19.952	0.7463	−116.257	−7.548	−0.5279	145.730
2	49.141	0.6469	0	−4.722	0.0677	0

Table VIII. Coefficients in Eqs. (9) and (10)

R32: $T_C = 351.56$ K, $\sigma = 0.152\%$

i	n_i	A_i	A_{ij}		
			$j=1$	$j=2$	$j=3$
0	0.056	237.3352	903.771	10.85674	−3.971753
1	0.515	43.18362	32094.814	−946.5879	26.75663
2	1.0	2387.694	−43941.716	1329.687	−1.400574
3	1.5	−631.4327	—	—	—

R152a: $T_C = 861.41$ K, $\sigma = 0.10\%$

i	n_i	A_i	A_{ij}		
			$j=1$	$j=2$	$j=3$
0	0.056	100.0	9311.400	−32.787	—
1	0.515	560.0	21151.000	55.140	—
2	1.0	1311.3	—	—	—
3	1.5	−47.69	—	—	—

R134a: $T_C = 374.27$ K, $\sigma = 0.120\%$

i	n_i	A_i	A_{ij}		
			$j=1$	$j=2$	$j=3$
0	0.056	130.6534	5412.091	−3.421151	0.377037
1	0.515	−344.8112	16378.544	34.6088	−2.47812
2	1.0	901.53	−215.971	−238.224	—
3	1.5	282.921	—	—	—
4	0.621	890.269	—	—	—

4.2.2. Speed of Sound

Experimental values of speed of sound in liquid refrigerants investigated were correlated with the following model:

$$\omega = \left[\omega_S^2 + 10^3 \sum_{i=0}^{2} \sum_{j=1}^{3} A_{ij} \tau^i (P - P_S)^j \right]^{1/3} \qquad (9)$$

where the temperature function for the speed of sound at phase equilibrium parameters (ω_S) was chosen accoding to [3]

$$\omega_S = \sum_{i=0}^{4} A_i \tau^{n_i} \tag{10}$$

and $\tau = 1 - T/T_C$; the unit for speed is traditional—$\mathrm{m \cdot s^{-1}}$.

The coefficients of Eqs. (9) and (10) and deviation σ are given in Table VIII. The standard deviation in the range of approximation (225 to 360 K, 0.1 to 20 MPa) equals 0.08%. Data in the liquid phase are correlated up to a pressure value of 16 MPa. Critical parameters as well as those of the boiling curve for each refrigerant investigated were calculated as described in Section 4.2.1.

5. CONCLUSION

In the present work, applying the experimental methods and research equipment which were used in the past while studying the working fluids of different kind, we obtained extensive new data on thermophysical and thermodynamic properties of a series of ozone-safe refrigerants. This study will help in effective choice of refrigerant and will provide the data needed for the design and manufacture of apparatus and techniques for industrial refrigerators. Our study is ongoing. Many data obtained are still to be processed, such as heat capacity, density and *PVT* relation, and thermal coefficients. As a result of this research, to be finished by the end of 1995, we will introduce the refrigerants and will develop the manufacturing technology. Some proposed refrigerants will be tested in existing equipment during 1995.

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