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Correlation and Prediction of Dense Fluid Transport Coefficients. VII. Refrigerants[1]

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A recently developed scheme, based on considerations of hard-sphere theory, is used for the simultaneous correlation of the coefficients of viscosity and thermal conductivity for the refrigerants R11, R12, R22, R32, R124, R125, R134a, R141b, and R152a in excellent agreement with experiment, over extended temperature and pressure ranges. Values for the roughness factors and correlations for the characteristic volume are presented. The overall average absolute deviations of the experimental viscosity and thermal conductivity measurements from those calculated by the correlation are 2.1 and 2.3%, respectively, over a temperature range from 200 to about 10 K below the critical temperature and a pressure range from saturation to about 40 MPa. Since the proposed scheme is based on recent and accurate density values, a Tait-type equation was also employed to correlate successfully the density of the refrigerants. The overall average absolute deviation of the experimental density measurements from those calculated by the correlation is ±0.08%.

KEY WORDS: density; high pressure; refrigerants; Tait equation; thermal conductivity; viscosity.

1. INTRODUCTION

In a series of recent papers [1-6], a scheme was developed for the simultaneous correlation of viscosity, self-diffusion, and thermal conductivity data over a wide range of temperatures and pressures, by means of a consistent set of values for the characteristic volume V_0. It was shown that the scheme provides a satisfactory correlation of dense fluid n-alkane transport coefficient data [1, 4], and consequently, it can be used with

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confidence to predict transport coefficients for these compounds under other conditions of temperature and pressure. The method has also been applied to simple organic molecular liquids [2] and has been extended to *n*-alkane mixtures [3], aromatic hydrocarbons [5], and *n*-alcohols [6]. It has been demonstrated that this scheme can lead to predictions of transport coefficients for these systems, at pressures up to 600 MPa, with an uncertainty of ±5%.

The scheme is based on the assumption that tranport coefficients of dense fluids, expressed in terms of the reduced volume V/V_0, are directly proportional to values given by the hard-sphere theory [7]. The proportionality factor, described as roughness factor R, accounts for molecular roughness and departure from molecular sphericity [1, 4].

As has been shown, in this scheme, the density of the liquid is the important parameter, and not the temperature and pressure. To overcome this, a generalized Tait-type equation was employed to correlate the density of *n*-alkanes [8] and *n*-alcohols [6]. This, in conjunction with the aforementioned scheme, allowed the calculation of the transport properties of *n*-alkanes and their mixtures and of *n*-alcohols as a function of temperature and pressure, rather than density.

In this paper, the general applicability of the scheme is further tested by its application to refrigerants R11, R12, R22, R32, R124, R125, R134a, R141b, and R152a. Furthermore, prior to the application of the scheme, accurate experimental density data are employed to develop a Tait-type equation for the correlation of their densities.

2. DENSITY

In the case of refrigerants, the Tait equation employed was of the form

$$\frac{\rho-\rho_s}{\rho} = C \log\left[\frac{B+P}{B+P_s}\right] \tag{1}$$

where ρ is the liquid density at corresponding pressure P, and ρ_s is the saturated liquid density at the corresponding saturation pressure, P_s. Parameter C has generally been found to be a constant, while B, however, definitely varies with temperature. In the case of *n*-alkanes [8] and *n*-alcohols [6], C was found to be a constant exactly equal to 0.2000, and B a quadratic function of temperature.

The saturated liquid density, ρ_s, and the saturation pressure, P_s, for R22, R32, R124, R125, R134a, and R152a employed, were obtained from the following equations proposed by McLinden [9]:

$$\ln(P_s/P_c) = a_1\tau/(1-\tau) + a_2\tau + a_3\tau^{1.89} + a_4\tau^3 \tag{2}$$

Table I. Coefficients and Critical Constants of Eqs. (1)–(4)

	R11	R12	R22	R32	R124	R125	R134a	R141b	R152a
a_1	−8.565442	−8.773648	−8.985153	−10.052244	−10.034418	−11.674144	−8.798572	−6.849090	−4.133708
a_2	1.725568	2.075277	2.125014	2.844032	2.870485	4.524166	1.379055	−0.403888	−3.135972
a_3	4.243231	3.854291	3.693854	4.108620	4.232259	4.567844	3.587903	4.356486	0.945576
a_4	−0.498819	1.156449	0.906855	3.450397	1.619401	6.904302	−2.390161	−7.680980	−11.934945
d_1	1.904827	1.430517	2.191954	1.667251	2.064566	1.959463	1.986361	3.186797	2.456208
d_2	0.579227	2.700950	0.046237	2.720934	0.387932	0.847479	0.677576	−4.845030	−1.665948
d_3	−0.435098	−3.558323	0.207547	−4.374005	−0.405756	−0.840846	−0.455932	7.734827	3.933187
d_4	0.642662	2.178127	0.397319	3.272961	0.715925	0.797769	0.592428	−3.530228	−2.045507
b_1 (MPa)	−236.049	587.350	741.502	189.897	2313.078	355.008	112.730	2654.771	320.404
b_2 (MPa)	1719.495	−1688.760	−2169.693	−315.564	−7776.805	−814.594	15.243	−10682.37	−664.683
b_3 (MPa)	−2990.746	1669.069	2191.708	119.590	8789.964	599.599	−336.680	14723.193	398.631
b_4 (MPa)	1539.050	−572.555	−769.649	0	−3334.695	−143.734	204.768	−6891.199	−58.587
T_c (K)	471.15	384.95	369.30	351.56	395.65	339.40	374.21	477.26	386.44
P_c (MPa)	4.403	4.129	4.988	5.830	3.634	3.631	4.056	4.230	4.520
ρ_c ($kg \cdot m^{-3}$)	559	560	513	430	560	572	515	461	368

Table II. Comparison of Calculated Density Values with Experimental Values

Fluid	First author	Ref. No.	Temp. range (K)	Press. max. (MPa)	No. of data	Av. dev. (%)	Max. dev. (%)
R11	Blanke[a]	12	254–380	31	59	0.08	0.22
R12	Blanke[a]	12	198-366	31	56	0.10	0.35
	Handel	13	200-300	8	42	0.06	0.20
R22	Blanke[a]	12	197-325	30	78	0.07	0.30
	Defibaugh[a]	14	263-353	7	81	0.04	0.26
	Handel	13	200-300	8	18	0.03	0.09
R32	Defibaugh[a]	15	240-338	7	187	0.08	0.24
R124	Kubota[a]	16	283-374	35	49	0.15	0.40
R125	Defibaugh[a]	17	275-324	7	34	0.03	0.09
R134a	Tillner-Roth[a]	18	243-363	16	202	0.09	0.32
	Klomfar[a]	19	200-310	26	52	0.13	0.35
	Morrison	20	278-358	6	58	0.09	0.16
	Piao	21	331-358	12	8	0.09	0.24
R141b	Defibaugh[a]	11	278-370	7	68	0.06	0.39
R152a	Tillner-Roth[a]	18	243-374	16	221	0.08	0.32
	Blanke	12	220-373	30	55	0.15	0.60
				Total	1268	0.08	0.60

[a] Author upon whom the correlation is preferentially based.

and

$$\rho_s/\rho_c = 1 + d_1\tau^{0.355} + d_2\tau^{2/3} + d_3\tau + d_4\tau^{4/3} \tag{3}$$

where T_c, P_c, and ρ_c are the critical temperature, pressure, and density, $\tau = (1 - T_r)$ and T_r $(= T/T_c)$ is the reduced temperature. For R11 and R12 we have employed the saturation values given by Platzer et al. [10], while for R141b the values of Defibaugh et al. [11] were used; we fitted them in the above form for consistency purposes. The coefficients a_i, d_i, and the critical parameters employed are all shown in Table I.

Equations (1), (2), and (3) were used to examine the temperature dependence of B, after it had been found that, similarly to the n-alkanes and n-alcohols, C could be taken as a constant, with an optimized value of 0.1800. The examination of all available experimental density data led to the following dependence of B on reduced temperature:

$$B = \sum_{i=1}^{4} b_i T_r^{i-1} \tag{4}$$

The coefficients b_i are also shown in Table I. Hence, Eqs. (1)–(4) form a consistent set by which the liquid density of the refrigerants can be calculated. In Table II, the experimental measurements [11–21] considered in this work and the temperature ranges and maximum pressures are shown. There are many investigators that have reported density measurements. In this work, however, it was peferred to consider only investigations that (a) are recent (the last 4 years), so that high-purity samples were available, (b) employed "properly" calibrated instruments, and (c) quote an uncertainty better than ±0.1%. An exception was made in the case of the density of R124, where the only available measurements were those of Kubota et al. [16] performed in 1988. The following points ought to be clarified in relation to the temperature ranges and maximum pressures shown in Table II (a) to include low-temperature experimental measurements of the density, Eqs. (2) and (3) were extrapolated when necessary, and (b) the highest temperature employed was restricted to about 10 K below the critical temperature. In the same table the average absolute deviation as well as the maximum absolute deviation for every data set in each refrigerant, are also given. The average absolute deviation of all 1268 data points considered is 0.08%, while the maximum absolute deviation is 0.60%.

3. VISCOSITY AND THERMAL CONDUCTIVITY

According to the proposed scheme [1–6], it was found that the reduced coefficients for viscosity η^* and thermal conductivity λ^*, defined as

$$\eta^* = 6.035 \times 10^8 \left[\frac{1}{MRT}\right]^{1/2} \eta V^{2/3} \tag{5}$$

$$\lambda^* = 1.936 \times 10^7 \left[\frac{M}{RT}\right]^{1/2} \lambda V^{2/3} \tag{6}$$

are functions of the reduced molar volume $V_r = (V/V_0)$, where V_0 is a characteristic molar volume of the liquid, weakly dependent on temperature. In the above equations, M represents the molecular mass ($\mathrm{kg \cdot mol^{-1}}$) and R the universal gas constant ($\mathrm{J \cdot mol^{-1} \cdot K^{-1}}$), while the molar volume V, temperature T, viscosity η, and thermal conductivity are expressed in $m^3 \cdot \mathrm{mol^{-1}}$, K, Pa · s, and $\mathrm{W \cdot m^{-1} \cdot K^{-1}}$, respectively. According to this scheme, the aforementioned functions were found to be universal for all liquids and equal to [1]:

$$\log\left[\frac{\eta^*}{R_\eta}\right] = 1.0945 - 9.26324V_r^{-1} + 71.0385V_r^{-2} - 301.9012V_r^{-3} + 797.69V_r^{-4} - 1221.977V_r^{-5} + 987.5574V_r^{-6} - 319.4636V_r^{-7} \tag{7}$$

$$\log\left[\frac{\lambda^*}{R_\lambda}\right] = 1.0655 - 3.538V_r^{-1} + 12.120V_r^{-2} - 12.469V_r^{-3} + 4.562V_r^{-4} \tag{8}$$

where parameters R_η and R_λ account for deviations from the behavior of smooth hard spheres [1]. In the case of pure *n*-alkanes [1], aromatic hydrocarbons [5], and *n*-alcohols [6], experimental measurements were used to calculate the temperature dependence of the characteristic molar volumes and the values of parameters R_η and R_λ. For nonpolar compounds [1–5] these parameters were found to be constant characteristic only of the liquid and the property. In the case of the polar *n*-alcohols [6], however, these parameters were found to be weak functions of the temperature. It should also be pointed out here that the above analysis, in all previous work, included equivalent relations for the self-diffusion coefficient. Since in the case of refrigerants no measurements of the self-diffusion coefficient exist, these relations have not been included.

Accurate measurements of viscosity and thermal conductivity coefficients were used to calculate the aforementioned parameters. R_η was found to be a constant near unity depending only on the fluid, whereas parameter R_λ was found at low temperatures to be a constant or a very weak function of temperature but increased with temperature when approaching the critical temperature. The equations obtained for parameter R_λ and the characteristic molar volume, V_0, of each refrigerant as a function of the reduced temperature, T_r, are

$$R_\lambda = \sum_{j=1}^{4} \theta_j T_r^{j-1} \tag{9}$$

$$V_0 = \sum_{i=1}^{4} v_i T_r^{i-1} \tag{10}$$

The coefficients θ_j and the parameter R_η are shown in Table III, whereas the coefficients v_i are presented in Table IV. In the calculation of these coefficients and parameters the main problem was the uncertainty involved in the experimental measurements themselves, since, for example, the experimental viscosity measurements of these refrigerants show discrepancies of about ±5% [22]. Hence, Eqs. (1)–(10) form a consistent set by which the density, the viscosity and the thermal conductivity can be

Table III. Parameters R_η and Coefficients θ_i of Eq. (9)

Fluid	R_η	θ_1	θ_2	θ_3	θ_4
R11	1.00	1.53	0	0	0
R12	1.60	39.0315	−143.0474	181.7211	−75.8405
R22	1.05	−5.0423	27.7171	−39.1609	18.5115
R32	1.25	−17.1399	73.7573	−96.8387	42.9216
R124	1.08	−5.8609	31.7968	−45.8863	22.2463
R125	1.35	−0.02981	2.3752	0	0
R134a	1.10	2.5041	−3.3481	2.7974	0
R141b	1.00	1.47	0	0	0
R152a	1.00	−0.2246	3.4732	−1.6137	0

calculated as a function of the temperature and pressure, or as a function of temperature only for saturation conditions.

In Table V a direct comparison of the available experimental data for the viscosity coefficient, with the values obtained by the scheme described by Eqs. (1)–(10) is presented. Only measurements performed over the last 4 years were considered. The reason for this is that older measurements in general showed systematically larger deviations from the recent ones, probably attributed to the impurity of samples, as discussed elsewhere [22]. To include measurements of the viscosity outside the range of the density correlation, the proposed density correlation was extrapolated. The extrapolated values were in good agreement with the values obtained by a corresponding-states or an equation-of-state approach. In the cases where extrapolation was employed, this is clearly marked in the table.

From the 1030 experimental data points used, only 53 (5.1%) were found to show deviations larger than 5%, while only 4 (0.4%) were found

Table IV. Coefficients v_i ($10^6\,\mathrm{m^3 \cdot mol^{-1}}$) of Eq. (10)

Fluid	v_1	v_2	v_3	v_4
R11	44.4517	99.27131	−206.4591	126.7803
R12	−566.9460	2384.226	−3073.981	1301.525
R22	46.4355	−27.14650	23.6760	−9.32477
R32	93.0184	−248.3420	305.6490	−132.4426
R124	24.6431	168.5865	−263.7055	124.5258
R125	80.7012	−108.8456	124.8455	−57.0415
R134a	69.8139	−91.4943	119.4357	−57.4219
R141b	84.4876	−87.6727	103.4928	−45.8241
R152a	111.1725	−237.9698	257.2758	−93.3449

Table V. Comparison of Calculated Viscosity Values with Experimental Values

Fluid	First author	Ref. No.	Temp. range (K)	Press. max. (MPa)	No. of data	Dev.		Av. dev. (%)	Max. dev. (%)
						5–10%	>10%		
R11	Assael	23	273-334	20	39	—		0.7	2.0
	Kumagai	30	273-354	Sat.	9	—	—	1.1	1.7
R12	Assael	23	273-334	16	39	—	—	1.5	4.1
	Kumagai	30	273-344	Sat.	8	—	—	3.0	4.9
R22	Assael	25	273-334	15	39	—	—	0.9	1.5
	Kumagai	30	273-324	Sat.	6	1	—	2.8	7.1
	Bivens	33	233-354	Sat.	13	—	—	1.7	3.3
	Diller	57	200-320	32	72	4	-	2.7	7.5
	Arnemann	37	241-334	Sat.	6	2	—	3.5	6.8
R32	Assael	24	273-314	16[a]	31	—	—	1.5	3.9
	Ripple	32	250-294	Sat.	10	—	—	1.5	4.6
	Bivens	33	253-344[a]	Sat.	6	—	—	3.6	4.9
	Oliveira	27	231[a]-344[a]	Sat.	19	3	2	4.6	13.5
R124	Assael	25	293-334	17	39	—	—	0.9	2.5
	Ripple	32	251[a]-314	Sat.	14	1	—	2.5	5.9
	Diller	36	240[a]-385[a]	55[a]	75	—	—	1.9	4.9
R125	Assael	25	273-314	15[a]	27	—	—	1.2	2.6
	Oliveira	27	251[a]-324	Sat.	12	7	1	6.1	12.8
	Ripple	32	250[a]-302	Sat.	15	—	—	0.7	2.2
	Bivens	33	253[a]-314	Sat.	4	—	—	1.9	3.1
	Diller	36	200[a]-325	31[a]	79	6	—	2.7	9.5
R134a	Assael	24	273-334	15	39	—	—	0.7	1.0
	Oliveira	26	237-344	51[a]	60	2	—	1.6	8.6
	Okubo	29	213-324	31[a]	63	—	—	3.2	4.9
	Kumagai	30	273-344	Sat.	8	1	1	3.2	11.2
	Ripple	32	250-302	Sat.	15	—	—	0.7	2.2
	Bivens	33	253-354	Sat.	6	—	—	2.6	4.5
	Diller	35	200-320	34[a]	88	22	—	3.8	9.9
	Arnemann	38	243-334	Sat.	6	1	—	4.2	9.2
R141b	Assael	23	273-334	19[a]	39		—	1.1	2.2
	Kumagai	31	273-354	Sat.	9	—	—	0.5	1.2
	Diller	35	200[a]-320	31[a]	42	2	—	2.7	8.1
R152a	Assael	23	273-334	18	39	—	—	0.7	1.6
	Kumagai	30	273-344	Sat.	8	—	—	1.9	4.9
	van der Gulik	28	243-373	Sat.	36	—	—	1.5	4.1
	Arnemann	37	242-353	Sat.	10	1	—	3.5	8.6
Total					1030	53	4	2.1	

[a] Employing extrapolation of the density equations.

Table VI. Comparison of Calculated Thermal Conductivity Values with Experimental Values

Fluid	First author	Ref. No.	Temp. range (K)	Press. max. (MPa)	No. of data	Dev. 5–10%	Dev. >10%	Av. dev. (%)	Max. dev. (%)
R11	Assael	39	253–334	22	40			0.9	1.9
	Yata	40	233[a]–438[a]	Sat.	12	1	—	2.8	6.5
	Kitazawa	41	211–323	54[a]	22	—	—	1.7	3.9
R12	Assael	39	253–334	23	45	—	—	0.8	3.7
	Yata	40	264–367	Sat.	10	—		2.8	4.8
R22	Assael	42	252–334	27	42	—	—	2.3	4.9
	Yata	40	234–355	Sat.	6	—	—	0.9	2.0
	Kim	43	223–324	21	25	—	—	1.8	3.6
	Bivens	33	233–364[a]	Sat.	14	—	—	2.2	4.2
	Tsvetkov	58	208–290	0.9	16	—	—	2.8	4.8
R32	Assael	61	252–314	18[a]	31	—	—	2.4	4.7
	Yata	59	253–325	30[a]	27	3	—	2.9	8.2
	Papadaki	45	205[a]–303	Sat.	10	4	—	4.5	8.7
	Bivens	33	254–345[a]	Sat.	12	2	—	2.7	6.4
	Ro	60	223[a]–324	20[a]	24	3	—	3.2	7.3
	Gross	46	233[a]–334	7	37	3	—	1.7	6.8
R124	Yata	47	257[a]–354	31	25	1	—	2.6	6.3
	Assael	61	252[a]–334	19	40	—	—	1.2	3.8
R125	Papadaki	45	225[a]–307	Sat.	7	1	—	3.7	8.5
	Assael	61	273–314	17[a]	23	—	—	2.7	4.0
	Yata	59	257[a]–305	31[a]	24	11	—	4.5	9.3
	Gross	62	253[a]–324	7	35	7	—	3.9	9.5
	Bivens	33	253[a]–329	Sat.	10	—	4	7.8	19.0
R134a	Assael	44	253–334	13	33	—	—	1.4	4.9
	Yata	63	253–334	31[a]	21	—	—	1.4	4.1
	Papadaki	48	240–307	Sat.	5	—	—	3.3	3.8
	Ueno	49	193[a]–354	31[a]	36	2	—	2.0	7.9
	Gross	50	261–355	Sat.	13	2	—	2.8	9.9
	Laesecke	51	203–343	68[a]	52	4	—	2.0	7.9
	Gross	52	253–354	7	39	9	1	5.1	14.1
	Perkins	53	204–304	6	7	—	—	1.9	3.3
	Gurova	54	200–300	Sat.	21	—	—	1.1	2.8
	Gurova	64	213–293	20	50	—	—	1.8	4.1
	Ro	60	223–324	20	24	—	—	0.7	1.8
	Bivens	33	253–349	Sat.	10	2	—	3.5	7.1
R141b	Papadaki	48	248[a]–304	Sat.	6	—	—	1.7	2.1
	Assael	61	253[a]–314	22[a]	31	—	—	0.6	1.5
	Yata	47	251[a]–393[a]	31[a]	29	1	1	2.8	10.5
	Gurova	54	200[a]–300	Sat.	21	—	—	2.0	3.3
R152a	Assael	55	253–334	23	42	—	—	1.1	3.7
	Yata	47	265–343	31	20	6	—	3.7	9.9
	Gross	56	253–314	7	24	—	—	2.5	3.8
	Kim	43	248–324	21	20	—	—	1.6	3.7
	Gross	52	253–364	7	43	4	—	3.6	8.8
	Mardolcar	34	211[a]–295	19	39	—	—	2.8	4.2
	Tsvetkov	58	189[a]–300	9	11	—	—	1.1	1.8
Total				1134	66	6	2.3		

[a] Employing extrapolation of the density equations.

to show deviations larger than 10%. It should be noted that of the 53 points, 22 are measurements performed by Diller et al. [35], which are much higher than all the rest. The rest of the points that deviate more than 5% are measurements that are systematically higher (or lower) than all other measurements. The average absolute deviation of all experimental points considered is 2.1%, which is excellent considering the fact that in most cases the temperature range is from 200 up to 10 K below the critical temperature and the pressures extend up to 50 MPa.

In Table VI a direct comparison of the available experimental data for the thermal conductivity, with the values obtained by the scheme described by Eqs. (1)-(10) is presented. Here some older measurements had to be included as there were very few recent ones. This implies that the scatter between the various data sets is expected to be larger. As already discussed, the density correlation was again extrapolated. From the 1134 experimental thermal conductivity measurements considered, 66 (5.8%) showed deviations larger than 5%, while only 6 of them (0.5%) showed deviations larger than 10%. The few points that deviate more than 5% are measurements that are systematically higher (or lower) than all the other measurements. Here, also, the agreement is very satisfactory, since in most cases the temperature range is from 200 up to 10 K below the critical temperature, and the pressures extend up to 70 MPa.

An advantage of the proposed scheme, as already discussed, is the fact that V_0 depends only on temperature. Hence, low-pressure measurements are sufficient for its evaluation. The pressure dependence enters only through the density. Hence, if accurate densities are available, the scheme can be easily extended to high pressures. An example of this is the case of R134a, where a very accurate density correlation exists up to 26 MPa. Extrapolation of this correlation up to 68 MPa produced values of the viscosity and thermal conductivity of R134a in excellent agreement with the experimental data. Furthermore, preliminary results of the application of the scheme to refrigerant mixtures, according to the mixing rules described elsewhere [3], have been very promising.

In summary, considering the extended temperature and pressure ranges covered, the power of this scheme is very satisfactory.

4. CONCLUSION

Viscosity and thermal conductivity coefficient data for nine refrigerants have been successfully correlated using a scheme developed for hydrocarbons based on a consideration of the hard-sphere theory of transport properties. Coupled with a Tait-type correlation for the density of these refrigerants, the scheme allows the calculation of the viscosity and thermal conductivity of these refrigerants as a function of temperature and pressure, with an uncertainty of about ±5%.

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