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Correlation and Prediction of the Transport Properties of Refrigerants Using Two Modified Rough Hard-Sphere Models[1]

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Two methods are presented for the correlation and prediction of the viscosities and thermal conductivities of refrigerants R11, R12, R22, R32, R124, R125, R134a, R141b, and R152 and their mixtures. The first (termed RHS1) is a modified rough-hard-sphere method based on the smooth hard-sphere correlations of Assael et al. The method requires two or three parameters for characterizing each refrigerant but is able to correlate transport properties over wide ranges of pressure and temperature. The second method (RHS2) is also a modified rough-hard-sphere method, but based on an effective hard-sphere diameter for Lennard–Jones (LJ) fluids. The LJ parameters and the effective hard-sphere diameter required in this method are determined from a knowledge of the density–temperature behavior of the fluid at saturation. Comparisons with the rough-hard-sphere method of Assael and co-workers (RHS3) are shown. We also show that the RHS2 method can be used to correlate as well as predict the transport properties of refrigerants.

KEY WORDS: viscosity; thermal conductivity; rough-hard-sphere; refrigerants; refrigerant mixtures.

1. INTRODUCTION

Refrigerants and refrigerant mixtures are widely used as working fluids in many industrial applications, such as refrigerators, heat pumps, and power

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plants. A knowledge of their transport properties is therefore of importance in the design and evaluation of these processes. Such knowledge is also of theoretical importance because it provides a framework for an understanding of intermolecular forces in refrigerant systems. Since it is unlikely that experimental measurements of transport properties under all conditions of interest can be found in the literature, reliable methods for their estimation are of considerable interest. One method that has been used successfully to correlate dense fluid transport properties was proposed by Assael et al. [1–5]. Their method (RHS3) is based on the rough-hard-sphere (RHS) theory and employs a characteristic volume V_o and coupling parameters (R_D, R_η, R_λ) for each substance to correlate self-diffusion, viscosity, and thermal conductivity over wide ranges of temperature and pressure. The method has been applied to *n*-alkanes [1], *n*-alkane mixtures [2], aromatic hydrocarbons [3], alkanols [4], and methane- and ethane-derived refrigerants [5]. A limitation of the method is that a complex series of calculations is required to obtain V_o and the coupling parameters. In this work, therefore, we propose a simplified and systematic way to evaluate these parameters and demonstrate the application of the simpler technique (RHS1) to refrigerants.

A second method (RHS2) described below is based on our earlier work [6] on using the Lennard–Jones (LJ) fluid to obtain the hard-sphere diameter σ_{LJ} and hence the characteristic volume V_o required in the calculations. The LJ parameters σ_{LJ} and ε_{LJ} for each fluid are determined from a knowledge of the density-temperature behavior of the fluid at saturation.

We have used the two methods described above to correlate both high- and low-(saturated) pressure experimental data, and to compare the results with those obtained using the method of Assael et al. Special attention was given to the ability of the two methods to extrapolate data.

2. THE ROUGH-HARD-SPHERE THEORY

The rough-hard-sphere concept was proposed by Chandler [7] and extended by Assael et al. [1–5] who showed that the reduced diffusivity D^*, viscosity η^*, and thermal conductivity λ^* of all fluids can be expressed as universal functions of the reduced molar volume $V_r(=V/V_o)$ as follows:

$$\log(D^*/R_D) = 3.285 - 31.74261\,V_r^{-1} + 133.0472\,V_r^{-2} - 285.1914\,V_r^{-3} + 298.1413\,V_r^{-4} - 125.2472\,V_r^{-5} \qquad (1)$$

$$\log(\eta^*/R_\eta) = 1.0945 - 9.26324V_r^{-1} + 71.0385V_r^{-2} - 301.9012V_r^{-3} + 797.69V_r^{-4} - 1221.977V_r^{-5} + 987.5574V_r^{-6} - 319.4636V_r^{-7} \qquad (2)$$

$$\log(\lambda^*/R_\lambda) = 1.0655 - 3.538V_r^{-1} + 12.120V_r^{-2} - 12.469V_r^{-3} + 4.562V_r^{-4} \qquad (3)$$

where R_D, R_η, and R_λ reflect the degree of coupling between translational and rotational motions of the molecules and, in general, empirically account for deviations from the behavior of smooth hard spheres. In Eqs. (1)–(3), the reduced diffusivity, viscosity, and thermal conductivity are defined as

$$D^* = 5.030 \times 10^8 (M/RT)^{0.5} DV^{-1/3} \qquad (4)$$

$$\eta^* = 6.035 \times 10^8 (1/MRT)^{0.5} \eta V^{2/3} \qquad (5)$$

$$\lambda^* = 1.936 \times 10^7 (M/RT)^{0.5} \lambda V^{2/3} \qquad (6)$$

where M is the molecular weight, R is the gas constant, T is the temperature, η is the viscosity, λ is the thermal conductivity, and V is the molar volume of the substance (all properties expressed as SI units).

The transport properties of a fluid at a given temperature and pressure can be calculated using Eqs. (1)–(6), provided that the coupling parameters R_D, R_η, and R_λ, and the characteristic volume V_o are known. Note that the volume V must also be available from experiment or from a pVT relation at these conditions.

3. CALCULATION OF PARAMETERS

The parameters R_D, R_η, R_λ and V_o are not equally significant at high pressures, as noted earlier by Dymond and Awan [8]. They found that the effect of nonspherical shape (and hence the coupling parameter) on diffusivity was negligible at high pressures and that R_D could be set to unity. Similar behavior was found in this work in the case of viscosities of spherical fluids at high pressures, which could be correlated satisfactorily when $R_\eta = 1$. On the other hand, diffusivities and viscosities were found to be very sensitive to changes in V_o. More emphasis was therefore placed on obtaining accurate values of V_o in the present work. Also, calculated thermal conductivities showed only a slight dependence on V_o. This parameter was therefore estimated from viscosity data. (Note that diffusivities of refrigerants were not available, so that this property could not be used to obtain V_o.)

The calculational procedure may be summarized as follows:

1. R_η was set equal to 1 or any other realistic value.
2. Experimental viscosities were used together with Eqs. (2) and (5) to calculate values of V_o, which were then fitted with a fourth-order polynomial in temperature. (This requires an iterative procedure using the secant method.)
3. The quantity $dy_1 = (\eta_{exp} - \eta_{cal})/\eta_{exp}$ was calculated.
4. V_o and experimental data on thermal conductivity were used to calculate values of R_λ, which were then fitted with a polynomial in temperature.
5. The term $dy_2 = (\lambda_{exp} - \lambda_{cal})/\lambda_{exp}$ was calculated.
6. Steps 1–5 were repeated until a minimum in $\sum dy_1^2 + \sum dy_2^2$ was obtained.

4. APPLICATION TO REFRIGERANTS

Four methane-based refrigerants and five ethane-based refrigerants were chosen for study because they had also been studied previously by Assael et al. [5]. Experimental viscosities of the nine refrigerants were obtained from the literature and consisted of 650 data at high pressures and 200 data at saturated pressure. The temperature range of the data was $T/T_c = 0.40$ to 0.94, and the volume V at each temperature was obtained either from the literature or from the Tait equation reported by Assael et al. [5].

Table I lists the results of our calculations of the viscosities of the nine refrigerants. Average absolute deviations (AAD%) and maximum absolute deviations (MAD%) between experimental and calculated viscosities are listed for both the RHS1 method and the RHS3 method of Assael et al. In general, both methods show excellent agreement between calculated and experimental values. The large values of MAD for the RHS3 method are a result of the wider temperature range of the data used for comparison in the present work.

Experimental thermal conductivities of the nine refrigerants were also obtained from the literature and consisted of 550 data at high pressures and 75 data at low pressures. The temperature range of the data was the same as for the viscosity. Table II lists the results of the comparisons for both the RHS1 and the RHS3 methods. Again, the results obtained using

Table I. Comparison of Calculated and Experimental Viscosities

	Data pts		RHS3			RHS1			RHS2			
			AAD (%)		MAD (%)	AAD (%)		MAD (%)	AAD (%)		MAD (%)	
System	HP[a]	LP[b]	HP	LP		HP	LP		HP	LP		Reference Nos.
R11	36	9	0.84	1.15	2.07	0.52	0.73	1.33	0.64	0.64	0.94	13, 20
R12	36	8	1.84	2.94	5.08	1.75	2.21	5.74	2.02	4.18	9.87	13, 20
R22	43	6	0.85	2.67	6.67	0.74	2.29	5.60	0.85	2.72	6.83	15, 20
R32	46	10	3.04	1.51	15.60	1.01	1.19	2.91	1.16	1.42	3.17	14, 17, 22
R124	124	39	4.17	4.43	41.84	2.92	2.00	12.13	2.96	3.74	14.61	15, 22, 24
R125	89	44	2.66	2.15	14.45	1.44	1.12	9.21	2.64	1.61	10.72	15, 17, 22, 25
R134a	148	52	2.44	4.07	17.03	3.41	2.36	16.67	3.75	2.15	17.40	14, 16, 19, 23
R141b	61	28	1.71	2.38	7.38	1.92	1.78	9.85	2.30	3.32	11.69	13, 21, 23
R152a	65	8	1.10	1.84	5.02	0.77	1.83	4.80	0.92	1.99	4.28	13, 18, 20
Average			2.03	2.57		1.61	1.72		1.92	2.42		

[a] Pressures above saturation.

[b] Saturation values.

Table II. Comparison of Calculated and Experimental Thermal Conductivities

			RHS3			RHS1			RHS2			
	Data pts		AAD (%)		MAD (%)	AAD (%)		MAD (%)	AAD (%)		MAD (%)	
System	HP[a]	LP[b]	HP	LP		HP	LP		HP	LP		Reference Nos.
R11	57	11	1.26	2.56	6.11	0.78	1.91	3.48	0.77	1.92	3.45	25–27
R12	40	10	0.78	6.58	28.42	1.16	2.12	3.62	1.51	2.75	5.02	25, 26
R22	62	5	2.17	1.06	5.56	0.53	1.54	2.56	0.52	1.89	3.06	26, 28, 29
R32	55	10	2.78	4.31	8.13	2.82	3.70	6.49	2.85	3.83	6.78	30, 36–38
R124	65	8	1.79	2.25	6.74	1.76	1.98	8.57	1.59	1.82	7.42	31, 38
R125	24	10	2.77	5.53	16.20	2.33	3.06	8.95	2.33	3.01	8.02	30, 38
R134a	113	9	2.31	3.52	9.48	2.33	1.75	7.43	2.26	1.71	7.21	30–34, 37
R141b	70	6	1.64	1.68	11.74	1.46	1.03	9.64	1.44	1.02	8.26	31, 32, 38
R152a	62	5	3.36	1.01	10.98	2.64	1.98	10.96	2.64	1.77	10.39	29, 31, 34, 35
Average			2.10	3.16		1.76	2.12		1.77	2.19		

[a] Pressures above saturation.
[b] Saturation values.

the two methods show good agreement with each other and with experimental values. It is interesting to see that although V_o was obtained from viscosity data, calculated values of the thermal conductivity at both high and low pressures are still very satisfactory.

Finally, V_o and R_λ were correlated as functions of temperature as follows:

$$V_o \times 10^6 = A_1 + A_2 T + A_3 T^2 \tag{7}$$

$$R_\lambda = B_1 + B_2 T + B_3 T^2 \tag{8}$$

where the coefficients A_i and B_i are listed in Table III.

5. EXTENSION TO REFRIGERANT MIXTURES

The RHS 1 method was extended to binary refrigerant mixtures using the mixing rules:

$$V_{o,m} = x_1 V_{o,1} + x_2 V_{o,2} \tag{9}$$

$$R_{\eta,m} = x_1 R_{\eta,1} + x_2 R_{\eta,2} \tag{10}$$

$$R_{\lambda,m} = x_1 R_{\lambda,1} + x_2 R_{\lambda,2} \tag{11}$$

where x_1 and x_2 are the mole fractions of components 1 and 2 and the subscript m denotes a mixture quantity. This mixing rule has no adjustable parameters and does not require experimental data for the pure components. It has been used successfully by Assael et al. to calculate viscosities and thermal conductivities of *n*-alkane mixtures over wide ranges of temperature and pressure.

The results of the application of the mixing rule to refrigerant mixtures are given in Table IV. Both the RHS1 method and the RHS3 method were compared and found to work well for thermal conductivities, although large errors were observed for the RHS3 method in the case of R12 + R22 because the temperature of the data (200 K) exceeded the fitting range of that correlation. Mixture viscosities, however, were not satisfactory. Both methods overpredicted the viscosities of R32 + R134a mixtures by as much as 7% and underpredicted the viscosities of R32 + R124 mixtures by as much as 13%. This may be due to the fact that each component has a strong dipole moment in the gas phase (1.98 D for R32, 2.06 D for R134a, and 1.47 D for R124) so that a more realistic mixing rule may be required.

Table III. Coefficients of Eqs. (7), (8), and (13)–(15)

System	R_η	A_1	$A_2 \times 10^2$	$A_3 \times 10^4$	B_1	$B_2 \times 10^2$	$B_3 \times 10^4$	σ_{LJ}	ε_{LJ}/k	D_1	D_2	D_3	E_1	E_2	E_3
R11	1.157	64.2171	−3.0060		1.8126	−0.2555	0.0652	4.9778	368.22	1.17775			1.73769	−0.70754	0.72920
R12	1.668	62.2981	−7.5906		0.8809	0.4021		4.7603	301.21	1.10841			2.18566	−1.74739	1.17223
R22	1.217	42.8546	−3.2715		1.0615	0.2142		4.3388	288.38	1.05793			1.90104	−1.23951	0.86858
R32	1.081	28.7302	−1.8940		1.0654	0.2462		3.8432	270.62	0.85195	0.12734		0.98171	0.65083	
R124	1.085	65.4159	−3.3840		1.2101	0.1284		4.9374	309.92	4.92547	−6.57959	2.95180	1.49591	0.29324	
R125	1.116	55.6437	−3.1591		3.4303	−1.5934	0.3499	4.6831	265.77	4.20784	−4.74695	1.92809	1.25585	0.64916	
R134a	1.248	53.7759	−3.9235		0.7265	0.3496		4.5826	292.12	4.80995	−6.39373	2.90635	1.09901	0.71446	
R141b	0.994	75.9654	−8.9035	1.0770	1.6299	−0.0561		5.0214	374.00	5.82544	−10.8581	6.39552	1.69667	−0.12541	
R152a	1.097	58.6224	−11.664	1.4562	0.7585	0.2779		4.4088	300.77	3.27308	−4.18262	1.98805	0.93081	0.66030	

Table IV. Comparison of Viscosities and Thermal Conductivities for Refrigerant Mixtures

			RHS3		RHS1		RHS2		
Mixture	*P*	Data	AAD (%)	MAD (%)	AAD (%)	MAD (%)	AAD (%)	MAD (%)	Ref. No.
				Viscosity					
R125a + R134a	LP[a]	15	1.55	2.95	3.84	5.11	3.00	4.56	32
R32 + R134a	LP	15	8.87	12.56	7.29	10.20	6.78	10.52	32
R32 + R124	LP	14	13.20	18.55	15.62	20.61	14.94	20.11	32
R22 + R152a	LP	29	3.29	10.07	2.76	9.17	2.74	9.98	39
				Thermal conductivity					
R22 + R152a	HP[b]	125	2.20	4.84	2.51	4.89	2.26	5.30	29[c]
R12 + R22	LP	24	5.69	30.72	2.01	5.52	2.63	6.47	40[c]
R32 + R134a	HP	120	2.54	10.86	2.72	6.60	2.17	5.78	41[c]

[a] Saturation pressures.

[b] Pressures above saturation.

[c] Mixure densities were estimated by $x_1 \rho_1 + x_2 \rho_2$, where x is the mole fraction.

6. CALCULATION OF V_o FROM THE LENNARD–JONES HARD-SPHERE DIAMETER

Since the Lennard–Jones potential behaves like the hard-sphere (HS) potential for dense fluids, the hard-sphere diameter σ_{HS} can be obtained from the Lennard–Jones parameters σ_{LJ} and ε_{LJ} provided that a proper correlation between these quantities is established. The hard-sphere diameter σ_{HS} can then be used to determine the characteristic volume V_o as follows:

$$V_o = (\sigma_{HS}^3/2^{0.5})\, N_A \tag{12}$$

where the term in parentheses is the HS close-packed volume [9] of each molecule and N_A is the Avogadro number.

Given V_o, the coupling parameters R_D, R_η, and R_λ can be obtained by fitting experimental data for the diffusivity, viscosity, and thermal conductivity as described previously. We call the method with parameters obtained via the LJ hard-sphere diameter the RHS2 method.

Several correlations for car σ_{HS}/σ_{LJ} were examined. The most reliable was that proposed by Heyes [10] and given by:

$$\sigma_{HS}/\sigma_{LJ} = 1.0217(1 - 0.0178T^{*-1.256})/T^{*1/12} \tag{13}$$

where $T^* = T/(\varepsilon_{LJ}/k)$. This equation was obtained by fitting the simulation results for transport properties of LJ fluids, with the power of 1/12 coming from the scaling behavior of soft-sphere fluids.

LJ parameters for the nine refrigerants studied were determined from a LJ equation of state [11] at $T/T_c = 0.70$ to 0.80 using saturated liquid density data. Two sets of data were used: densities from the Tait equation reported by Assael et al. and densities from NIST tables [12]. The parameters obtained from the Tait equation were in excellent agreement with those using densities from the NIST tables, and averaged values are listed in Table III.

Experimental viscosities of the nine refrigerants were chosen at $T^* = 0.5$ to 1.2, corresponding to $T/T_c = 0.39$ to 0.94 ($T_c/(\varepsilon_{LJ}/k) \approx 1.28$). The values in Table III were used together with Eqs. (12) and (13) to obtain the characteristic volume V_o for each refrigerant at a given temperature. Experimental viscosity data were then used to obtain the coupling parameter R_η for each refrigerant. It was found that R_η is constant for R11, R12, and R22, a linear function of temperature for R32, but a quadratic function of temperature for the ethane-based refrigerants. The results are given in Table III.

Similarly, it was found that R_λ is a linear function for R32 and ethane-based refrigerants, but a quadratic function for R11, R12, and R22. Table III lists the results from the various methods. In general, the results from the different methods are comparable.

The coupling parameters were correlated as follows:

$$R_\eta = D_1 + D_2 T^* + D_3 T^{*2} \tag{14}$$

$$R_\lambda = E_1 + E_2 T^* + E_3 T^{*2} \tag{15}$$

where the coefficients D_i and E_i are listed in Table III.

Results for refrigerant mixtures using the mixing rule given in Eqs. (9)–(11) ware presented in Table IV.

7. APPLICATION TO HALOGENATED METHANES

Five halogenated methanes were chosen to test the RHS3 further. Dymond [13] found that for the halogenated methanes, the translational-rotational coupling is in the order: R10 > R11 > R13 > R14. Therefore, it is important to test whether any of the methods described above can predict such a trend.

LJ parameters and average values of R_η and R_λ were obtained as described above and are given in Table V. The diameter σ_{LJ} decreases by 0.20 Å for each substitution from R10 to R14 except between R13 and R14, whereas ε_{LJ}/k changes by -60 K with each substitution, and $T_c/(\varepsilon_{LJ}/k) = 1.28$ to 1.26. In the case of the viscosity, R_η shows a good trend with each substitution from Cl to F except the last one (from R13 to R14). This may be because of experimental uncertainty. In the case of the thermal conductivity, the method exhibits excellent trends.

Table V. Parameters for Halogenated Methanes

				RHS2		RHS3		RHS1		
Liq.	T_c (K)	σ_{LJ} (Å)	ε_{LJ}/k (K)	R_η [a]	R_λ [a]	R_η	R_λ [a]	R_η	R_λ [a]	R_D [b]
R10	556.35	5.1654	434.85	1.32	1.77					0.54
R11	471.20	4.9768	368.03	1.20	1.67	1.0	1.5	1.2	1.6	0.64
R12	384.95	4.7593	301.16	1.13	1.63	1.6	2.0	1.7	2.1	
R13	302.01	4.5028	237.39	1.08	1.58					0.9
R14	227.50	4.1776	179.92	1.11	1.52					1.0

[a] R_η and R_λ are averaged values.

[b] R_D from Dymond [9].

8. SUMMARY

Two approaches for obtaining the parameters of the RHS method for transport properties of dense fluids are described. The first approach (RHS1) determines the characteristic volume V_0 and coupling parameter R_η for each substance from viscosity data. R_λ is then obtained from thermal conductivity data using the same value of the characteristic volume. The second approach (RHS2) determines V_0 from the effective hard-sphere diameter of Lennard–Jones fluids and the Heyes relationship between the Lennard–Jones diameter and the hard-sphere diameter. The LJ parameters required in the calculations were determined from saturated densities at $T/T_c = 0.75$. The coupling parameters were found to be independent of pressure for both methods. The RHS2 method has the advantages of simplicity and of yielding parameters which exhibit the correct trends.

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