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Correlation for Viscosity and Thermal Conductivity of Liquid Halogenated Ethane Refrigerants[1]

M. J. Assael,[2,4] N. K. Dalaouti,[2] J. H. Dymond,[3] and E. P. Feleki[2]

The viscosity and thermal conductivity of liquid halogenated ethane refrigerants from about 200 K to near the critical temperature, at saturation and also at pressures up to 50 MPa, are shown to be satisfactorily correlated on the basis of a scheme developed by Dymond and Assael from consideration of hard-sphere theory.

KEY WORDS: correlation; hard spheres; liquid; refrigerants; thermal conductivity; viscosity.

1. INTRODUCTION

Recent accurate measurements of the liquid density and viscosity of refrigerants make it possible to carry out a more extensive test of the application of the Assael and Dymond scheme [1] than earlier tests [2], and the results are described here for halogenated ethane refrigerants.

2. TRANSPORT PROPERTIES OF DENSE FLUIDS

The Dymond and Assael scheme [1] for correlation of dense-fluid experimental viscosity and thermal conductivity data over wide ranges of pressure and temperature was developed on the basis of hard-sphere theory, taking into consideration experimental results for simple fluids. In

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[2] Chemical Engineering Department, Aristotle University, Thessaloniki 54006, Greece.

[3] Department of Chemistry, University of Glasgow, Glasgow G12 8QQ, United Kingdom.

[4] To whom correspondence should be addressed.

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this scheme, a reduced experimental viscosity η^* and a reduced experimental thermal conductivity λ^* are introduced by means of the equations

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

$$\lambda^* = 1.936 \times 10^7 \lambda V^{2/3} (M/RT)^{1/2} / R_\lambda \tag{2}$$

where V is the molar volume, T is the absolute temperature, M is the molar mass, and R is the universal gas constant. R_η and R_λ are roughness factors which were introduced for molecular fluids to account for effects of nonspherical shape.

It was found that these reduced transport properties were each functions of only the reduced volume V/V_o, where V_o is a characteristic volume which corresponds in a hard-sphere system to the volume of close packing. For convenience, the universal curves were expressed by

$$\log \eta^* = \sum_{i=0}^{7} a_{\eta i} (V_o/V)^i \tag{3}$$

$$\log \lambda^* = \sum_{i=0}^{4} a_{\lambda i} (V_o/V)^i \tag{4}$$

with values of the coefficients $a_{\eta i}$ and $a_{\lambda i}$ given in Table I.

In the earlier application of this scheme to refrigerant viscosity and thermal conductivity data [2], it was found that the values for the R_η parameter which gave the optimum data fit did not vary with compound structure in a regular manner. However, for six of the nine compounds considered, R_η was in the range 1.0 to 1.1, and was exactly equal to 1 in three

Table I. Coefficients $a_{\eta i}$ and $a_{\lambda i}$

i	$a_{\eta i}$	$a_{\lambda i}$
0	1.0945	1.0655
1	−9.26324	−3.538
2	71.0385	12.120
3	−301.9012	−12.469
4	797.69	4.562
5	−1221.9770	—
6	987.5574	—
7	−319.4636	—

cases. In view of this result and the absence of high-pressure data necessary to define R_η or R_λ more closely, we have investigated the possibility that R_η can be taken as equal to 1 for the correlation of viscosity data for all halogenated ethane refrigerants.

Application of Eq. (3) to viscosity data for a given compound at a specified temperature at different densities gives practically identical values for the characteristic volume V_o. When this has been repeated for all temperatures of viscosity measurement, the set of V_o values can be fitted by an equation in temperature. Then, Eq. (4) with these V_o values is applied to experimental thermal conductivity values on the same compound to derive values for the roughness factor R_λ. In the previous application, Assael et al. [2] found that, in all but two cases, R_λ was temperature dependent; similar behavior had been found by Assael et al. [3] for alcohols.

The liquid halogenated ethane refrigerants investigated in this paper are R113, R114, R115, R123, R123a, R124, R125, R133a, R134a, R141b, R142b, R143a, and R152a. Since the aforementioned scheme depends upon the density values employed (as molar volume) and for consistency purposes, it was decided to employ saturation density correlations proposed in literature. Hence, for the refrigerants R113, R114, and R115, the correlation proposed by Platzer et al. [4] was employed, while for refrigerants R123, R124, R125, R134a, R141b, R142b, R143a, and R152a, the correlation by Sato et al. [5] was used. Only in the case of R123a and R133a were the measured densities employed. The form of equation of the saturation density ρ employed in all these correlations is

$$\rho = \rho_\mathrm{c}\left[1 + \sum_{i=1}^{5} d_i \left(1 - \frac{T}{T_\mathrm{c}}\right)^{e_i}\right] \tag{5}$$

The critical temperature T_c, the critical density ρ_c, coefficients d_i, and powers e_i, as well as the ranges of applicability, are shown in Table II. The uncertainty of the density calculated by this equation is less than $\pm 0.1\%$, which in some cases is much better than that employed in the measurements. It should be pointed out, however, that the range of applicability of this equation results in a restriction in the application of the scheme to wider range experimental data.

In Table III references for recent accurate viscosity measurements and the experimental temperature range considered in this work are shown. Capillary viscometer measurements of Phillips and Murphy [6] include some refrigerants for which no other viscosity data are available, and so an attempt has been made [7] to correct their data by relating their results to recent accurate values from other references.

Table II. Critical Constants and Coefficients of Eq. (5) for Density

	R113	R114	R115	R123
T range (K)	250–470	290–400	200–340	200–450
T_c (K)	487.25	418.83	353.10	456.86
ρ_c ($kg \cdot m^{-3}$)	576	580	613.5	555
d_1	1.757368	1.801754	1.711552	2.350495
d_2	0.766357	0.704496	0.606856	−0.15959
d_3	0.401929	0.464355	0.760639	0.546187
d_4	0	0	0	0
d_5	0	0	0	0
e_1	1/3	1/3	1/3	0.375
e_2	2/3	2/3	2/3	0.6
e_3	3.0	3.0	3.0	1.3
e_4	0	0	0	0
e_5	0	0	0	0

	R124	R125	R134a	R141b
T range (K)	200–390	220–340	200–370	230–340
T_c (K)	395.35	339.165	374.30	480.00
ρ_c ($kg \cdot m^{-3}$)	566	568	508	460
d_1	1.853	1.4055	2.451	2.262
d_2	0.8089	1.96371	0.44031	0.5078
d_3	0	−1.57237	0	0
d_4	0	0.97421	0	0
d_5	0	0	0	0
e_1	0.33	0.3	0.38	0.38
e_2	0.897	2/3	1.6	1.6
e_3	0	1.0	0	0
e_4	0	4/3	0	0
e_5	0	0	0	0

	R142b	R143a	R152a
T range (K)	230–340	230–340	200–380
T_c (K)	410.29	346.25	386.44
ρ_c ($kg \cdot m^{-3}$)	446	434	368
d_1	2.349	1.06001	1.9992
d_2	0.4544	3.43587	−0.0824
d_3	0	−3.87356	0.18307
d_4	0	2.25883	1.1538
d_5	0	0	−0.4648
e_1	0.38	0.3	0.338
e_2	1.7	2/3	0.838
e_3	0	1.0	0.8915
e_4	0	4/3	1
e_5	0	0	1.3915

Table III. Comparison of Calculated Viscosities with Experimental Values

	T (K)	P (MPa)	No. of data	Deviations >5%	Deviations Max	σ (%)	Refs.
R113	273–353	Sat	9	—	0.7	0.4	8
R114	200–372	Sat	26	—	3.4	1.9	6, 8, 9
R115	200–303	Sat	8	—	2.4	1.3	6
R123	200–353	Sat	29	—	4.9	1.4	8, 10
	200–353	1.2–6	44	4	7.9	2.8	10, 23
R123a	273–353	Sat	9	—	1.0	0.7	8
R124	270–365	Sat	29	—	5.0	2.3	11–13
	270–365	1.5–56	60	1	5.5	2.3	11, 13
R125	176–310	Sat	86	12	7.0	3.1	11–15, 22
	200–420	3.5–53	109	7	7.3	2.6	11, 13
R133a	225–342	Sat	9	—	2.2	1.3	6
R134a	200–343	Sat	89	16	8.4	3.2	8, 10, 12, 14, 16–18
	200–370	0.9–53	169	16	8.0	2.9	10, 17, 24
R141b	270–353	Sat	24	6	7.4	4.2	10, 19, 20
	270–333	0.8–27	40	—	4.6	1.3	10, 20
R142b	239–353	Sat	25	—	2.9	1.3	9, 19
R143a	256–305	Sat	12	—	4.4	2.4	8, 12
R152a	254–373	Sat	78	3	7.5	3.0	6, 8, 9, 12, 20, 21
	273–333	1.5–15	32	—	2.0	1.0	20

The application of Eq. (3) to the viscosity data of references given in Table III, including the data of Phillips and Murphy [6] corrected as above, produced values for V_o which were fitted to the equation

$$V_o = \sum_{i=1}^{3} v_i (T/T_c)^{i-1} \tag{6}$$

Values for the coefficients v_i are given in Table IV. Viscosities at saturation pressures calculated from Eqs. (1), (3), and (5) with V_o values from Eq. (6) agree well with the experimental data as shown in Table III. In that table, in addition to the number of data points whose experimental viscosity deviates by more than 5% from the correlated value, the maximum deviation and the standard deviation of the fit are also shown. Where there are deviations greater than 5%, these occur with individual data sets (9 points of Ripple and Mattar [12] for R125; 13 points of Diller et al. [10] for R134a and 6 points for R141b) which differ from other results.

The method has been applied to viscosities at elevated pressures—there are no additional parameters (densities quoted were employed). The

Table IV. Coefficients v_i of Eq. (6) for V_o and c_i of Eq. (7) for the Thermal Conductivity Factor R_λ[a]

	v_1	v_2	v_3	c_1	c_2
R113	85.02	−9.75	0	1.53	0
R114	82.10	−27.67	15.44	1.82	1.14
R115	66.93	−6.00	0	1.68	0.55
R123	75.69	−25.00	12.11	1.55	0
R123a	69.50	−6.87	0	—	—
R124	68.27	−15.83	0	1.70	0.70
R125	55.00	−8.55	0	1.84	0.95
R133a	57.89	−7.31	0	—	—
R134a	50.74	−7.48	0	1.74	0.74
R141b	78.81	−51.50	31.32	1.48	0
R142b	66.78	−37.19	20.64	1.44	0
R143a	45.03	−3.46	0	—	—
R152a	47.63	−16.91	5.67	1.65	0.75

[a] Units of the v_i are $10^{-6}\,\mathrm{m^3 \cdot mol^{-1}}$.

results are also shown in Table III. Differences greater than 5% occur with the data of Diller et al. [10] for R134a (12 points) and R124 (4 points) and of Diller and Peterson [11] for R125 (7 points). This disagreement is in part attributed to uncertainties in the density values employed by the investigators [5].

With values for V_o given by Eq. (6) with coefficients from Table II, Eq. (4) was applied to the thermal conductivity data given in the references in Table V. Values for the thermal conductivity roughness parameter R_λ showed a small temperature dependence for some compounds, which was expressed as

$$R_\lambda = c_1 + c_2[(T/T_c) - 1] \tag{7}$$

Values for the coefficients c_1 and c_2 are listed in Table IV. Agreement with the selected measurements, as seen in Table V, is very satisfactory, not only at saturation pressure, but also at elevated pressures. The fit is restricted to temperatures less than about 0.85 times the critical temperature. Above this temperature, the experimental values are increasingly greater than the calculated values due to neglect of the critical enhancement in this approach. The effect decreases at a given temperature as the density of the fluid increases, and closer agreement is found at higher densities.

Table V. Comparison of Calculated Thermal Conductivities with Experimental Values

				Deviations			
	T (K)	P (MPa)	No. of data	>5%	Max	σ (%)	Refs.
R113	254–380	Sat	8	—	3	1.5	25
R114	290–387	Sat	5	—	3.1	2.0	25
R115	234–320	Sat	7	—	3.6	2.5	25
R123	200–354	Sat	49	—	2.8	2.3	29–32
	270–354	0.1–28	88	—	4.0	2.5	26–28, 30, 31
R124	234–354	Sat	13	—	4.9	2.2	25, 33
	253–354	0.6–30	52	—	4.6	1.8	25, 33
R125	220–300	Sat	29	1	6.4	2.8	14, 32–34, 36
	254–300	0.3–16	50	2	6.0	3.8	33, 36–39
R134a	200–350	Sat	54	4	9.8	3.5	14, 29–32, 34, 40
	200–350	1–68	300	18	9.8	3.4	27, 28, 30, 31, 41–43, 45, 46
R141b	240–333	Sat	11	—	2.9	1.7	33, 35
	253–369	0.1–22	35	—	3.3	1.5	33, 37, 48
R142b	223–323	Sat	5	—	3.0	1.0	50
	290–370	0.4–20	28	—	4.3	1.5	49
R152a	223–333	Sat	17	—	3.4	1.8	30, 44, 50
	190–333	0.6–30	126	1	5.6	2.8	30, 44, 47, 48, 50, 51

3. CONCLUSION

The universal equations of Assael and Dymond provide a very satisfactory correlation of viscosities and thermal conductivities both along the saturation line and at elevated pressures for liquid halogenated ethane refrigerants. They may be used for interpolation and also for modest extrapolation of data.

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