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DYNAMIC VISCOSITY AND THERMAL CONDUCTIVITY PREDICTION OF REFRIGERANTS AND REFRIGERANT MIXTURES

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abstract

Prediction methods to evaluate liquid dynamic viscosity and thermal conductivity along the saturation line are presented in this paper for pure and mixed refrigerants. The thermal conductivity correlation presented here is an extension to the new refrigerants of an equation previously proposed by one of the authors. The viscosity correlation has been improved upon a previous version in order to make it a real predictive method. The accuracy of the two correlations is checked with the most recent experimental data available in literature. The absolute deviations between estimated and experimental data for pure fluids are generally less than 5% (mean) and 10% (maximum) for the thermal conductivity correlation and are generally less than 6% (mean) and 13% (maximum) for viscosity. Both prediction methods are applied to refrigerant mixtures as well.

1. INTRODUCTION

Accurate prediction of transport properties of refrigerants is growing more and more important as a number of new pure fluids and mixtures appear to replace CFCs. Actually, experimental data for such fluids do not exist at all or they are given in very small temperature ranges and, often, at only just one temperature level; hence the necessity of reliable prediction methods. Since theoretical studies based on statistical mechanics produce acceptable results only for very simple fluids and in any case are not useful for engineering purposes, it is necessary to turn to empirical or semiempirical equations based on simple physical hypothesis.

Correlations of this kind are used in this paper to predict liquid dynamic viscosity and thermal conductivity along the saturation line for several refrigerants and refrigerant mixtures; most of these refrigerants have been presented only recently and therefore lack thermal properties data. The correlations are able to estimate the transport properties with good accuracy in wide ranges of temperature, and, in addition, are straightforward and easy to use.

The formula that predicts thermal conductivity in the liquid saturated state has been proposed by one of the present paper authors /1, 2/ and its accuracy is checked here through comparison with published experimental data; most of these data have appeared in literature only recently and refer to refrigerants proposed as promising CFCs substitutes. Mixtures behaviour is estimated with the method presented earlier in /3/ and checked by comparison with new experimental data as well.

Dynamic viscosity in the liquid saturated state was related with reduced temperature by

means of a correlation proposed by the authors in /4,5,6/; one of the three parameters contained in the formula had to be calculated by means of reliable experimental data. The correlation was not, therefore, a real prediction method, even if only one experimental datum was enough to estimate accurately the fluid behaviour from the melting point to beyond the boiling point. In the present work this drawback has been overcome, and an actual predictive equation is given: the new formula can predict liquid viscosities in the reduced temperature range 0.40-0.75.

2. THERMAL CONDUCTIVITY

Pure fluids

The correlation proposed has the form, /1,2/:

$$\lambda = A \cdot \frac{(1 - T_r)^{0.38}}{T_r^{1/6}} \quad (1)$$

where λ thermal conductivity, [$\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$],
 T temperature, [K],
 T_c critical temperature, [K],
 T_r T/T_c , reduced temperature,
 A constant, [$\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$].

The constant A has three main features: it is characteristic of each liquid, it is practically temperature independent and depends on the molecular structure. The value of A can be either calculated by linear regression from experimental data (only one experimental datum at $T_r \approx 0.55$ can be sufficient), or estimated, through the knowledge of fluid physical properties, with:

$$A = A^* \cdot \frac{T_c^\alpha}{M^\beta} \quad (2)$$

where M molecular weight,
 A^* constant whose value is:
 = 0.494 for refrigerants R10 to R14 and R110 to R160
 = 0.562 for refrigerants R20 to R23
 α = 0.166667
 β = 0.5

Comparisons between estimated thermal conductivities and experimental data are reported in Table I for several pure refrigerants. When using coefficients A calculated from experimental data, absolute general mean deviation is 2.5% and maximum deviation is 8.8%. When using coefficients A predicted from eq. (2) general mean and maximum deviations are 4.9% and 14.6% respectively.

Table I - Comparison between experimental and estimated thermal conductivities of pure refrigerants

Refrig.	formula	M	T _c [K]	T _r exp. range	Source of exp. data	constant A of eq. (1) calculated from experimental data			constant A of eq. (1) estimated with eq. (2)		
						A	δ _{mean} %	δ _{max} %	A	δ _{mean} %	δ _{max} %
R10	CCl ₄	153.82	556.3	0.46-0.65	/7,8/	.1220	2.0	3.7	.1142	6.0	8.7
R11	CCl ₃ F	137.37	471.1	0.36-0.74	/9/	.1199	2.6	4.9	.1176	2.9	4.8
R12	CCl ₂ F ₂	120.91	385.0	0.37-0.89	/9/	.1218	3.0	4.2	.1212	2.8	4.8
R13	CCIF ₃	104.46	302.0	0.49-0.90	/9/	.1256	5.0	5.1	.1252	5.0	5.1
R13b1	CBrF ₃	148.92	340.1	0.50-0.79	/10/	.1062	4.3	8.8	.1070	5.5	13.4
R20	CHCl ₃	119.38	536.4	0.41-0.65	/9/	.1401	1.6	2.7	.1466	3.3	5.2
R21	CHCl ₂ F	102.92	451.6	0.33-0.72	/9/	.1473	3.0	4.4	.1534	4.5	8.4
R22	CHClF ₂	86.46	369.3	0.43-0.86	/9,11/	.1585	4.0	6.4	.1619	4.1	11.2
R23	CHF ₃	70.02	299.0	0.50-0.77	/10/	.1884	2.2	3.9	.1737	7.8	11.2
R113	CCl ₂ FCClF ₂	187.39	487.3	0.49-0.90	/9/	.1017	2.7	4.9	.1012	2.6	5.3
R114	CClF ₂ CClF ₂	170.94	418.9	0.44-0.86	/9/	.0995	1.4	2.8	.1034	3.9	5.9
R114b2	CBrF ₂ CBrF ₂	259.83	487.6	0.34-0.66	/9,12/	.0831	0.7	1.5	.0860	3.5	5.0
R115	CClF ₂ CF ₃	154.47	353.2	0.49-0.85	/9/	.1037	1.7	2.5	.1057	2.3	4.0
R116	CF ₃ CF ₃	138.01	292.8	0.61-0.93	/9/	.1199	4.4	7.0	.1084	9.4	14.4
R123	CHCl ₂ CF ₃	152.93	456.9	0.56-0.78	/13,14/	.1080	1.3	2.8	.1109	3.6	5.5
R123a	CHClFCClF ₂	153	462.8	0.67-0.74	/14/	.1048	0.3	0.8	.1111	6.0	6.8
R124	CHClFCF ₃	136.47	395.6	0.78-0.88	/14/	.1152	2.0	4.2	.1146	2.0	4.8
R134a	CF ₃ CH ₂ F	102.03	374.2	0.70-0.89	/13,14, 15,16/	.1479	2.3	4.6	.1313	11.1	14.6
R142b	CH ₃ CClF ₂	100.49	410.2	0.35-0.85	/17/	.1356	4.1	5.2	.1343	3.9	6.1
R152a	CHF ₂ CH ₃	66.05	386.4	0.42-0.77	/18/	.1774	1.9	2.8	.1641	7.5	10.1
general mean						2.5			4.9		

δ_{mean}(%)=abs([(Σ(λ_{exp}-λ_{calc})/λ_{exp})/n]·100; δ_{max}(%)=max of [(λ_{exp}-λ_{calc})/λ_{exp}]-100; λ_{exp}= experimental thermal conductivity value; λ_{calc}= estimated thermal conductivity value; n= number of experimental points.

Mixtures

The authors proposed /3/ the following general correlation for the binary organic liquid mixtures:

$$\lambda = \left(A_1 x_1^2 + A_2 x_2^2 + 2.20 \sqrt{\frac{A_1^3}{A_2}} x_1 x_2 \right) \left(\frac{(1 - T_r)^{0.38}}{T_r^{1/6}} \right) \quad (3)$$

where: x₁, x₂ molar fractions of the two components.

A₁, A₂ coefficients A of eq. (1), with A₁ ≤ A₂.

In /3/ the correlation (3) was validated against experimental data of 50 organic mixtures and an absolute general average deviation of 2.5% and a maximum deviation of 13.1% were found out. In this work eq. (3) has been exploited to compare experimental and estimated thermal

conductivities of mixtures R502 as reported in Table II. To the authors knowledge no other reliable experimental data exist in literature for liquid thermal conductivity of refrigerant mixtures.

Table II - Comparison between experimental and estimated thermal conductivities of refrigerant mixtures

mixtures								
mixture	comp. 1	% weight	comp. 2	% weight	T _r exp. range	Source of exp. data	δ _{mean} %	δ _{max} %
azeotropic mixtures								
R502	R22	0.488	R115	0.512	0.85-0.90	/14/	2.9	7.0

3. DYNAMIC VISCOSITY

Pure fluids

A correlation of this type has been recently proposed /4,5,6/:

$$\frac{1}{\mu} = \frac{A}{C - T_r} - B \quad (4)$$

where: μ dynamic viscosity, [mPa·s],

A, B constants, characteristic of the fluid, [mPa⁻¹·s⁻¹],

C constant, dimensionless.

The parameter C depends on the fluid molecular structure; it has the same value for all the refrigerants of the methane series and the same value for all the refrigerants of the ethane series. A is related to the most important physical properties of the fluid and can be evaluated with simple monomial expressions; the evaluation of B, on the other hand, requires the knowledge of experimental viscosity data; even only one reliable datum would be sufficient.

The analysis of many reliable experimental data recently appeared in literature over wide temperature ranges make possible an important improvement of equation (4): a proper choice of the value of factor C allows to assume the value of A to be equal to the value of B with very good approximation. In particular, C=1.30 for methane series refrigerants, and C=1.35 for ethane series refrigerants; the equation (4) becomes then as follows:

$$\frac{1}{\mu} = A \cdot \left(\frac{1}{C - T_r} - 1 \right) \quad (5)$$

Table III shows the comparison between measured viscosities and viscosities calculated with eq. (5), where A values are obtained by regression of experimental data. In this case the correlation validity ranges from the melting point to T_r=0.8. The experimental sources choice has been made on the basis of measurements reliability and of width of investigated temperature range. Mix of viscosity measurements performed by different workers were avoided and for each fluid viscosity values from only one author were utilized. This is because often the difference

between two μ values obtained with different techniques is too high with respect to the two claimed accuracies.

Table III - Comparison between experimental and calculated viscosities of pure refrigerants

Refrig.	formula	T_c	T_r	Source of exp. data	C	A calc. from exp. data	$\delta_{\text{mean}}\%$	$\delta_{\text{max}}\%$
		[K]	range					
R10	CCl ₄	556.3	0.53-0.65	/19/	1.30	3.9496	5.6	10.6
R11	CCl ₃ F	471.1	0.44-0.75	/20/	1.30	4.6866	3.5	9.4
R12	CCl ₂ F ₂	385.0	0.52-0.81	/20/	1.30	5.4785	3.0	6.2
R13	CClF ₃	302.0	0.33-0.99	/21/	1.30	6.5097	1.3	5.9
R13b1	CBF ₃	340.1	0.53-1.00	/10/	1.30	4.9607	4.1	10.7
R20	CHCl ₃	536.4	0.39-0.66	/20/	1.30	5.2557	1.6	3.9
R21	CHCl ₂ F	451.6	0.46-0.77	/20/	1.30	5.5696	3.2	8.5
R22	CHClF ₂	369.3	0.54-0.73	/20/	1.30	5.8974	2.8	5.1
R23	CHF ₃	299.0	0.63-0.86	/20/	1.30	6.0713	2.6	3.8
R30	CH ₂ Cl ₂	510	0.41-0.73	/20/	1.30	5.9250	2.4	6.7
R31	CH ₂ ClF	426.6	0.45-0.74	/20/	1.30	6.2212	2.2	4.6
R32	CH ₂ F ₂	351.6	0.57-0.82	/20/	1.30	5.4335	1.0	3.7
R40	CH ₃ Cl	416.3	0.66-0.82	/22/	1.30	8.0351	0.7	1.8
R113	CCl ₂ FCClF ₂	487.3	0.56-0.73	/23/	1.35	4.3987	4.5	9.0
R114	CClF ₂ CClF ₂	418.9	0.65-0.84	/23/	1.35	4.9978	1.7	2.9
R115	CClF ₂ CF ₃	353.2	0.53-0.88	/24/	1.35	5.0207	5.6	11.5
R123	CHCl ₂ CF ₃	456.9	0.60-0.77	/23/	1.35	5.5220	1.4	2.8
R123a	CHClFCClF ₂	458.4	0.60-0.77	/23/	1.35	5.3333	1.9	3.8
R133a	CH ₂ ClCF ₃	395.6	0.52-0.79	/20/	1.35	5.9812	2.0	3.8
R134a	CF ₃ CH ₂ F	374.2	0.67-0.92	/25/	1.35	5.8600	0.9	1.5
R142b	CH ₃ CClF ₂	410.2	0.59-0.81	/26/	1.35	5.6899	4.9	9.5
R152a	CHF ₂ CH ₃	386.4	0.52-0.82	/20/	1.35	8.3482	1.2	3.1
R160	CH ₂ CH ₂ Cl	460.4	0.55-0.68	/27/	1.35	9.5695	3.4	6.7
R50	CH ₄	190.4	0.50-1.00	/28/	1.20	3.3043	1.3	3.0
R170	CH ₃ CH ₃	305.4	0.31-0.98	/29/	1.22	9.6448	2.3	6.6
R290	CH ₃ CH ₂ CH ₃	369.8	0.24-0.81	/29/	1.26	8.8591	1.1	3.3
R600	CH ₃ CH ₂ CH ₂ CH ₃	425.2	0.31-0.66	/30/	1.27	8.1922	1.3	2.6
R600a	CH(CH ₃) ₃	425.2	0.28-0.69	/30/	1.30	8.0262	3.4	7.5
general mean							2.5	

$\delta_{\text{mean}}(\%) = \text{abs} \{ (\sum(\mu_{\text{exp}} - \mu_{\text{calc}}) / \mu_{\text{exp}}) / n \} \cdot 100$; $\delta_{\text{max}}(\%) = \text{max of } [(\mu_{\text{exp}} - \mu_{\text{calc}}) / \mu_{\text{exp}}] \cdot 100$; μ_{exp} = experimental viscosity value; μ_{calc} = estimated viscosity value; n = number of experimental points.

But, to make the correlation a real predictive one, a method to calculate the coefficient A without any experimental datum has to be given. The link between A and the physical properties of the fluid can be well expressed by the relationship:

$$A = h \cdot \frac{T_c^\alpha \cdot V_c^\beta}{M^{\gamma} \cdot T_b^\delta} \quad (6)$$

where: V_c critical volume, [cm³·gmole⁻¹],
 T_b normal boiling temperature (at 101.325 kPa), [K],
 $h, \alpha, \beta, \gamma, \delta$ constants.

The values of the constants in eq. (6) have been obtained for the methane and ethane series with the mean squares method, as shown in Table IV. It must be noted that eq. (6) with the constants of Table IV can be used only with halogenated hydrocarbon refrigerants. More appropriate formulations have to be studied for methane, ethane, propane and n- and iso-butane, which have to be considered as belonging to the family of alkanes.

Table IV - Coefficients of eq. (6)

refrigerants	h	α	β	γ	δ
methane series	11.46125	6.147103	1.200513	0.69352	7.35470
ethane series	9.15130	0.367119	0.528526	0.82114	0.29033

In Table V the same comparisons of Table III are reported, but this time the A values for the different fluids are estimated with eq. (6). Of course deviations are higher when A is estimated rather than regressed from experimental data, but an absolute general mean deviation of 5% and maximum deviations usually less than 13% are still satisfactory for engineering purposes. Very few higher δ_{max} are due to experimental data near the melting point. At this point has to be noted that the prediction methods available in literature /2/ use more complicated techniques and do not reach better accuracies, where the method proposed in the present paper requires only the knowledge of easily available physical properties as M, T_b , T_c , and V_c .

Further comparisons have been developed between estimated values and reliable experimental data recently published but not used to implement the method. These comparisons are shown in Table VI. The comparison between estimates on R134a and data obtained in /31/ showed mean and maximum deviations of 14.4 and 32% respectively and are not reported in Tab VI.

Mixtures

The following mixing rule has been used here /6/:

$$A_{mix} = A_1 x_1 + A_2 x_2 \quad (7)$$

where A_1, A_2 are the A coefficients of eq. (5) for the mixture components, with $A_1 \leq A_2$, and x_1, x_2 are the molar fractions.

Table V - Comparison between experimental and estimated viscosities of pure refrigerants

Refrig.	formula	M	T _c	T _b	V _c	C	A estimated with eq. (6)	δ _{mean} %	δ _{max} %
			[K]	[K]	cm ³ mol ⁻¹				
methane series									
R10	CCl ₄	153.82	556.3	349.9	275.9	1.30	4.3540	9.3	18.9
R11	CCl ₃ F	137.37	471.1	269.9	247.8	1.30	4.9875	6.0	14.9
R12	CCl ₂ F ₂	120.91	385.0	245.2	216.7	1.30	5.4785	3.0	6.1
R13	CClF ₃	104.46	302.0	193.2	180.4	1.30	6.3047	3.2	9.4
R13b1	CBrF ₃	148.92	340.1	215.3	195.9	1.30	5.1037	3.7	13.2
R20	CHCl ₃	119.38	536.4	334.3	238.9	1.30	4.8765	7.8	9.9
R21	CHCl ₂ F	102.92	451.6	282.1	196.4	1.30	5.1707	8.0	11.4
R22	CHClF ₂	86.46	369.3	232.4	165.6	1.30	5.7410	3.8	5.9
R23	CHF ₃	70.02	299.0	191	132.7	1.30	5.9250	3.2	6.2
R30	CH ₂ Cl ₂	84.93	510	313	189.5	1.30	5.5646	6.6	9.5
R31	CH ₂ ClF	68.48	426.6	264.1	158.5	1.30	6.0754	3.1	5.5
R32	CH ₂ F ₂	52.02	351.6	221.5	120.8	1.30	5.8870	3.2	7.5
R40	CH ₃ Cl	50.49	416.3	249.1	138.9	1.30	8.4631	4.7	5.8
ethane series									
R113	CCl ₂ FCCLF ₂	187.39	487.3	320.8	304.2	1.35	4.6430	5.7	13.8
R114	CClF ₂ CClF ₂	170.94	418.9	276.2	293.8	1.35	4.8567	2.9	5.6
R115	CClF ₂ CF ₃	154.47	353.2	235.2	251.8	1.35	4.7872	7.4	11.9
R123	CHCl ₂ CF ₃	152.93	456.9	300.2	278.1	1.35	5.2088	6.0	8.2
R123a	CHClFCClF ₂	152.93	458.4	301.0	284	1.35	5.2683	2.2	4.2
R133a	CH ₂ ClCF ₃	118.49	395.6	279.2	239.5	1.35	5.9384	2.2	3.1
R134a	CF ₃ CH ₂ F	102.03	374.2	247.0	198	1.35	5.9673	1.8	3.2
R142b	CH ₃ CClF ₂	100.49	410.2	263.4	231	1.35	6.6516	14.5	22.6
R152a	CHF ₂ CH ₃	66.05	386.4	248.2	181	1.35	8.2214	2.0	2.8
R160	CH ₃ CH ₂ Cl	64.51	460.4	285.5	199	1.35	9.0250	6.1	13.2
general mean								5.1	

Table VI - Comparison between estimated viscosities and experimental data not used to implement the prediction method

Refrig.	T _r exp. range	Source of exp. data	δ _{mean} %	δ _{max} %
R123	0.61-0.72	/14/	6.7	8.3
R123a	0.61-0.72	/14/	11.0	12.9
R124	0.70-0.74	/14/	5.4	6.8
R134a	0.67-0.76	/14/	1.6	3.1
R141b	0.58-0.74	/14/	1.6	6.4
R123	0.42-0.70	/31/	3.7	8.5
R141b	0.37-0.67	/31/	1.6	6.5

Comparisons between experimental and estimated viscosities for azeotropic and non azeotropic mixtures are shown in Table VII. The estimation has been carried on with eq. (5), (6) and (7). Absolute general mean deviation is 4.4% and absolute maximum deviations are generally less than 10%.

Table VII - Comparison between experimental and estimated viscosities of refrigerant mixtures

Mixture	comp. 1	% weight	comp. 2	% weight	T_r exp. range	Source of exp. data	C	A estimated from eq. (7)	δ_{mean} %	δ_{max} %
azeotropic mixtures										
R500	R12	0.738	R152a	0.262	0.53-0.74	/20/	1.320	6.559	3.9	7.6
R502	R22	0.488	R115	0.512	0.56-0.76	/20/	1.318	5.388	4.3	6.5
R503	R23	0.401	R13	0.599	0.64-0.73	/20/	1.300	5.892	4.2	6.1
R504	R32	0.482	R115	0.518	0.60-0.73	/20/	1.313	5.595	16.8	17.9
non-azeotropic mixtures										
R22/R114	R22	0.101	R114	0.899	0.62-0.72	/32/	1.341	5.017	2.4	4.1
	R22	0.199	R114	0.801	0.62-0.72	/32/	1.334	5.148	6.9	9.6
	R22	0.299	R114	0.701	0.63-0.74	/32/	1.327	5.261	3.0	7.2
	R22	0.399	R114	0.601	0.65-0.75	/32/	1.322	5.359	3.1	3.9
	R22	0.499	R114	0.501	0.65-0.71	/32/	1.317	5.443	4.0	5.7
	R22	0.600	R114	0.400	0.66-0.72	/32/	1.313	5.518	5.6	7.3
	R22	0.699	R114	0.301	0.73-0.78	/32/	1.309	5.583	4.9	5.9
	R22	0.805	R114	0.195	0.67-0.78	/32/	1.305	5.644	5.5	6.4
	R22	0.900	R114	0.100	0.68-0.79	/32/	1.303	5.694	5.1	5.9
R22/R152a	R22	0.198	R152a	0.802	0.64-0.77	/32/	1.342	7.828	2.4	5.3
	R22	0.403	R152a	0.597	0.64-0.77	/32/	1.333	7.377	2.6	7.5
	R22	0.600	R152a	0.400	0.65-0.78	/32/	1.323	6.897	4.1	5.1
R22/R142b	R22	0.101	R142b	0.899	0.59-0.72	/32/	1.344	6.546	4.0	6.9
	R22	0.299	R142b	0.701	0.60-0.74	/32/	1.333	6.350	7.0	11.6
	R22	0.401	R142b	0.599	0.61-0.75	/32/	1.328	6.253	3.3	5.8
	R22	0.502	R142b	0.498	0.61-0.76	/32/	1.323	6.160	4.0	6.1
	R22	0.599	R142b	0.401	0.64-0.76	/32/	1.318	6.074	1.6	2.2
	R22	0.700	R142b	0.300	0.64-0.77	/32/	1.313	5.986	4.3	6.3
	R22	0.799	R142b	0.201	0.65-0.73	/32/	1.309	5.903	1.0	1.4
	R22	0.900	R142b	0.100	0.65-0.73	/32/	1.304	5.820	2.8	4.3
general mean									4.4	

4. CONCLUSIONS

Prediction methods to estimate liquid thermal conductivity and dynamic viscosity in the saturated state for pure and mixed refrigerants are presented in this work and validated against experimental data.

The thermal conductivity correlation presented here is an extension to the new refrigerants of an equation previously proposed by one of the authors and its accuracy is checked with the most recent experimental data available in literature. The pure fluids comparisons show that absolute general mean deviations between the calculated values and the values of experimental origin are usually less than 5%, whereas the maximum deviations are generally less than 10%. Accuracy validation for mixtures has been performed for just one azeotropic mixture showing a mean deviation of 2.9% and a maximum deviation of 7%.

The viscosity correlation has been improved from a previous version in order to make it a real predictive method. The new method proposed in this work allows to predict dynamic viscosity in the reduced temperature range 0.40-0.75; only the knowledge of easily available physical properties as M , T_b , T_c , and V_c is necessary to run the method. The comparisons used to check the method accuracy showed mean deviations between calculated and reliable experimental values generally less than 6% and maximum deviations usually less than 13%. The same method was applied to mixtures using appropriate mixing rules and, when checked, mean and maximum deviations usually less than 5% and 10% were noticed.

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