A New Approach to the Evaluation of Transport Properties of Azeotropic and Quasi-Azeotropic Refrigerant Mixtures[1]

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Azeotropic and quasi-azeotropic mixtures of organic compounds could become the most effective candidates as replacement fluids in refrigeration devices and heat pumps. Following the development of effective prediction formulas for several families of pure organic compounds, in this paper the evaluation of transport properties of liquid mixtures is approached from a rather different point of view. Azeotropic and near-azeotropic mixtures are treated as pure compounds rather than as a combination of several pure substances. This is now possible, having developed a single, specialized formula for both the liquid thermal conductivity and the dynamic viscosity. The prediction method requires the knowledge of only a few equilibrium properties of the mixture to be analyzed and thus is a simple and powerful tool for the evaluation of alternative refrigerants. Each formula has been tested against experimental data and shows deviations below those required for engineering purposes. Important results have also been achieved on applying the same equations to quasi-azeotropic mixtures with deviations comparable to those for azeotropic mixtures.

KEY WORDS: azeotropic mixtures; dynamic viscosity; prediction method; quasi-azeotropic mixtures; refrigerants; thermal conductivity.

1. INTRODUCTION

Following the ban of many widely used CFCs, the primary concern of the refrigeration industry has been to develop alternative refrigerants. These

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new alternatives must have low ozone depletion potential and require minimal changes when adopted as working fluids in existing equipment. For some CFCs other "ad hoc" single-component refrigerants were developed and produced, while in many other cases it was necessary to introduce mixtures.

Mixtures can be roughly grouped as azeotropes and zeotropes (i.e., non-azeotropes). Azeotropes are mixtures of fluids whose composition in the vapor phase and in the liquid phase does not change when in equilibrium. On the other hand, zeotropes are associated with composition and temperature glides or shifts during phase changes. In some cases, the temperature glide (i.e., the difference between its temperature at the dew point and its temperature at the bubble point) is quite negligible and the mixture is said to be a "quasi-azeotrope," a "near-azeotrope," or "azeotrope-like." The boundaries between azeotropes, near-azeotropes, and zeotropes cannot be easily stated. However, a mixture with a temperature glide smaller than 1°C is generally judged to be a near-azeotrope and a mixture with a temperature glide greater than 5°C is almost certainly a zeotrope [1].

During the past years we have developed prediction methods to evaluate transport properties of refrigerants in their saturated liquid state. The related formulas required the knowledge of a few equilibrium properties and were effective over a wide range of temperatures. Each formula could be used to evaluate the thermal conductivity or the dynamic viscosity of each compound belonging to a refrigerant series or to other organic compound series. Namely, we developed specialized formulas for methane-series refrigerants, ethane-series refrigerants, alkanes, and aromatic compounds. Few differences existed between the equations related to the first two families, and we were already aware about the possibility to develop a single equation for all the refrigerants belonging to the two series. However, this approach was not justified for pure refrigerants since we already had a small number of constants and a further reduction would have led to higher deviations.

An azeotropic mixture can be regarded, in terms of $P-V-T$ behavior, as a pure substance. We can determine a normal freezing point temperature and a normal boiling point temperature as well as critical point physical properties. Since these properties are, namely, the only parameters to be introduced in our formulas, together with the molecular mass, it was possible to evaluate the transport properties of azeotropic mixtures as they were pure substances [2]. We just needed to develop new formulas to evaluate transport properties of both methane-series and ethane-series refrigerants and apply them to azeotropic mixtures. For a quasi-azeotropic mixture, we can still evaluate its properties at the normal boiling point and at the critical point, for example, by means of a refrigerant database such

as REFPROP [3]. We can also use such databases to evaluate the same properties for zeotropes.

As a result, in this paper we propose new formulas to evaluate liquid thermal conductivity in the reduced temperature range from the normal freezing point to near the critical point and the liquid dynamic viscosity in the reduced temperature range from 0.5 to 0.9. We validated the prediction methods against both pure refrigerant data and mixture data available in the literature. Mean deviations between predicted results and experimental data are usually below 6% for thermal conductivity and usually below 8% for dynamic viscosity. Since mixtures were not considered during the fitting of constants, it was possible to test the predictive capability of the method. The method, as it is, should be considered as a tool for an "a priori" investigation on the applicability of new refrigerant mixtures.

2. PREDICTION METHODS

Our prediction method for the liquid thermal conductivity of refrigerant mixtures is based on the semi-empirical equation already proposed by present authors for pure refrigerants [4] and for other organic compounds [5]:

$$\lambda = B(1 - \tfrac{3}{4} T_r) \tag{1}$$

where λ is the thermal conductivity [10^3 W $\cdot$ m^{-1} $\cdot$ K^{-1}], T_r is the reduced temperature T/T_c, and the constant B was linked with some thermophysical properties in the form:

$$B = B^* \cdot T_c^\alpha \cdot P_c^\beta \cdot M^\gamma \tag{2}$$

where P_c is the critical pressure and M is the molecular mass. The constant B^* and the exponents α, β, and γ assumed different values for the methane-series refrigerants and the ethane-series refrigerants as listed in Table I [4].

We modified Eq. (2) to reach a single formula for both the ethane-series and the methane-series refrigerants taking into account that the critical pressure should not be included in the formulas since related data for

Table I. Coefficients in Eq. (2)

	B^*	α	β	λ
Methane series	0.4	1/4	1/3	$-3/4$
Ethane series	2.8	$-1/6$	1/6	$-1/2$

mixtures are not easily available. The knowledge that a single equation was able to estimate the thermal conductivity of all the refrigerants with reasonable errors was a good indication that our approach was justified. We found that the new equation:

$$B = 0.85 T_c^{1/3} M^{-3/4} \tag{3}$$

could be used to predict the liquid thermal conductivity of all the refrigerants with acceptable typical mean deviations, with respect to experimental data, of 5% as shown in Table II (where $AAD(\%) = [100\Sigma abs(\lambda_{calc}/\lambda_{exp} - 1)]/n$ and $MAD(\%) = 100$ max of $[abs(\lambda_{calc}/\lambda_{exp} - 1)]$, with λ_{exp} and λ_{calc}, respectively, the experimental and the estimated liquid thermal conductivity values and n the number of experimental points).

Table II. Investigated Pure Compounds for Thermal Conductivity (Equilibrium Properties Are Taken from Refs. 3, 28, 29)

Compound	Reference(s)	M	T_b (K)	T_c (K)	B (10^3 W·m^{-1}·K^{-1})	AAD (%)	MAD (%)
R112	[18]	203.83	366.0	551.0	0.1221	12.51	13.54
R113	[6, 7]	187.38	320.7	487.5	0.1274	6.42	14.20
R114	[6–8]	170.92	276.8	418.8	0.1331	1.60	4.48
R115	[6, 7, 9]	154.47	235.2	353.2	0.1396	1.33	3.13
R116	[18]	138.01	194.9	293.0	0.1472	5.05	5.50
R123	[8, 10–14]	152.93	301.9	456.9	0.1468	3.45	5.00
R123a	[8]	152.93	301.2	461.1	0.1470	1.15	1.90
R124	[6–8, 15]	136.47	260.0	395.6	0.1561	2.54	4.86
R125	[8, 14, 16, 17]	120.02	224.7	339.3	0.1675	2.71	16.04
R132b	[18]	134.94	320.0	493.2	0.1633	6.11	8.89
R133a	[18]	118.49	279.2	432.0	0.1761	6.08	8.06
R134a	[8, 10–13, 19–24]	102.03	247.1	374.3	0.1923	6.05	10.01
R141b	[15, 22]	116.95	304.9	477.3	0.1808	4.92	14.76
R142b	[15, 23]	100.49	263.4	409.6	0.1975	10.20	13.33
R152a	[11, 15, 25]	66.050	248.5	386.7	0.2679	8.80	15.73
R10	[6]	153.82	349.9	556.3	0.1510	13.15	15.02
R11	[6–8, 26]	137.37	296.9	471.2	0.1599	3.72	6.01
R12	[6–8, 26]	120.91	243.4	385.0	0.1701	3.77	10.08
R13	[6, 7]	104.46	191.7	302.0	0.1823	7.02	12.90
R13B1	[6, 7]	148.91	215.5	340.2	0.1426	2.46	6.28
R20	[6]	119.38	334.3	536.4	0.1815	9.53	12.63
R21	[6]	102.93	282.1	451.7	0.1971	1.37	5.00
R22	[6–8, 25, 26]	86.47	232.3	369.3	0.2173	1.39	5.96
R23	[6, 27]	70.01	191.0	299.1	0.2457	3.37	6.50
R30	[6]	84.93	313.0	510.0	0.2324	6.25	7.05
R31	[6]	68.48	263.9	430.0	0.2654	6.79	8.71
R32	[16, 18]	52.02	221.6	351.4	0.3154	12.12	13.87

For the dynamic viscosity, the starting point was the equation [30]

$$\frac{1}{\mu} = A \left(\frac{1}{C - T_r} - 1 \right) \tag{4}$$

where μ is the dynamic viscosity [mPa·s], A is a constant [mPa^{-1}·s^{-1}], C is a constant sensitive to the molecular structure, and T_r is the reduced temperature T/T_c.

For the constant C a new single value of 1.35 seems to be applicable to both the methane and the ethane series of refrigerants, provided that the constant A is calculated by means of the new formula,

$$A = hM^\alpha T_{br}^\beta \tag{5}$$

Table III. Investigated Pure Compounds for Dynamic Viscosity (Equilibrium Properties Are Taken from Refs. 3, 28, 29)

Compound	Reference(s)	M	T_b (K)	T_c (K)	C	A (mPa^{-1}·s^{-1})	AAD (%)	MAD (%)
R113	[32]	187.38	320.7	487.5	1.35	4.9114	8.39	13.75
R114	[32]	170.92	276.8	418.8	1.35	5.2224	2.79	7.12
R115	[33]	154.47	235.2	353.2	1.35	5.5546	5.24	9.08
R123	[32, 34, 35]	152.93	301.9	456.9	1.35	5.5619	3.18	10.98
R123a	[32]	152.93	301.2	461.1	1.35	5.7453	5.43	8.93
R124	[36, 37]	136.48	260.0	395.6	1.35	5.8062	2.18	5.14
R125	[36, 38]	120.02	224.7	339.3	1.35	5.8687	3.88	7.74
R133a	[36]	118.49	279.2	432.0	1.35	6.3137	5.40	8.90
R134a	[32, 37–43]	102.03	247.1	374.3	1.35	6.1654	4.10	14.28
R141b	[32, 44]	116.95	304.9	477.3	1.35	6.5442	0.58	2.21
R142b	[45]	100.49	263.4	410.4	1.35	6.7080	7.78	8.37
R143a	[32]	84.04	225.9	346.3	1.35	6.6973	13.58	13.77
R152a	[32, 36, 45, 46]	66.05	248.5	386.7	1.35	7.4220	9.74	14.52
R10	[47]	153.82	349.9	556.3	1.35	6.3868	9.76	14.54
R11	[32, 36, 46]	137.37	296.9	471.2	1.35	6.5350	4.89	11.96
R12	[32, 36, 46]	120.91	243.4	385.0	1.35	6.6823	5.52	16.07
R13	[48]	104.46	191.7	302.0	1.35	6.8543	14.67	14.54
R13B1	[32, 48]	148.91	215.5	340.2	1.35	6.3101	3.40	10.97
R20	[32, 36]	119.38	334.3	536.4	1.35	6.9851	2.71	5.52
R21	[36]	102.93	282.1	451.7	1.35	7.2037	6.31	13.28
R22	[322, 36, 49]	86.47	232.3	369.3	1.35	7.3761	1.81	5.99
R23	[36]	70.01	191.0	299.1	1.35	7.4472	5.71	10.18
R30	[36]	84.93	313.0	510.0	1.35	7.9461	6.10	12.80
R31	[36]	68.48	263.9	430.0	1.35	8.3858	5.12	12.09
R32	[36–39, 41]	52.02	221.6	351.4	1.35	8.3104	5.04	10.18
R40	[50]	50.49	249.1	416.3	1.35	9.7274	2.51	5.43

where M is the molecular mass and T_{br} is the reduced normal boiling-point temperature T_b/T_c.

This new equation contains only two terms: the reduced temperature at the normal boiling point and the molecular mass. The presence of M in the correlation is essential to take into account both the mass and the strong relation between molecular structure and liquid dynamic viscosity [31], having set the factor C to a single value.

Equations (4) and (5) have been tested against a large number of pure refrigerants, and it was found that it is effective over the reduced temperature range 0.5–0.9. Results are shown in Table III (AAD and MAD have the same meaning as for thermal conductivity).

3. APPLICATION TO MIXTURES

Azeotropic mixtures can be regarded as pure refrigerants provided that a single reliable formula can be separately applied to each component with satisfactory results. The same approach can be applied to near-azeotropic mixtures with comparable results. For both azeotropic and near-azeotropic mixtures, only the molecular mass, the critical temperature, and the temperature at the normal boiling point are easily available. For zeotropes we found that our prediction method could be applied by introducing as the normal boiling point temperature the average of the dew point temperature and bubble point temperature.

We applied Eq. (1) with the constant B evaluated by means of Eq. (3) to azeotropic, near-azeotropic, and small temperature-glide nonazeotropic mixtures as listed in Table IV. We obtained good results, with the deviations equal to or less than those we got for pure refrigerants. The same results were obtained for the dynamic viscosity of refrigerant mixtures listed in Table V. This seems to justify that a general equation for the prediction of transport properties of pure refrigerants could be used to evaluate the same properties of azeotropic mixtures and small temperature glide zeotropes.

Again, it should be noted that, for zeotropes, we introduced a "boiling-point-like" temperature defined as the average between the normal dew-point temperature and normal bubble-point temperature. This temperature generally corresponds to the one calculated as the boiling-point temperature when REFPROP [3] is used to evaluate zeotropes.

4. RESULTS

Results are shown in Tables IV and V. Table IV shows deviations between predicted thermal conductivity values and experimental data,

Table IV. Investigated Mixtures for Thermal Conductivity (Properties Are Taken from Refs. 1, 3, 28, 29, 54)

Refrigerant mixture	ASHRAE code	Reference	Weight fract.		M	T_b (T_{bubble}/glide) (K)	T_c (K)	B (10^3 W·m^{-1}·K^{-1})	AAD (%)	MAD (%)
			1st comp.	2nd comp.						
R22/125/290	R402B	[41]	0.600	0.380	98.372	(226.05/2.3)	367.95	0.1971	6.91	12.14
R125/143A/R134a	R404A	[41]	0.440	0.520	100.6	(226.65/0.8)	355.8	0.1927	6.00	7.84
R32/R125/R134A	R407D	[41]	0.300	0.100	88.83	(226.48/7.4)	373.6	0.2133	1.86	6.19
R32/125	R410A	[52]	0.500		86.02	(220.35/<0.1)	358.1	0.2170	7.93	8.99
R12/R152a	R500	[33]	0.738		99.31	239.7	378.7	0.1966	6.35	16.81
R22/R115	R502	[33, 51]	0.486		111.6	227.8	355.4	0.1782	5.01	12.32
R13/R23	R503	[33]	0.401		87.28	184.5	292.7	0.2075	4.52	14.96
R32/R115	R504	[33]	0.482		79.2	266.2	339.6	0.2288	2.01	5.42
R32/R134a		[41]	0.250		89.54	(232.75/7.2)	377.5	0.2124	1.57	3.68
R23/116	R508	[53]	0.460		106.5	(185.15/<0.1)	298.6	0.1793	6.79	9.23
R32/125		[41]	0.600		79.22	221.01 (est.)	359.6	0.2309	3.20	4.32

Table V. Investigated Mixtures for Dynamic Viscosity (Properties Are Taken from Refs. 1, 3, 28, 29, 54)

| Refrigerant mixture | ASHRAE code | Reference | Weight fract. | | M | T_b (T_{bubble}/glide) (K) | T_c (K) | A (mPa^{-1}·s^{-1}) | AAD (%) | MAD (%) |
			1st comp.	2nd comp.						
R22/125/290	R402B	[41]	0.600	0.380	98.372	(226.05/2.3)	367.95	7.5042	7.74	14.32
R125/143A/R134a	R404A	[41]	0.440	0.520	100.6	(226.65/0.8)	355.8	6.8472	14.79	23.22
R32/R125/R134A	R407D	[41]	0.300	0.100	88.83	(226.48/7.4)	373.6	7.8273	4.74	9.51
R32/125	R410A	[52]	0.500		86.02	(220.35/<0.1)	358.1	7.8896	7.69	15.93
R12/R152a	R500	[36]	0.738		99.31	239.7	378.7	6.9998	5.28	8.34
R22/R115	R502	[36]	0.486		111.6	227.8	355.4	6.5586	6.69	14.13
R13/R23	R503	[36]	0.401		87.28	184.5	292.7	7.3158	3.67	10.06
R32/R115	R504	[36]	0.482		79.2	266.2	339.6	7.3065	6.84	10.31
R32/R134a		[41]	0.250		89.54	(232.75/7.2)	377.5	8.0029	4.45	8.25
R23/116	R508	[53]	0.460		106.5	(185.15/<0.1)	298.6	7.0899	6.13	10.28
R32/125		[41]	0.600		79.22	221.01 (est.)	359.6	8.0497	11.84	19.37
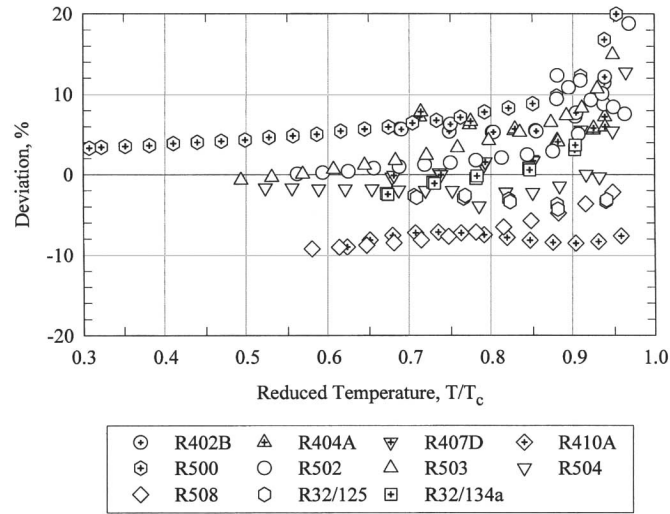
Fig. 1. Deviations between predicted thermal conductivities and experimental values for some azeotropic and quasi-azeotropic mixtures.
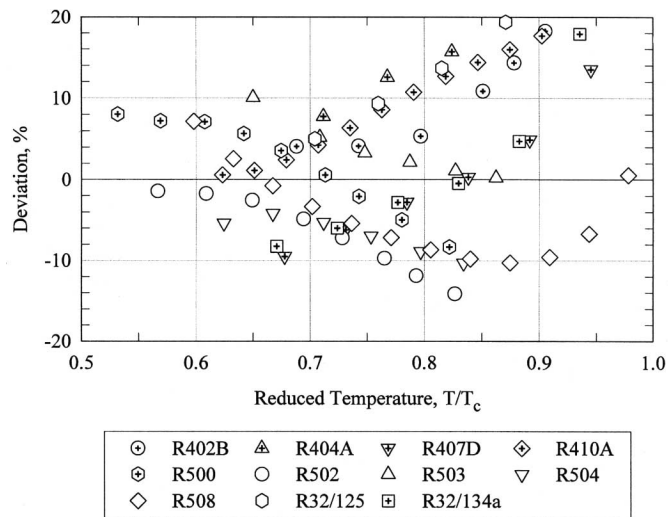

Fig. 2. Deviations between predicted dynamic viscosities and experimental values for some azeotropic and quasi-azeotropic mixtures.

while Table V refers to dynamic viscosity. As we expected, best results were achieved for the thermal conductivity. This is due to the greater dependence of dynamic viscosity on the molecular structure of compounds. This dependence was taken into account by factor C, which has now been set to a single value for all refrigerants thus leading to an expected increase of the deviations.

For pure refrigerants (Table III), mean deviations range from few to 10%, while maximum deviations are usually around 10%. Exceptions are those for R13 and R143a, where average deviations are around 14%. However, such deviations are constant within the reduced temperature range. For thermal conductivity, we have, in general, better results, but some compounds show average deviations greater than 10%. We have not yet analyzed such deviations. However, we think that the predictive capability of the prediction method for mixtures should not be affected by errors of the same magnitude. This can result from such a "balance" between components, which seems to smooth the behavior in terms of deviations.

5. CONCLUSIONS

The method for the evaluation of transport properties of liquid mixtures presented in this paper shows a good reliability and a reasonably good precision.

A statistical analysis of the sensitivity of prediction errors with respect to the temperature glide of the mixtures is currently being studied. In the near-future this analysis could lead to the application of the same approach to mixtures whose organic components belong to certain refrigerant families.

ACKNOWLEDGMENTS

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