A NEW APPROACH TO THE EVALUATION OF TRANSPORT PROPERTIES OF AZEOTROPIC AND QUASI-AZEOTROPIC REFRIGERANT MIXTURES[1]

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ABSTRACT

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 a combination of several pure substances. Both for liquid thermal conductivity and dynamic viscosity this is now possible having developed a single, specialised formula for each property.

The prediction method requires the knowledge of few equilibrium properties of the mixture to be analysed thus becoming a simple and powerful tool for exhaustive analysis of alternatives Each formula has been tested against experimental data leading to deviations far below that required for engineering purposes. Important results have also been achieved on applying the same equations to quasi-azeotropic mixtures being the deviations comparable to those of Azeotropic Mixture.

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

INTRODUCTION

Following the ban of many widely used CFCs, the primary concern of refrigeration industry has been to develop alternative refrigerants. These new alternatives had both to have low ozone depletion potential and to require minimal changes when adopted as working fluid in existing machinery. For some old CFCs other "ad hoc" single component refrigerants were developed and produced while in many other cases it was due to introduce mixtures.

Mixtures can be roughly grouped in azeotropes and zeotropes (i.e. non-azeotropes). Azeotropes are mixtures of fluids whose composition in the vapour phase and in the liquid phase does not change when in equilibrium. On the other hand, to zeotropes are associated 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 bubble point) is quite negligible and the mixture is said to be a "quasi-azeotrope" or "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 past years we developed prediction methods to evaluate transport properties of refrigerants in their saturated liquid state. The related formulas required the knowledge of few equilibrium properties and were effective in wide range of temperatures. Each formula was able 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 specialised formulas for: methane series refrigerants, ethane series refrigerants, alkanes and aromatic compounds. Few differences there were between the equations related to the first two families and we were already conscious about the possibility to develop a single equation for all the refrigerants belonging to the two

data and predicted values was not justified by a negligible reduction of the number of constants.

An azeotropic mixture can be regarded, in terms of P-V-T behaviour, as a pure substance. We can evaluate 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, we conceived the opportunity to evaluate transport properties as they were pure substances [2]. We just needed to develop those new formulas able 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 and we did it.

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 near to the critical point and the liquid dynamic viscosity in the reduced temperature range from 0.5 to 0.9. We validated prediction methods against both pure refrigerant data and mixture data available in literature. Mean deviations between prediction results and experimental data are usually below 6% for thermal conductivity and usually below 8% for dynamic viscosity. The predictive capability of the method was also tested having not considered the mixtures during the extraction of constants. It must be pointed out that we developed the equations we present as final result as a tool for an “a priori” investigation on the range of applicability of refrigerant mixtures. In fact the main feature of our method is its being simple, handy and reliable rather than its overall precision.

THE 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]:

$$\lambda = B \cdot \left(1 - \frac{3}{4} T_r\right) \qquad (1)$$

where λ is the thermal conductivity [W/mK·10^3], T_r is the reduced temperature T/T_C and the constant B was linked with some thermophysical properties in the form:

$$B = B^* \cdot T_c^{a} \cdot P_c^{b} \cdot M^{g} \qquad (2)$$

being P_c the critical pressure and M the molecular mass. The constant B^* and the exponents a, b, and g assumed different values for the methane series refrigerants and the ethane series refrigerants as listed in Table 1 [4].

We modified Eq. (2) to reach a single formula both for ethane series and methane series refrigerants taking into account that the critical pressure should not be present inside formulas being related data for mixtures 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 guarantee that our approach was, at least, worth to be developed. We found that the new equation:

$$B = 0.85 \cdot T_c^{1/6} \cdot M^{-3/4} \qquad (3)$$

was able to evaluate liquid thermal conductivity of all the refrigerants with errors still acceptable being typical mean deviations, with respect to experimental data, within 5% as shown in Table 2 (where AAD(%) = [Σabs($\lambda_{calc}/\lambda_{exp}$-1)]/n·100, MAD(%) = max of [abs($\lambda_{calc}/\lambda_{exp}$-1)]·100, being λ_{exp} and λ_{calc} respectively the experimental and the estimated liquid thermal conductivity values and n the number of experimental points).

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

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

where μ is the dynamic viscosity [mPa·s], A is a constant [mPa^{-1}·s^{-1}], C is a constant

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

$$A = h \cdot M^{-0.25} \cdot T_{br}^{-2.85} \qquad (5)$$

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 inside the correlation is essential to take into account both the mass itself and the strong relation between molecular structure and liquid dynamic viscosity [31], having set the factor C to a sole 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 3 (AAD and MAD have the same meaning as for thermal conductivity).

APPLICATION TO MIXTURES

After the first research step we reached some preliminary statements for what concerns transport properties of 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 or easily calculable. For what concerns zeotropes we found that our prediction method could be applied by introducing as normal boiling point temperature the average between dew point temperature and bubble point temperature.

We applied the Eq. (1) with the constant B evaluated by means of Eq. (3) to azeotropic, near-azeotropic and low temperature glide non-azeotropic mixtures as listed in Table 4. We obtained good results, being deviations equal or less of those we got for pure refrigerants. The same results were obtained for dynamic viscosity of refrigerant mixtures listed in Table 5. This seems to prove that a general equation for the prediction of transport properties of refrigerants could be able to evaluate the same properties of azeotropic mixtures and low temperature glide zeotropes.

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

RESULTS

Results are shown in Table 4 and Table 5. Table 4 shows deviations between predicted thermal conductivity and experimental data while Table 5 refers to dynamic viscosity. As we expected best results were achieved in the first case. This is due to the greater dependency of dynamic viscosity on the molecular structure of compounds. This dependency was taken into account by factor C which has now been set to a sole value for all refrigerants thus leading to the expected deviations.

For pure refrigerants (Table 3), mean deviations range from few percentage units to a ten percent while maximum deviations are usually around 10%. Unfortunate exceptions are those of R13 and R143a whose average deviations are around 14%. However such deviations are constant within the reduced temperature range. For thermal conductivity, we have, in general, quite better results but some compounds show average deviations greater than 10%. At the moment we have not analysed such deviations. However we think that the predictive capability of the prediction method for mixtures should not be

“balance” between components which seems to smooth the behaviour in terms of deviations.

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.. Having approached Azeotropic, and near Azeotropic mixtures as pure compounds the resulting formulas are really handy and require the knowledge of very few, easily available data thus becoming a simple and powerful tool for exhaustive analysis of alternatives Each formula has been tested against experimental data leading to deviations below that required for engineering purposes. Important results have also been achieved on applying the same equations to quasi-azeotropic mixtures and low temperature glide zeotropes being the deviations comparable to those of Azeotropic Mixture.

A statistical analysis about the sensitivity of prediction errors with respect to the temperature glide of the mixture is currently being carried out. In the near future this analysis could lead to the application of the same approach to all the mixtures whose organic components belong to determined refrigerant families.

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Halocarbon refrigerants of the	B^*	a	b	g
methane series	0.4	1/4	1/3	-3/4
ethane series	2.8	-1/6	1/6	-1/2

Table 1 - Coefficients of Eq. (2)

Compound		M	T_b [K]	T_c [K]	B	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] [7] [8] [26]	137.37	296.9	471.2	0.1599	3.72	6.01
R12	[6] [7] [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

Table 2: Investigated pure compounds for thermal conductivity
(equilibrium properties are taken from [3] [28] [29]).

Compound		M	T_b [K]	T_c [K]	C	A	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	[32] [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

Table 3: Investigated pure compounds for dynamic viscosity
(equilibrium properties are taken from [3] [28] [29]).

Refrigerant Mixture	ASHRAE code	References	Weight Fract. 1st comp.	Weight Fract. 2nd comp.	M	T_b (T_{bubble}/glide)	T_c	B	AAD [%]	N
R22/125/290	R402B	[41]	0.600	0.380	98.372	(226.05/2.3)	367.95	0.1971	6.91	1
R125/143A/R134a	R404A	[41]	0.440	0.520	100.6	(226.65/0.8)	355.8	0.1927	6.00	
R32/R125/R134A	R407D	[41]	0.300	0.100	88.83	(226.48/7.4)	373.6	0.2133	1.86	
R32/116	R410A	[52]	0.500		86.02	(220.35/<0.1)	358.1	0.2170	7.93	
R12/R152a	R500	[33]	0.738		99.31	239.7	378.7	0.1966	6.35	1
R22/R115	R502	[33] [51]	0.486		111.6	227.8	355.4	0.1782	5.01	1
R13/R23	R503	[33]	0.401		87.28	184.5	292.7	0.2075	4.52	1
R32/R115	R504	[33]	0.482		79.2	266.2	339.6	0.2288	2.01	
R32/R134a	R508	[41]	0.250		89.54	(232.75/7.2)	377.5	0.2124	1.57	
R23/116	R508B	[53]	0.460		106.5	(185.15/<0.1)	298.6	0.1793	6.79	
R32/125		[41]	0.600		79.22	221.01 (estim.)	359.6	0.2309	3.20	

Table 4: Investigated mixtures for thermal conductivity
(properties are taken from [1] [3] [28] [29] [54]).

Refrigerant Mixture	ASHRAE code	References	Weight Fract. 1st comp.	Weight Fract. 2nd comp.	M	T_b (T_{bubble}/glide)	T_c	A	AAD [%]	N
R22/125/290	R402B	[41]	0.600	0.380	98.372	(226.05/2.3)	367.95	7.5042	7.74	1
R125/143A/R134a	R404A	[41]	0.440	0.520	100.6	(226.65/0.8)	355.8	6.8472	14.79	2
R32/R125/R134A	R407D	[41]	0.300	0.100	88.83	(226.48/7.4)	373.6	7.8273	4.74	[illegible]
R32/116	R410A	[52]	0.500		86.02	(220.35/<0.1)	358.1	7.8896	7.69	1
R12/R152a	R500	[36]	0.738		99.31	239.7	378.7	6.9998	5.28	[illegible]
R22/R115	R502	[36]	0.486		111.6	227.8	355.4	6.5586	6.69	1
R13/R23	R503	[36]	0.401		87.28	184.5	292.7	7.3158	3.67	1
R32/R115	R504	[36]	0.482		79.2	266.2	339.6	7.3065	6.84	1
R32/R134a	R508	[41]	0.250		89.54	(232.75/7.2)	377.5	8.0029	4.45	[illegible]
R23/116	R508B	[53]	0.460		106.5	(185.15/<0.1)	298.6	7.0899	6.13	1
R32/125		[41]	0.600		79.22	221.01 (estim.)	359.6	8.0497	11.84	1

Table 5: Investigated mixtures for dynamic viscosity
(properties are taken from [1] [3] [28] [29] [54]).

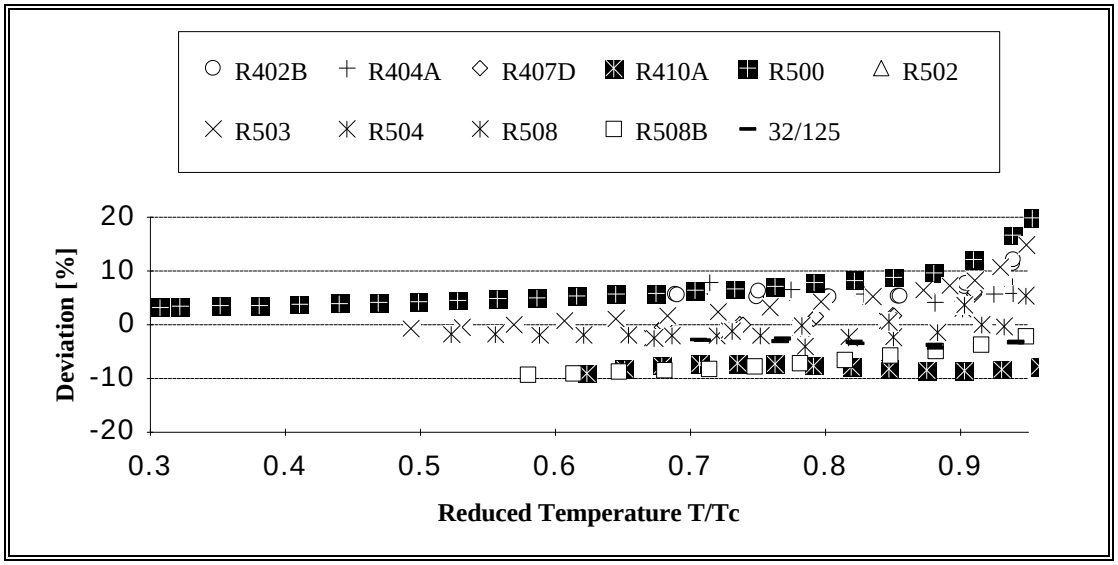

Fig. 1 - Deviation between predicted thermal conductivities and experimental values for some azeotropic and quasi-azeotropic mixtures

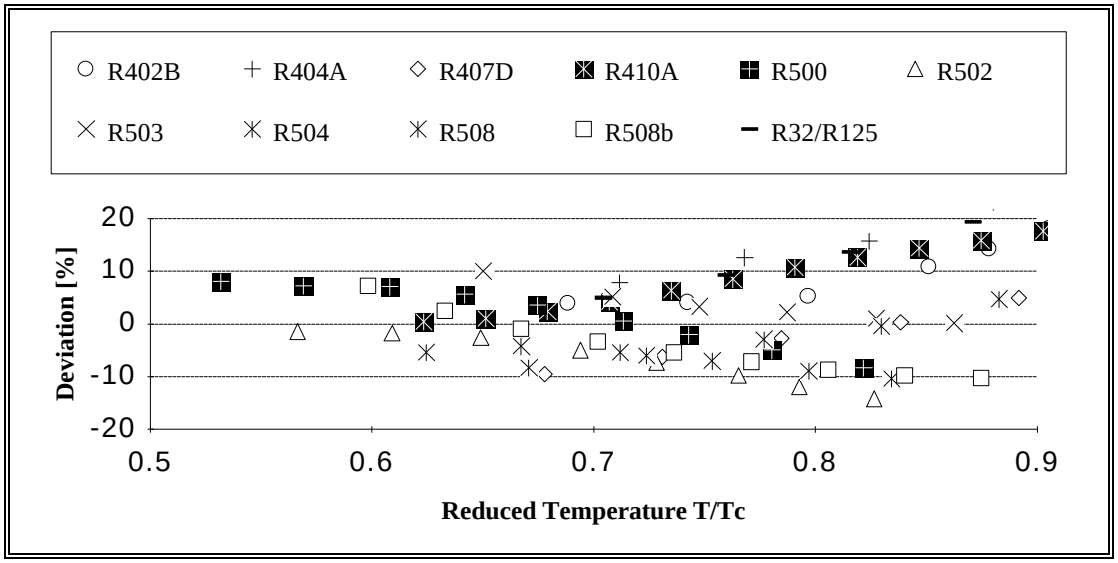

Fig. 2 - Deviation between predicted dynamic viscosities and experimental values for