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DYNAMIC VISCOSITY OF REFRIGERANTS AND REFRIGERANT MIXTURES: A PREDICTION METHOD.

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Abstract

A correlation relating the dynamic viscosity with reduced temperature Tr=T/Tc is provided. The correlation is useful for pure refrigerant fluids and their binary mixtures in the temperature range from about the normal melting point to well above the normal boiling point (in some cases up to near the critical point) at atmospheric pressure or along the saturation line.

The correlation contains three different parameters A, B and C: C is dimensionless and depends simply on the number of carbon atoms, while A and B are measured in cP^{-1} and are given by simple expressions for each group of refrigerant fluids (compounds with the structure of methane, compounds with the structure of ethane and so on).

Suitable mixing rules are provided in order to evaluate A, B and C for the binary mixtures when A, B and C are known for the pure components.

The mean deviations between predicted and experimental dynamic viscosity data are generally less than 3% and the maximum deviations usually do not exceed 7% in the above pointed out temperature range.

The experimental dynamic viscosity data used to evaluate A, B, C are clearly indicated (any mixing of data due to different authors is avoided in reaching this goal), but the check of the correlation is developed by taking into account all the most recent and accurate experimental dynamic viscosity data.

VISCOSITE DYNAMIQUE DES FRIGORIGENES ET MELANGES DE FRIGORIGENES. METHODE DE PREVISION.

RESUME : Les auteurs proposent une corrélation entre la viscosité dynamique et la température réduite : Tr=T/Tc. Cette corrélation est utile pour les frigorigènes purs et leurs mélanges binaires à des températures allant d'environ du point de fusion normal à une température bien supérieure au point d'ébullition normal (dans certains cas jusqu'au voisinage du point critique) à la pression atmosphérique ou le long de la ligne de saturation.

La corrélation contient trois paramètres différents : A, B et C. C est sans dimensions et dépend simplement du nombre d'atomes de

carbone, tandis que A et B sont mesurés en cP^{-1} et sont donnés par des expressions simples pour chaque groupe de frigorigènes (composés dérivés du méthane, composés dérivés de l'éthane, etc).

Des règles de mélange appropriées sont indiquées de façon à évaluer A, B et C pour les mélanges binaires quand on connaît A, B et C pour les composants purs.

Les écarts moyens entre les viscosités dynamiques prévues et expérimentales sont généralement inférieurs à 3 % et les écarts maximaux ne dépassent habituellement pas 7 % dans le domaine de température indiqué ci-dessus.

Les valeurs expérimentales de la viscosité dynamique utilisées pour évaluer A, B et C sont clairement indiquées (tout mélange de valeurs provenant d'auteurs différents est évité pour atteindre ce but), mais la vérification de la corrélation est effectuée en tenant compte des valeurs expérimentales de la viscosité dynamique les plus récentes et les plus précises.

DYNAMIC VISCOSITY OF REFRIGERANTS AND REFRIGERANT MIXTURES: A PREDICTION METHOD.

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INTRODUCTION

The fluid dynamic viscosity is a very important physical property, the knowledge of which is basic in the fundamental study of many transfer processes and fluid flow.

In this paper an empirical correlation is provided to predict the dynamic viscosity of refrigerants, particularly hydrocarbons and their binary mixtures, in the saturated liquid state. The investigated range starts from the normal melting point ending well above the normal boiling point which, in terms of reduced temperature, corresponds up to Tr=0.9.

Viscosity prediction of refrigerants is an important matter mainly for two reasons:

(a) many of the liquid viscosity experimental data that are available for hydrocarbons are rather sparse along the saturation line, especially near the melting and above the normal boiling points;

(b) the theoretical or empirical correlations that can be found in literature exhibit severe limitation in their use. The theoretical relations are usually valid only for some particular liquids, while for others prediction is unsatisfactory; in addition, some correlations make use of physical quantities that are difficult to evaluate with acceptable accuracy. On the other hand, empirical or semiempirical correlations are generally inaccurate over the whole liquid range [1].

By focusing the attention to the empirical correlations, improved clarification arises by dividing the existing correlations roughly in two main groups: in the first, there are correlations that might be considered as modifications of the Andrade exponential form [2] (Orrick and Erbar [2], Van Velzen, Cardozo and Langenkamp [3,4],); they all use structural-sensitive parameters and hold for certain homologous series of compounds within a range of validity that usually does not exceed the reduced temperature of 0.75. The correlations of the second group involve concepts dealing with the Law of Corresponding States (such as Przezdziecki and Sridhar correlation [5]) but they show inadequate accuracy in the vicinity of the normal melting point.

Some other sophisticated equations (as the equation of Letsou and Stiel [6]) are restricted only to relatively high temperatures (Tr>0.75); thus, any attempt in matching the estimated data across the connecting region 0.74<Tr<0.76 would turn critical.

The mean deviations of the predicted values from accurate experimental data for such correlations are usually greater than 10% exceeding in some cases even 50%.

Particular attention to the correlation purposely proposed by Srinivasan and Murthy for hydrocarbon refrigerants should be drawn. The correlation relates the reduced fluidity ϕd to a particular reduced temperature Td by means of two constants a and b (characteristic for any hydrocarbon) requiring for their estimation viscosity experimental data:

$$\phi d = a + b\,Td \tag{1}$$

The above reduced quantities are evaluated from the following expressions:

$$1/\phi d = \mu d = \mu Vc^{2/3} / [M(Tc - Tf)]^{1/2} \tag{2}$$

$$Td = (T - Tf)/(Tc - Tf) \tag{3}$$

where:

T = absolute temperature
Tf = normal melting point
Tc = critical temperature
Vc = critical molal volume
M = molecular weight
μ = dynamic viscosity

When introducing the proper values of a and b in equation (1), the standard error for each compound is smaller than 5%, while assuming constant "averaged" values for the whole group of the refrigerants R10, R11, R12, R13, R20, R21, R22, R23, R500, the standard error rapidly increases to about 12.6%.

In order to overcome some of the difficulties that are involved in predicting dynamic viscosity, a simple and accurate empirical correlation is provided in this work. Among the requirements which the correlation had to meet, priority in accuracy over a wide range of the reduced temperature variation, has been given.

DYNAMIC VISCOSITY PREDICTION OF SUBSTANCES IN THE LIQUID STATE

The starting point of the analysis is the empirical correlation suggested by the Authors referred to [8], that relates in a very simple form the dynamic viscosity to the reduced temperature by means of three parameters characteristic for any substance:

$$1/\mu = A/(C - Tr) - B \qquad (4)$$

The correlation was derived from the Batschinski equation [9] which has been modified by Hildebrand [10, 11] as follows:

$$1/\mu = D\ (V - Vo)/Vo \qquad (5)$$

where V is the molar volume while Vo and D were assumed as characteristic constants for any substance although they were sensitive to temperature [12]. The molal volume V in equation (5) was replaced by an appropriate relation involving the reduced temperature and, by introducing some simple assumptions, the equation was furtherly simplified to reach the final form (4).

An extensive check was initially carried out throughout 25 refrigerant fluids [8], successively increased up to 50 organic compounds, inorganic compounds and elements [13] as alkanes and aromatics, water, ammonia and carbon dioxide, n-hydrogen, nitrogen, oxygen, argon and bromine. The investigation was developed through the following steps:

- several set of experimental data of dynamic viscosity were carefully selected from literature [3, 14, 15, 16, 17] on the basis of accuracy and experimental methodology;
- the characteristic parameters A, B and C of equation (4) were evaluated for each substance with an optimization procedure;
- mean and standard deviations between predicted and experimental data were computed for the whole group of substances.

The main results which the analysis yielded were:

- the dimensionless parameter C was almost temperature-independent but sensitive to the molecular structure; typical values were found to range from 1.0 to 2.0;
- the A and B parameters were in turn insensitive to temperature with a very good approximation, but their range of variation was quite wider: from 3 cP^{-1} to 400 cP^{-1} for A and from 4 cP^{-1} to 200 cP^{-1} for B;
- the mean deviations between predicted and experimental values were spread within the range 3-10% when the reduced temperature varied from the reduced melting point up to about 0.9.

Although the above results were generally satisfactory, it should be observed that no precise means or computation rules for evaluating the A, B and C parameters were indicated. From this standpoint, therefore, the correlation itself did not strictly represent a complete prediction method.

The correlation we here propose fulfills this objective at least for both the hydrocarbon refrigerants and their binary mixtures.

The correlation parameters A, B and C for hydrocarbon refrigerants

As pointed out in the preceding section, the dimensionless parameter C seems to be sensitive to the molecular structure. To check this property, methane and ethane were investigated mainly for the following reasons:

- the availability of very accurate experimental data throughout the entire liquid region [18, 19]; Haynes [18] and Diller-Saber [19] claimed that the experimental accuracy is within ±2%;
- many of the hydrocarbon refrigerants exhibit a molecular structure very similar to that of methane or ethane, arising by simply substituting one or more atoms of hydrogen with bromine, chlorine or fluorine atoms. Thus, it seemed very reasonable to assume for the whole group of hydrocarbons the same value of the dimensionless C parameter evaluated for methane or ethane.

The resulting value of C that derives from processing the experimental data for both methane and ethane, was equal to 1.4 and the corresponding values of A and B were, respectively, 23.9002 cP^{-1} and 20.9204 cP^{-1} for methane, 14.9851 cP^{-1} and 12.8136 cP^{-1} for ethane. The maximum deviations between predicted and experimental data were below 2.5% from the normal reduced melting point to nearly Tr=0.9.

Assuming then C=1.4 for the entire investigated group of the hydrocarbon refrigerants, the unknown parameters A and B were computed through a least-square linear regression routine by using as set of variables 1/μ and 1/(1.4-Tr). Proper screening of experimental data was necessary in order to eliminate any uncertainty that would had compromised the optimum fitting of data: any mix of viscosity measurements performed by different authors, or either "recommended" or "estimated" values, was carefully avoided.

In Table I there are listed, for each investigated compound, the molecular weight, the normal melting and boiling points, the critical temperature and the experimental reduced temperature range together with the sources of information.

Table II is referred to seven binary mixtures for which the weight and molar fractions are also shown.

Furthermore, Table I lists the values of the A, B and C parameters for the investigated substances and the temperature ranges where the correlation (4) ensures satisfactory results. Mean and maximum deviations between predicted and experimental values are also shown. Finally, Table II provides for the selected mixtures the same information as in Table I.

As it can be seen from Tables I and II, the agreement between predicted and experimental data is very good if one considers that the accuracy of the measurements is generally better than ±3% for pure refrigerants and ±5% for their azeotrope mixtures. Nevertheless, a further improvement of the correlation should require proper guidance for determining the parameters A and B from the knowledge of related physical quantities.

Table I indicates that, if methane and ethane are excluded from the list, all the values of A lie within a narrow range from 5.8453 cP^{-1} (R13B1) to 8.8083 cP^{-1} (R32). There is an experimental evidence in this circumstance since the measured values for all the refrigerants are close enough to each other and their magnitude is well above that of methane or ethane.

At this stage, it seemed reasonable to attempt the search for a relationship between A and some related physical quantity such as the molecular weight, the normal boiling point or the critical temperature. The best accordance was obtained from the following relation:

$$A=10.02\ Tb^{1.2342}/(Tc^{0.8927}\ M^{0.4051}) \qquad (6)$$

which can be used for all the hydrocarbons listed in Table I with the exception of methane and ethane. The standard error that occur when A is evaluated by equation (6), is equal to 4.9%.

The use of an analogous relationship for B would give rise, as it can be easily verified when both the errors affecting A and B are added, to markedly enhanced errors in equation (4) because of it form. A better approach for determining the unknown B is to keep using equation (4), but with C=1.4 and A evaluated by equation (6). Now, since the B parameter is very little sensitive to temperature, only few selected experimental data will suffice although the computation can be satisfactorily performed with just one experimental value of viscosity.

The results obtained by following this methodology are gathered in Table III, where the final values of B to be used directly in equation (4) are shown together with the mean and maximum deviations: the former is generally less than 2.5% while the latter does not usually exceed 5% (the highest deviation is -12.9% and corresponds to R152, but this value clearly arises from a high-scattered measurement that also affect significantly the mean deviation).

The correlation (4) also shows a satisfactory agreement when it is used to predict the dynamic viscosity of refrigerants other than the investigated ones. For instance, the predicted viscosity of CH_3Cl or R40 (for which Tb=248.93 K, Tc=416.23 K and M=50.488, and hence A=8.5082 cP^{-1}, C=1.4 while the value of B=6.9139 cP^{-1} is computed from the viscosity measurement of 0.221 cP [17] at T=273.15 K) is 0.1607 cP at 313.15 K and 0.1882 cP at the temperature of 293.15 K against the experimental values of 0.152 cP and 0.183 cP, which show deviations of about 5.7% and 2.8% respectively. Very similar results may be obtained for other refrigerants as R133, R142 and so on.

Table I - General table of pure refrigerants with molecular weights M, normal melting Tf and boiling Tb points, critical temperatures Tc, ranges $\Delta T_{exp.}$ of μ experimental data, ranges $\Delta T_{val.}$ of validity of eq. (4), A, B and C parameters of eq. (4), mean Δ% and maximum $\Delta\%_M$ deviations between calculated and experimental μ values, and references.

Refr.	Chem. Form	M	Tf, K	Tb, K	Tc, K	$\Delta T_{exp.}$, K	$\Delta T_{val.}$, K	A, 1/cP	B, 1/cP	C	Δ%	$\Delta\%_M$	Ref.
R10	CCl_4	153.80	250.00	349.70	556.40	273-373	273-373	6.8190	6.7777	1.4	1.0	4.0	[17]
R11	CCl_3F	137.40	162.00	296.97	471.15	209-352	209-352	6.4291	5.9891	1.4	1.5	-2.9	[20]
R12	CCl_2F_2	120.90	115.40	243.40	385.00	202-312	202-312	6.0925	5.2213	1.4	2.0	-5.4	[20]
R13	$CClF_3$	104.50	92.00	191.70	302.00	192-272	192-272	6.0708	4.7654	1.4	0.6	1.5	[20]
R13B1	$CBrF_3$	148.90	105.15	215.40	340.15	246-301	246-301	5.8453	4.8649	1.4	0.4	-1.1	[21]
R20	$CHCl_3$	119.40	209.60	334.30	536.40	210-353	210-353	6.8923	6.3323	1.4	0.6	1.4	[22]
R21	$CHCl_2F$	102.90	138.00	282.00	451.60	208-347	208-347	6.6348	5.8954	1.4	2.5	7.4	[22]
R22	$CHClF_2$	86.50	113.00	232.40	369.20	201-299	201-299	6.5890	5.6482	1.4	1.6	4.6	[22]
R23	CHF_3	70.00	117.95	191.12	299.05	190-257	190-257	6.9230	5.8824	1.4	0.2	0.4	[22]
R30	CH_2Cl_2	84.90	178.10	313.00	510.00	208-374	208-374	7.2713	6.5572	1.4	2.7	9.1	[22]
R31	CH_2CLF	68.50	----	264.05	426.59	192-315	192-315	7.8468	7.1284	1.4	2.1	6.5	[22]
R32	CH_2F_2	52.00	----	221.45	357.26	200-287	200-287	8.8083	8.6737	1.4	0.9	2.6	[22]
R50	CH_4	16.04	90.70	111.70	190.60	95-190	95-170	23.9002	20.9204	1.4	0.8	1.8	[18]
R113	CCl_2FCClF_2	187.40	238.16	320.73	487.26	250-400	250-400	5.6641	5.6972	1.4	1.4	3.8	[23]
R114	$CCLF_2CCLF_2$	170.90	179.30	276.90	418.90	198-331	198-331	5.8908	5.8109	1.4	1.5	5.1	[20]
R115	$CCLF_2CF_3$	154.50	167.00	234.00	353.20	199-303	199-303	5.8544	5.6579	1.4	1.3	2.1	[20]
R152a	CH_3CHF_2	66.05	156.20	248.40	386.60	200-318	200-318	8.6897	8.0537	1.4	2.0	5.8	[20]
R170	CH_3CH_3	30.07	89.90	184.50	305.40	95-300	95-275	14.9851	12.8136	1.4	0.9	2.2	[19]

Table II - Table of the investigated mixtures with weight and molar fractions.

Mixture	Composition	P1%	P2%	X1	X2	M	Tb, K	Tc, K	$\Delta T_{exp.}$, K	$\Delta T_{val.}$, K	A_m, 1/cP	B_m, 1/cP	C_m	Δ%	$\Delta\%_M$	Ref.
R500	R12/R152a	73.80	26.20	0.606	0.394	99.30	239.65	379.00	201-311	201-311	7.0174	6.2738	1.4	2.4	-6.9	[20]
R502	R22/R115	48.80	51.20	0.630	0.370	111.66	227.55	360.00	201-294	201-294	6.3592	5.6896	1.4	1.6	-4.7	[20]
R503	R23/R13	40.10	59.90	0.500	0.500	87.25	184.45	300.50*	191-252	191-252	8.9469	8.2971	1.4	0.4	0.9	[20]
R504	R32/R115	48.20	51.80	0.734	0.266	79.22	215.85	356.20*	212-284	212-284	8.5395	7.9487	1.4	0.7	-1.8	[20]
R31/R114 azeotrope		55.10	44.90	0.754	0.246	93.71	260.65	424.70*	203-312	203-312	7.7436	7.1768	1.4	1.6	-3.8	[20]
R115/R152a azeotrope		83.90	16.10	0.690	0.310	127.10	231.15	363.55*	197-300	197-300	7.0503	6.8073	1.4	1.6	-4.3	[20]
R32/R12 azeotrope		74.25	25.75	0.870	0.130	60.94	219.45	360.86*	202-286	202-286	8.1574	7.0431	1.4	0.9	-2.8	[20]

(*) Critical Temperature calculated by Kay's rule [1]

Evaluation of the correlation parameters for binary mixtures

For the investigated mixtures the value of C_m=1.4 was obviously retained in equation (4) and, following the same regression analysis and computations as performed for the hydrocarbon refrigerants, new correlation parameters A_m and B_m were obtained. Table II summarizes the comparison results between predicted and experimental data, in terms of mean and maximum deviations, which indicate that equation (4) could be employed satisfactorily also for mixtures. To turn now the correlation of pratical use, the A_m and B_m parameters must be evaluated independently from regression routines. The following suggestions are hence recommended:

- to evaluate A_m as a mole fraction average from the corresponding values A_1 and A_2, which are computed by equation (6):

$$A_m = A_1\ X1 + A_2\ X2 \tag{7}$$

where X1 and X2 indicate the molar fractions;

- once A_m is determined, to evaluate B_m in the same way as described before, i.e., by employing few selected experimental data of viscosity or even only one if necessary;

Table IV shows these values of B_m together with the mean and maximum deviations between predicted and experimental data.

CONCLUSIONS

The following conclusions can be drawn from this study:

- accurate prediction of dynamic viscosity in terms of reduced temperature can be carried out by means of equation (4);
- the correlation itself may be used satisfactorily for organic and inorganic compounds, elements, as well as for hydrocarbon refrigerants and their binary azeotrope mixtures;
- the correlation is valid along the saturation line, from the reduced normal melting point to well above the reduced normal boiling point, i.e., up to Tr=0.9;
- the dimensionless parameter C (or C_m) seems structure-sensitive and hence a unique value for all the compounds exhibiting similar molecular-structure may be set. For the investigated hydrocarbon refrigerants, the optimum value is 1.4;
- the correlation parameter A (or A_m) may be evaluated as a function of molecular weight, of the normal boiling point and the critical temperature. For hydrocarbon refrigerants this relationship is expressed by equation (6) while for their binary mixtures, in addition to equation (6) that must be used for the components, equation (7) is also needed;
- once A (or A_m) is determined and C is set equal to 1.4, the correlation parameter B (or B_m) may be evaluated from equation (4) by selecting few experimental data of viscosity or even only one if necessary;
- the mean deviations between predicted and experimental data are of the same order of magnitude of the best measurement accuracies.

Table III - Pure halocarbons refrigerants: mean Δ% and maximum $\Delta\%_M$ deviations between the dynamic viscosity data calculated by equations (4) and (6) and the experimental ones; B is calculated through a few (even only one) experimental μ values.

Refr.	Chem. Form.	B, 1/cP	Δ%	$\Delta\%_M$
R10	CCl_4	6.2161	2.8	3.9
R11	CCl_3F	5.8432	2.2	-6.1
R12	CCl_2F_2	5.4124	1.9	-3.5
R13	$CClF_3$	4.8332	0.5	-1.1
R13B1	$CBrF_3$	4.2716	1.1	-3.1
R20	$CHCl_3$	6.3213	0.7	-1.7
R21	$CHCl_2F$	6.2591	1.9	-4.2
R22	$CHClF_2$	6.2198	1.8	3.3
R23	CHF_3	6.3543	1.1	2.2
R30	CH_2Cl_2	6.9970	2.1	-4.8
R31	CH_2ClF	7.2104	1.8	-5.4
R32	CH_2F_2	8.0234	1.3	3.5
R113	CCL_2FCClF_2	6.0659	1.1	5.5
R114	$CClF_2CClF_2$	5.8069	1.5	5.3
R115	$CClF_2CF_3$	5.5811	1.8	-5.4
R152a	CH_3CHF_2	7.2724	3.7	-12.9

Table IV - Binary refrigerant mixtures: mean Δ% and maximum $\Delta\%_M$ deviations between the dynamic viscosity data calculated by equations (4), (6) and (7) and the experimental ones; B_m is calculated through a few (even only one) experimental μ values.

Mixture	Composition	B_m, 1/cP	Δ%	$\Delta\%_M$
R500	R12/R152a	6.2183	2.4	-7.4
R502	R22/R115	5.9697	1.6	2.4
R503	R23/R13	4.9363	5.9	10.5
R504	R32/R115	6.7060	2.6	-7.3

Mixture	Composition	B_m, 1/cP	Δ%	$\Delta\%_M$
R31/R114 azeotrope		6.8000	2.7	-5.4
R115/R152a azeotrope		6.0581	3.9	-12.5
R32/R12 azeotrope		6.9157	1.0	-3.5

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