International Journal of Thermophysics, Vol. 7, No. 5, 1986

A New Correlation for the Viscosity of Gaseous Fluorocarbon Refrigerants

K. Nagaoka,[1] Y. Tanaka,[2] H. Kubota,[2] and T. Makita[2]

Received November 29, 1985

A new generalized correlation is presented for the low-pressure gaseous viscosity of fluorocarbon refrigerants. The following empirical equation is obtained based on the most reliable experimental data for 16 fluorocarbons:

$$\eta\xi = (0.5124T_r - 0.0517)^{0.82} Z_c^{-0.81}$$

where η is the viscosity in μPa · s and ξ is the viscosity parameter defined using the critical temperature T_c in K, the critical pressure P_c in MPa, and the molar mass M in g · mol^{-1} as follows:

$$\xi = T_c^{1/6} M^{-1/2} P_c^{-2/3}$$

The applicable ranges are $0.6 < T_r < 1.8$ and $0.253 < Z_c < 0.282$. The availability of the correlating equation for both pure fluorocarbons and their mixtures has been investigated based on the experimental data of these authors and those in the literature. It is found that the present correlation is useful for the prediction of the viscosity of pure fluorocarbons and their binary mixtures at atmospheric pressure with mean deviations less than 1.6%.

KEY WORDS: dimensional analysis; fluorocarbon; gas mixture; generalized correlation; principle of corresponding states; refrigerant; viscosity.

1. INTRODUCTION

Recently fluorocarbons are used as the working fluids not only in refrigeration processes but also in various energy systems for the utilization of low-quality energies such as solar, geothermal, ocean-thermal, and waste heats from industries. Therefore, it is essential to accumulate reliable

---

[1] The Graduate School of Science and Technology, Kobe University, Kobe 657, Japan.
[2] Department of Chemical Engineering, Kobe University, Kobe 657, Japan.

0195-928X/86/0900-1023$05.00/0

experimental data and to establish predictive methods for the thermophysical properties of fluorocarbons. As for the viscosity, although the experimental data for pure fluorocarbons are comparatively abundant, the experimental data for fluorocarbon mixtures are scarce except for a limited number of azeotropic mixtures. There is no reliable predictive method for the viscosity of both pure fluorocarbons and their mixtures. Therefore, it would be important for engineering use to establish simple predictive methods for the gaseous viscosity.

In the present work, applying the dimensional analysis proposed by Thodos and his co-workers [1], a new generalized correlation is presented based on the most reliable viscosity data collected. The availability of this correlation for both pure fluorocarbons and their mixtures is discussed.

2. DIMENSIONAL ANALYSIS FOR THE VISCOSITY CORRELATION

Thodos and his co-workers [1–3] developed a series of generalized correlations for the transport properties of fluids based on the principle of corresponding states by means of dimensional analysis. According to them, the viscosity of gases at low density is expressed by the following equations:

$$\eta \cdot \xi = k T_r^{\,m} Z_c^{\,n} \tag{1}$$

$$\xi = T_c^{1/6} M^{-1/2} P_c^{-2/3} \tag{2}$$

where η is the viscosity in μPa · s, T_c is the critical temperature in K, T_r is the reduced temperature, P_c is the critical pressure in MPa, and M is the molar mass in $\mathrm{g \cdot mol^{-1}}$. The quantity ξ is the viscosity parameter characteristic to each substance and its physical meaning is supposed to be the reciprocal of the critical viscosity η_c. The quantity Z_c is the compressibility factor at the critical point and is a measure of the polarity of molecules. If the unknown constants k, m, and n in Eq. (1) are determined empirically, it is possible to calculate the viscosity of any dilute gas.

Thodos and his co-workers classified substances into several groups and determined the coefficients of Eq. (1) for each group. The viscosity of each group was correlated successfully with a mean deviation of about 2%. However, as the correlations of Thodos et al. were based on experimental data obtained before 1959, considerable uncertainties would be included not only in the viscosity data but also in the values of the critical constants. Therefore, it is not desirable at present to use their correlation without any critical evaluation, especially in the case of the prediction of viscosity of fluorocarbons.

However, the Rayleigh's dimensional analysis method is considered to be effective for the establishment of a new correlation of the viscosity data. Recently, the authors measured the viscosity of five binary gaseous mixtures of fluorocarbons containing R12, R13, R13B1, R14, R22, and R152a at atmospheric pressure [4]. In the analysis of the results, it is found that the method of dimensional analysis is useful as a correlation method. Therefore, it seems worthwhile to revise the empirical coefficients of Eq. (1) based on the latest and most reliable viscosity data and critical constants of gaseous fluorocarbon refrigerants.

3. CORRELATION FOR PURE FLUOROCARBONS

In the present work, 16 sets of the most reliable experimental data and correlated values for the viscosity of gaseous fluorocarbons have been selected from the literature [5–14] as shown in Table I. The new equation thus obtained is as follows:

$$\eta\xi = (0.5124T_r - 0.0517)^{0.82}\, Z_c^{-0.81} \tag{3}$$

where the effective ranges are $0.6 < T_r < 1.8$ and $0.253 < Z_c < 0.282$. Equation (3) reproduces 116 data points adopted with a mean deviation of 1.62% and a maximum deviation of 4.97%. Table II shows the results of the present correlation and a comparison with the deviations calculated by the equation of given by Stiel and Thodos [3]. Figures 1 and 2 illustrate the relations of $\eta\xi Z_c^{0.81}$ versus T_r for eight methane derivatives and for five ethane derivatives and others, respectively. It is found that Eq. (3) is useful for the prediction of the viscosity of pure gaseous fluorocarbons, although systematic deviations are found only in R115.

4. EXTENSION TO FLUOROCARBON MIXTURES

As is generally known, the principle of corresponding states is also applicable for gaseous mixtures. It is observed in many thermophysical properties of fluids that a mixture behaves as a hypothetical pure component with a set of pseudocritical constants. The prediction of viscosity of mixtures was studied by Dean and Stiel [15], who showed that the viscosity of mixtures can be calculated with mean deviations of about 2%. In this work, the applicability of Eq. (3) to fluorocarbon mixtures has been investigated.

In order to apply Eq. (3) to mixtures, it is required to obtain the pseudocritical constants of mixtures at every composition. In this work, the pseudocritical constants were calculated by three methods proposed by

Table I. Physical Constants and Sources of Viscosity Data for Fluorocarbons

Fluorocarbon	M ($g \cdot mol^{-1}$)	T_c (K)	P_c (MPa)	Z_c	ξ	No. of data points	Temp. (K)	Ref. No.
R11 (CCl_3F)	137.368	471.2	4.41	0.279	0.08850	6	298–423	5
R12 (CCl_2F_2)	120.914	384.95	4.125	0.279	0.09536	8	248–423	6
R13 ($CClF_3$)	104.459	302.0	3.92	0.282	0.10194	9	223–423	7
R13B1($CBrF_3$)	148.910	340.2	3.97	0.281	0.08636	7	298–448	8
R14 (CF_4)	88.005	227.6	3.74	0.276	0.10932	9	234–423	5
R21 ($CHCl_2F$)	109.923	451.6	5.17	0.271	0.09132	7	303–423	9
R22 ($CHClF_2$)	86.469	369.2	4.977	0.267	0.09881	8	248–423	10
R23 (CHF_3)	70.014	298.8	4.84	0.259	0.10800	9	230–423	7
R113 (CCl_2FCClF_2)	187.376	487.3	3.41	0.277	0.09044	6	323–423	19
R114 ($CClF_2CClF_2$)	170.922	418.8	3.26	0.275	0.09516	8	298–423	19
R115 ($CClF_2CF_3$)	154.467	353.2	3.157	0.271	0.09940	6	273–398	19
R142b (CH_3CClF_2)	100.496	410.2	4.12	0.279	0.10580	6	298–423	12
R152a (CH_3CHF_2)	66.050	386.6	4.50	0.253	0.12184	7	273–423	12
RC318 (cyclo-C_4F_8)	200.031	388.37	2.777	0.279	0.09667	7	273–423	9
R500[a]	99.31	378.7	4.43	0.281	0.10007	7	273–423	13
R502[b]	111.6	355.37	4.27	0.277	0.09883	6	273–398	14

[a] R12 and R152a azeotropic mixture.

[b] R22 and R115 azeotropic mixture.

Table II. Deviation of Generalized Correlation

Fluorocarbon	Deviation (%)			
	Present work, Eq. (3)		Ref. 3	
	Mean	Max.	Mean	Max.
R11	1.21	1.93	5.18	6.42
R12	1.77	3.48	4.55	8.25
R13	1.36	3.37	4.92	5.86
R13 B1	2.27	2.76	5.36	5.43
R14	1.08	2.27	6.94	8.49
R21	0.82	1.15	3.73	5.17
R22	0.77	1.37	2.87	3.73
R23	1.86	2.96	3.66	5.38
R113	0.99	1.60	5.40	6.71
R114	2.30	2.82	7.16	8.36
R115	4.68	4.97	9.40	10.22
R142b	1.47	2.20	3.62	3.73
R152a	1.29	2.70	3.24	4.21
RC318	2.44	3.73	7.38	9.31
R500	1.44	2.06	3.90	4.02
R502	0.42	0.53	5.59	6.07

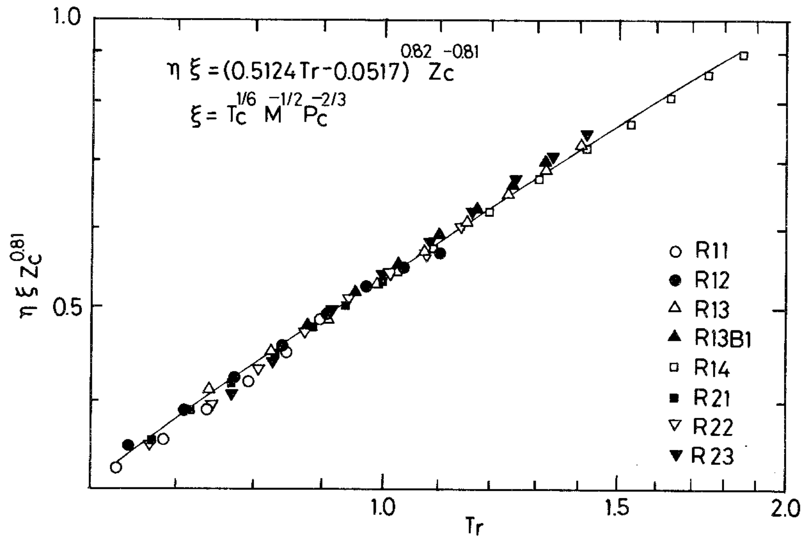

Fig. 1. Relation between T_r and $\eta\xi Z_c^{0.81}$ for methane-derivative refrigerants.

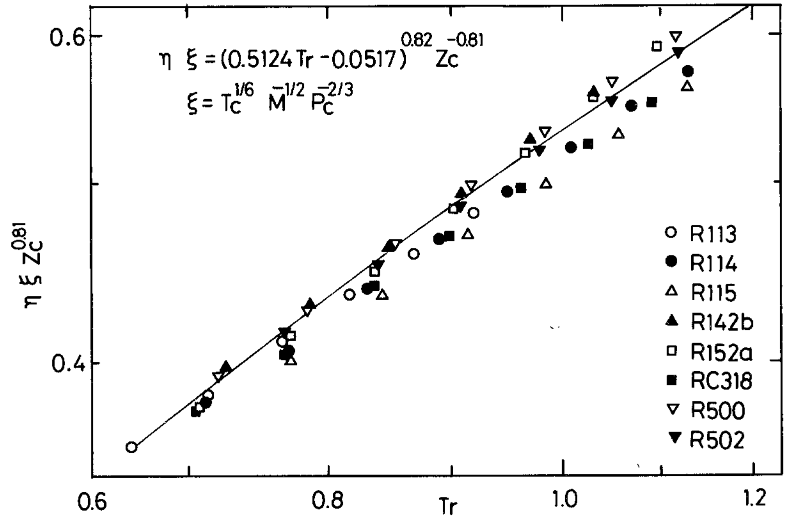

Fig. 2. Relation between T_r and $\eta\xi Z_c{}^{0.81}$ for ethane-derivative and other refrigerants.

Kay [16], Prausnitz and Gunn [17], and Joffe [18]. The molar masses of mixtures were determined by the mole fraction average method and the viscosity parameters of mixtures were determined for each composition by the following equation:

$$\xi_m = T_{cm}{}^{1/6}\left(\sum_i x_i M_i\right)^{-1/2} P_{cm}{}^{-2/3} \tag{4}$$

where the subscript m denotes the values of mixtures.

The predicted viscosity values thus obtained were compared with our experimental data [4] and those of Srichand et al. [19]. The results of comparison are summarized in Table III for six binary gaseous mixtures. Figure 3 indicates the deviations at 298.15K using the pseudocritical constants calculated by Joffe's method. In the systems except R22 + R14, the predicted values agreed satisfactorily with the experimental data, with mean deviations of about 0.3% and maximum deviations within 1.4%. Regarding the calculation methods of pseudocritical constants, no remarkable difference was found among the three methods. On the other hand, it is noticeable that the deviations in the R22 + R14 mixture are considerably larger than those in the other systems. This mixture consists of polar and nonpolar molecules and the critical temperature of R14 is considerably lower than those of other fluorocarbons. The large deviation

Table III. Prediction of Viscosity of Mixtures

System	Temp. (K)	Method	Deviation (%) Mean	Max.	Ref. No.
R22 + R12	298–323	Kay	0.22	0.72	4
		Prausnitz	0.31	0.72	
		Joffe	0.37	0.72	
R22 + R13	298–323	Kay	0.25	0.37	4
		Prausnitz	0.33	0.56	
		Joffe	0.25	0.37	
R22 + R13B1	298–323	Kay	0.38	0.94	4
		Prausnitz	0.46	0.94	
		Joffe	0.38	0.94	
R22 + R14	298–323	Kay	2.66	4.76	4
		Prausnitz	2.22	4.11	
		Joffe	2.03	3.86	
R22 + R152a	298–323	Kay	0.44	1.12	4
		Prausnitz	0.30	0.89	
		Joffe	0.34	0.97	
R22 + R12	Near 300	Kay	0.32	0.82	19
		Prausnitz	0.94	1.37	
		Joffe	0.59	1.00	

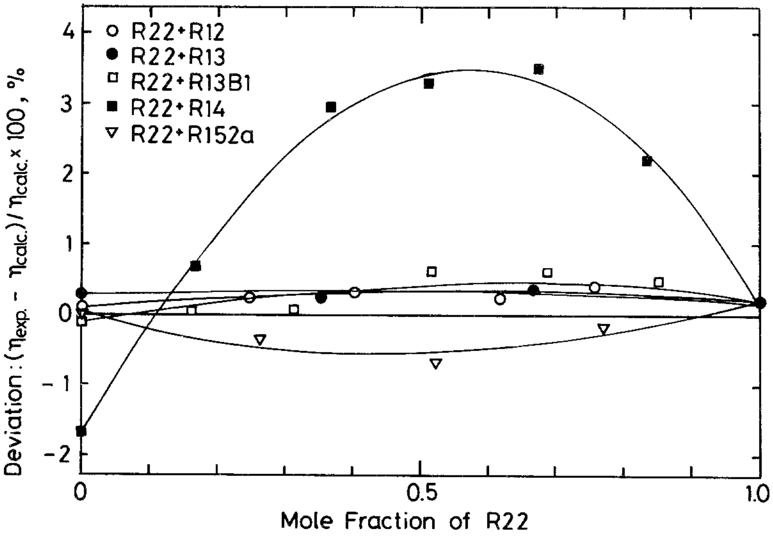

Fig. 3. Deviation between experimental and estimated viscosity of mixtures at 298.15 K.

mentioned above would be due to the large difference of physical properties between the component gases.

In order to express the viscosity of an R22 + R14 mixture satisfactorily, a separate correlation has been made and the coefficients in Eq. (1) are determined, using the pseudocritical values calculated by the Joffe method, as follows:

$$\eta\xi = (0.5751 T_r - 0.3362)^{0.3711} Z_c^{-0.7544} \tag{5}$$

Equation (5) is found to reproduce the viscosity of the R22 + R14 mixture with a mean deviation of 0.62% and a maximum deviation of 1.70%.

5. CONCLUSION

A new empirical equation is presented for the prediction of low-pressure gaseous viscosity of fluorocarbon refrigerants. The equation could be applicable to both pure gases and binary gaseous mixtures for engineering use. However, in the case of mixtures which consist of polar and nonpolar components, this generalized method would be less reliable, as in the case of the R22 + R14 system. Therefore, it is concluded that the application of this method to the mixtures should be limited to the systems consisting of fluorocarbons with similar physical properties.

REFERENCES

1. L. I. Stiel and G. Thodos, *AIChE J.* **7**:611 (1961).
2. L. W. Flynn and G. Thodos, *J. Chem. Eng. Data* **6**:457 (1961).
3. L. I. Stiel and G. Thodos, *AIChE J.* **8**:229 (1962).
4. K. Nagaoka, Y. Tanaka, H. Kubota, and T. Makita, Preprint of the Tokushima Meeting of the Society of Chemical Engineers of Japan, 5 (1984).
5. Y. Tanaka, *Refrigeration* **52**:34 (1977).
6. Japanese Association of Refrigeration, *Thermophysical Properties of Refrigerant 12* (JAR, Tokyo, 1981).
7. Japan Society of Mechanical Engineers, *Thermophysical Properties of Fluids* (JSME, Tokyo, 1983).
8. M. Takahashi, C. Yokoyama, and S. Takahashi, Preprint of the 25th High Pressure Conference of Japan, 226 (1984).
9. American Society of Heating, Refrigerating and Air-Conditioning Engineers Inc., *Thermophysical Properties of Refrigerants* (ASHRAE, New York, 1976).
10. Japanese Association of Refrigeration, *Thermophysical Properties of Refrigerant 22* (JAR, Tokyo, 1975).
11. M. Takahashi, C. Yokoyama, and S. Takahashi, *Kagaku Kogaku Ronbunshu* **15**:155 (1985).
12. M. Takahashi and S. Takahashi, Preprint of the 23rd High Pressure Conference of Japan, 208 (1982).

13. M. Takahashi, C. Yokoyama, and S. Takahashi, Preprint of the 24th High Pressure Conference of Japan, 264 (1983).
14. M. Takahashi and H. Iwasaki, Preprint of the 15th High Pressure Conference of Japan, 57 (1973).
15. D. E. Dean and L. I. Stiel, *AIChE J.* **11**:526 (1965).
16. W. B. Kay, *Ind. Eng. Chem.* **28**:1014 (1936).
17. J. M. Prausnitz and R. D. Gunn, *AIChE J.* **4**:430 (1958).
18. J. Joffe, *Ind. Eng. Chem.* **39**:837 (1947).
19. M. Srichand, M. A. Tirunarayanan, and A. Ramachandran, *ASHRAE J.* **12**:61 (1970).