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Viscosity of Gaseous Mixtures of HCFC-22 + HCFC-142b at Pressures to 6.3 MPa

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Abstract The gaseous viscosity of non-azeotropic mixtures of HCFC-22 and HCFC-142b was measured by an oscillating disk viscometer of the Maxwell type from 298.15 K to 423.15 K and at pressures up to 6.3 MPa. The viscosity at approximately atmospheric pressure was predicted with a maximum deviation of 1.84 % and an average deviation of 0.61 % by the Sutherland-Thiesen equation, coefficients of which were determined by the Brokaw method. An empirical equation was developed for the viscosity as a function of composition, temperature, and density. This equation reproduced the observed viscosity with a maximum deviation of 5.61 % and an average deviation of 1.14 %.

Keywords HCFC-22 · HCFC-142b · Sutherland-Thiesen equation · Viscosity

1 Introduction

The viscosity of gases, like the thermal conductivity and diffusion coefficient, is an important thermophysical property in chemical engineering and in molecular physics. Accurate experimental data of the gaseous viscosity are used to obtain information about intermolecular forces and more practically to design unit operations in chemical engineering.

Recently prediction models for transport properties, including viscosity and thermal conductivity of dilute and/or dense fluid mixtures, have been developed using schemes based on molecular physics developed by Kestin, Wakeham, Vesovic, and others [1–5], which will be described as the Vesovic–Wakeham method in this article,

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and by Moghadasi et al. [6,7]. The method of Moghadasi et al. can be applied only for gaseous mixtures of HCFCs at normal pressure and will be modified to predict viscosities of dense fluid mixtures of HCFCs in the future. The Vesovic–Wakeham method can be used to predict transport properties of dense fluid mixtures at moderately low uncertainty with the use of the density dependence of the viscosity of the pure fluids, the viscosity of the fluid mixture at normal pressure, and the density of the dense fluid mixture. To evaluate applicability of the Vesovic–Wakeham method thoroughly, accurate experimental viscosity data of gaseous mixtures consisting of molecules having different types of molecular shape and polarity have been required.

In our previous studies, we have reported experimental viscosity data of pure and mixed dense fluids, of pure HCFCs, of mixed HCFCs, and of some mixtures containing non-polar and polar fluids [8–26]. As part of a continuing study of the viscosity of dense fluid mixtures, measurements of the viscosity of gaseous mixtures of HCFC-22 (chlorodifluoromethane) + HCFC-142b (1-chloro-1,1-difluoroethane) at (298.15, 323.15, 348.15, 373.15, 398.15, and 423.15) K over a pressure range from 0.1 MPa to 6.27 MPa are reported in this paper. The reference data used for pure HCFC-22 and HCFC-142b were taken from our previous studies [10,13].

2 Experimental

The viscosity was measured with an oscillating-disk viscometer of the Maxwell type. The gas density at the experimental conditions of the viscosity measurements was measured with a high-pressure gas pipette. The density values have an estimated uncertainty of $0.1\,\mathrm{kg \cdot m^{-3}}$. In order to minimize the uncertainty due to the density value, the REFPROP program [27] was also used to determine the gas density. Use of the REFPROP program for the density determination provides a more reliable base for future studies of multicomponent mixtures. The experimental apparatus and procedures were the same as those described in previous studies [8,9,13,17]. The apparatus constant of the viscometer at the experimental temperature and pressure conditions was determined by using the viscosity data of nitrogen taken from Stephan et al. [28] and the nitrogen gas density data from Jacobsen and Stewart [29]. There are more recent and certainly improved studies of the viscosity and density of nitrogen [30,31]. We obtained the apparatus constant by using these data and found the new data will not change the apparatus constant in the present experimental region. The samples were prepared in a sample vessel by first charging the less volatile constituent gas (HCFC-142b) and then the more volatile one (HCFC-22). When the sample gas mixture was loaded into the viscometer vessel, the temperatures of the sample vessel and the connecting tubing between the sample vessel and the viscometer vessel were kept at about 423 K to prevent condensation. Temperature and pressure values have an uncertainty of ±0.01 K and ±0.5 kPa, respectively. The compositions of the sample mixtures were determined by weighing. The uncertainty of the composition determination was estimated to be less than 10^{-4} mole fraction. The estimated uncertainty of the present viscosity data is within ±0.5 % from the consideration of the errors in the measurements of the amplitude and period of oscillation, the temperature, the pressure, and the composition.

HCFC-22 and HCFC-142b were supplied by Asahi Glass Co.Ltd. The purity of both samples, certified by the supplier, was approximately 99.9 mol%. The samples were further purified by distillation several times before preparing mixture samples.

3 Results and Discussion

The experimental results are presented in Table 1. For comparison purposes, both the density values from the gas pipette measurement and from the REFPROP program are listed. As can be seen, agreement is quite good. It should be pointed out that the deviations between calculated density values and experimental results increased rapidly with increases in density at 398.15 K and 423.15 K and at densities above about 180 $\text{kg}\cdot\text{m}^{-3}$ for the mixture with an HCFC-22 mole fraction of 0.2615. This may suggest that these experimental regions are close to the critical point of the mixture. Excluding the experimental conditions at 398.15 K and 423.15 K and at densities above about 180 $\text{kg}\cdot\text{m}^{-3}$ for the mixture with an HCFC-22 mole fraction of 0.2615, the agreement between REFPROP and experimental density is as follows: a maximum deviation of 1.4 %, an absolute average deviation of 0.26 %, and an average deviation of −0.01 %. Based on the reason mentioned above, the density values from the REFPROP program will be used in the data analysis. The viscosity at normal pressure (0.1 MPa) is plotted against the mole fraction of HCFC-22 in Fig. 1. High-pressure viscosity versus pressure or density plots for the mixture with an HCFC-22 mole fraction of 0.5169 are shown in Figs. 2 and 3, respectively. As shown in Fig. 2a, a negative initial-density dependence was observed at 298.15 K and 323.15 K for this mixture. The other two mixtures also show the negative initial-density dependence at 298.15 K and 323.15 K. This type of initial-density dependence has been observed for pure fluids and gaseous mixtures at low temperature [8–18,20–26,32–42]. The viscosity in the zero-density limit, which has a theoretical basis as the standard value for viscosity analysis, can be obtained by extrapolation. Great care should be required for extrapolation in the case that the viscosity isotherm showing a negative initial-density dependence. In the present study, the number of data in the low-density region is not sufficient to accurately determine the viscosity in the zero-density limit by extrapolation. Therefore, the viscosity at 0.1 MPa was used as the standard value in the following analysis.

The normal pressure viscosity was compared with that calculated by the Sutherland-Thiesen equation [43]:

$$\eta^0 = \frac{\eta_1^0}{1 + \phi_{12}(y_2/y_1)} + \frac{\eta_2^0}{1 + \phi_{21}(y_1/y_2)} \tag{1}$$

where η_i^0 is the viscosity of component i, y_i is the mole fraction of component i, ϕ_{12}, ϕ_{21} are interaction coefficients, and subscripts 1 and 2 represent HCFC-22 and HCFC-142b, respectively. When values of the coefficient were determined with the Brokaw method [44], the deviations were 0.61 % on average and 1.84 % at maximum. The parameters of the Stockmayer potential for HCFC-22 and HCFC-142b required for the Brokaw method were determined in our previous studies [10,13] and are shown in Table 2. The lines in Fig. 1 show the calculated results from the Sutherland-Thiesen

Table 1 Experimental results for the viscosity of HCFC-22 + HCFC-142b mixtures

P (MPa)	ρ_{exp} ($\mathrm{kg \cdot m^{-3}}$)	ρ_{REF} ($\mathrm{kg \cdot m^{-3}}$)	η (μPa · s)
HCFC-22 (0.2615) + HCFC-142b (0.7385)			
298.15 K			
0.1018	4.070	4.072	11.09
0.2056	8.450	8.441	11.04
0.2992	12.59	12.60	11.02
0.3573	15.25	15.30	11.03
0.4064	17.60	17.66	11.00
323.15 K			
0.1018	3.732	3.734	12.06
0.1024	3.755	3.757	12.07
0.2000	7.459	7.470	12.03
0.2994	11.38	11.40	12.01
0.3966	15.36	15.40	12.00
0.4929	19.48	19.54	11.99
0.5933	23.99	24.06	11.99
0.6870	28.43	28.51	12.02
0.7937	33.76	33.88	12.11
348.15 K			
0.1021	3.463	3.462	12.95
0.1990	6.847	6.839	12.94
0.2969	10.36	10.35	12.93
0.4462	15.92	15.91	12.94
0.5908	21.56	21.55	12.97
0.7369	27.57	27.56	12.97
0.8873	34.14	34.11	13.01
1.036	40.96	41.03	13.02
1.182	48.22	48.32	13.07
1.334	56.48	56.56	13.12
1.489	65.73	65.84	13.25
373.15 K			
0.1022	3.224	3.224	13.77
0.2982	9.612	9.610	13.80
0.4936	16.27	16.27	13.81
0.6952	23.40	23.49	13.81
0.8767	30.25	30.33	13.87
0.8898	30.77	30.83	13.88
1.085	38.57	38.63	13.94
1.281	46.94	46.97	14.03
1.483	56.20	56.20	14.13
1.689	66.30	66.42	14.26

Table 1 continued

P (MPa)	ρ_{exp} (kg · m^{-3})	ρ_{REF} (kg · m^{-3})	η (μPa · s)
1.897	77.72	77.77	14.39
2.115	91.26	91.15	14.61
2.299	104.3	104.1	14.86
2.467	117.9	117.7	15.16
398.15 K			
0.1016	2.997	2.997	14.72
0.1020	3.006	3.007	14.70
0.2789	8.354	8.354	14.75
0.4680	14.28	14.26	14.77
0.6541	20.27	20.27	14.82
0.9089	28.88	28.88	14.85
1.153	37.53	37.58	14.93
1.397	46.73	46.78	15.04
1.608	55.16	55.20	15.10
1.821	63.97	64.20	15.25
2.031	73.54	73.65	15.42
2.251	84.17	84.28	15.60
2.442	94.09	94.21	15.76
2.637	105.1	105.2	15.98
2.835	117.2	117.3	16.29
3.101	136.0	135.8	16.73
3.322	154.3	153.7	17.24
3.481	170.2	168.6	17.73
3.698	196.3	193.4	18.58
3.854	222.4	216.3	19.53
3.998	261.2	245.1	21.01
423.15 K			
0.1021	2.829	2.829	15.79
0.3940	11.13	11.14	15.81
0.6865	19.80	19.84	15.88
0.9786	28.81	28.92	15.94
1.273	38.54	38.52	16.04
1.569	48.60	48.69	16.18
1.878	59.81	59.92	16.32
2.158	70.63	70.73	16.50
2.436	81.74	82.14	16.71
2.797	98.10	98.13	17.03
3.152	115.5	115.4	17.43
3.440	131.6	130.8	17.85

Table 1 continued

P (MPa)	ρ_{exp} (kg · m^{-3})	ρ_{REF} (kg · m^{-3})	η (μPa · s)
3.680	145.8	144.8	18.22
3.921	160.9	160.2	18.68
4.227	184.1	182.0	19.39
4.504	207.1	204.7	20.23
4.740	230.5	226.9	21.20
4.956	254.1	250.4	22.20
5.183	285.1	279.4	23.61
5.286	294.5	294.4	24.39
HCFC-22 (0.5169) + HCFC-142b (0.4831)			
298.15 K			
0.1015	3.896	3.896	11.64
0.1771	6.906	6.907	11.63
0.2487	9.867	9.853	11.60
0.3239	13.12	13.06	11.59
0.3915	16.21	16.04	11.58
0.4531	19.06	18.86	11.57
323.15 K			
0.1030	3.663	3.630	12.62
0.1900	6.762	6.788	12.61
0.2762	10.03	10.01	12.59
0.3734	13.80	13.75	12.59
0.4732	17.99	17.73	12.58
0.5755	22.07	21.98	12.57
0.6706	26.30	26.10	12.58
0.7710	30.74	30.64	12.59
0.8671	35.23	35.19	12.60
348.15 K			
0.1014	3.305	3.305	13.54
0.1986	6.548	6.548	13.55
0.3454	11.61	11.6	13.55
0.4908	16.98	16.79	13.55
0.6372	22.25	22.24	13.54
0.7817	27.87	27.85	13.58
0.9302	33.93	33.90	13.65
1.077	40.31	40.20	13.65
1.262	48.67	48.66	13.70
1.408	56.00	55.83	13.77
1.552	63.84	63.42	13.86

Table 1 continued

P (MPa)	ρ_{exp} $(\mathrm{kg \cdot m^{-3}})$	ρ_{REF} $(\mathrm{kg \cdot m^{-3}})$	η $(\mathrm{\mu Pa \cdot s})$
373.15 K			
0.1015	3.079	3.079	14.51
0.2943	9.101	9.089	14.49
0.4900	15.46	15.43	14.52
0.6862	22.09	22.04	14.54
0.879	28.79	28.83	14.58
1.080	36.34	36.25	14.64
1.277	43.98	43.90	14.76
1.472	51.98	51.94	14.80
1.666	60.38	60.34	14.86
1.862	69.61	69.44	15.02
2.065	80.02	79.59	15.09
2.295	92.76	92.19	15.37
2.448	102.0	101.4	15.42
398.15 K			
0.1018	2.886	2.889	15.47
0.3457	9.975	9.989	15.48
0.5915	17.39	17.42	15.53
0.8347	25.01	25.08	15.57
1.081	33.05	33.17	15.62
1.326	41.40	41.59	15.72
1.569	50.20	50.36	15.84
1.824	59.84	60.06	15.96
2.121	71.87	72.10	16.16
2.376	82.87	83.19	16.32
2.634	94.96	95.24	16.55
2.884	107.4	107.9	16.80
3.195	124.9	125.2	17.16
3.432	139.6	139.9	17.52
3.714	159.3	159.6	18.08
3.961	179.5	179.6	18.65
4.222	204.9	204.6	19.52
4.446	231.6	231.1	20.48
423.15 K			
0.1022	2.723	2.725	16.31
0.3462	9.354	9.366	16.34
0.6420	17.67	17.69	16.39
0.9358	26.24	26.28	16.46
1.230	35.16	35.23	16.57
1.537	44.54	44.99	16.63

Table 1 continued

P (MPa)	ρ_{exp} (kg · m^{-3})	ρ_{REF} (kg · m^{-3})	η (µPa · s)
1.832	54.69	54.81	16.77
2.158	65.98	66.21	16.94
2.444	76.64	76.76	17.11
2.740	88.10	88.28	17.36
3.055	101.2	101.3	17.65
3.389	116.1	116.1	17.91
3.703	131.2	131.0	18.30
3.992	146.2	145.9	18.74
4.325	165.2	164.5	19.26
4.634	184.7	183.4	19.88
4.895	203.4	201.0	20.48
5.173	223.6	221.6	21.24
5.380	240.7	238.4	21.87
5.612	261.9	258.9	22.95
5.795	280.3	276.6	23.57
HCFC-22 (0.7502) + HCFC-142b (0.2498)			
298.15 K			
0.1017	3.754	3.756	12.19
0.1980	7.440	7.441	12.17
0.2916	11.16	11.16	12.12
0.3848	15.02	15.00	12.12
0.4773	18.96	18.97	12.10
0.5713	23.23	23.19	12.09
323.15 K			
0.1022	3.468	3.468	13.20
0.1990	6.842	6.841	13.20
0.2973	10.36	10.36	13.17
0.3987	14.09	14.10	13.18
0.5100	18.34	18.34	13.20
0.6181	22.66	22.61	13.20
0.7247	26.96	26.98	13.18
0.8284	31.41	31.41	13.20
0.9379	36.25	36.29	13.22
348.15 K			
0.1016	3.189	3.19	14.27
0.2007	6.368	6.366	14.23
0.3443	11.09	11.09	14.22
0.4908	16.07	16.06	14.24
0.6382	21.43	21.25	14.23
0.7825	26.50	26.51	14.25

Table 1 continued

P (MPa)	ρ_{exp} (kg · m^{-3})	ρ_{REF} (kg · m^{-3})	η (μPa · s)
0.9256	31.97	31.93	14.28
1.090	38.36	38.43	14.30
1.240	44.53	44.65	14.38
1.392	51.21	51.28	14.43
1.392	51.21	51.28	14.43
1.541	58.10	58.14	14.50
1.683	64.70	65.06	14.57
373.15 K			
0.1021	2.984	2.985	15.18
0.2458	7.266	7.269	15.17
0.3901	11.68	11.68	15.20
0.5834	17.74	17.75	15.21
0.7828	24.22	24.25	15.26
0.9785	30.82	30.87	15.30
1.173	37.60	37.72	15.35
1.361	44.52	44.62	15.42
1.565	52.28	52.46	15.50
1.760	60.20	60.33	15.59
1.952	68.25	68.49	15.68
2.190	79.10	79.26	15.86
2.398	89.07	89.37	16.02
2.604	99.88	100.2	16.18
2.789	110.3	110.6	16.38
2.988	121.8	122.9	16.65
398.15 K			
0.1006	2.753	2.751	16.05
0.4048	11.29	11.29	16.05
0.7799	22.29	22.32	16.12
1.036	30.15	30.20	16.19
1.374	40.98	41.08	16.28
1.667	50.88	51.02	16.42
2.016	63.22	63.56	16.57
2.376	77.16	77.41	16.79
2.813	95.48	95.73	17.11
3.115	109.3	109.6	17.41
3.403	123.5	123.8	17.84
3.825	146.7	147.1	18.27
4.086	162.7	163.3	18.65
4.343	180.2	180.9	19.18

Table 1 continued

P (MPa)	ρ_{exp} ($\mathrm{kg \cdot m^{-3}}$)	ρ_{REF} ($\mathrm{kg \cdot m^{-3}}$)	η ($\mathrm{\mu Pa \cdot s}$)
4.642	203.6	204.1	19.91
4.840	218.9	221.6	20.50
423.15 K			
0.1011	2.597	2.599	16.82
0.3907	10.18	10.19	16.87
0.6855	18.14	18.17	16.97
1.026	27.65	27.72	17.02
1.368	37.56	37.70	17.11
1.706	47.76	47.98	17.24
2.055	58.73	59.09	17.31
2.415	70.79	71.13	17.50
2.719	81.34	81.79	17.69
3.159	97.70	98.15	17.97
3.481	110.3	110.9	18.29
3.855	126.4	126.7	18.63
4.203	142.0	142.3	18.94
4.560	159.4	159.6	19.47
4.897	176.4	177.2	19.97
5.301	200.1	200.2	20.57
5.605	219.3	219.0	21.21
5.936	242.2	241.3	22.02
6.270	267.2	265.9	22.99

equation combined with the Brokaw method. The deviations of the experimental viscosities from those calculated were 1.15 % on average and 2.47 % at maximum when the coefficients, ϕ_{12}, ϕ_{21} were determined with the Wilke method [43]. Therefore, we recommend the Brokaw method to determine values of the coefficients in the Sutherland-Thiesen equation for the HCFC-22 + HCFC-142b system.

The viscosity under pressure was expressed by the following empirical equation:

$$\eta - \eta^0 = (a_1 + a_2 T_\mathrm{r}^3)\rho_{\mathrm{exr}} + (a_3 + a_4 T_\mathrm{r}^3)\rho_{\mathrm{exr}}^2 + (a_5/T_\mathrm{r})\rho_{\mathrm{exr}}^4 + a_6\rho_{\mathrm{exr}}^6 \tag{2}$$

where η and η_0 denote the viscosity at high pressure and at normal pressure, respectively. The values of η_0 were obtained from Eq. 1. T_r and ρ_{exr} are a reduced temperature and a reduced density which are defined as follows:

$$T_\mathrm{r} = T/T_\mathrm{c} \tag{3}$$

$$\rho_{\mathrm{exr}} = (\rho - \rho_0)/\rho_\mathrm{c} \tag{4}$$

where ρ and ρ_0 are the density at high pressure and at normal pressure, respectively, which are determined with the REFPROP program. The pseudo-critical temperature

Fig. 1 Viscosity of HCFC-22 + HCFC-142b mixtures at 0.1 MPa. Solid lines are the results from Eq. 1 with the Brokaw method

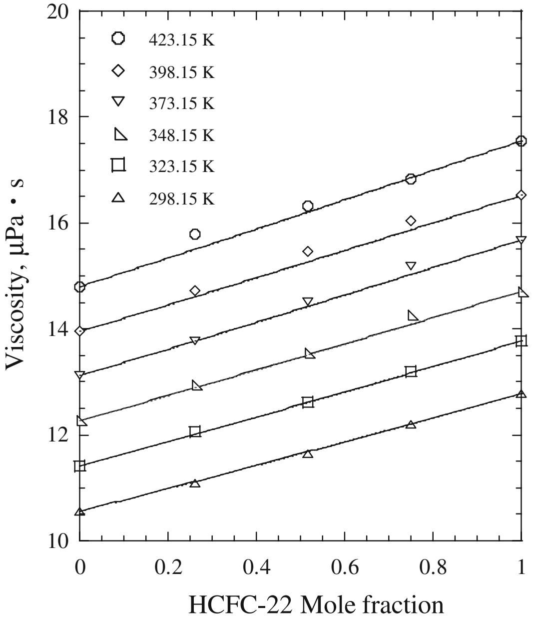
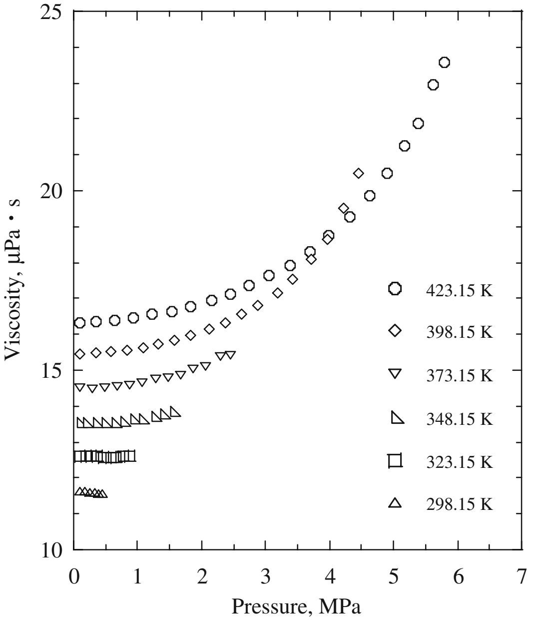


Fig. 2 Viscosity of HCFC-22 (0.5169) + HCFC-142b (0.4831) mixture as a function of pressure



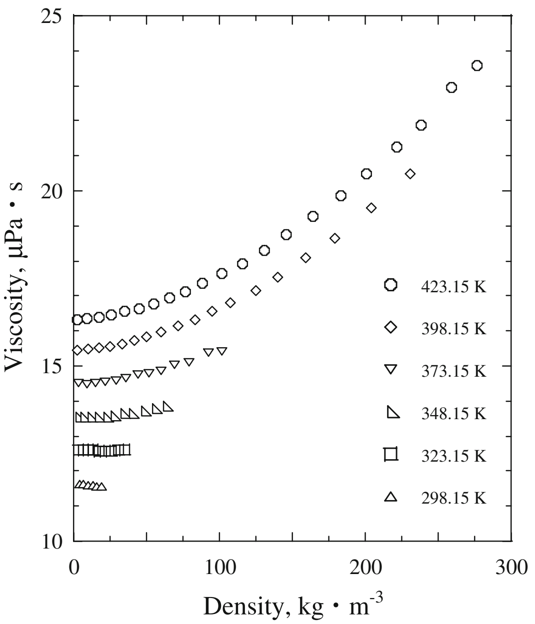


Fig. 3 Viscosity of HCFC-22 (0.5169) + HCFC-142b (0.4831) mixture as a function of density

of the mixture T_c and the pseudo-critical density of the mixture ρ_c are defined as follows:

$$T_c = y_1 T_{c1} + y_2 T_{c2} \tag{5}$$

$$\rho_c = y_1 \rho_{c1} + y_2 \rho_{c2} \tag{6}$$

The critical constants used in this study were taken from JAR Thermodynamic Tables [45] and are listed in Table 2. The values of constants $a_i (i = 1 \text{ to } 6)$ in Eq. 2 were determined using a least-squares fit and the experimental data for mixtures shown in this paper along with the pure component data in our previous studies [8,13]. The values of the constants are as $a_1 = -3.66208$, $a_2 = 4.54825$, $a_3 = 23.9247$, $a_4 = -1.53720$, $a_5 = -24.9089$, and $a_6 = 29.7747$. The deviations of experimental viscosities from Eq. 2 are shown in Fig. 4. It was found that this equation reproduced the

Table 2 Potential parameters for Sutherland-Thiesen equation and physical properties for HCFC-22 and HCFC-142b

	HCFC-22	HCFC-142b
Molar mass	83.468	100.495
T_c (K)	369.30	410.26
ρ_c $(\mathrm{kg \cdot m^{-3}})$	515	446
μ (D)	1.4	2.1
ε/k (K)	206	362
σ (nm)	0.499	0.499
δ	0.277	0.359

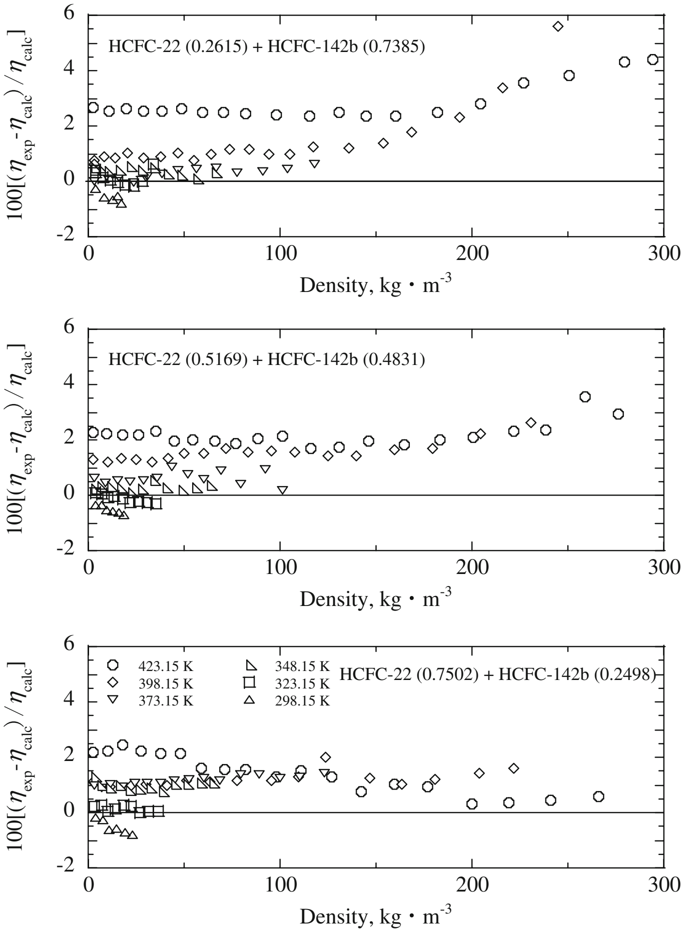


Fig. 4 Deviations of experimental viscosities from Eq. 2 for HCFC-22 + HCFC-142b mixtures

experimental values with a maximum deviation of 5.61 % and an average deviation of 1.14 % for densities less than 300 kg · m^{-3}.

4 Conclusion

In this article we report experimental viscosity values for HCFC-22 + HCFC-142b mixtures. The viscosity of the gas mixture at normal pressure can be represented with the Sutherland-Thiesen equation with the use of the Brokaw method. The viscosity under pressure can be reproduced as a function of reduced temperature and reduced density with an empirical equation. The density values were determined from the REFPROP program. Simple mixing rules are applied to the critical temperature and critical density.

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