# Viscosity of Gaseous Mixtures of HFC-125 + Propane from 298.15 to 423.15 K at Pressures to 6.7 MPa

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This paper reports experimental results for the viscosity of gaseous mixtures of HFC-125 (pentafluoroethane) + propane. The measurements were carried out with an oscillating-disk viscometer of the Maxwell type at temperatures from 298.15 to 423.15 K. The viscosity was measured for two mixtures containing 50.11 and 75.03 mol% HFC-125 in propane. The viscosity at normal pressure was analyzed with the extended law of corresponding states developed by Kestin et al., and the scaling parameters were obtained for unlike-pair interactions between HFC-125 and propane. The modified Enskog theory developed by Vesovic and Wakeham was applied to predict the viscosity for the binary gaseous mixtures under pressure. From comparisons between experimental results and calculated values of the HFC-125+ propane system, it should be concluded that the Vesovic-Wakeham method gives reliable predictions for the viscosity of a gaseous mixture containing both polar and nonpolar compounds.

**KEY WORDS:** corresponding states; Enskog theory; HFC-125; mixture model; propane; viscosity

## **1. INTRODUCTION**

Potential alternatives for chlorofluorocarbons (CFCs) and hydrochlorofluorocarbons (HCFCs) include hydrofluorocarbons (HFCs), such as HFC-125 (pentafluoroethane), HFC-134a (1,1,1,2-tetrafluoroethane), HFC-143a (1,1,1-trifluoroethane), and HFC-32 (difluoromethane), and their binary and/or ternary mixtures. Natural refrigerants such as carbon dioxide, ammonia, and propane have also been received considerable

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attention as alternative fluids. To realize optimum performance as working fluids in refrigeration cycles, blended refrigerants of an HFC and a natural refrigerant should have the possibility to become a good solution.

Transport properties, such as viscosity and thermal conductivity, of the alternative refrigerants influence the economical feasibility of heat exchangers which can show close to the theoretical efficiency of the thermodynamic cycle with the CFCs. Therefore, reliable prediction methods for the transport properties of mixtures of HFCs and natural refrigerants are required to establish the process design methodology for a search of the optimum operating conditions for refrigeration systems using mixed refrigerants.

In our previous studies, we measured the gaseous viscosity of HFC-32, HFC-134a, HFC-143a, HFC-125 [1–4], HFC-125+HFC-134a [5], HFC-125+HFC-32 [6], and HFC-134a+HFC-32 [7] systems. As part of a continuing study of the viscosity of dense fluid systems containing HFCs and natural refrigerants, measurements of the viscosity of gaseous mixtures of HFC-125+propane, made at 298.15, 323.15, 348.15, 373.15, 398.15, and 423.15K over a pressure range from 0.1 to 6.7 MPa, are reported in this paper. The reference data used for pure HFC-125 were quoted from our previous study [4] and for propane from the recommended values by Vogel et al. [8]. The viscosity data at 0.1 MPa were used to determine the scaling parameters of the unlike-pair interactions between HFC-125 and propane from the extended law of corresponding states developed by Kestin et al. [9]. The viscosity data under pressure were analyzed with the extended Enskog theory developed by Vesovic and Wakeham [10].

## 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 measurement was measured with a high-pressure gas pipette. The experimental apparatus and procedures were the same as those described in previous studies [11–16]. 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. [17] and the nitrogen gas density data from Jacobsen et al. [18]. The REFPROP program was used to determine the gas density [19]. The samples were prepared in a sample vessel by first charging the less volatile constituent gas (HFC-125) and then the more volatile one (propane). When the sample gas mixture was loaded into the viscometer vessel, the temperature of the sample vessel and the connecting tubing between the sample vessel

and the viscometer vessel were thermostatted 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%.

The HFC-125 and propane were supplied by Asahi Glass Co. Ltd. and by Takachiho Chem. Co. Ltd., respectively. The purity of both samples, certified by the suppliers, 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 for the viscosity and density of the HFC-125+ propane system are presented in Table I. The viscosity values of the mixture of 0.5011 HFC-125+0.4989 propane are shown in Figs. 1 and 2. As seen in Fig. 1, the curves as a function of pressure intersect for the isotherms from 373.15 to 423.15 K, but the curves as a function of density do not, as seen in Fig. 2. Almost the same behavior was observed in the other mixture and also for the pure HFCs and the binary HFC systems (HFC-125+ HFC-134a, HFC-125+ HFC-32, and HFC-134a+HFC-32) in our previous studies [1–6].

The viscosities of gaseous mixtures at 0.1 MPa,  $\eta_0$ , are plotted as a function of mole fraction in Fig. 3, in which the viscosities of HFC-125 [4] and propane [8] are also shown. As can be seen in Fig. 3, the shape of the curves for  $\eta_0$  is slightly monotonically convex upwards in the present experimental temperature range.

The extended law of corresponding states for the transport properties was applied to determine the scaling parameters for the binary interaction between HFC-125 and propane. The equations used are the same as given by Kestin et al. [9]. The scaling parameters of HFC-125 and propane are determined from a least-squares fit to the viscosity data of HFC-125 reported previously [4] and recommended viscosity values of propane by Vogel et al. [8]. The optimum values of the scaling parameters between the HFC-125 and propane pair interaction are determined with the use of the viscosity data measured in this study. The values of the scaling parameters obtained are shown in Table II. As can be seen, the values of the unlike-pair interaction scaling parameters deviate significantly from a simple mixing rule, which may be due to the characteristics of the interaction between polar (HFC-125) and nonpolar (propane) molecules. The

## Viscosity of Gaseous Mixtures of HFC-125 + Propane

T (K)	P (MPa)	$\rho(\mathrm{kg}\cdot\mathrm{m}^{-3})$	$\eta \left(\mu Pa \cdot s\right)$	T (K)	P (MPa)	$\rho(\mathrm{kg}\cdot\mathrm{m}^{-3})$	$\eta \left( \mu \cdot \mathbf{Pa} \cdot \mathbf{s} \right)$		
$X_{HFC-125} = 0.7503$									
298.15	0.1012	4.185	12.194	348.15	2.4924	114.3	15.547		
	0.1979	8.300	12.230		2.6606	125.2	15.834		
	0.2946	12.54	12.208		2.8047	135.2	16.044		
	0.3915	16.92	12.217		2.9288	144.2	16.289		
	0.4923	21.62	12.229		3.0794	155.9	16.661		
	0.5891	26.31	12.246		3.2306	168.6	16.972		
	0.6890	31.32	12.287		3.3832	182.5	17.395		
	0.8035	37.32	12.287						
	0.9134	43.68	12.346	373.15	0.1010	3.313	15.052		
	1.0124	55.10	12.480		0.2200	7.275	15.058		
	1.2313	62.86	12.573		0.3897	13.04	15.102		
					0.6377	21.72	15.153		
323.15	0.1022	3.887	13.191		0.8778	30.43	15.253		
	0.1990	7.650	13.168		1.1418	40.38	15.374		
	0.2949	11.46	13.200		1.4774	53.64	15.518		
	0.3911	15.37	13.212		1.7170	63.57	15.656		
	0.5194	20.73	13.225		1.9611	74.11	15.828		
	0.6363	25.76	13.249		2.1852	84.20	16.051		
	0.7664	31.55	13.262		2.4159	95.04	16.227		
	0.8832	36.93	13.339		2.6312	105.6	16.406		
	1.0051	42.74	13.394		2.9503	122.1	16.789		
	1.1251	48.68	13.460		3.2099	136.4	17.136		
	1.2497	55.10	13.514		3.4212	148.7	17.485		
	1.3712	61.63	13.612		3.6846	164.9	17.979		
	1.4863	68.10	13.675		3.9694	183.7	18.553		
	1.6189	75.93	13.788		4.2274	202.1	19.131		
	1.7361	83.23	13.874		4.4110	216.0	19.643		
	1.8644	91.71	14.039		4.6077	231.8	20.195		
	1.9921	100.7	14.201		4.8162	249.7	20.916		
	2.1016	109.0	14.352		5.0018	266.7	21.695		
	2.2116	118.0	14.520		5.1572	281.8	22.287		
	2.3492	130.2	14.823		5.3059	296.9	23.051		
348.15	0.1014	3.571	14.089	398.15	0.1018	3.125	15.886		
	0.1997	7.092	14.072		0.2625	8.132	15.938		
	0.3882	14.02	14.164		0.4886	15.33	16.018		
	0.5886	21.65	14.242		0.7334	23.34	16.057		
	0.7508	28.04	14.268		0.9770	31.53	16.154		
	0.9784	37.36	14.324		1.2183	39.89	16.222		
	1.1738	45.73	14.427		1.4766	49.09	16.355		
	1.3747	54.71	14.554		1.7125	57.76	16.471		
	1.5657	63.66	14.676		1.9443	66.52	16.662		
	1.7702	73.72	14.824		2.1768	75.56	16.724		
	1.9639	83.76	14.985		2.3889	84.05	16.936		
	2.1304	92.85	15.108		2.6224	93.65	17.152		
	2.3355	104.7	15.377		2.8889	105.0	17.342		

Table I. Experimental Viscosity Values for the HFC-125+Propane System

T (K)	P (MPa)	$\rho(\mathrm{kg}\cdot\mathrm{m}^{-3})$	$\eta \left(\mu Pa \cdot s\right)$	T (K)	P (MPa)	$\rho(\mathrm{kg}\cdot\mathrm{m}^{-3})$	$\eta \left( \mu \cdot \mathbf{Pa} \cdot \mathbf{s} \right)$
398.15	3.1783	117.7	17.649	298.15	0.3961	13.91	11.233
	3.4057	128.1	17.939		0.4994	17.83	11.234
	3.6249	138.4	18.167		0.5911	21.43	11.237
	3.8227	147.9	18.439		0.6874	25.34	11.247
	3.9929	156.4	18.677		0.7890	29.64	11.276
	4.2254	168.2	19.021		0.9282	35.84	11.314
	4.5239	184.0	19.467		1.0251	40.40	11.384
	4.6907	193.1	19.835				
	4.8046	199.5	20.020	323.15	0.1018	3.146	12.143
	5.0409	213.0	20.512		0.2139	6.692	12.129
	5.2245	223.8	20.868		0.3958	12.64	12.152
	5.4624	238.3	21.500		0.5928	19.38	12.188
	5.8015	259.7	22.396		0.7889	26.45	12.248
					0.9838	33.86	12.278
423.15	0.1024	2.955	16.711		1.1818	41.86	12.412
	0.1946	5.640	16.721		1.3749	50.19	12.550
	0.3364	9.814	16.761		1.6677	64.07	12.719
	0.5701	16.81	16.825		1.8626	74.36	12.894
	0.8129	24.24	16.920		1.9857	81.42	13.000
	1.0558	31.85	16.956		2.0714	86.64	13.144
	1.2549	38.21	17.063				
	1.4489	44.52	17.152	348.15	0.1011	2.893	12.952
	1.6400	50.84	17.239		0.1984	5.725	12.990
	1.9064	59.85	17.399		0.3055	8.898	12.968
	2.1282	67.51	17.518		0.4016	11.80	12.958
	2.3759	76.25	17.663		0.5925	17.71	13.065
	2.5915	84.02	17.807		0.7848	23.89	13.106
	2.9193	96.12	18.076		0.9860	30.61	13.183
	3.1601	105.2	18.242		1.1784	37.31	13.258
	3.2893	110.2	18.360		1.3746	44.45	13.354
	3.5521	120.5	18.589		1.5712	51.94	13.497
	3.8136	131.0	18.858		1.7723	60.00	13.618
	4.0615	141.1	19.159		1.9609	67.97	13.740
	4.2985	151.0	19.435		2.1399	75.95	13.865
	4.4926	159.2	19.623		2.2979	83.37	14.035
	4.6898	167.7	19.883		2.5010	93.52	14.304
	4.9025	177.1	20.230		2.6956	104.0	14.509
	5.0700	184.5	20.424		2.9325	117.9	14.936
	5.2858	194.3	20.774		3.1117	129.6	15.236
	5.4933	203.8	21.079				
	5.6722	212.1	21.388	373.15	0.1015	2.706	13.759
	5.8873	222.3	21.750		0.2255	6.062	13.809
					0.3977	10.82	13.827
					0.5856	16.15	13.862
X <sub>HFC-12</sub>	5 = 0.5011						
298.15	0.1020	3.428	11.222		0.7814	21.85	13.926
	0.1982	6.753	11.211		0.9805	27.83	14.010
	0.3026	10.47	11.207		1.1742	33.82	14.067

 Table I.
 (continued)

T(K)	P (MPa)	$\rho(\mathrm{kg}\cdot\mathrm{m}^{-3})$	$\eta \left(\mu Pa \cdot s\right)$	T(K)	P (MPa)	$\rho(\mathrm{kg}\cdot\mathrm{m}^{-3})$	$\eta \left( \mu \cdot \mathbf{Pa} \cdot \mathbf{s} \right)$
373.15	1.3705	40.09	14.133	398.15	3.8428	121.7	16.806
	1.5604	46.35	14.223		4.0624	131.8	17.052
	1.7539	52.95	14.335		4.3296	142.3	17.439
	1.9494	59.84	14.473		4.6523	156.8	17.924
	2.1253	66.26	14.576		5.0141	174.0	18.531
	2.3189	73.58	14.686		5.1796	182.2	18.849
	2.5081	81.01	14.872		5.3742	192.1	19.345
	2.7084	89.19	15.080				
	2.9614	100.1	15.357	423.15	0.1013	2.376	15.247
	3.2170	111.7	15.675		0.2005	4.724	15.313
	3.4299	121.9	15.927		0.3906	9.282	15.373
	3.6131	131.2	16.242		0.5862	14.06	15.369
	3.7373	137.8	16.418		0.7819	18.92	15.430
	3.9609	150.2	16.852		0.9602	23.42	15.422
	4.1716	162.7	17.303		1.1757	28.97	15.522
	4.3770	175.8	17.773		1.4217	35.44	15.648
	4.5761	189.3	18.330		1.6588	41.82	15.726
	4.7603	202.7	18.886		1.8985	48.42	15.844
	4.8783	211.8	19.315		2.1422	55.28	15.977
					2.3817	62.18	16.125
398.15	0.1014	2.530	14.474		2.5758	67.88	16.229
	0.2006	5.033	14.541		2.9031	77.75	16.431
	0.3484	8.813	14.570		3.1488	85.35	16.630
	0.5410	13.83	14.632		3.4128	93.73	16.851
	0.7832	20.31	14.669		3.6514	101.5	17.049
	1.0287	27.06	14.793		3.9039	109.9	17.290
	1.2727	33.96	14.881		4.1386	117.8	17.558
	1.5200	41.18	14.968		4.5337	131.7	17.922
	1.7481	48.03	15.111		4.8227	142.1	18.227
	2.0370	56.99	15.212		5.0737	151.4	18.578
	2.2741	64.60	15.398		5.3573	162.2	18.972
	2.4903	71.75	15.479		5.7011	175.6	19.473
	2.7774	81.58	15.684		6.0491	189.5	19.999
	3.0597	91.62	15.923		6.2706	198.6	20.381
	3.3368	101.9	16.260		6.4945	208.0	20.797
	3.6099	112.4	16.503		6.7102	217.1	21.291

Table I.(continued)

calculated results with the parameters in Table II are shown as the solid lines in Fig. 3. The average absolute deviation between the experimental viscosity results and the calculated values is 0.13%, and the maximum deviation is 0.41%. For comparison, the calculated results using the method of Wilke [20] are shown as dotted lines. The extended law of



Fig. 1. Viscosity of the binary gaseous mixture of 0.5011 HFC-125+0.4989 propane as a function of pressure.

corresponding states gives better results at high temperatures than does the Wilke method.

The viscosity under pressure is analyzed with the extended Enskog theory developed by Vesovic and Wakeham (V-W method) [10]. In the V-W method, we need the equations for the viscosity of pure constituent gases at 0.1 MPa and under pressure and for mixture gases at 0.1 MPa. The viscosities at 0.1 MPa are obtained from the extended law of corresponding states described above. The viscosity equation of pure HFC-125 used is the same as reported previously [5,6]. The viscosity of propane is calculated with an excess viscosity equation expressed as follows:

$$\eta - \eta_0 = a_1(\rho - \rho_0) + a_2(\rho - \rho_0)^2 + a_3(\rho - \rho_0)^4 + a_4(\rho - \rho_0)^6$$
(1)



Fig. 2. Viscosity of the binary gaseous mixture of 0.5011 HFC-125+0.4989 propane as a function of density.

and

$$a_1 = a_{10} + a_{11}/T + a_{12}T^3 \tag{2}$$

$$a_2 = a_{20} + a_{21}T^3 \tag{3}$$

$$a_3 = a_{30} + a_{31}/T \tag{4}$$

$$a_4 = a_{40} + a_{41}/T \tag{5}$$

where  $\eta$  is the viscosity under pressure in  $\mu$ Pa·s,  $\eta_0$  is the gas viscosity at 0.1 MPa in  $\mu$ ·Pa·s,  $\rho$  is the gas density under pressure in kg·m<sup>-3</sup>,  $\rho_0$ is the gas density at 0.1 MPa in kg·m<sup>-3</sup>, T is the absolute temperature in K, and  $a_{ij}$ 's are adjustable parameters. The values of  $a_{ij}$  for propane are determined by a least-squares fit using the recommended viscosity values



Fig. 3. Viscosity of the gaseous mixtures of HFC-125+ propane at 0.1 MPa.

 Table II.
 Scaling Parameters for the HFC-125 + Propane System

i – j	$arepsilon/{ m k}~({ m K})$	σ (nm)
HFC-125 – HFC-125 <sup><i>a</i></sup>	235.85	0.52600
Propane – Propane <sup><i>b</i></sup>	250.12	0.50772
HFC-125 – Propane <sup><i>c</i></sup>	257.40	0.50392

<sup>a</sup>Least-squares fit with the viscosity data for HFC-125 from Ref 4.

<sup>b</sup>Least-squares fit with the viscosity data for propane from Ref. 8.

<sup>c</sup>Least-squares fit with the viscosity data for HFC-125+ propane system from this study.

of Vogel et al. [8]. The temperature range of the viscosity values used is from 280 to 420 K with densities up to 210 kg  $\cdot$  m<sup>-3</sup>, which is almost the same for the HFC-125 + propane system. The values of  $a_{ij}$  for propane are shown in Table III. The viscosity equation of HFC-125 was described previously [5,6]. It should be noted that Eqs (1)–(5) are not applicable outside the temperature and density ranges of the least-squares fit. Vogel et al. [8] developed a reference correlation equation of the propane viscosity, which can be used over a wide range of temperature and pressure. Their correlation includes a zero-density correlation and an initial-density dependence correlation. Both correlations, however, have never been investigated for HFC-125 due to the lack of precise viscosity data in the low-density region. Furthermore, in the present analysis based on the V-W method, we chose the viscosities at 0.1 MPa as the reference values both for the pure compounds and binary mixtures. Therefore, we used Eqs (1)-(5) to calculate the viscosity values of propane instead of the correlation of Vogel et al. While Eqs (1)–(5) represent the experimental viscosity values with an average absolute deviation of less than 0.05% for propane in our experimental region, as shown in Table IV, it should not be used outside the temperature and/or density ranges of our experimental results. In the V-W method, the mean free path shorting factor,  $\alpha_{ii}$ , and the switch-over density are obtained from the following relations:

$$(\mathrm{d}\eta_i/\mathrm{d}\rho)|_T = \eta_i/\rho \tag{6}$$

In the lower temperature range below 348.15 K, the switch-over densities at which Eq. (6) is valid are much higher than the maximum density of the present experimental conditions. Therefore, Eqs (1)–(5) should not be applicable to Eq. (6). Thus, the Lee-Thodos (LT) viscosity correlation [21] was applied to Eq. (6). In the LT correlation, we used the extended law of corresponding states to calculate the viscosity at 0.1 MPa and treated the triple-point temperature as the adjustable parameter in order to improve

 Table III.
 Constants in Eqs (1)–(5) for Propane

<i>a</i> <sub>10</sub>	$2.832701 \times 10^{-2}$
$a_{11}$	-9.505498
<i>a</i> <sub>12</sub>	$8.863486 \times 10^{-11}$
$a_{20}$	$4.730655 \times 10^{-4}$
$a_{21}$	$-2.959302 \times 10^{-12}$
$a_{30}$	$3.869643 \times 10^{-8}$
<i>a</i> <sub>31</sub>	$-1.716098 \times 10^{-5}$
$a_{40}$	$-9.443454 \times 10^{-13}$
<i>a</i> <sub>41</sub>	$4.088699 \times 10^{-10}$

substance	n.d. <sup>a</sup>	BIAS (%) <sup>b</sup>	AAD (%) <sup>c</sup>	MAX (%) <sup>d</sup>
Propane	116	-0.018	0.050	0.404

Table IV. Correlation Results of Eq. (1) for Propane

<sup>*a*</sup>n.d.: number of data.

<sup>b</sup>BIAS (%) =  $\sum 100(\eta_{exp} - \eta_{cal})/\eta/$  n.d.

<sup>c</sup>AAD (%) =  $\sum |(\eta_{exp} - \eta_{cal})|\eta_{cal}|/$  n.d.

<sup>d</sup>MAX (%) = maximum of  $100|(\eta_{exp} - \eta_{cal})|\eta_{cal}$ .

the agreement between the experimental viscosity and correlated values. The optimum values of the triple-point temperatures were 144.88 K for HFC-125 and 168.65 K for propane.

As for the mixture viscosity calculations, the pseudo-radial distribution function  $\chi_{ij}$  for species i and j in the mixture should be evaluated. Kestin and Wakeham [22] proposed an equation for  $\chi_{ij}$  with the use of the density expansion equation for the radial distribution function of a hard-sphere fluid mixture. Since they used the density expansion equation truncated after second-order density terms, the performance of the  $\chi_{ij}$ equation in the high density region is unclear. To overcome this problem, we proposed a new method to calculate  $\chi_{ij}$  from the exact theoretical equation for the radial distribution functions for the hard-sphere fluid mixture proposed by Carnahan and Starling [23]. As shown by Vesvic and Wakeham [10], the pseudo-radial distribution function for pure gases,  $\chi_i$ , can be obtained from the pure-component viscosity by application of the hard-sphere expression for the viscosity of a pure gas [Eqs (6) and (7) in Ref. 10]. We assumed that  $\chi_i$  is equal to the Carnahan-Starling radial distribution function of a pure hard-sphere fluid i as follows:

$$\chi_i = \frac{1}{(1-\xi_3)} + \frac{3\xi_3^2}{2(1-\xi_3)^2} + \frac{\xi_3^3}{2(1-\xi_3)^3}$$
(7)

where  $\xi_3$  is a reduced density defined by  $(1/6)\pi\rho N_{AB}d_i^3$ ,  $\rho$  is a molar density in mol·cm<sup>-3</sup>, N<sub>AV</sub> is Avogadro's number in mol<sup>-1</sup>, and  $d_i$  is a hard-sphere diameter for species i in cm. Once the value of  $\chi_i$  was obtained from the pure-component viscosity data,  $d_i$  can be determined from solving Eq. (7). Equation (7) can be expressed as a cubic equation in  $\xi_3$ , as shown in the following equation:

$$\xi_3^3 - 3\xi_3^2 + \left(3 - \frac{1}{2\chi_i}\right)\xi_3 + \left(\frac{1}{\chi_i} - 1\right) = 0$$
(8)



**Fig. 4.** Deviations of the present viscosity results from values calculated by the V-W method with the mixing rule of Eqs. (7)-(14). (Dev (%) =  $100(\eta_{exp} - \eta_{cal})/\eta_{cal}$ )

Since Eq. (8) has only a single real root under the present experimental conditions, the hard-sphere diameter value can be obtained from the following analytical equation:

$$\xi_3 = \frac{\pi}{6}\rho d_i^3 = \left(-\frac{n}{2} + \sqrt{D}\right)^{1/3} + \left(-\frac{n}{2} - \sqrt{D}\right)^{1/3} \tag{9}$$

where

$$D = \frac{n^2}{4} + \frac{m^3}{27} \tag{10}$$

$$n = \frac{27}{2\chi_i} \tag{11}$$

$$m = -\frac{1}{2\chi_i} \tag{12}$$

Once the hard-sphere diameter for every constituent species in the mixture is determined from Eqs (9)–(12), the pseudo-radial distribution functions for species i and j in the mixture were obtained from the Carnahan-Starling radial distribution function of hard-sphere mixtures [23]. For the case of a binary mixture of species i and j, the following equation is valid:

$$\chi_{ij} = \frac{1}{(1-\xi_3)} + \frac{3(d_i d_j)\xi_2}{(d_i + d_j)(1-\xi_3)^2} + \frac{2(d_i d_j)^2 \xi_3^2}{(d_i + d_j)^2 (1-\xi_3)^3}$$
(13)

The reduced density  $\xi_k$  (k = 2, 3) for the binary mixture (i + j) is defined as follows:

$$\xi_{\mathbf{k}} = \left(\frac{1}{6}\right) \pi \rho \mathbf{N}_{\mathbf{A}\mathbf{V}} \left(x_i d_i^k + x_j d_j^k\right) \tag{14}$$

where  $x_i$  is the mole fraction of species i. Since the Carnahan-Starling equation can give superior results for the thermophysical properties for highly dense hard-sphere fluids, it is reasonable to consider that we can extrapolate  $\chi_{ij}$  ( $\rho$ , T) to the higher density region.

Comparisons between the calculated results from the Vesovic-Wakeham method with the original mixing rule for  $\chi_{ij}$  [22] and those with the mixing rule of Eqs (7)–(14) are shown in Table V. The deviations of the two mixtures from the two mixing rules for  $\chi_{ij}$  ( $\rho$ , T) are almost the same. This suggests that the mixng rule for  $\chi_{ij}$  ( $\rho$ , T) used in this study is almost equivalent to the original mixing rule of Kestin and Wakeham [22] for the HFC-125+ propane system at the present experimental conditions. The ability of the V-W method to represent the density and temperature dependence of the viscosity for the binary mixture of an HFC and propane may depend mainly on the viscosity correlations used for the pure constituents and also on the viscosity correlations at normal pressures for the HFC+ propane mixture.

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X <sub>HFC-125</sub>	n.d. <sup>a</sup>	Original mixing rule for $\chi_{ij}$ BIAS (%) <sup>b</sup> AAD (%) <sup>c</sup> MAX (%) <sup>d</sup>			Mixing rule of Eqs. (7)–(14) BIAS (%) AAD (%) X (%)			
0.7503	129	0.535	0.602	2.60	0.534	0.601	2.59	
0.5011	117	0.714	0.760	2.39	0.713	0.759	2.39	

 Table V. Prediction
 Results
 for
 the
 VW
 Method
 with
 Two
 Mixing
 Rules

 for
 the
 HFC-125 + Propane
 System

<sup>a</sup>n.d. : number of data

<sup>b</sup>BIAS (%) =  $\sum 100(\eta_{exp} - \eta_{cal})/\eta/$  n.d.

<sup>c</sup>AAD (%) =  $\overline{\sum} 100 |(\eta_{exp} - \eta_{cal})| \eta_{cal} / n.d.$ 

<sup>d</sup>MAX (%) = maximum of  $100|(\eta_{exp} - \eta_{cal})|/\eta_{cal}$ 

From the present results and also our previous results, we can conclude that the V-W method is applicable not only for polar+polar mixtures (such as HFC mixtures) but also for polar+nonpolar mixtures (such as the HFC+propane mixture). To verify this conclusion, it is desirable to measure the viscosity of other gaseous mixtures consisting of polar+nonpolar constituents.

### 4. CONCLUSION

In this paper, we report experimental results for the gaseous viscosity of the HFC-125 + propane system. The scaling parameters for the molecular interaction between HFC-125 and propane were determined with extended corresponding state theory and viscosity data at 0.1 MPa. Viscosity values of the gaseous mixture under pressure were predicted with the modified Enskog theory developed by Vesovic-Wakeham. In the Vesovic-Wakeham method, we applied our method to calculate the pseudoradial distributions based on the exact equation for the radial distribution function for hard-sphere fluids developed by Carnahan and Starling. From comparisons between the calculated values and the experimental viscosity results, both the present method and the original one proposed by Wakeham and Kestin give almost the same results at the present experimental conditions. We can conclude that the Vesovic-Wakeham method should be a reliable method for viscosity calculations for mixtures of HFC and propane under pressure for the case when the viscosities of pure HFC and propane under pressure and of gaseous HFC+ propane mixtures at normal pressure are available and also the density of gas mixtures under pressure can be determined with high accuracy.

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