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Viscosity Prediction for Natural Gas Mixtures[1]

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The viscosity of multicomponent natural gas-mixtures containing hydrocarbons (C1 through C7) is predicted by modifying a previously published dilute-gas viscosity model and extending its applicability to a wide range of temperature and pressure conditions including liquid and gas states. Nitrogen, oxygen, carbon dioxide, and helium are also included among components of mixtures for which published viscosity data are available. The approach takes advantage of currently available formulations and models for the density and viscosity of pure fluid constituents of natural gases. The predicted viscosity is compared with available data in both gas and liquid regions. Comparisons of calculated values to the available measurements of viscosity of natural gas mixtures and of binary, ternary, and quaternary mixtures of constituent fluids are summarized to illustrate the accuracy of the predicted values.

KEY WORDS: natural gas; prediction; viscosity.

1. INTRODUCTION

This paper reports a new development in a series of studies of comprehensive models for transport properties of pure fluids and mixtures covering a wide range of fluid states. The motivation for this study is the need for a model for transport properties to serve as a companion to the generalized mixture model for equilibrium thermodynamic properties reported by Lemmon and Jacobsen [1]. The ultimate goal is a versatile package of

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computer programs capable of calculating all fluid properties of interest to the design engineer over a wide range of temperatures and pressures including liquid and gas states. The application to natural gas mixtures has been stimulated by the continued increasing demand for natural gas as an energy resource.

One-fluid corresponding-states methods are most often used to estimate the viscosity of natural gases. Among the generally accepted methods, reduced viscosity charts [2] have been used by natural gas engineers for several decades. On these charts, the ratio of the viscosity of natural gas to its viscosity at the same temperature and at 1 atm is related to the pseudo-reduced temperature and pseudo-reduced pressure of the mixture. The viscosity of gas mixtures at low pressures is obtained from the viscosity of the constituent pure fluids by applying mixing rules. The format of generalized charts is not compatible with modern computer process design and analysis. The Herning and Zipperer [3] mixture viscosity equation has long been used for dilute gas mixture viscosity. The extended corresponding states (ECS) method [4] assumes the viscosity of a mixture can be treated as that of a pseudo-pure fluid corresponding to a specified state of a selected reference fluid. Shape factors are introduced to modify the behavior of the fluid at the reduced states of the mixture and reference fluid. The ECS method has been used for hydrocarbons and has been extended to polar refrigerant mixtures. Other attempts to predict the mixture viscosity over wide ranges of conditions include using equations of state to represent the viscosity of mixtures.

In contrast to the one-fluid corresponding-states methods, this paper reports a method for viscosity prediction for natural gases and constituent mixtures based on the viscosity of the pure components. The dilute-gas viscosity equation of Herning and Zipperer [3] has been extended to wide temperature and pressure ranges. The viscosity of a mixture is obtained by using the equation of Herning and Zipperer [3] with the viscosity of constituent pure fluids at states corresponding to the mixture temperature and pressure. Formulations for the viscosity and density of the pure fluid constituents are required. Published equations of state for pure fluids are used in this work to calculate the density of the pure fluids at the corresponding temperature and pressure as defined in the following section. Published formulations from the literature are used for the viscosity of pure methane, ethane, propane, carbon dioxide, nitrogen, helium, and oxygen. The viscosities of pure *n*-butane, isobutane, pentane, hexane, and heptane are estimated using a two-reference fluid corresponding-states method given by Teja and Rice [5]. The predicted viscosity for natural gases and constituent mixtures is compared with available experimental data in both gas and liquid regions to confirm the accuracy of the calculated values.

A summary of comparisons of viscosity calculated using our approach to available data sets is given. A brief analysis of the experimental viscosity data for natural gas is also included.

2. VISCOSITY PREDICTION FOR NATURAL GASES AND CONSTITUENT MIXTURES

2.1. Viscosity Model for Natural Gases and Constituent Mixtures

Herning and Zipperer [3] proposed the following relation for the viscosity of a gaseous mixture at low pressure:

$$\eta_m^0 = \frac{\sum_{i=1}^{n} x_i \eta_i^0 \sqrt{M_i}}{\sum_{i=1}^{n} x_i \sqrt{M_i}} \tag{1}$$

where η_m^0 is the viscosity of a gaseous mixture at low pressure, η_i^0 is the viscosity of component i at the pressure and temperature of the mixture, and x_i and M_i are the mole fraction and molar mass, respectively, of component i. Equation (1) has been used for viscosity estimation for natural gases at atmospheric pressure by various authors including Carr et al. [6] and Lohrenz et al. [7].

In the present work we extend this simple model to all temperature and pressure ranges of natural gases for which viscosity data are available from the literature. The viscosity of a natural gas at any temperature and pressure is expressed as

$$\eta_m(T, P) = \frac{\sum_{i=1}^{n} x_i \eta_i(T_i^{\mathrm{cor}}, \rho_i^{\mathrm{cor}}) \sqrt{M_i}}{\sum_{i=1}^{n} x_i \sqrt{M_i}} \tag{2}$$

where $\eta_m(T, P)$ is the mixture viscosity at temperature T and pressure P, and $\eta_i(T_i^{\mathrm{cor}}, \rho_i^{\mathrm{cor}})$ is the viscosity of pure component i at corresponding temperature T_i^{cor} and corresponding density ρ_i^{cor}. The corresponding temperature T_i^{cor}, corresponding density ρ_i^{cor}, and corresponding pressure P_i^{cor} are given as follows:

$$T_i^{\mathrm{cor}} = T T_{c,i} / T_{c,m} \tag{3}$$

$$\rho_i^{\mathrm{cor}} = \rho_i(T_{c,i}^{\mathrm{cor}}, P_{c,i}^{\mathrm{cor}}) \tag{4}$$

$$P_i^{\mathrm{cor}} = P P_{c,i} / P_{c,m} \tag{5}$$

where $T_{c,i}$ and $P_{c,i}$ are the critical temperature and critical pressure of component i, and $T_{c,m}$ and $P_{c,m}$ are the pseudo-critical temperature and pseudo-critical pressure of the mixture, which are obtained using the mixing rules given by Teja and Thurner [8].

To obtain the mixture viscosity at any temperature and any pressure, we first calculate the corresponding density ρ_i^{cor} using an equation of state for each pure component *i*. Then we use the individual viscosity representation for each pure fluid to obtain the viscosity of that fluid at the corresponding temperature T_i^{cor} and corresponding density ρ_i^{cor}. The mixture viscosity is then calculated using Eq. (2).

2.2. Density and Viscosity Formulations for Pure Components

For most pure components in natural gases, equations for the viscosity dependence on density and temperature, and the density dependence on temperature and pressure, are available in the literature. Table I lists the sources of the formulations used for calculating the density and the viscosity of pure fluids. Table II gives the statistical analysis of the viscosity representations for the pure fluids compared to available viscosity data. Table II shows that the viscosity formulations for the principal components in natural gases, e.g., methane, ethane, propane, and nitrogen exhibit absolute average deviations between 1 and 5 percent of measured values. The maximum deviations are as large as ± 10 percent for some fluids. These values are included here to illustrate the estimated accuracy of each pure fluid correlation, and to establish a basis for estimating the accuracy of the mixture correlation.

3. COMPARISON OF PREDICTED MIXTURE VISCOSITY WITH DATA

3.1. Viscosity Data from the Literature

There is a paucity of viscosity data available for natural gases in the recent literature. The compositions of the associated natural gases are listed in Table III, as well as the compositions of other multicomponent mixtures. The ranges of pressure, density, temperature, and viscosity for these systems are listed in Table IV. Data on the viscosity of binary mixtures have also been collected from the literature. The data ranges for the binary systems are given in Table V, along with the results of viscosity predictions.

3.2. Comparison Results

The predicted mixture viscosities have been compared with literature data. Tables IV and V show the statistical analysis of the comparisons using the following parameters:

Table I. Sources of Viscosity and Density Formulations for Pure Constituents of Natural Gases

Fluid	Source of density formulation	Source of viscosity formulation
Methane (CH_4)	Setzmann and Wagner [9]	Friend et al. [17]
Ethane (C_2H_6)	Friend et al. [10])	Friend et al. [10]
Propane (C_3H_8)	Younglove and Ely [11]	Vogel et al. [18]
n-Butane (n-C_4H_{10})	Younglove and Ely [11]	Teja and Rice (C_2H_6 and C_3H_8 as reference fluids)
i-Butane (i-C_4H_{10})	Younglove and Ely [11]	Teja and Rice [5] (C_2H_6 and C_3H_8 as reference fluids)
Pentane (C_5H_{12})	Starling [12]	Teja and Rice [5] (C_3H_8 and n-C_4H_{10} as reference fluids)
Hexane (C_6H_{14})	Starling [12]	Teja and Rice [5] (C_3H_8 and n-C_4H_{10} as reference fluids)
Heptane (C_7H_{16})	Starling [12]	Teja and Rice [5] (C3H8 and n-C_4H_{10} as reference fluids)
Carbon dioxide (CO_2)	Span and Wagner [13]	Fenghour and Wakeham [19]
Nitrogen (N_2)	Span et al. [14]	Shan et al. [20]
Helium (He)	McCarty and Arp [15]	McCarty [21]
Oxygen (O_2)	Schmidt and Wagner [16]	Shan et al. [20]

Table II. Comparison between Viscosity Calculated by Models and Experimental Data for Pure Fluids

								Range of Properties			
Ref.	No. of points	AAD	Bias	SD	RMS	MaxDev	No. of points dev > 10%	P (MPa)	ρ (mol · dm^{-3})	T (K)	η (μPa · s)
Methane											
Huang et al. [22]	114	2.344	0.366	2.877	2.887	9.704	5	0.0–35	0.0–28.5	103–273	5–206
Boon et al. [23]	8	2.474	−2.256	1.844	2.84	−4.662	0	0.0–0.1	26.1–28	91–114	112–200
Giddings et al. [24]	100	0.277	0.089	0.38	0.388	−1.509	0	0.1–55	0.0–18.5	283–411	10.7–39
Diller [25]	116	0.964	0.49	1.335	1.417	6.093	0	0.6–33	1.0–28.6	100–300	9–219
Carmichael et al. [26]	103	0.727	0.494	0.818	0.952	3.172	0	0.1–36	0.0–15.9	278–478	10.5–31
Carr [27]	77	0.996	−0.5	1.07	1.174	−2.676	1	0.1–55	0.0–17.8	294–367	10.8–36
Van der Gulik et al. [28]	26	3.022	2.098	2.708	3.376	5.725	4	0.1–300	0.0–27.7	298–298	11–116
Overall	544	1.183	0.274	1.749	1.768	9.704	10	0.0–300	0.0–28.6	91–478	5–219
Ethane											
Carmichael and Sage [29]	226	0.804	0.05	1.118	1.116	4.5	0	0.1–36	0.0–15	300–478	9.4–83.4
Diller and Saber [30]	164	0.845	0.188	1.077	1.09	3.635	0	0.0–32	0.3–22	95–320	9.4–1130
Diller and Ely [31]	71	1.066	−0.008	1.393	1.383	5.075	3	1.7–61	0.7–16	292–500	10.4–100
Eakin et al. [32]	209	1.545	−0.445	2.188	2.228	−8.855	8	0.1–69	0.0–17	294–511	9–111
Kestin et al. [33]	5	1.435	−0.02	1.775	1.588	−2.734	0	0.0	0.0	301–477	9–15
Overall	675	1.07	−0.072	1.56	1.56	−8.855	11	0.0–69	0.0–22	95–511	9–1130

Propane										
Huang et al. [22]	36	0.35	−0.138	0.531	0.541	−1.52	0	0.0–35	12.0–15	173–273	126–554
Kestin et al. [33]	5	0.696	0.696	0.352	0.764	0.952	0	0.1	0.0	299–478	8–12.9
Giddings et al. [25]	78	0.527	0.264	0.813	0.85	−5.413	1	0.1–55	0.0–13	278–378	7–186.3
Overall	119	0.481	0.159	0.751	0.765	−5.413	1	0.0–55	0.0–15	173–478	7–554
Butane											
Kestin et al. [33]	5	0.427	0.417	0.322	0.507	0.79	0	0.1	0.0	299–478	7.5–11.8
Carbon Dioxide											
Herreman et al. [34]	111	5.223	5.089	2.272	5.569	9.994	15	1.0–19	14.2–27	219–303	55–242
Van Dyke [35]	11	0.844	−0.844	0.048	0.846	−0.918	0	0.1	0.0	296	15
Diller and Ball [36]	82	4.173	4.173	1.663	4.488	8.566	2	0.7–31	0.3–27	220–320	16–273
Diller and Ely [31]	59	4.468	4.432	1.883	4.81	9.666	0	1.7–59	0.7–24	293–500	17–140.5
Michels et al. [37]	185	1.305	1.018	1.632	1.918	8.128	39	0.9–72	0.4–26.8	273–348	14–219
Padua et al. [38]	65	2.654	2.573	1.642	3.045	6.338	0	6–100	16.7–27	260–300	63–240
Overall	513	3.219	3.042	2.509	3.942	9.994	56	0.1–172	0.0–27	219–500	14–273
Helium											
Gracki et al. [39]	35	2.75	−2.75	1.924	3.341	−6.84	0	0.5–17	0.0	183–298	14–20
Nitrogen											
Overall (19 sources)	677	1.214	0.21	1.928	1.938	8.821	14	0.0–401	0.0–31	67–3273	5–348
Oxygen											
Overall (16 sources)	406	2.247	−0.502	3.142	3.178	−9.819	40	0.0–79	0.0–39	70–3273	6–444

Table III. Compositions of Natural-Gas and Multicomponent Systems (Mole Fraction)

System	Ref. No.	CH_4 (1)	C_2H_6 (2)	C_3H_8 (3)	n-C_4H_{10} (4)	i-C_4H_{10} (5)	n-C_5H_{12} (6)	n-C_6H_{14} (7)	n-C_7H_{16} (8)	CO_2 (9)	N_2 (10)	He (11)	O_2 (12)
NG1	[40]	0.8630	0.0680	0.0240	0.0048	0.0043	0.0022	0.0010	0.0004	0.0320			
NG2	[40]	0.7170	0.1400	0.0830	0.0190	0.0077	0.0039	0.0009	0.0001	0.0140	0.0140	0.0003	
NG3	[40]	0.8070	0.0870	0.0290	0.0170		0.0013	0.0006	0.0003	0.0090	0.0480	0.0003	
NG4	[40]	0.9150	0.0310	0.0140	0.0050	0.0067	0.0028	0.0026	0.0008	0.0170	0.0055		
NG5	[27]	0.7160	0.2770	0.0030							0.0040		
NG6	[27]	0.7350	0.2570	0.0020							0.0060		
NG7	[27]	0.7310	0.0610	0.0340	0.0060	0.0020					0.1580	0.0080	
NG8	[27]	0.9560	0.0360	0.0050							0.0030		
NG9-1	[41]	0.9859								0.0031	0.0094		0.0016
NG9-2	[41]	0.9840	0.0100								0.005		0.001
NG9-3	[41]	0.9667	0.0264							0.0031	0.0009		0.003
NG9-4	[41]	0.9647	0.0203								0.0134		0.0016
NG9-5	[41]	0.9568	0.0093							0.0016	0.0275		0.0048
NG9-6	[41]	0.9473	0.0264							0.003	0.0189		0.0044
NG9-7	[41]	0.9052	0.0456							0.0021	0.0429		0.0042
NG9-8	[41]	0.8928	0.0536							0.0015	0.0491		0.0030
NG9-9	[41]	0.8904	0.0563							0.0021	0.0468		0.0044
NG9-10	[41]	0.8882	0.0320							0.0016	0.0768		0.0014

NG9-11	[41]	0.8720	0.0758							0.0030	0.0462		0.003
NG9-12	[41]	0.8286	0.1651							0.0031	0.0016		0.0016
NG9-13	[41]	0.8481	0.1426							0.0015	0.0063		0.0015
NG9-14	[41]	0.8191	0.1751							0.0031	0.0011		0.0016
NG9-15	[41]	0.7750	0.1470							0.0030	0.075		
NG9-16	[41]	0.7592	0.1076							0.0881	0.0341		0.0110
NG9-17	[41]	0.7396	0.2489							0.0015	0.007		0.0030
NG9-18	[41]	0.6990	0.2110							0.0010	0.086		0.0030
NG9-19	[41]	0.6727	0.2443							0.0345	0.0391		0.0094
NG9-20	[41]	0.6631	0.3170							0.0047	0.0121		0.0031
NG9-21	[41]	0.6081	0.0522							0.0015	0.3367		0.0015
NG9-22	[41]	0.5878	0.3349							0.0077	0.0603		0.0093
NG9-23	[41]	0.5102	0.4069							0.0031	0.0706		0.0092
NG9-24	[41]	0.1782	0.6139							0.0077	0.1893		0.011
NG9-25	[41]	0.0238	0.8857							0.0094	0.067		0.0141
Tern1	[42]	0.5203	0.2918	0.1879									
Tern2	[42]	0.5401	0.2916		0.1683								
Tern3	[42]	0.4928		0.2487	0.2585								
Tern4	[42]		0.3763	0.3901	0.2336								
Quar1	[42]	0.3655	0.2424	0.2695	0.1226								
Tern5	[43]	0.5285								0.2713	0.2002		
Tern6	[43]	0.2941								0.1953	0.5106		
Tern7	[43]	0.1758								0.5588	0.2654		
Tern8	[43]	0.3115								0.3456	0.3429		

Table IV. Viscosity Prediction Results and Data Ranges for Natural Gases and Multicomponent Systems

System	Ref. No.	No. of points	AAD	Bias	SD	RMS	MaxDev	No. of Points Dev > 10%	P (MPa)	ρ (mol · dom^{-3})[a]	T (K)	η (μPa · s)
NG1	[40]	29	0.85	0.64	0.81	1.02	2.26	0	1.4–27.6	0.4–8.6	311–444	12.6–22.6
NG2	[40]	33	1.60	0.35	1.91	1.91	5.11	0	5.5–55.2	1.7–16.5	311–444	15.3–51.0
NG3	[40]	26	2.58	2.43	1.73	2.96	4.93	0	1.4–17.2	0.5–4.8	344–444	13.0–19.9
NG4	[40]	27	3.37	3.37	1.86	3.83	8.23	1	2.8–55.2	0.8–16.9	311–444	14.0–40.7
NG5	[27]	7	2.53	−2.53	0.45	2.56	−3.36	0	0.8–10.4	0.3–4.5	299–339	10.5–15.0
NG6	[27]	46	1.67	−1.49	1.79	2.31	−9.50	1	0.1–69.2	0.04–18.6	298 356	10.4–54.3
NG7	[27]	35	1.62	0.97	1.61	1.86	−3.95	0	0.1–66.1	0.04–18.3	299–394	11.8–45.2
NG8	[27]	33	0.98	−0.83	0.79	1.14	−2.60	0	0.1–58.4	0.03–17.8	303–398	11.0–38.2
NG9	[41]	25	3.54	−1.96	3.93	4.31	8.91	3	0.2	0.04	289	9.6–12.9
Tern1	[42]	3	0.28	−0.18	0.44	0.40	−0.69	0	0.1	0.03–0.04	298–468	9.8–14.6
Tern2	[42]	3	0.22	−0.13	0.24	0.23	−0.34	0	0.1	0.03–0.04	298–468	9.6–14.4
Tern3	[42]	3	0.30	−0.07	0.40	0.33	−0.45	0	0.1	0.03–0.04	298–468	9.0–13.6
Tern4	[42]	3	0.17	0.10	0.21	0.19	0.30	0	0.1	0.03–0.04	298–468	8.3–12.8
Quar1	[42]	3	0.17	−0.07	0.26	0.22	−0.36	0	0.1	0.03–0.04	298–468	9.1–13.8
Tern5	[43]	3	5.36	−5.36	0.33	5.36	−5.68	0	0.1	0.03–0.04	298–473	14–21
Tern6	[43]	3	2.30	−2.30	0.36	2.32	−2.70	0	0.1	0.03–0.04	298–473	16–23
Tern7	[43]	3	3.99	−3.99	0.41	4.00	−4.46	0	0.1	0.03–0.04	298–473	15–23
Tern8	[43]	3	4.39	−4.39	0.35	4.40	−4.77	0	0.1	0.03–0.04	298–473	15–22
Overall		288	1.92	0.03	2.55	2.54	−9.50	5	0.1–69.2	0.03–18.6	289–473	8–54

[a] Densities of mixtures are calculated from the mixture density model by Lemmon and Jacobsen [1].

$$\%\mathrm{Dev} = 100 \left(\frac{\eta_{\mathrm{Exp}} - \eta_{\mathrm{Pred}}}{\eta_{\mathrm{Exp}}} \right) \tag{6}$$

$$\mathrm{AAD} = \frac{1}{n} \sum_{i=1}^{n} |\%\mathrm{Dev}_i| \tag{7}$$

$$\mathrm{Bias} = \frac{1}{n} \sum_{i=1}^{n} (\%\mathrm{Dev}_i) \tag{8}$$

$$\mathrm{SD} = \sqrt{\frac{1}{n-1} \sum_{i=1}^{n} (\%\mathrm{Dev}_i - \mathrm{Bias})^2} \tag{9}$$

$$\mathrm{RMS} = \sqrt{\frac{1}{n-1} \sum_{i=1}^{n} (\%\mathrm{Dev}_i)^2} \tag{10}$$

$$\mathrm{MaxDev} = \max\{|\%\mathrm{Dev}_i|\} \tag{11}$$

where η_{Exp} is the experimental viscosity and η_{Pred} is the predicted viscosity.

Details of the available viscosity measurements including the estimated accuracy are given in the references listed in Tables IV and V and are not repeated here. These data are represented generally within an AAD of 0.2–5.4% with maximum deviations of $\pm 9.5\%$. These appear to be consistent with the accuracies of the pure fluid representations given in Table II. While we are aware that recent developments in kinetic theory have resulted in a number of methods superior in accuracy to this approach, particularly for moderate and high pressures, we have used this model to establish a baseline for further analysis both of the experimental data and of the model. The accuracy of this model for the calculation of viscosity of hydrocarbon mixtures at low to moderate pressures is comparable to that of other models available in the literature. We also recognize that the current model does not include binary interaction parameters to account for the presence of carbon dioxide and nitrogen. The comparisons in Tables IV and V indicate that the uncertainties of calculated viscosities of mixtures containing carbon dioxide and nitrogen are larger than those of other mixtures. These deviations for mixture viscosity may be caused by the relative simplicity of the model and its inability to represent the complex behavior of these mixtures. However, the relatively large deviations of calculated viscosities for pure carbon dioxide from experimental values, as listed in Table II, are also a probable cause of inaccuracies in the calculated viscosity for mixtures containing carbon dioxide.

Table V. Viscosity Prediction Results and Data Ranges for Binary Systems

Ref.	No. of points	AAD	Bias	SD	RMS	MaxDev	No. of points dev > 10%	P (MPa)	ρ ($\mathrm{mol \cdot dm^{-3}}$)	T (K)	Composition x_1
CH_4 (1)–C_2H_6 (2)											
Abe et al. [42]	15	0.43	−0.17	0.61	0.62	−1.46	0	0.1	9.6–15.5	298–468	0.26–0.75
Diller [44]	326	2.31	−0.20	3.10	3.11	9.91	10	1.5–34.9	9.9–736	100–300	0.35–0.69
CH_4 (1)–C_3H_8 (2)											
Giddings [24]	282	1.39	1.12	1.74	2.07	8.81	0	0.1–55	8.9–125	311–411	0.22–0.79
Abe et al. [42]	15	0.35	0.27	0.30	0.39	0.65	0	0.1	8.7–15	298–468	0.28–0.78
Huang et al. [45]	140	2.62	1.92	2.66	3.27	8.04	0	3.4–34.5	11–589	123–311	0.22–0.75
CH_4 (1)–n-C_4H_{10} (2)											
Kestin and Yata [46]	41	0.30	0.28	0.23	0.36	0.87	0	0.1–0.7	7.2–11	293–303	0.36–0.84
Abe et al. [42]	15	0.47	0.30	0.48	0.55	1.07	0	0.01	8–14	298–468	0.16–0.74
CH_4 (1)–CO_2 (2)											
Kestin and Yata [46]	48	4.45	−4.45	3.38	5.57	−8.86	0	0.1–2.6	11–15	293–303	0.14–0.67
DeWitt and Thodos [47]	132	6.21	−6.21	1.78	6.45	−9.92	7	3.4–69	14–86	323–474	0.2–0.8
Kestin and Ro [43]	25	5.33	−5.12	3.38	6.10	−8.6	0	0.1	13–23	298–473	0.2–0.7
CH_4 (1)–N_2 (2)											
Bzowski et al. [48]	26	2.98	2.98	0.73	3.07	5.32	0	0.1	10–78	200–3272	0.5
Diller [49]	276	4.28	−3.01	4.33	5.26	−9.97	61	1.6–33.6	8–173	100–300	0.3–0.7
Kestin and Ro [43]	25	2.15	2.14	1.11	2.40	3.36	0	0.1	12–25	298–473	0.2–0.8
C_2H_6 (1)–C_3H_8 (2)											
Abe et al. [42]	17	0.62	0.40	0.59	0.70	1.17	0	0.1	8–14	298–468	0.34–0.86

C_2H_6 (1)–n-C_4H_{10} (2)										
Abe et al. [42]	15	0.40	0.29	0.43	0.51	1.30	0	0.1	7–14	298–468	0.19–0.84
C_2H_6 (1)–CO_2 (2)										
Diller et al. [50]	205	7.12	−5.62	4.85	7.38	−9.97	163	2.1–36.8	12–229	210–320	0.26–0.75
Diller and Ely [31]	148	4.49	−3.65	3.69	5.17	−9.92	55	1.7–61	12–100	319–500	0.3–0.7
Abe et al. [51]	10	0.60	−0.57	0.46	0.72	−1.47	0	0.1	12–20	298–468	0.3–0.6
C_2H_6 (1)–N_2										
Bzowski et al. [48]	21	6.21	6.21	0.16	6.21	6.46	0	0.1	11–38	250–1273	0.5
Abe et al. [51]	10	6.16	6.16	0.56	6.18	6.93	0	0.1	12–21	298–468	0.3–0.7
C_3H_8 (1)–n-C_4H_{10} (2)										
Abe et al. [42]	19	0.65	0.65	0.46	0.79	1.82	0	0.1	7 –12	298–468	0.22–0.8
C_3H_8 (1)–CO_2 (2)										
Abe et al. [51]	10	1.64	1.64	0.32	1.67	2.17	0	0.1	10 -19	298–468	0.3–0.7
C_3H_8 (1)–N_2 (2)										
Abe et al. [51]	10	7.71	7.71	0.56	7.72	8.53	0	0.1	11 -19	298–468	0.3–0.6
n-C_4H_{10} (1)–CO_2 (2)										
Abe et al. [51]	12	3.20	3.20	0.49	3.23	3.95	0	0.1	10–17	298–468	0.3–0.6
n-C_4H_{10} (1)–N_2 (2)										
Abe et al. [51]	9	7.90	7.90	0.79	7.94	9.03	0	0.1	14–20	298–468	0.1–0.5
C_7H_{16} (1)–N_2 (2)										
Carmichael and Sage [52]	2	5.0	5.0	1.33	5.09	5.94	0	0.01–0.02	10–16	344–344	0.15–0.48
CO_2 (1)–N_2 (2)										
Bzowski et al. [48]	25	3.30	−3.30	0.91	3.41	−4.01	0	0.1	14–93	250–3272	0.5
Kestin and Ro [43]	28	2.43	−2.43	0.72	2.53	−3.95	0	0.1	15–38	298–873	0.2–0.8

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[a] Densities of mixtures are calculated from the mixture density model by Lemmon and Jacobsen [1].

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