Viscosity of Gas Mixtures at Normal Pressures: Binary Polar-Nonpolar Systems

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An interaction model is presented for the prediction of viscosities at normal pressures of gas mixtures containing a polar and a nonpolar constituent. This model is based on the principle of corresponding states and requires T_R, the viscosity parameter $\xi = T_c^{1/6}/M^{1/2}P_c^{2/3}$ for each component, and the value of z_c for the polar component.

Viscosity measurements available in the literature for several binary systems have been used to establish interaction coefficients which have been correlated to obtain relationships capable of predicting them. This method has been applied to 14 systems for temperatures ranging from 288° to 523°K. to produce an average deviation of 2.64% for 356 experimental values.

Methods for the prediction of viscosity of gas mixtures at normal pressures (approximately 1 atm.) have been presented in the literature by Brokaw (1), Cheung, Bromley, and Wilke (3), Curtiss and Hirschfelder (4), Dean and Stiel (5), Herning and Zipperer (7), Mason and Monchick (11), Sutherland (15), Wilke (19), and Yoon and Thodos (20). The development of these methods has been largely associated with the kinetic theory of nonpolar spherical molecules in their dilute gaseous state.

For polar-nonpolar gas mixtures, Mason and Monchick (11) and Cheung, Bromley, and Wilke (3) modify their developments for nonpolar gases to account for the nature of the polar constituent in the mixture. The present study attempts to extend the application of the corresponding states theorem as used by Yoon and Thodos (20) for the viscosity of nonpolar gas mixtures to include polar constituents.

A comprehensive literature search for the procurement of viscosity data of gas mixtures containing polar components showed that such information was limited to binary mixtures containing one polar and one nonpolar constituent. The only exception includes a ternary system with air as a nonpolar constituent. No experimental data could be found for polar-polar binary gas mixtures. Consequently, this study is limited to the analysis and interpretation of viscosity data of available polar-nonpolar binary mixtures. The paucity of viscosity data for polar gas mixtures extends even to polar-nonpolar binary mixtures and limits the polar constituent to be hydrogen chloride, sulfur dioxide, ammonia, and hydrogen sulfide. These four substances are strongly polar as indicated by their significant dipole moments (12). Substances such as carbon dioxide and nitrous oxide possessing weak dipole moments can be considered essentially nonpolar. Experimental information for polar-nonpolar gas mixtures is reported only by Chakraborti and Gray (2), Iwasaki et al. (9), Jung and Schmick (10), and Trautz and co-workers (16 to 18).

TREATMENT OF EXPERIMENTAL DATA

The analysis developed in this study represents an extension of the method outlined by Yoon and Thodos (20) for nonpolar gas mixtures. Their method considers a mixture to be a pure substance and utilizes T'_c, the pseudocritical temperature of the mixture, to establish T_R in order to obtain $\mu_m^* \xi_m$, the viscosity modulus for the mixture. The viscosity parameter of the mixture ξ_m is predicted by them

with the interaction model

$$\xi_m - \xi' = \frac{x_i x_j}{A_{ij} + B_{ij} x_i} \tag{1}$$

where the pseudoviscosity parameter $\xi' = \xi_i x_i + \xi_j x_j$ in which x_i represents the mole fraction of the component i having the larger value of ξ, that is, $\xi_i > \xi_j$ and A_{ij} and B_{ij} are interaction coefficients specific to the system.

For pure gases at normal pressures, Stiel and Thodos (13, 14) developed relationships between the viscosity modulus $\mu^* \xi z_c^\alpha$ and T_R. The relationships for polar and nonpolar gases are presented in Figure 1. For nonpolar gases and hydrogen, Yoon and Thodos (20) have developed the following expressions:

Nonpolar gases ($\alpha = 0$):

$$\mu^* \xi z_c^\alpha \times 10^5 = 46.10 \, T_R^{0.618}$$
$$- 20.4 \, e^{-0.449 \, T_R} + 19.4 \, e^{-4.058 \, T_R} + 1 \tag{2}$$

Hydrogen ($\alpha = 0$):

$$\mu^* \xi z_c^\alpha \times 10^5 = 47.65 \, T_R^{0.657}$$
$$- 20 e^{-0.858 \, T_R} + 19 e^{-3.995 \, T_R} + 1 \tag{3}$$

Stiel and Thodos (14) developed expressions for polar gases with and without hydrogen bonding. Their relationship for polar gases without hydrogen bonding may be modified for the sake of uniformity and is presented along with the relationship for polar gases exhibiting hydrogen bonding.

Polar gases—no hydrogen bonding ($\alpha = 0.667$):

$$\mu^* \xi z_c^\alpha \times 10^5 = [33.8 \, T_R - 5.16]^{0.80} \quad \text{for } T_R < 2.0 \tag{4}$$

Polar gases—hydrogen bonding ($\alpha = 1.25$):

$$\mu^* \xi z_c^\alpha \times 10^5 = 7.55 \, T_R - 0.55 \quad \text{for } T_R < 2.0 \tag{5}$$

In order to establish the viscosity modulus $(\mu^* \xi z_c^\alpha)_m$ for a polar-nonpolar mixture, it becomes necessary to establish first this modulus for the pure components at the temperature of the mixture. For this temperature, the corresponding reduced temperatures of these pure components become T_{Ri} and T_{Rj}. The corresponding values of $(\mu^* \xi z_c^\alpha)_i$ and $(\mu^* \xi z_c^\alpha)_j$ are then established from pure component values, if available, or directly from the relationships of Figure 1. A straight line interpolation is then postulated for mixtures between the points representing the polar and nonpolar constituents located on the log-log coordinates of Figure 1.
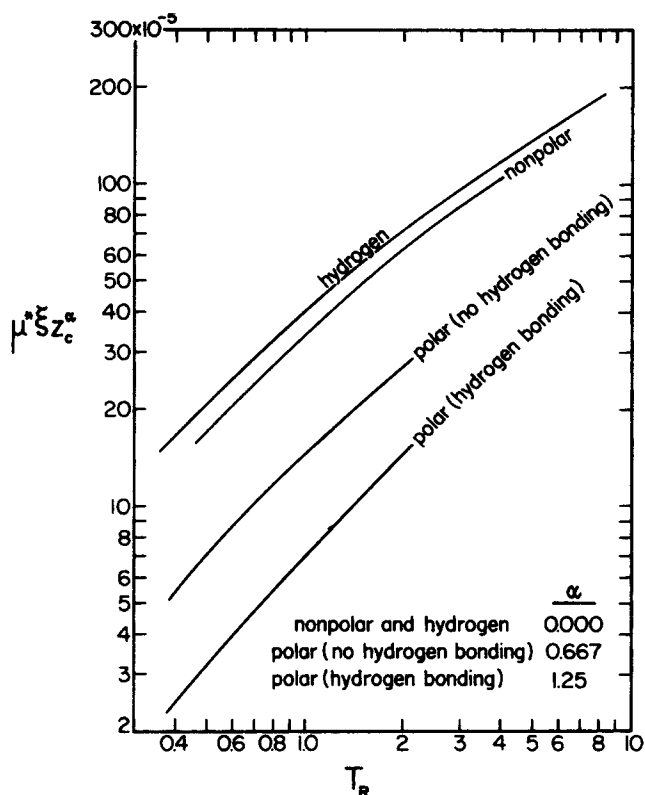
Fig. 1. Relationships between $\mu^* \xi z_c^\alpha$ and T_R for polar and non-polar gases at normal pressures (1 atm.).

This straight line may be represented in equation form as follows:

$$\log \, (\mu^* \, \xi z_c^\alpha)_m = m\log \, T_R' + \log \, k \tag{6}$$

where $m = \log \, [(\mu^* \, \xi z_c^\alpha)_i/(\mu^* \, \xi z_c^\alpha)_j]/\log \, [T_{Ri}/T_{Rj}]$ and $k = (\mu^* \, \xi z_c^\alpha)_i/T_{Ri}^m$. For the mixture the pseudocritical temperature $T_c' = T_{ci}x_i + T_{cj}x_j$ is used to obtain T_R as T/T_c'. This value of T_R, when substituted into Equation (6), establishes $(\mu^* \, \xi z_c^\alpha)_m$ for the mixture. For this viscosity modulus the value of the exponent α lies between α_i and α_j. However, evaluation of the numerical value of this exponent is not necessary. Values of $(\mu^* \, \xi z_c^\alpha)_m$, when divided by a measured viscosity, will produce the viscosity parameter $(\xi z_c^\alpha)_m$. This parameter should be dependent on composition and independent of temperature.

The validity of this approach has been tested on a number of polar-nonpolar binary systems. This approach is illustrated for the hydrogen-hydrogen chloride system for which the viscosity measurements of Trautz and Narath (17) are presented in Figure 2. Since hydrogen chloride is a polar substance exhibiting hydrogen bonding, the relationship corresponding to this substance should be used along with that for hydrogen. These relationships are given in Figure 1 and are represented by Equations (5) and (3), respectively. However, since pure component values of μ^* are reported by Trautz and Narath (17), these values were used to establish directly the viscosity moduli, $(\mu^* \, \xi z_c^\alpha)_i$ and $(\mu^* \, \xi z_c^\alpha)_j$, using $\xi_i = 0.230$ and $z_{ci}^0 = (0.305)^0 = 1.00$ for hydrogen and $\xi_j = 0.0231$ and $z_{cj}^0 = (0.266)^{1\cdot25} = 0.191$ for hydrogen chloride. For each temperature, a different straight line in Figure 1 was used to obtain the interpolated $(\mu^* \, \xi z_c^\alpha)_m$ values for each of the compositions. These values, when divided by the corresponding experimental viscosities, produce the values for the viscosity parameter $(\xi z_c^\alpha)_m$. Table 1 includes the results obtained for 21.0°,
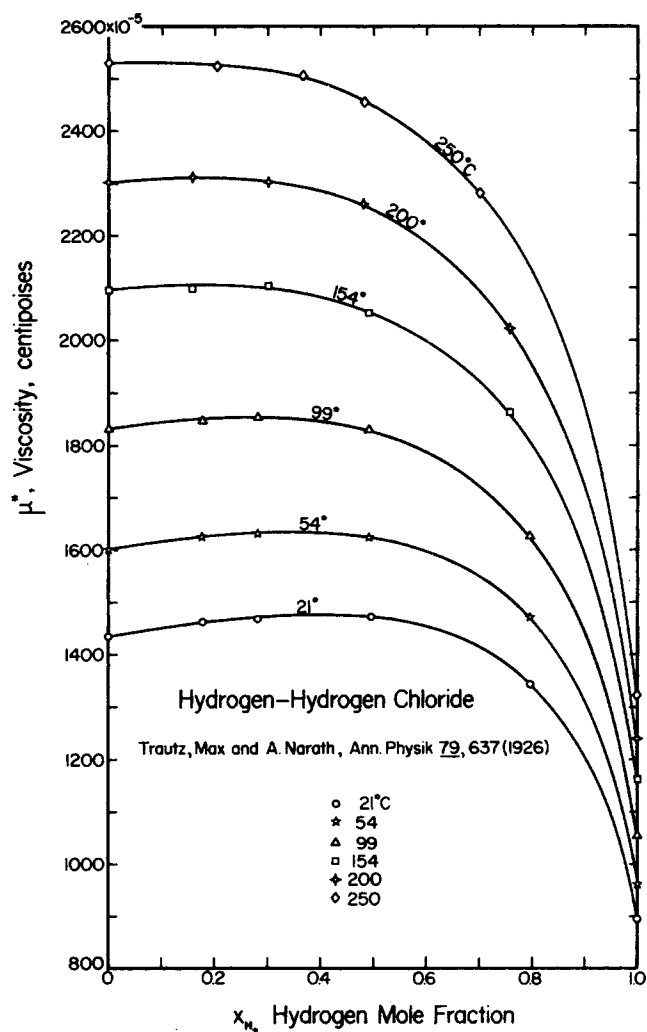

Fig. 2. Isothermal viscosity-composition relationships for the hydrogen-hydrogen chloride system at 1 atm. and temperatures ranging from 21° to 250°C.
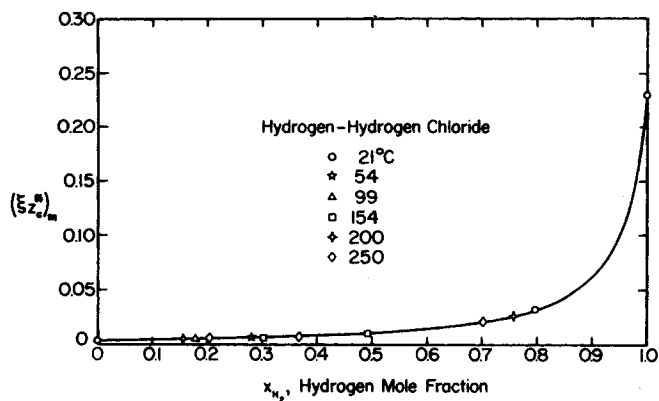

Fig. 3. Relationship between viscosity parameter $(\xi z_c^\alpha)_m$ and composition for the hydrogen-hydrogen chloride system.

99.0°, and 250.0°C. These calculated values of $(\xi z_c^\alpha)_m$ may be seen to be independent of temperature and dependent only on composition. Values of $(\xi z_c^\alpha)_m$ versus composition produce the nonlinear relationship of Figure 3. The fact that this parameter is temperature independent is consistent with the theorem of corresponding states and the next step therefore is to establish a capability for its prediction only from composition.

TABLE 1. VISCOSITY PARAMETERS $(\xi z_c^\alpha)_m$ CALCULATED FROM THE EXPERIMENTAL MEASUREMENTS OF TRAUTZ AND NARATH FOR THE HYDROGEN-HYDROGEN CHLORIDE SYSTEM AT 1 ATM. AND 21° TO 250°C.

Mole fraction		T'_c, °K.	T'_R	μ_m, cp.	$(\mu^* \, \xi \, z_c^\alpha)_m$	$(\xi \, z_c^\alpha)_m$
x_{H_2}	x_{HCl}					
21.0°C. (294.2°K.)						
0.0000	1.0000	324.5	0.907	1434×10^{-5}	6.32×10^{-5}	0.0044
0.1780	0.8220	272.7	1.079	1461	8.26	0.0056
0.2821	0.7179	242.4	1.214	1469	9.89	0.0067
0.4958	0.5042	180.1	1.633	1471	15.57	0.0106
0.7969	0.2031	92.4	3.183	1342	43.19	0.0322
1.0000	0.0000	33.3	8.835	895	205.9	0.2300
99.0°C. (372.2°K.)						
0.0000	1.0000	324.5	1.147	1832	8.08	0.0044
0.1780	0.8220	272.7	1.365	1848	10.49	0.0057
0.2821	0.7179	242.4	1.536	1855	12.50	0.0067
0.4908	0.5092	181.6	2.050	1831	19.25	0.0105
0.7969	0.2031	92.2	4.039	1629	52.78	0.0324
1.0000	0.0000	33.3	11.177	1055	242.7	0.2300
250.0°C. (523.2°K.)						
0.0000	1.0000	324.5	1.612	2530	11.57	0.0044
0.2053	0.7947	264.7	1.976	2527	15.00	0.0059
0.3688	0.6312	217.1	2.410	2507	20.01	0.0080
0.4822	0.5178	184.1	2.842	2454	25.42	0.0104
0.7009	0.2991	120.4	4.346	2281	47.08	0.0206
1.0000	0.0000	33.3	15.712	1322	304.1	0.2300

To relate the viscosity parameter $(\xi z_c^\alpha)_m$ to the composition of the binary mixture, an interaction model of the form proposed by Yoon and Thodos (20) has been adopted. In order to account for the nonlinearity of the $(\xi z_c^\alpha)_m$ relationship, the excess of this parameter has been linked to composition as follows:

$$(\xi z_c^\alpha)_m - (\xi z_c^\alpha)' = \frac{x_i x_j}{A_{ij} + B_{ij} x_i} \qquad (7)$$

where $(\xi z_c^\alpha)' = x_i (\xi z_c^\alpha)_i + x_j (\xi z_c^\alpha)_j$, x_i, and x_j represent the composition of the nonpolar and polar gas, respectively, and A_{ij} and B_{ij} are interaction coefficients specific to the system. Rearranging Equation (7) gives

$$\frac{x_i x_j}{(\xi z_c^\alpha)_m - (\xi z_c^\alpha)'} = A_{ij} + B_{ij} x_i \qquad (8)$$

The validity of Equation (7) is tested by plotting $x_i x_j / [(\xi z_c^\alpha)_m - (\xi z_c^\alpha)']$ versus x_i to obtain a straight line of intercept A_{ij} and slope B_{ij}.

For the hydrogen-hydrogen chloride system at 99.0°C., values of the group $x_i x_j / [(\xi z_c^\alpha)_m - (\xi z_c^\alpha)']$ have been plotted against composition in Figure 4 to produce for this system a linear relationship, having an intercept $A_{ij} = -4.54$ and a slope $B_{ij} = 4.36$. This approach was applied to the binary systems for which viscosity values were available and the interaction coefficients resulting from this analysis are summarized in Table 2.

CRITERION FOR POLARITY

The amount of interaction between polar and nonpolar molecules will depend not only on the extent of polarity of the polar constituent, but also on the nature of the nonpolar component. To account for polarity, the dimensionless modulus

$$\delta^* = \frac{1}{2} \frac{\mu^2}{\varepsilon \sigma^3} \qquad (9)$$

has been used as a measure of deviation from nonpolar behavior (8). In Equation (9), ε and σ are the Lennard-Jones

TABLE 2. INTERACTION COEFFICIENTS AND PURE COMPONENT PARAMETERS FOR BINARY POLAR-NONPOLAR GAS MIXTURES

i-j	ξ_i	ξ_j	z_{cj}	$\xi_j z_{cj}^\alpha$	$\xi_i - \xi_j z_{cj}^\alpha$	Actual A_{ij}	Actual B_{ij}	Calculated A_{ij}	Calculated B_{ij}
H_2—HCl	0.230	0.0231	0.266	0.00441	0.22559	-4.54	4.36	-4.543	4.361
H_2—NH_3	0.230	0.0284	0.242	0.00482	0.22518	-4.56	4.38	-4.558	4.379
H_2—SO_2	0.230	0.0189	0.269	0.00788	0.22212	-4.67	4.51	-4.670	4.510
A—SO_2	0.0275	0.0189	0.269	0.00788	0.01962	-58.0	40.9	-57.97	40.44
A—NH_3	0.0275	0.0284	0.242	0.00482	0.02268	-45.2* / $-48.9^\dagger$	34.1* / $37.2^\dagger$	-49.72	35.48
N_2—NH_3	0.0407	0.0284	0.242	0.00482	0.03588	-30.8	24.5	-30.56	23.75
O_2—NH_3	0.0301	0.0284	0.242	0.00482	0.02528	-43.7	32.7	-44.31	32.20
N_2O—SO_2	0.0227	0.0189	0.269	0.00788	0.01482	-115.2	50.4	-109.1	52.37
N_2O—NH_3	0.0227	0.0284	0.242	0.00482	0.01788	-69.6	36.0	-85.75	44.04
CO_2—HCl	0.0224	0.0231	0.266	0.00441	0.01799	-84.0	42.0	-85.09	43.80
CO_2—SO_2	0.0224	0.0189	0.269	0.00788	0.01452	-114.5	52.5	-112.0	53.38
CH_4—SO_2	0.0464	0.0189	0.269	0.00788	0.03852	-32.0	22.6	-31.98	22.37
CH_4—NH_3	0.0464	0.0284	0.242	0.00482	0.04158	-27.9‡ / $-27.9^\dagger$	20.6‡ / $20.0^\dagger$	-28.99	20.99
C_2H_4—NH_3	0.0356	0.0284	-0.242	0.00482	0.03078	-42.6	26.2	-42.67	27.09

*Iwasaki, Kestin, and Nagashima (9).
†Chakraborti and Gray (2).
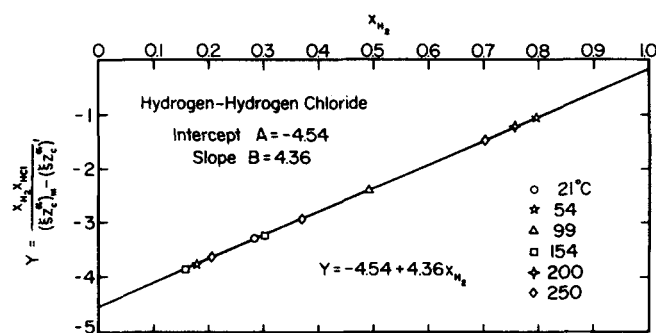‡Jung and Schmick (10).

Fig. 4. Linear relationship between Y and x_{H_2} for the hydrogen-hydrogen chloride system.

force constants and μ represents the dipole moment in debyes. In order to render Equation (9) more generally usable, the Lennard–Jones force constants have been replaced by the corresponding critical constants to produce the dimensionless polarity parameter (6)

$$\Delta^* = 4363 \frac{\mu^2}{v_c T_c} \qquad (10)$$

The dimensionless parameter Δ^* has the advantage of requiring the use of easily accessible critical constants, and therefore this parameter was adopted to account for the deviation from nonpolar behavior. For the polar substances encountered in this study using the dipole moments reported by McClellan (12) the following values of Δ^* result:

	μ, debyes	T_c, °K.	v_c, cc./g.-mole	Δ^*
NH_3	1.47	405.5	72.5	0.3207
SO_2	1.61	430.7	122	0.2152
HCl	1.07	324.6	86.8	0.1773
H_2S	0.98	373.6	97.7	0.1148
Freon-12	0.55	384.7	218	0.0157
CO_2	0.18	304.2	94.0	0.0049
N_2O	0.18	309.7	96.3	0.0047

This parameter indicates that NH_3, SO_2, HCl, and H_2S are strongly polar in comparison with Freon-12 (CCl_2F_2), CO_2, and N_2O, and that the dipole moment alone is insufficient to represent this property properly. Therefore this parameter can be used to differentiate between polar substances and those, which for all practical purposes may be considered to be nonpolar.

CORRELATION OF INTERACTION COEFFICIENTS

A cursory study of the interaction coefficients for the systems presented in Table 2 indicated that the viscosity parameter of the nonpolar constituent ξ_i affected A_{ij} and B_{ij} more than that of the polar constituent ξ_j. Yoon and Thodos (20) demonstrated for nonpolar substances the dependence of these interaction coefficients upon $\xi_i - \xi_j$. As suggested by Figure 1, the viscosity parameters for polar substances become either $\xi_j z_{cj}^{3/4}$ or $\xi_j z_{cj}^{1/3}$, while those for nonpolar substances remain as $\xi_i = \xi_i z_{ci}^0$. Therefore the difference $\xi_i z_{ci}^0 - \xi_j z_{cj}^{3/4}$ may be used when the polar constituent forms hydrogen bonds and $\xi_i z_{ci}^0 - \xi_j z_{cj}^{1/3}$ when it does not. These differences can be expressed in general as $\xi_i - \xi_j z_{cj}^\alpha$. This system parameter also accounts properly for the dominance of ξ_i over ξ_j in its relation to A_{ij} and B_{ij}.

Attempts to relate these interaction coefficients with the system parameter $\xi_i - \xi_j z_{cj}^\alpha$ showed that systems containing hydrogen are unique in themselves and therefore have

to be treated separately. This behavior is consistent with that observed by Yoon and Thodos (20) for nonpolar hydrogen-containing systems.

HYDROGEN SYSTEMS

Linear relationships resulted for the three hydrogen systems when the interaction coefficients A_{ij} and B_{ij} were plotted against the reciprocal of $\xi_i - \xi_j z_{cj}^\alpha$. These relationships can be expressed in equation form as follows:

$$A_{ij} = -\frac{1.843}{\xi_i - \xi_j z_{cj}^\alpha} + 3.627 \qquad (11)$$

$$B_{ij} = \frac{2.153}{\xi_i - \xi_j z_{cj}^\alpha} - 5.1826 \qquad (12)$$

NONHYDROGEN SYSTEMS

The interaction coefficient A_{ij} for the nonhydrogen systems of Table 2, when related to the reciprocal of $\xi_i - \xi_j z_{cj}^\alpha$ on log-log coordinates, produced the two distinct linear relationships presented in Figure 5. The characteristic difference in these relationships resides in the fact that the nonpolar constituents appear to be simple in one case (A, N_2, O_2) and complex in the other $(N_2O, CO_2, CH_4, C_2H_4)$. The simple substances A, N_2, O_2 are monatomic and diatomic and are characterized with low polarizabilities, $\alpha_{pi} < 20$. On the other hand, the complex nonpolar constituents, N_2O, CO_2, CH_4, and C_2H_4, are polyatomic and are characterized with higher polarizabilities, $\alpha_{pi} > 20$. For the nonpolar substances encountered in this study, the polarizabilities (2, 8) are as follows:

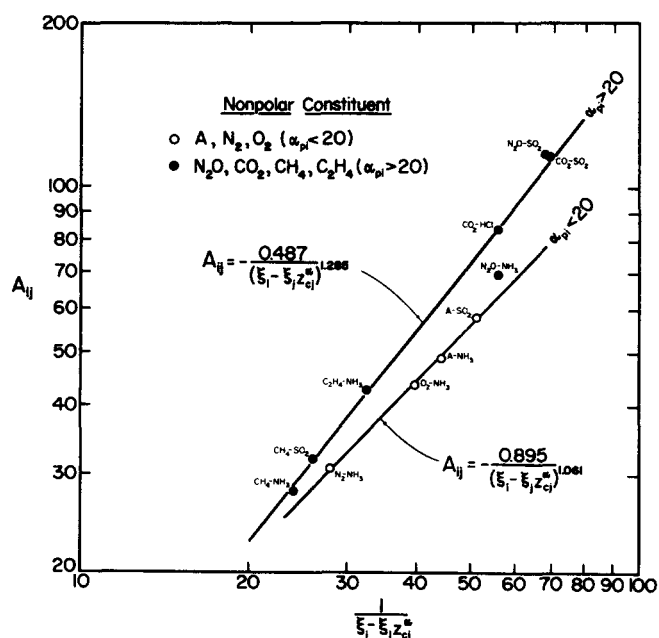|	α_{pi}, cc.
H_2	7.9×10^{-25}
O_2	16.0
A	16.2
N_2	17.6
CH_4	26.0
CO_2	26.5
N_2O	30.0
C_2H_4	42.6

Fig. 5. Relationships between A_{ij} and the reciprocal of $\xi_i - \xi_j z_{ci}^\alpha$ for nonhydrogen systems.
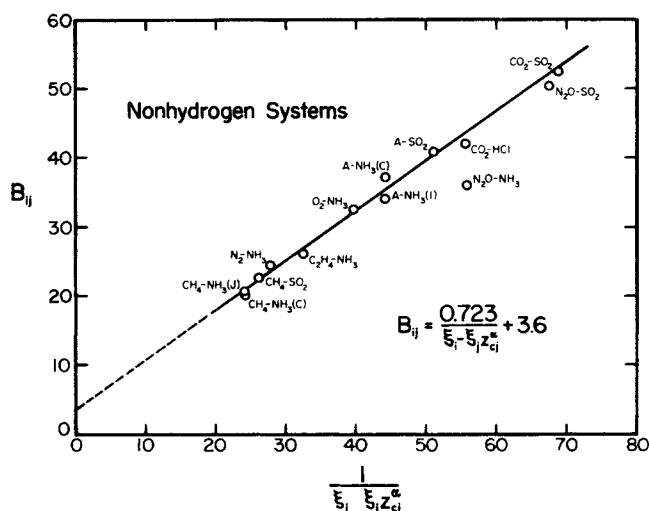
Fig. 6. Relationship between B_{ij} and the reciprocal of $\xi_i - \xi_j z_{cj}^{\alpha}$ for nonhydrogen systems.

For the simple substances ($\alpha_{pi} < 20$), the dependence of A_{ij} on $\xi_i - \xi_j z_{cj}^{\alpha}$ is given by

$$A_{ij} = -\frac{0.895}{[\xi_i - \xi_j z_{cj}^{\alpha}]^{1.061}} \tag{13}$$

and for the complex substances ($\alpha_{pi} > 20$)

$$A_{ij} = -\frac{0.487}{[\xi_i - \xi_j z_{cj}^{\alpha}]^{1.285}} \tag{14}$$

The values of the interaction coefficient B_{ij} presented in Table 2 have been plotted against the reciprocal of $\xi_i - \xi_j z_{cj}^{\alpha}$ using rectilinear coordinates to establish the linear relationship of Figure 6. This relationship can be expressed in equation form as follows:

$$B_{ij} = \frac{0.723}{\xi_i - \xi_j z_{cj}^{\alpha}} + 3.6 \tag{15}$$

Equations (11), (12), (13), (14), and (15) enable the calculation of the interaction coefficients A_{ij} and B_{ij} for binary systems containing a polar and a nonpolar constituent. For each of the 14 binary systems examined, interaction coefficients have been calculated and these are included in Table 2.

APPLICATION OF METHOD

The background developed for the estimation of the viscosity of gas mixtures has been applied to the systems included in this study. In order to illustrate the procedure, the following example is presented.

Example: Viscosity measurements for the methane-sulfur dioxide system at 1 atm. and temperatures of 35° and 80°C. are reported in the literature by Chakraborti and Gray (2). Calculate the viscosity of the mixtures at these conditions and compositions used by them. Pertinent constants for the components are as follows:

	CH$_4$	SO$_2$
z_c	0.289	0.269
T_c, °K.	191.1	430.7
ξ	0.0464	0.0189

The polar constituent SO$_2$, does not exhibit any hydrogen bonding and therefore its viscosity parameter becomes

$$\xi_j z_{cj}^{\alpha} = 0.0189 (0.269)^{0.667} = 0.00788$$

Therefore the system parameter is

$$\xi_i - \xi_j z_{cj}^{\alpha} = 0.0464 - 0.00788 = 0.03852$$

Upon substituting into Equations (14) and (15), the interaction coefficients for this system become

$$A_{ij} = -31.98 \quad \text{and} \quad B_{ij} = 22.37$$

For a composition of $x_{CH_4} = 0.608$ and $x_{SO_2} = 0.392$, the excess of the viscosity parameter according to Equation (7) becomes

$$(\xi z_c^{\alpha})_m - (\xi z_c^{\alpha})' = \frac{(0.608)\,(0.392)}{-31.98 + 22.37\,(0.608)} = -0.01297$$

where

$(\xi z_c^{\alpha})' = 0.608\,(0.0464)\,(0.289)° + 0.392\,(0.0189)\,(0.269)^{0.667}$

$= 0.608\,(0.0464) + 0.392\,(0.00788) = 0.03130$

Therefore for this mixture

$$(\xi z_c^{\alpha})_m = 0.03130 - 0.01297 = 0.01833$$

In order to obtain the value of $(\mu^* \, \xi z_c^{\alpha})_m$ for the mixture at 80°C., pure component values must be established using Equations (2) and (4). At 80°C. for pure methane ($T_R = 1.848$), Equation (2) yields the value of $\mu^* \, \xi z_c° = 59.50 \times 10^{-5}$ and for pure sulfur dioxide ($T_R = 0.820$), Equation (4) yields $\mu^* \, \xi z_c^{0.667} = 12.09 \times 10^{-5}$. Then for this system, the slope and intercept of Equation (6) become

$$m = \left(\log \frac{59.50 \times 10^{-5}}{12.09 \times 10^{-5}}\right)\Bigg/\left(\log \frac{1.848}{0.820}\right) = 1.9605$$

$$k = (59.50 \times 10^{-5})/(1.848)^{1.9605} = 17.606 \times 10^{-5}$$

For this composition $T'_c = 0.608\,(191.1) + 0.392\,(430.7) = 285.0°K$. and consequently $T'_R = 353.2/285.0 = 1.239$. Substituting into Equation (6)

$$\log (\mu^* \, \xi z_c^{\alpha})_m = 1.9605 \log 1.239 + \log 17.606 \times 10^{-5}$$

For this mixture at 80°C.

$$(\mu^* \, \xi z_c^{\alpha})_m = 27.17 \times 10^{-5}$$

and therefore $\mu^* = 27.17 \times 10^{-5}/0.01833 = 1482 \times 10^{-5}$ centipoise. For this composition at 80°C. Chakraborti and Gray report an experimental value of $\mu^* = 1469 \times 10^{-5}$ centipoise.

Similar calculations have been carried out for the other compositions used by Chakraborti and Gray. These calculated viscosities, on comparison with corresponding measurements at 35° and 80°C., produce an average deviation of 1.07% for 15 points.

RESULTS AND DISCUSSION

Calculations similar to those used for methane-sulfur dioxide mixtures have been carried out for the remaining systems of this study. For each system the average deviation resulting from calculated and experimental values is presented in Table 3. The overall average deviation for 356 points representing the 14 binary systems of this study is 2.64%. In this comparison all available data for polar-nonpolar binary mixtures have been included, with the exclusion of none. It should be noted, however, that discrepancies exist between viscosity measurements carried out by different investigators for the same system. These discrepancies exist even for pure component values and point out the need for the establishment of more accurate

System	Temp., °C.	No. of points	Avg. Dev., %	Source of viscosity data
H_2—HCl	21,54,99,154,200,250	24	2.84	(17)
H_2—NH_3	20,100,200,250	23	2.87	(16)
H_2—SO_2	17,45,70,92,124,159,199	38	2.39	(18)
A—SO_2	25,35,80	27	3.36	(2)
A—NH_3	20,30	13	5.44	(9)
	25,35,80	26	2.29	(2)
N_2—NH_3	20,100,200,250	20	2.92	(16)
O_2—NH_3	20,100,200	15	2.49	(16)
CH_4—SO_2	35,80	15	1.07	(2)
CH_4—NH_3	14,46	9	3.43	(10)
	25,35,80	27	1.69	(2)
C_2H_4—NH_3	20,100,200,250	24	3.13	(16)
N_2O—SO_2	25,35,80	28	0.90	(2)
N_2O—NH_3	25,35,80	27	5.52	(2)
CO_2—SO_2	25,35,80	31	1.40	(2)
CO_2—HCl	18,40	9	0.42	(10)
		356	2.64	

measurements. Despite such differences, no values were discarded in obtaining the above deviations. Stiel and Thodos point out that the average deviations to be expected for the prediction of pure component values, as indicated by Equations (2) to (5), is 1.77% for nonpolar gases (13) and 2.59% for polar gases (14). However, for the pure components used in the systems of this study, the average deviations encountered by Stiel and Thodos range from 1.05 for methane to 1.92% for hydrogen chloride. The overall average deviation of 2.64% for this study includes inherently these expected deviations since the pure component values were calculated using Equations (2) to (5). Therefore the method developed in this study has the ability to predict viscosities of binary polar-nonpolar gas mixtures with an expected deviation of 2.64%, without any knowledge of experimental pure component viscosities.

To calculate viscosities of binary polar-nonpolar mixtures, a FORTRAN program was developed and used on a CDC-6400 digital computer. A printout of this program is available elsewhere (6).

The only experimental viscosities relating to ternary mixtures are the three air systems with ammonia, hydrogen chloride, and hydrogen sulfide (10) at essentially room temperature. In this case air may be treated as a nitrogen-oxygen mixture. This information is insufficient to permit an extension to include the treatment of multicomponent systems at 1 atm. and varying temperatures.

ACKNOWLEDGMENT

NOTATION

A,B = interaction coefficients, Equation (7)
k = constant, Equation (6)
m = slope, Equation (6)
P_c = critical pressure, atm.
R = gas constant
T = temperature, °K.
T_c = critical temperature, °K.
T_c' = pseudocritical temperature of mixture, $\sum_i x_i T_{ci}$, °K.

T_R = reduced temperature, T/T_c
T_R' = pseudoreduced temperature of mixture, T/T_c'
x = mole fraction
v_c = critical volume, cc./g.-mole
z_c = critical compressibility factor, $P_c v_c/RT_c$

Greek Letters

α = exponent, ranging from 0 to 1.25
α_{pi} = polarizability of nonpolar constituent, cc.
δ^* = polarity modulus, Equation (9), dimensionless
Δ^* = polarity parameter, Equation (10), dimensionless
ε = maximum energy of attraction for Lennard-Jones potential, erg
σ = collision diameter for Lennard-Jones potential, A.
μ = dipole moment, debyes
μ^* = viscosity at atmospheric pressure, centipoise
ξ = viscosity parameter, $T_c^{1/6}/M^{1/2}P_c^{2/3}$
$(\xi z_c^\alpha)'$ = pseudoviscosity parameter, $\sum_i x_i(\xi z_c^\alpha)_i$

Subscripts

i = nonpolar component
j = polar component
ij = system consisting of i and j
m = mixture

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