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# Correlation and Extrapolation Scheme for the Composition and Temperature Dependence of Viscosity of Binary Gaseous Mixtures: Carbon Dioxide + Ethane[1]

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Experimental viscosity data of ethane, carbon dioxide, and three mole fractions of the binary system carbon dioxide + ethane in the temperature range $293.15 \leqslant T \leqslant 633.15$ K and in the density range $0.01 \leqslant \rho \leqslant 0.05 \text{ mol} \cdot \text{L}^{-1}$ reported earlier were evaluated simultaneously to find out a useful correlation and extrapolation scheme for the viscosity of binary systems in the range of moderate densities. A procedure based on the ideas of the modified Enskog theory has been found to give the best results. Dependent on temperature, the collision diameters related to the equilibrium radial distribution function at contact are fitted to viscosity values of the pure substances and of at least one mixture. The results are compared with experimental data from the literature. A recommendation is given concerning the density range in which the first density contribution to the viscosity coefficient of the system carbon dioxide + ethane is sufficient to be included.

KEY WORDS: binary gaseous mixture; carbon dioxide; ethane; transport properties; viscosity.

## 1. INTRODUCTION

In a recently published paper [1], experimental results for the viscosity of the binary system carbon dioxide + ethane have been reported at three mole fractions $x_{CO_2} = 0.7398$, 0.50000, and 0.2500, respectively. These data

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were evaluated together with new experimental data for ethane [2] and carbon dioxide [3].

All these measurements were carried out in an all-quartz oscillating-disk viscometer which has been described in detail in Refs. 4–6. The viscometer was calibrated for large ranges of the boundary layer thickness at room temperature by means of reference values of the viscosity coefficient of argon, krypton as well as nitrogen. After three or four series of measurements the calibration of the viscometer was checked by an additional measurement on argon or nitrogen. The uncertainty of the experimental data has been estimated to be ±0.15% near room temperature, increasing up to ±0.3% at 633.15 K, whereas the reproducibility is even better (±0.1%) covering the whole temperature range.

The viscosity of pure substances as well as of mixtures can be represented by density expansions,

$$\eta_i = \eta_i^{(0)} + \eta_i^{(1)}\rho + \eta_i^{(2)}\rho^2 + \cdots \tag{1}$$

$$\eta_{\mathrm{mix}} = \eta_{\mathrm{mix}}^{(0)} + \eta_{\mathrm{mix}}^{(1)}\rho + \eta_{\mathrm{mix}}^{(2)}\rho^2 + \cdots \tag{2}$$

The kinetic theory of dilute gases [7, 8] gives the theoretical background for the zero-density viscosity coefficients $\eta_i^{(0)}$ and $\eta_{\mathrm{mix}}^{(0)}$. Whereas in the case of a pure substance the Rainwater–Friend theory [9–11] can be used to describe the initial density dependence of the viscosity $\eta_i^{(1)}$ at a relatively high level, this theory has not been extended to mixtures. Thus, the calculation and prediction of the viscosity of gaseous mixtures have to be restricted to the extensions of the Enskog theory to binary mixtures by Thorne [12] and to multicomponent mixtures by Tham and Gubbins [13]. Lopez de Haro et al. [14] examined in detail the differences between the standard Enskog theory (SET) due to Tham and Gubbins and the revised Enskog theory (RET) due to van Beijeren and Ernst [15]. They found that there are no differences in the case of the viscosity of multicomponent mixtures following from the SET and RET. The extensions of the Enskog theory for multicomponent mixtures represent practical tools for the treatment of the composition and density dependence of the viscosity of a real dense gaseous mixture. In accordance with the ideas of the modified Enskog theory for pure substances, the hard-sphere quantities are replaced by suitably chosen real dense-gas quantities. Kestin et al. [16] developed a method on this basis for calculating the viscosity of dense gas mixtures in large density ranges using a pseudo-radial distribution function at contact of the colliding spheres.

The present paper reports a new scheme for the calculation and prediction of the viscosity of binary mixtures according to the modified Enskog theory in the range of the first density contribution. An extrapolation to higher densities allows conclusions concerning the density range in which the first density correction gives reasonably good results.

## 2. THEORY

Since the theory of the density dependence of the viscosity for a binary gaseous mixture has recently been discussed [1, 17–19], here only the essential features are outlined.

In the case of a pure gas consisting of hard spheres, the coefficient $\eta_i^{(0)}$ of Eq. (1) is given in the first approximation of the Chapman–Enskog solution of the Boltzmann equation by

$$\eta_i^{(0)}=\frac{5}{16}\frac{(\pi m_i kT)^{1/2}}{\pi\sigma_i^2} \tag{3}$$

where $m_i$ is the molecular mass, $T$ the temperature, $k$ Boltzmann's constant, and $\sigma_i$ the diameter of a hard sphere. Then the first density correction $\eta_i^{(1)}$ is

$$\eta_i^{(1)}=\eta_i^{(0)}\left(\frac{4}{5}b_i-\chi_i^{(1)}\right)=\eta_i^{(0)}B_{\eta_i} \tag{4}$$

with

$$b_i=\frac{2}{3}\pi N_{\mathrm{Av}}\sigma_i^3 \tag{5}$$

$$\chi_i=1+\chi_i^{(1)}\rho+\cdots=1+\frac{b_i}{c_i}\rho+\cdots \tag{6}$$

$$\chi_i^{(1)}=\frac{5}{12}\pi N_{\mathrm{Av}}\sigma_i^3 \tag{7}$$

$b_i$ and $c_i$ are the second and third pressure virial coefficients for hard spheres. $N_{\mathrm{Av}}$ is Avogadro's constant, $B_{\eta_i}$ the second viscosity virial coefficient, and $\chi_i$ the equilibrium radial distribution function at contact of hard spheres.

The zero-density and the linear-in-density contributions in the formula for the viscosity of a binary gas mixture are evaluated from

$$\eta_{\mathrm{mix}}=-\begin{vmatrix}H_{11} & H_{12} & y_1\\ H_{21} & H_{22} & y_2\\ y_1 & y_2 & 0\end{vmatrix}\Bigg/\begin{vmatrix}H_{11} & H_{12}\\ H_{21} & H_{22}\end{vmatrix}+\kappa_{\mathrm{mix}} \tag{8}$$

with the $H_{ij}$, $y_i$, and $\chi_{ij}$ given by the density series

$$H_{ij} = H^{(0)}_{ij} + H^{(1)}_{ij}\rho + \cdots \tag{9}$$

$$y_i = x_i + y^{(1)}_i \rho + \cdots \tag{10}$$

$$\chi_{ij} = 1 + \chi^{(1)}_{ij}\rho + \cdots \tag{11}$$

In the case of a mixture of hard spheres the first two terms in Eq. (2) are identified with

$$\eta^{(0)}_{\text{mix}} = [x_1^2 H^{(0)}_{22} + x_2^2 H^{(0)}_{11} - 2x_1 x_2 H^{(0)}_{12}][H^{(0)}_{11} H^{(0)}_{22} - H^{(0)2}_{12}]^{-1} \tag{12}$$

$$\begin{aligned}\eta^{(1)}_{\text{mix}} = {} & [x_1^2 H^{(1)}_{22} + x_2^2 H^{(1)}_{11} - 2x_1 x_2 H^{(1)}_{12} + 2x_1 y^{(1)}_1 H^{(0)}_{22} + 2x_2 y^{(1)}_2 H^{(0)}_{11} \\ & -2x_1 y^{(1)}_2 H^{(0)}_{12} - 2x_2 y^{(1)}_1 H^{(0)}_{12} - \eta^{(0)}_{\text{mix}}(H^{(0)}_{11} H^{(1)}_{22} + H^{(1)}_{11} H^{(0)}_{22} - 2H^{(0)}_{12} H^{(1)}_{12})] \\ & \times [H^{(0)}_{11} H^{(0)}_{22} - H^{(0)2}_{12}]^{-1}\end{aligned} \tag{13}$$

The leading term of the bulk viscosity $\kappa_{\text{mix}}$ is quadratic in density. Here

$$H^{(0)}_{ii} = \frac{x_i^2}{\eta^{(0)}_i} + \sum_{j=1, j\neq i}^{2} \frac{2x_i x_j}{\eta^{(0)}_{ij}} \frac{m_i m_j}{(m_i + m_j)^2} \left(\frac{5}{3A^*_{ij}} + \frac{m_j}{m_i}\right) \tag{14}$$

$$H^{(1)}_{ii} = \frac{x_i^2 \chi^{(1)}_{ii}}{\eta^{(0)}_i} + \sum_{j=1, j\neq i}^{2} \frac{2x_i x_j \chi^{(1)}_{ij}}{\eta^{(0)}_{ij}} \frac{m_i m_j}{(m_i + m_j)^2} \left(\frac{5}{3A^*_{ij}} + \frac{m_j}{m_i}\right) \tag{15}$$

$$H^{(0)}_{ij}(j \neq i) = -\frac{2x_i x_j}{\eta^{(0)}_{ij}} \frac{m_i m_j}{(m_i + m_j)^2} \left(\frac{5}{3A^*_{ij}} - 1\right) \tag{16}$$

$$H^{(1)}_{ij}(j \neq i) = -\frac{2x_i x_j \chi^{(1)}_{ij}}{\eta^{(0)}_{ij}} \frac{m_i m_j}{(m_i + m_j)^2} \left(\frac{5}{3A^*_{ij}} - 1\right) \tag{17}$$

$$\eta^{(0)}_{ij} = \frac{5}{16} \frac{(\pi m_{ij} kT)^{1/2}}{\pi \sigma^2_{ij}} \tag{18}$$

$$m_{ij} = \frac{2m_i m_j}{(m_i + m_j)^2} \tag{19}$$

$$\sigma_{ij} = \frac{1}{2}(\sigma_i + \sigma_j) \tag{20}$$

$$\chi^{(1)}_{ij} = \zeta_3 + \frac{3\sigma_{ii}\sigma_{jj}}{2\sigma_{ij}} \zeta_2, \qquad \zeta_k = \frac{\pi}{6} N_{\text{Av}} \sum_{i=1}^{2} x_i \sigma^k_{ii} \tag{21}$$

$$y^{(1)}_i = \frac{4}{5} x_i \sum_{j=1}^{2} \frac{m_j}{m_i + m_j} x_j b_{ij}, \qquad b_{ij} = \frac{2}{3}\pi N_{\text{Av}} \sigma^3_{ij} \tag{22}$$

The interaction viscosity $\eta_{ij}^{(0)}$ is that of a hypothetical one-component hard-sphere fluid with the mass $m_{ij}$ and the diameter $\sigma_{ij}$ in the limit of zero density. $A_{ij}^{*}$ represents a dimensionless ratio of collision integrals and is exactly unity for hard spheres. It should be noted that in the case of a gas of hard spheres only a single diameter $\sigma_{ij}$ is necessary for the description of $b_{ij}$, $\chi_{ij}$, and $\eta_{ij}^{(0)}$.

In the adaptation of the Enskog theory of hard spheres to real molecules, it is assumed that the transport coefficients have the same functional form as in the case of a hard-sphere gas. Thus, the hard-sphere quantities can be replaced by real-gas quantities. This requires us to distinguish among three real-gas diameters $\sigma_i^{\eta}$ for $\eta_i^{(0)}$, $\sigma_i^{b}$ for $b_i$, and $\sigma_i^{\chi}$ for $\chi_i^{(1)}$. The quantity $\sigma_i^{\eta}$ is commonly replaced by experimental values for the zero-density viscosity coefficient $\eta_i^{(0)}$. Furthermore, the pressure $P$ in the equation of state of a gas of hard spheres is replaced by the thermal pressure $T(\partial P/\partial T)_\rho$ of the real gas. On this condition it follows immediately [20] that

$$b_i = B_i + T\frac{dB_i}{dT} \tag{23}$$

$$c_i = C_i + T\frac{dC_i}{dT} \tag{24}$$

where $B_i$ and $C_i$ are the second and third pressure virial coefficients of the real gas. According to this theory $\sigma_i^{b}$ and $\sigma_i^{\chi}$ can be determined directly from $B(T)$ and $C(T)$ data by means of

$$\sigma_i^{b} = \left(\frac{3}{2\pi N_{\mathrm{Av}}} b_i\right)^{1/3} \tag{25}$$

$$\sigma_i^{\chi} = \left(\frac{12}{5\pi N_{\mathrm{Av}}} \frac{b_i}{c_i}\right)^{1/3} \tag{26}$$

For binary gaseous mixtures, the hard-sphere quantities have also to be identified as real-gas quantities. Then $\sigma_{ij}^{\eta}$ is replaced by experimentally obtained values for the interaction viscosity in the limit of zero density $\eta_{ij}^{(0)}$, whereas $\sigma_{ij}^{b}$ is calculated from interaction pressure virial coefficients $B_{ij}(T)$. Unfortunately, $\sigma_{ij}^{\chi}$ cannot be obtained by means of second and third pressure virial coefficients because four different $C_{ijk}(T)$ values would have to be reduced to three $\sigma_{ij}^{\chi}$ and $\chi_{ij}^{(1)}$ values, respectively. But, it is possible either to fit only the $\sigma_i^{\chi}$ to the experimental data and to calculate the $\sigma_{ij}^{\chi}$ according to the mixing rule, expressed by Eq. (20), or to fit the $\sigma_i^{\chi}$ and the $\sigma_{ij}^{\chi}$ simultaneously to the experimental data.

## 3. EVALUATION

To deduce a useful and practicable correlation and prediction scheme the following three methods were investigated.

1. The composition dependence of the first density correction of the equilibrium radial distribution function at contact $\chi_{ij}^{(1)}$ is neglected.
2. The viscosity of the mixture is predicted only from the pure-gas quantities $\sigma_i^\chi$ including the mixing rule, Eq. (20).
3. The $\sigma_i^\chi$ are determined by simultaneously fitting to the experimental data of the pure substances and of only one mixture. In this connection $\sigma_{ij}^\chi$ is implicitly fitted using the mixing rule, Eq. (20).

It should be stressed that in all cases the zero-density viscosity of the pure substances $\eta_i^{(0)}$, as well as the interaction viscosity of the binary mixture $\eta_{ij}^{(0)}$, have to be known in order to replace $\sigma_i^\eta$ and $\sigma_{ij}^\eta$ and that the diameters $\sigma_i^b$ as well as $\sigma_{ij}^b$ are calculated from $B(T)$ data via Eqs. (23) and (25). The temperature functions $b_i$ and $b_{ij}$ for ethane, carbon dioxide, and the binary system are illustrated in Fig. 1. The increasing values of $\sigma_i^b$ at temperatures higher than 500 K (see Table I) are related to the peculiar behavior of the $b_i$ curve of ethane that is caused by the experimental $B(T)$ values at higher temperatures.

As already discussed in Ref. 1 the neglect of the composition dependence of the first density correction of the equilibrium radial distribution function at contact $\chi_{ij}^{(1)}$ [see Eq. (21)] resulted in relatively large positive

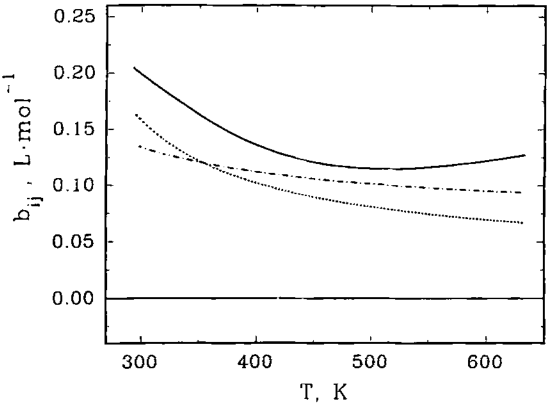

**Fig. 1.** Temperature dependence of $b_{ij}$ calculated from $B(T)$ data according to the modified Enskog theory. (—) Ethane; (···) carbon dioxide; (·—·—·) carbon dioxide + ethane.

**Table I.** Diameters $\sigma_i^b$ and $\sigma_{ij}^b$ According to the Modified Enskog Theory and Diameters $\sigma_i^\chi$ and $\sigma_{ij}^\chi$ Fitted to Experimental Viscosity Data

| Temperature $T$ (K) | Collision diameter (nm)[a] | | | | | |
|---|---|---|---|---|---|---|
| | $\sigma_i^b$ | $\sigma_j^b$ | $\sigma_{ij}^b$ | $\sigma_i^\chi$ | $\sigma_j^\chi$ | $\sigma_{ij}^\chi$ |
| 298.15 | 0.5418 | 0.5020 | 0.4742 | 0.5330 | 0.4942 | 0.5136 |
| 293.15 | 0.5452 | 0.5075 | 0.4763 | 0.5381 | 0.5035 | 0.5208 |
| 323.15 | 0.5242 | 0.4783 | 0.4650 | 0.5050 | 0.4636 | 0.4843 |
| 353.15 | 0.5034 | 0.4567 | 0.4563 | 0.4682 | 0.4360 | 0.4521 |
| 383.15 | 0.4848 | 0.4402 | 0.4494 | 0.4287 | 0.4178 | 0.4233 |
| 423.15 | 0.4658 | 0.4233 | 0.4420 | 0.3896 | 0.3977 | 0.3936 |
| 453.15 | 0.4565 | 0.4133 | 0.4375 | 0.3671 | 0.3851 | 0.3761 |
| 483.15 | 0.4514 | 0.4048 | 0.4337 | 0.3598 | 0.3720 | 0.3659 |
| 513.15 | 0.4498 | 0.3974 | 0.4303 | 0.3592 | 0.3590 | 0.3591 |
| 543.15 | 0.4511 | 0.3910 | 0.4274 | 0.3684 | 0.3463 | 0.3573 |
| 573.15 | 0.4546 | 0.3853 | 0.4248 | 0.3843 | 0.3319 | 0.3581 |
| 603.15 | 0.4596 | 0.3802 | 0.4225 | 0.4027 | 0.3136 | 0.3582 |
| 633.15 | 0.4657 | 0.3756 | 0.4205 | 0.4255 | 0.2866 | 0.3561 |

[a] $i$ = ethane, $j$ = carbon dioxide.

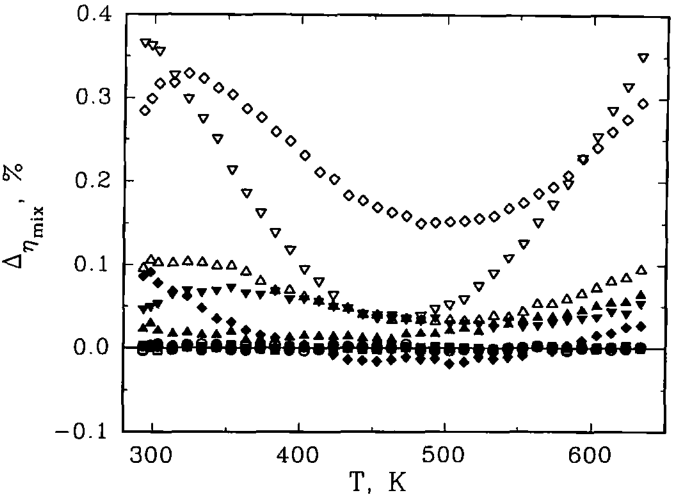

**Fig. 2.** Deviations of calculated viscosity data from experimental $\eta_{mix}$ data according to the second method. Filled symbols: (●) ethane, (■) carbon dioxide, (▼) $x_{CO_2} = 0.2500$, (◆) $x_{CO_2} = 0.5000$, and (▲) $x_{CO_2} = 0.7398$, all at 0.01 mol · L$^{-1}$. Open symbols: same as filled symbols, all at 0.05 mol · L$^{-1}$. $\Delta_{\eta_{mix}} = 100(\eta_{mix,exp} - \eta_{mix,calc})/\eta_{mix,calc}$.

values for $\chi_{ii}^{(1)}$ and in physically unreasonably negative values for $\chi_{ij}^{(1)}$ (see Fig. 4 in Ref. 1).

The prediction of the viscosity of the three binary mixtures from only pure-substance quantities according to the second method requires the calculation of the diameters $\sigma_i^\chi$ by means of Eqs. (4)–(7) with

$$\sigma_i^\chi = \left[\frac{12}{5\pi N_{\mathrm{Av}}}\left(\frac{4}{5}b_i - \frac{\eta_i^{(1)}}{\eta_i^{(0)}}\right)\right]^{1/3} \tag{27}$$

where $b_i$ is calculated via Eq. (23). The quantity $\sigma_{ij}^\chi$ is obtained according to the mixing rule, Eq. (20). This procedure leads to small deviations of the calculated values from the experimental $\eta_{\mathrm{mix}}$ data in the case of a very low density ($0.01\ \mathrm{mol \cdot L^{-1}}$) and to relatively higher deviations at a density of $0.05\ \mathrm{mol \cdot L^{-1}}$, as shown in Fig. 2. It is noted that the deviations are systematically positive, with two maxima at both temperature extrema, and that the small deviations of the mixture with the mole fraction $x_{\mathrm{CO_2}} = 0.7398$ are coincidental.

The best results were obtained by the third method, in which $\sigma_i^\chi$ are fitted to the experimental data of the pure substances and to only one mixture, while the mixing rule Eq. (20) is used for $\sigma_{ij}^\chi$. The mixture with the mole fraction $x_{\mathrm{CO_2}} = 0.5000$ was chosen to be included. A fit of all three diameters ($\sigma_i^\chi, \sigma_j^\chi, \sigma_{ij}^\chi$) to the experimental viscosity data of the two pure substances and of the three mixtures failed numerically because of the small influence of $\sigma_{ij}^\chi$. Values of $\sigma_i^b$ and $\sigma_{ij}^b$ according to the modified Enskog theory and $\sigma_i^\chi$ and $\sigma_{ij}^\chi$ fitted to the experimental data in the described manner are presented at selected temperatures in Table I. The deviatons of the calculated values from the experimental viscosities of the pure substances and of the three mixtures are shown in Fig. 3. The first density correction of the equilibrium radial distribution function at contact $\chi_{ij}^{(1)}$, resulting from the fitted $\sigma_i^\chi$ and $\sigma_{ij}^\chi$ via Eq. (21), are compared with the results of the first method in Fig. 4. It turns out that the composition dependence of $\chi_{ij}^{(1)}$ is small but not negligible. When we calculate the total value of the radial distribution function at contact $\chi_{ij}$ in the density range under consideration and extrapolate to zero density, the $\chi_{ij}$ values for all isotherms are $0.99999 \leqslant \chi_{ij} \leqslant 1.00001$. This shows that the scheme works well.

Finally, by means of the results of the third method, extrapolated viscosity values were calculated for densities up to $16\ \mathrm{mol \cdot L^{-1}}$ at 320 and 500 K and compared with experimental data of Diller and Ely [21]. It emerges from Fig. 5 that an extrapolation up to $1\ \mathrm{mol \cdot L^{-1}}$ leads to deviations within $\pm 3\%$. For the system under discussion, it is recommended to include the next term of Eq. (2), quadratic in density, to extrapolate to densities higher than $0.6\ \mathrm{mol \cdot L^{-1}}$.

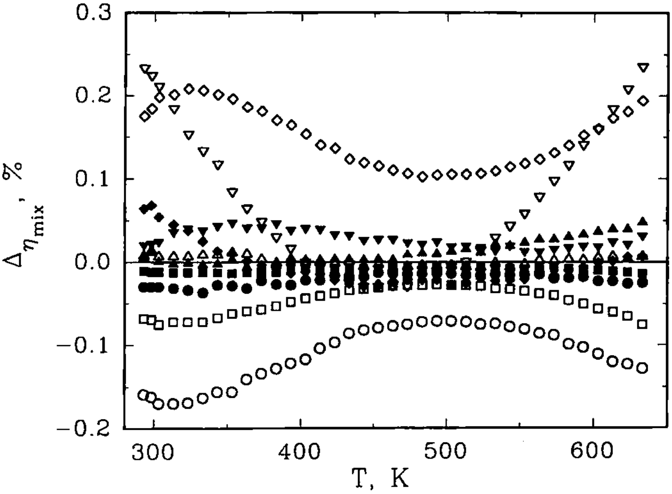

**Fig. 3.** Deviations of calculated viscosity data from experimental $\eta_{mix}$ data according to the third method. Filled symbols: (●) ethane, (■) carbon dioxide, (▼) $x_{CO_2} = 0.2500$, (◆) $x_{CO_2} = 0.5000$, and (▲) $x_{CO_2} = 0.7398$, all at 0.01 mol · L$^{-1}$. Open symbols: same as filled symbols, all at 0.05 mol · L$^{-1}$. $\Delta_{\eta_{mix}} = 100(\eta_{mix, exp} - \eta_{mix, calc})/\eta_{mix, calc}$.

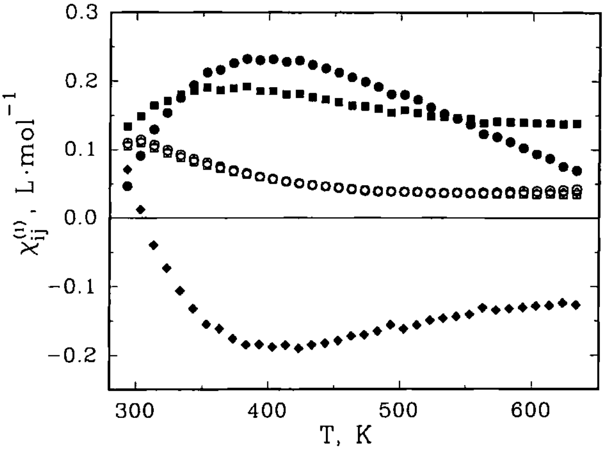

**Fig. 4.** Results for the temperature dependence of the first density correction of the equilibrium radial distribution function at contact $\chi_{ij}^{(1)}$. Filled symbols: (●) ethane, (■) carbon dioxide, and (◆) carbon dioxide + ethane, first method. Open symbols: same as filled symbols, third method.

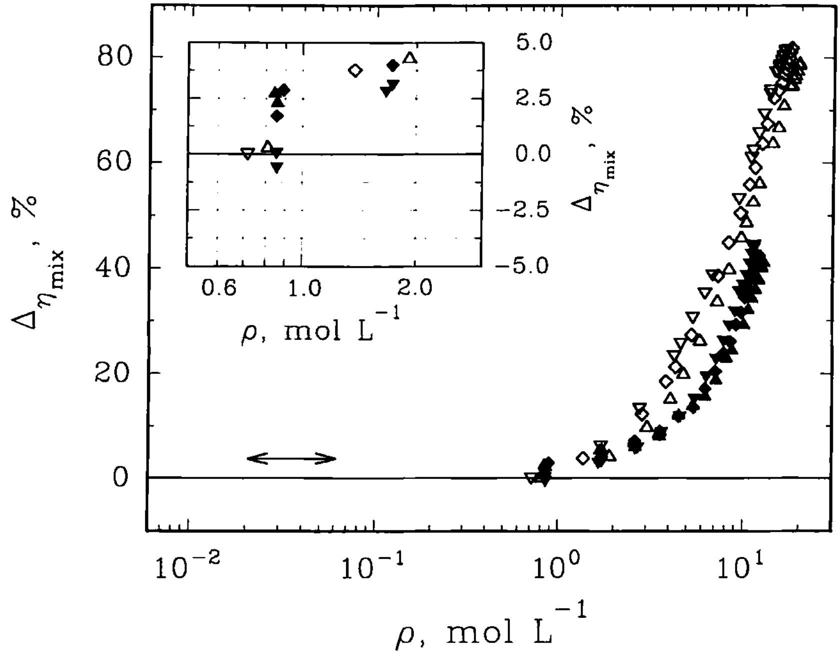

**Fig. 5.** Deviations of experimental viscosity data at 320 and 500 K of Diller and Ely [21] from the extrapolated values of the present paper for the system carbon dioxide + ethane. Open symbols: (▽) $x_{CO_2} = 0.25166$, (◇) $x_{CO_2} = 0.49245$, and (△) $x_{CO_2} = 0.73978$, all at 320 K. Filled symbols: same as open symbols, all at 500 K. (↔) Density range of experimental data used in the correlation. $\Delta_{\eta_{mix}} = 100(\eta_{mix,\,Diller} - \eta_{mix,\,calc})/\eta_{mix,\,Diller}$.

## 4. CONCLUSIONS

A new correlation and extrapolation scheme for simultaneously evaluating viscosity data of binary mixtures and pure substances in the limit of the first density correction has been developed. Three methods were tested.

In these schemes experimentally based values for the viscosity of the two pure substances and for the interaction viscosity, both in the limit of zero density, as well as experimental data for the second pressure virial coefficients of the pure substances and for the interaction second pressure virial coefficient are used to replace the hard-sphere diameters related to the viscosity and the second pressure virial coefficient. The schemes differ with regard to the treatment of the hard-sphere diameters pertaining to the first density correction of the equilibrium radial distribution function at contact.

For the system under discussion, carbon dioxide + ethane, a procedure with a fit of the diameters $\sigma_i^\chi$ to the experimental data of the pure substances and of at least one mixture at moderate densities, with the mixing rule, Eq. (20), for hard-sphere diameters, leads to the best results.

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