Viscosity Measurements on Carbon Monoxide, Nitrogen up to 900 Atmospheres and Correlation to Mass Diffusion

At high pressures the viscosity of gaseous mixtures can be measured with great accuracy. For this reason, viscosity data can offer a profitable means of checking other transport properties of the mixture, such as mass diffusivity, which are difficult to measure accurately.

The present paper is aimed to check the experimental mass diffusivity data for the carbon monoxide-nitrogen mixture, which have been obtained by the dynamic method up to 600 atm. (1, 2). The application of this method at high pressure is relatively recent; it presents some advantages with respect to the discontinuous method but also some operational difficulties. Thus, a check of the results was needed. To this aim, viscosity measurements have been carried out on the same mixture and the correlation between viscosity and mass diffusivity has been derived by Enskog's theory. The carbon monoxide-nitrogen mixture has been chosen, because the molecular parameters of its components are very similar so that the correlation between the two transport properties approximates that given by Enskog for single gases.

CORRELATION BETWEEN VISCOSITY AND MASS DIFFUSIVITY

In the mixture of components A and B, the binary coefficient of mass diffusion D_{AB} , at pressure P can be expressed by:

$$D_{AB} = (D_{AB})_1 \frac{Z}{P X_{AB}}$$
(1)

Z is the compressibility factor of the mixture. X_{AB} is a function of P, depending on the diameters of molecules A and B (3). $(D_{AB})_1$ denotes the diffusion coefficient calculated at P = 1 atm. by the dilute gas theory, at the same temperature of D_{AB} .

In turn, $(D_{AB})_1$ is related to the interaction term, $(\eta_{AB})_1$ (5), which appears in the equation for the mixture viscosity, $(\eta_m)_1$:

$$(\eta_m)_1 = (\eta_m)_1 (\eta_A, \eta_B, \eta_{AB}, x_A, x_B)_1$$
 (2)

As in Equation (1), index 1 refers to the state at P = 1 atm., $(\eta_A)_1$ and $(\eta_B)_1$ being the viscosities of the single components (5) and x_A , x_B the mole fractions.

For the mixture of $A = \text{carbon monoxide and } B = \text{nitro$ $gen at temperature } T = 323$, 1°K. of our measurements, Equation (2) in the form given by Waldmann (6) leads to:

$$\left(\frac{d(\eta_m)_1}{dx_B}\right)_{x_A \to 1} = 1.998 \left[(\eta_{AB})_1 - (\eta_A)_1\right]; \\ \left(\frac{d(\eta_m)_1}{dx_B}\right)_{x_B \to 1} = 2.000 \left[(\eta_B)_1 - (\eta_{AB})_1\right] \quad (3)$$

Table 1 also shows that $(\eta_A)_1 < (\eta_{AB})_1 < (\eta_B)_1$ and that $(\eta_A)_1 \simeq (\eta_B)_1$. According to Equation (3), the interaction term, $(\eta_{AB})_1$, can be simply determined by:

$$(\eta_{AB})_1 \simeq \frac{(\eta_A)_1 + (\eta_B)_1}{2}$$
 (4)

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TABLE 1. VISCOSITY VALUES CALCULATED BY THE DILUTE GAS THEORY (4)

$P = 1$ atm.; $T = 323.1^{\circ}$ K.	A = CO	$B = N_2$	$CO \cdot N_2$
Collision integral (7) Molecular diameter, Å. (7)	0.989 3.706	$0.998 \\ 3.681$	0.9925 3.693
Viscosity, 10 ⁶ poise	187.0	187.8	187.6

At high pressures, a correlation of type (2) is no longer available but we find that the linear dependence of η_m on the mixture composition is substantially fulfilled, with $\eta_A \cong \eta_B$. Hence, the term η_{AB} can still be determined from the experimental values η_A , η_B by an equation of type (4), while the calculation can be carried out by an equation which approximates that given by Enskog for the viscosity of single gases (8):

$$\eta_{i} = (\eta_{i})_{1} \left[\frac{1}{X_{i}} + 0.800 \left(\frac{b_{0}}{\overline{V}} \right)_{i} + 0.761 \left(\frac{b_{0}}{\overline{V}} \right)_{i}^{2} X_{i} \right]$$
$$i = A; B \quad (5)$$

In Equation (5), b_{0i} and \overline{V}_i are the second virial coefficient and the molar volume, respectively, of component *i*. The term X_i is analogous to X_{AB} of Equation (1).

By putting i = AB in Equation (5), combination of Equations (1) and (5) gives:

$$\eta_{AB} \simeq \frac{P}{\alpha \ TZ} \left[D_{AB} + 0.800 \frac{b_0 (D_{AB})_1}{RT} + 0.761 \left(\frac{b_0 (D_{AB})_1}{RT} \right)^2 \frac{1}{D_{AB}} \right]$$
(6)

$$\alpha = 3.52 \ \frac{n_d}{\Omega_v} \tag{7}$$

where Ω_d , Ω_v denote the generalized collision integrals for mass diffusivity and for viscosity, respectively, of the pair *AB*.

When $P \rightarrow 1$, one can recognize in Equation (6) the correlation between viscosity and mass diffusivity as given by Chapman-Enskog (9). When P increases, the decreasing of D_{AB} makes the last term in square brackets more and more important until finally it predominates. In this case Equation (6) predicts an inverse proportionality between viscosity and mass diffusivity as for liquids.

In order to apply Equation (6), specification of b_0 is needed. To this aim, we consider the virial coefficients of the components carbon monoxide and nitrogen and we compare them, after checking how the original formulation of such terms is modified at high pressures.

THE INTERMEDIATE AND HIGH PRESSURE REGIONS

Each of these regions corresponds to a different definition of the terms b_{0i} and X_i , which appear in Equations (5) and (6), the limit between the two regions depending upon the particular gas.

In the intermediate region, according to the original definition of X_i :

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