

Viscosity of the Quaternary Gaseous Mixture He-N<sub>2</sub>-O<sub>2</sub>-CO<sub>2</sub>

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**Synopsis.** The kinetic theory expressions for multi-component gas mixtures are used in conjunction with the law of corresponding states formulated by Kestin, Ro and Wakeham to generate the viscosities of the quaternary gaseous mixture He-N<sub>2</sub>-O<sub>2</sub>-CO<sub>2</sub>. The calculated viscosities are compared with direct measurements performed in a capillary flow apparatus.

The extended law of corresponding states of Kestin, Ro, and Wakeham<sup>1-3</sup>) has provided the correlation of low-density equilibrium and transport properties of the monatomic gases and their mixtures with an uncertainty commensurate with the experimental accuracy. In an earlier paper<sup>4</sup>) it has been established that the viscosity of the quaternary mixture N<sub>2</sub>-O<sub>2</sub>-CO<sub>2</sub>-N<sub>2</sub>O conforms to the extended law of corresponding states. In this work the kinetic-theory expressions for multicomponent gas mixture are used in conjunction with the law of corresponding states and the appropriate scaling parameters  $\sigma_{ij}$  and  $\epsilon_{ij}$  to generate the viscosities of He-N<sub>2</sub>-O<sub>2</sub>-CO<sub>2</sub> mixture. The calculated viscosities are compared with direct measurements made by a capillary flow method at temperatures 0 and 40 °C. For the viscosity we can employ the known formula<sup>5</sup>)

$$\mu(T) = (-1) \begin{vmatrix} H_{11} & H_{12} \cdots H_{1n} & X_1 \\ H_{21} & H_{22} \cdots H_{2n} & X_2 \\ \vdots & \vdots & \vdots \\ H_{n1} & H_{n2} \cdots H_{nn} & X_n \\ X_1 & X_2 \cdots X_n & 0 \end{vmatrix} \begin{vmatrix} H_{11} & H_{12} \cdots H_{1n} \\ H_{21} & H_{22} \cdots H_{2n} \\ \vdots & \vdots \\ H_{n1} & H_{n2} \cdots H_{nn} \end{vmatrix}^{-1} \quad (1)$$

where  $X_i$  is the fraction of component  $i$ , and the element  $H_{ij}$  are written as:

$$H_{ii} = \frac{X_i}{\mu_i} + \sum_{k=1}^n \frac{2X_i X_k}{\mu_{ik}} \frac{M_i M_k}{(M_i + M_k)^2} \left( \frac{5}{3A_{ik}^*} + \frac{M_k}{M_i} \right), \quad (1a)$$

$$H_{ij} = - \frac{2X_i X_j}{\mu_{ij}} \frac{M_i M_j}{(M_i + M_j)^2} \left( \frac{5}{3A_{ij}^*} - 1 \right) \quad i \neq j. \quad (1b)$$

Here  $M_i$  represents the molecular weight of species  $i$  in the mixture. The interaction viscosity  $\mu_{ij}$  is determined from:

$$\mu_{ij} = 5/16(k M_{ij} T/\pi N)^{1/2} (1/\sigma_{ij}^2 \Omega_{22}), \quad (2)$$

where  $M_{ij} = 2M_i M_j / (M_i + M_j)$  and  $\sigma_{ij}$  is a distance scaling parameter.

It should be mentioned that the recently determined scaling parameters for rare-gas pairs by Kestin and co-workers perform a discrepancy by a factor of 1.016 in the distance of zero potential.<sup>6,7</sup>)

The best empirical expressions for the universal functions of the single parameter  $T^* = k_B T / \epsilon_{ij}$  determined by Kestin *et al.* are given as:

$$\Omega_{11} = \Omega^{(1,1)*} = \exp [0.347 - 0.444 (\ln T^*) + 0.093 (\ln T^*)^2 - 0.010 (\ln T^*)^3], \quad 0.5 < T^* < 25, \quad (3)$$

$$\Omega_{22} = \Omega^{(2,2)*} / f_\mu = \exp [0.45667 - 0.53955 (\ln T^*) + 0.18265 (\ln T^*)^2 - 0.03629 (\ln T^*)^3 + 0.00241 (\ln T^*)^4], \quad 1 < T^* < 90, \quad (4)$$

where  $f_\mu$  is higher-order correction to the viscosity and:

$$A^* = \Omega^{(2,2)*} / \Omega^{(1,1)*} \simeq \Omega_{22} / \Omega_{11}.$$

The quantities  $\Omega_{22}$  and  $A_{ij}^*$  are functionals of the potential energy function describing the interaction of molecular pairs. Equations 1–4 together with a knowledge of the scaling parameters  $\sigma_{ij}$  and  $\epsilon_{ij}$  characteristic of the  $i$ - $j$  interaction permit the evaluation of the viscosity  $\mu$  of He-N<sub>2</sub>-O<sub>2</sub>-CO<sub>2</sub> at any composition and any desired temperature.

With the aid of the scaling factors, the universal functions provide us with a sharp criterion which permits us to disqualify guessed analytic forms of the pair potential. The (11-6-8) potential proposed by Hanley and Klein<sup>8</sup>) is acceptable as a correlator data (for  $1 \leq T^* \leq 20$ ), even this potential fails for helium at high reduced temperatures. For the temperature range considered here, we are able (within the precision of the experimental data) to neglect the quantum effects for the present gaseous mixtures.

## Experimental

The measurements reported here were made with a constant-volume gas viscometer similar to that of McCoubrey and Singh.<sup>9</sup>) The viscometer bulb and capillary were thermostated in a water bath except at 0 °C where an ice-slush was used. The bath was controlled to within  $\pm 0.1$  °C. Pressures were measured with a travelling microscope reading to 0.05 mm and the times with a precision stopwatch (calibrated in tenths of a second). The apparatus constant at each temperature was determined from the wellknown Poiseuille equation modified for slip correction. The apparatus constant at each temperature was determined using nitrogen and checked with oxygen (whose viscosities are known). The pressure range in these measurements was 22–11 cm mercury.

Nitrogen was supplied by the Matheson Co. Oxygen and helium were obtained from Fluka Co. Carbon dioxide was

TABLE 1. VISCOSITY OF QUATERNARY MIXTURES

| Compn (mole fraction) |                |                |                 | Temp<br>°C | Viscosity/ $\mu$ P |       |              |
|-----------------------|----------------|----------------|-----------------|------------|--------------------|-------|--------------|
| He                    | N <sub>2</sub> | O <sub>2</sub> | CO <sub>2</sub> |            | Exptl              | Calcd | Deviation(%) |
| 0.10                  | 0.50           | 0.25           | 0.15            | 0          | 178.5              | 176.3 | 1.25         |
|                       |                |                |                 | 40         | 197.2              | 197.0 | 0.10         |
| 0.25                  | 0.25           | 0.25           | 0.25            | 0          | 182.6              | 185.8 | -1.72        |
|                       |                |                |                 | 40         | 208.6              | 207.9 | 0.34         |

supplied by the Chemical Co., Shiraz, Iran. The minimum purities were 99%. For each mixture, viscosity results at each temperature were averaged from at least three runs.

### Results and Discussion

The experimental results for He-N<sub>2</sub>-O<sub>2</sub>-CO<sub>2</sub> are given in Table 1. The viscosity of the two quaternary mixtures can be calculated with the aid of Eqs. 1—4 and scaling parameters, for the gases He, N<sub>2</sub>, O<sub>2</sub>, and CO<sub>2</sub>, taken from Ref. 10. Evaluation of the viscosity is made by using computer program developed by Kestin and co-workers. The scheme presented here may be used for the generation of viscosity for any mixtures of these same gases.

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