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INITIAL DENSITY DEPENDENCE OF GAS VISCOSITY[a]

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The gas viscosity depends on pressure or density and was expressed as a sum of the viscosity which arose from the effect of molecular association and that which was caused from the effect of transfer of the momentum by the molecular collision. The initial density dependence of gas viscosity, $\alpha' = \eta_0^{-1}(\partial\eta/\partial\rho)_{\rho\to0}$, was obtained from the experimental viscosity values and was compared with the theoretically calculated α'-values, where η is viscosity of gas; ρ density; and η_0 the viscosity extrapolated to zero density.

For a nonpolar gas, the experimental α'-values were obtained from the viscosity data of eleven kinds of gases. The effect of molecular association on the gas viscosity was slight, and the contribution from collisional transfer was correlated by use of T^*, where $T^*=T/\epsilon k^{-1}$; T is absolute temperature; ϵ the potential parameter; and k the Boltzman constant. The improved α'-values were in better agreement with the experimental ones than the original theoretical ones.

For a polar gas, the experimental α'-values were obtained from the viscosity data of NH_3, SO_2, H_2O, and three hydrocarbon halides. The effect of molecular association on the gas viscosity was extremely larger than that of a nonpolar gas. The theoretically calculated α'-values were correlated by use of the polarity parameter, δ. The improved α'-values were in good agreement with the experimental ones except H_2O.

Introduction

The gas viscosity η depends on pressure P or density ρ and can be approximately expressed as a function of P or ρ at constant temperature as follows:

$$\eta=\eta_0(1+\alpha P+\cdots\cdots) \qquad (1)$$

or

$$\eta=\eta_0(1+\alpha'\rho+\cdots\cdots) \qquad (2)$$

where η_0 is the viscosity extrapolated to zero pressure (or density), $\alpha=\eta_0^{-1}(\partial\eta/\partial P)_{P\to0}$ the initial pressure dependence of viscosity, and $\alpha'=\eta_0^{-1}(\partial\eta/\partial\rho)_{\rho\to0}$ the initial density dependence of viscosity[1]. The viscosities of nonpolar-polar gaseous mixtures under pressures containing ammonia as a polar gas, i.e., the nitrogen-ammonia system[1], the hydrogen-ammonia system[2], and the argon-ammonia system[3] were measured, and the effect of the nonpolar gases upon α' of ammonia was investigated. α' of the

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nonpolar gas-ammonia systems increased, consequently, with a rise in temperature and in the mole fraction of the nonpolar gases and became almost constant above a certain concentration of the nonpolar gases regardless of temperature. Moreover, it was concluded in the three systems that the larger the molecular weight of the nonpolar gases was, the stronger the influence of the nonpolar molecules upon α' of ammonia was. In this papes α' of pure nonpolar and polar substances have theoretically been considered.

Initial Density Dependence of Gas Viscosity

It is well known[4] that the transport coefficients of gases, such as viscosity, thermal conductivity, and diffusion, depend on pressure or density.

For nonpolar gases, as the pressure coefficient of viscosity, $(\partial\eta/\partial P)_T$, decreases with a rise in temperature, the temperature coefficient of viscosity, $(\partial\eta/\partial T)_P$, is positive at the lower pressure region and is negative at the higher pressure region. Thus the viscosity isotherms cross one another in the relation between the gas viscosity and pressure. However, in the relation between the gas viscosity and density the distances between each viscosity isotherm are nearly equal and the isotherms do not cross. Accordingly, it is easier to treat the gas viscosity as a function of density than pressure.

For polar gases, the viscosity decreases with an increase in density bolow a certain temperature and increases with an increase in density in the higher temperature region[1,5~7]. Above the critical temperature, it is supposed that the viscosity behavior similar to that of nonpolar gases can be observed.

In order to obtain α' from the experimental viscosity values, five kinds of the polynomials of density (third, fourth, fifth, sixth, and nineth degree equation) were tested. In the case that there are enough numbers of accurately measured points in the low density region, a linear or quadratic equation represents adequately the experimental values. However, the coefficients of five kinds of polynomials were determined from the experimental viscosity values of, for examples, nitrogen[1], hydrogen[2] and argon[3], because the way of taking measured points is various depending upon the data. The results of the determination at 100°C among them are shown in Table 1.

Compared Table 1 with the figures, *i. e.*, Fig. 1 in Ref. 2 and Fig. 1 in Ref. 3, it can be seen that the lower order equation can better reproduce the low pressure (density) region rather than the higher order equation though the higher order equation expresses well whole measured points. There are some reports[8~13] about the coefficients of the polynomials. Hanley *et al.*[10] have recommended the

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Table 1 Coefficients of polynomials

gas	T/℃	n	η_0	order number 1	2	3	4	5	6	7	8	9	stand. dev.
N_2	100	23	211.2	106.3	907.9	-106×10							0.149
			211.3	94.1	150.1×10	-102×10^{2}	435×10^{3}						0.150
			211.2	113.6	7.89	299×10^{2}	394×10^{3}	167×10^{4}					0.151
			211.2	140.3	-293.0×10	150×10^{3}	-260×10^{4}	204×10^{5}	-596×10^{6}				0.153
			211.3	75.3	434.9×10	-132×10^{3}	139×10^{4}	696×10^{5}	225×10^{6}	-125×10^{7}	141×10^{8}	-496×10^{8}	0.172
H_2	100	15	104.8	30.7	200.4×10^{3}	-106×10^{4}							0.040
			104.8	157.1	-679.1×10^{3}	180×10^{6}	-126×10^{7}						0.034
			104.8	$-$ 62.6	177.0×10^{3}	-763×10^{4}	133×10^{6}	-784×10^{7}					0.021
			104.8	-199.2	396.2×10^{3}	-205×10^{6}	471×10^{8}	-485×10^{10}	182×10^{11}				0.017
			104.8	-218.8	409.7×10^{3}	-197×10^{6}	388×10^{8}	-275×10^{10}	137×10^{12}	-531×10^{14}	752×10^{15}	-313×10^{17}	0.020
A_r	100	12	272.9	118.4	451.3	-250							0.189
			272.8	123.0	298.7	129×10	-481×10						0.200
			272.8	130.2	$-$ 83.2	820×10	-550×10^{3}	126×10^{5}					0.213
			272.7	192.3	-474.8×10	134×10^{3}	-158×10^{4}	858×10^{5}	176×10^{7}				0.213
			272.8	154.8	-162.8×10	351×10^{3}	-527×10^{4}	-380×10^{6}	348×10^{7}	-100×10^{9}	511×10^{9}	125×10^{10}	0.294

density function which involves the logarithmic term. The density function is regarded as the higher order equation, as the term which involves logarithm summarizes higher order terms. If the viscosity measurements were accurately made and there were several measured values containing some data points in the low density range, it was concluded that the cubic equation reproduced sufficiently the experimental values because the standard deviations of the equations from the experimental values were comparable.

$$\eta=\eta_0(1+\alpha'\rho+\beta'\rho^2+\gamma'\rho^3) \tag{3}$$

Each coefficient of Eq. (3) was obtained by means of the least-squares method.

The initial pressure dependence of gas viscosity, α, was theoretically considered by Stogryn and Hirschfelder[11]. The gas viscosity was expressed as a sum of the viscosity which arose from the effect of molecular association and that which was caused from the effect of transfer of the momentum by the molecular collision. Since α or α' was the coefficient of the first order term of Eq. (1) or (2), only the dimer formation was considered in molecular association. When assuming the gas to be a mixture of monomers and dimers, η can be represented as

$$\eta=\eta_{\rm dm}+\eta_{\rm c} \tag{4}$$

where $\eta_{\rm dm}$ is the viscosity of the mixture of dimers and monomers, and $\eta_{\rm c}$ is the viscosity which was influenced by the collisional transfer. $\eta_{\rm dm}$ can be obtained from Chapman-Enskog's theory in terms of the contribution of dimers to the second virial coefficient and $\eta_{\rm c}$ can be obtained by extending Enskog's theory by means of the second virial coefficient.

The second virial coefficient $B(T)$ can by written as[12]

$$B(T)=B_{\rm f}(T)+B_{\rm b}(T)+B_{\rm m}(T) \tag{5}$$

where $B_{\rm f}(T)$ denotes the contribution attributed to the collisions between free molecules; $B_{\rm b}(T)$ results from the equilibrium constant for the formation of bound double molecules; and $B_m(T)$ results from the equilibrium constant for the formation of metastable double molecules. All these quantities are expressed as a function of the intermolecular potential function. Expressing the relation of the dimer formation as

$$2X \rightleftharpoons X_2, \tag{6}$$

the equilibrium constant of the dimerization reaction $K(T)$ is given by[12]

$$K(T) = n_d V / n_m^2 = -[B_b(T) + B_m(T)] = -B_d(T), \tag{7}$$

where n_d and n_m are the numbers of moles of dimers and monomers in the volume V. The quantities of $B_b(T)$ and $B_m(T)$ are expressed by complicated integrals, and are tablated in reduced form, $B_b^\star(T)$ and $B_m^\star(T)$, in Ref. 12 for a nonpolar gas and as $B_d^\star(T)$ in Ref. 13 for a polar gas, where $B_b^\star(T) = B_b(T)/b_0$; $B_m^\star(T) = B_m(T)/b_0$; $B_d^\star(T) = B_d(T)/b_0$; $b_0 = \frac{2}{3}\pi N\sigma^3$; N is Avogadro's number; and σ is the potential parameter. The $B_d^\star(T)$ values for the Stockmayer potential are shown in Fig. 1 as a function of polarity parameter δ and the reduced temperature T^*, where $T = T^*/\varepsilon k^{-1}$; T is absolute temperature; ε is the potential parameter; and k is the Boltzman constant. For $\delta = 0$, the $B_d^\star(T)$ values become those for the Lennard-Jones potential.

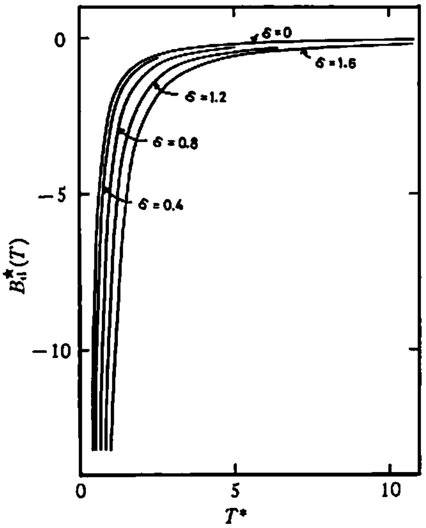

Fig. 1 $B_d^\star(T)$ value for the Stockmayer potential

The final expression for the initial pressure dependence of viscosity was obtained as follows[11]:

$$\frac{RT\alpha}{b_0} = -B_d^\star(T)\left[0.5113 + 0.2481\left(\frac{6D_{dm}}{D_{mm}} - 4.5455\frac{D_{mm}}{D_{dm}}\right)\right]$$
$$+0.1750\left[B^\star(T) + T^*\left(\frac{dB^\star}{dT^*}\right)\right] \tag{8}$$

where

$$\frac{D_{dm}}{D_{mm}} = \frac{\sqrt{3}}{2}\left(\frac{\sigma_{mm}}{\sigma_{dm}}\right)^2 \frac{\Omega_{mm}^{\star(1,1)}}{\Omega_{dm}^{\star(1,1)}} \tag{9}$$

the D's are diffusion coefficients and $\Omega^{\star(1,1)}$'s are the reduced collision integrals for diffusion; the

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subscripts dm and mm denote dimer-monomer and monomer-monomer interactions, respectively; and R is gas constant. Eq. (9) was obtained by Chapman-Enskog's theory. The left hand side of Eq. (8) can be expressed by the use of α' instead of α in the low density range as follows:

$$\frac{M\alpha'}{b_\circ}=-B_{\mathrm{d}}^{\star}(T)\left[0.5113+0.2481\left(\frac{6D_{\mathrm{dm}}}{D_{\mathrm{mm}}}-4.5455\frac{D_{\mathrm{mm}}}{D_{\mathrm{dm}}}\right)\right]$$

$$+0.1750\left[B^{\star}(T)+T^{*}\left(\frac{dB^{\star}}{dT^{*}}\right)\right] \qquad (10)$$

where M is molecular weight of a gas. The comparison between the experimental values of α' obtained from Eq. (3) and the theoretical ones calculated from Eq. (10) will be made below.

Comparison of experimental and theoretical initial density dependence of viscosity

Initial density dependence of viscosity for nonpolar gas: Among the experimental values of the gas viscosity under pressures, eleven kinds of nonpolar gases, of which viscosities were precisely determined in the low density region, were adopted, that is, $N_2$1,14~16), $O_2$14,17,18), $H_2$2,16,17,19), $D_2$14), He14~16,20,21), Ne14,21,22), Ar3,14~16,20~22), Kr14,17,23), Xe14), $CO_2$14,17,22~24) and $CH_4$17,25,26). Each coefficient of Eq. (3) was obtained by use of the viscosity data mentioned above. Figure 2 shows the

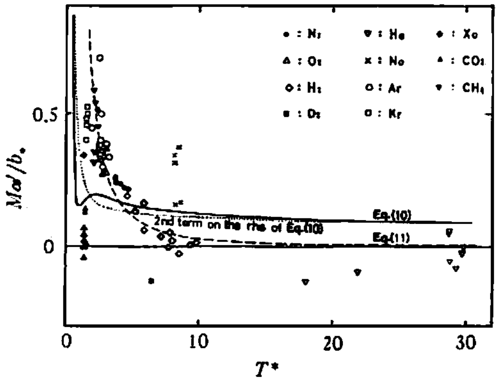

Fig. 2 Comparison of experimental value of $M\alpha'/b_\circ$ with original theoretical one obtained from Eq. (10) and corrected one from Eq. (11) for nonpolar gas

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relation between $M\alpha'/b_0$ and T^*. On the whole, the values of $M\alpha'/b_0$ decreases with an increase in T^*-value and becomes almost zero at high temperature.

The Lennard-Jones potential function was applied to the theoretical calculation of α' of a nonpolar gas. The force constants for the Lennard-Jones potential used in this study are listed in Table 2. Stogryn *et al.*[28] determined empirically two parameters, $\varepsilon_{dm}/\varepsilon_{mm}=1.32$ and $\sigma_{dm}/\sigma_{mm}=1.04$, in the computation of the initial pressure dependence of thermal conductivity, β. The experimental values of β were in good agreement with the theoretical ones calculated by use of the empirical ones. Therefore, the same numerical values were used in the calculation of the right hand side (the rhs) of Eq. (10). The calculated values of $M\alpha'/b_0$ are shown by a solid line in Fig. 2. It can be seen from the figure that the calculated values do not agree with the experimental ones. In order to obtain the appropriate value of two parameters, the theoretical ones of α' calculated by substituting various numerical ones for $\varepsilon_{dm}/\varepsilon_{mm}$ and σ_{dm}/σ_{mm}. When $\varepsilon_{dm}/\varepsilon_{mm}=1.01$ and $\sigma_{dm}/\sigma_{mm}=1.02$, the theoretical α'-values represented the experimental ones below $T^*=5$, but the theoretical ones were much the same to the solid line in Fig. 2 above $T^*=10$. The parameters which represented the experimental α'-values on the whole temperature range could not be found. Since it was considered that the effect of dimers on the viscosity was a little for nonpolar gases, only the second term on the right hand side of Eq. (10), which expressed the effect of the collisional transfer, was taken up and plotted by a dotted line in Fig. 2. As shown in the figure, a difference between the solid line and the dotted one is not so large. Namely, it is concluded that the first term on the right hand side of Eq. (10) contributes hardly to the gas viscosity compared with the second term. Then, the first term is neglected and the second term is correlated by use of T^* so as to agree the experimetnal α'-values as follows:

Table 2 The force constants for the Lennard-Jones

Gas	ε/k [K]	σ [10^{-10}m]	Reference
N_2	79.8	3.73_2	2
O_2	113	3.433	4
H_2	38.0	2.93_2	2
D_2	39.3	2.948	4
He	10.2	2.576	4
Ne	35.7	2.789	27
Ar	116	3.44_2	3
Kr	190	3.61	4
Xe	229	4.055	4
CO_2	213	3.897	4
CH_4	144	3.796	4

$$\frac{M\alpha'}{b_0}=0.1750\left[B^\star(T)+T^*\left(\frac{dB^\star}{dT^*}\right)\right]\left[10(T^*)^{-\frac{3}{2}}\right] \tag{11}$$

The corrected term in Eq. (11) will be able to be assigned a theoretical significance. The improved

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curve of $M\alpha'/b_0$ obtained from Eq. (11) is shown by a dashed line in Fig. 2. The improved values are in better agreement with the experimental ones of α' than those obtained from Eq. (10) except carbon dioxide and neon.

Hanley *et al.*[29] obtained the coefficients of first order term of density by the use of the modified Enskog theory with the second and third virial coefficients for the Lennard-Jones potential, and showed the good results. However, the difference between the theoretical and experimental values (argon, oxygen and hydrogen) became larger as temperature increased for $T^* > 10$ though the both values are in reasonable agreement for $T^* < 10$. Besides, the theory of Hanley *et al.* was not extended to a polar gas.

Initial density dependence of viscosity for polar gas: α' of polar gases was treated by the same way of thinking as α' of nonpolar gases. However, few papers have been published on the viscosity of polar gases under pressures. Especially, the viscosity data from which α' can be obtained are very few, i. e., ammonia[1,5,30], sulfur dioxide[6], and water vapor[31~34]. The coefficients of Eq. (3) were determined from the viscosity data for polar gases as well as for nonpolar gases.

The theoretical values of α' of a polar gas were also calculated from Eq. (10). In the calcutation the Stockmayer potential function was used and it was assumed that all the relative orientations between two dipoles were in equal probability at a molecular collision. The theoretically calculated α'-values of a polar gas decreased with an increase in the polarity parameter, δ, when the two parameters, $\varepsilon_{dm}/\varepsilon_{mm}$ and σ_{dm}/σ_{mm}, were constant. In analogy with the case of a nonpolar gas, the parameters which expressed the experimental values of $M\alpha'/b_0$ in the temperature range could not be obtained. So, the parameters which were semi-empirically determined[13] were used in the calculation, that is,

$$\varepsilon_{dm}/\varepsilon_{mm}=1.32, \qquad \sigma_{dm}/\sigma_{mm}=1.16.$$

The force constants for the Stockmayer potential are presented in Table 3.

Table 3 The force constants for the Stockmayer potential

Gas	δ	ε/k [K]	σ $[10^{-10}\mathrm{m}]$	Reference
NH_3	0.70	358	3.15	1
SO_2	0.42	347	4.04	4
H_2O	1.0	775	2.52	4
CCl_2F_2	0.03	210	5.518	
$CClF_3$	0.04	196	5.002	
$CHClF_2$	0.28	196	5.035	

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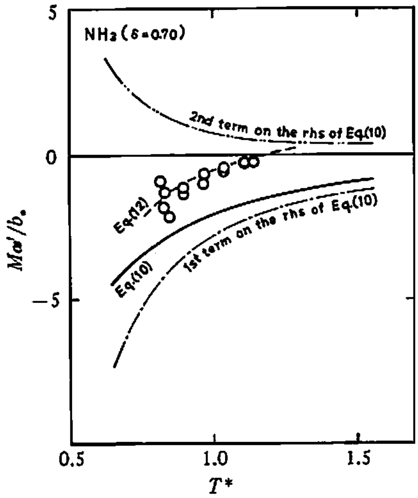

Fig. 3 Comparison of experimental value of $M\alpha'/b_0$ with original theoretical one obtained from Eq. (10) and corrected one from Eq. (12) for ammonia

The theoretical values of $M\alpha'/b_0$ of ammonia obtained from Eq. (10) are shown by a solid line in Fig. 3, in comparison with the experimental ones denoted by the symbol O. The theoretical curve was lower than the experimental values, and the similar behavior was seen in the case of the other polar gases. As expressed by Eq. (4), it was considered that the viscosity of polar gases was divided into two parts. The first term on the right hand side of Eq. (10), which expresses the contribution from the dimer-monomer mixtures, is denoted by a dot-dash-line, and the second term, which expresses the contribution from the collisional transfer, is denoted by a two-dot-dash-line in Fig. 3. It is clear from the figure that the effect of molecular association of polar substances on the gas viscosity is extremely larger than that of nonpolar substances, even if judging from the experimental values of $M\alpha'/b_0$ the contribution of molecular association is actually smaller than that obtained from the theoretical calculation. In the case of a polar gas, the right hand side of Eq. (10) is correlated by use of δ and T^* as follows:

$$\frac{M\alpha'}{b_0} = -B_{\mathrm{d}}^{\star}(T)\left[0.5113+0.2481\left(\frac{6D_{\mathrm{dm}}}{D_{\mathrm{mm}}}-4.5455\frac{D_{\mathrm{mm}}}{D_{\mathrm{dm}}}\right)\right]\frac{10}{(T^*)^{2.3}}\log(1+\sqrt[6]{\delta})$$
$$+0.1750\left[B^{\star}(T)+T^*\left(\frac{\mathrm{d}B^{\star}}{\mathrm{d}T^*}\right)\right]\frac{10}{(T^*)^{1.5}} \qquad (12)$$

The corrected term in Eq. (12) will be also able to be assigned a theoretical significance. For $\delta=0$, Eq. (12) is equal to Eq. (11). The improved curve obtained from Eq. (12) is denoted by a dashed line in Fig. 3 and is in better agreement with the experimental values than the original theoretical curve obtained from Eq. (10) in the experimental conditions.

The above-described method was also applied to sulfur dioxide and water vapor, and in addition applied to three hydrocarbon halides, dichlorodifluoromethane (R-12)[35], chlorotrifluoromethane (R-13)[36] and chlorodifluoromethane (R-22)[37], which were measured by Iwasaki *et al.* Figure 4 shows

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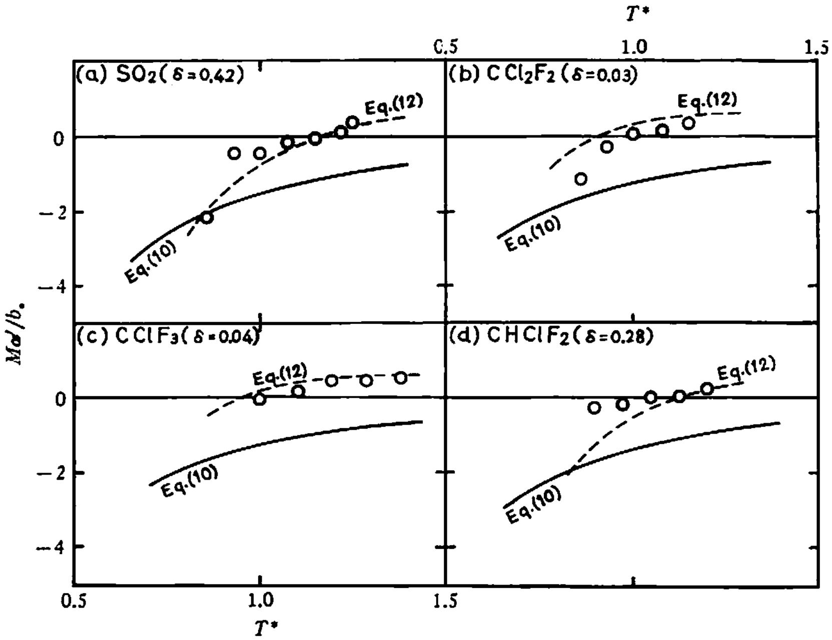

Fig. 4 Comparison of experimental and calculated value of $M\alpha'/b_*$ for sulfur dioxide (a), R-12 (b), R-13 (c), and R-22 (d)

the original theoretical values obtained from Eq. (10) (denoted by solid lines) and the improved ones obtained from Eq. (12) (by dashed lines) in comparison with the experimental values for SO_2, CCl_2F_2 (R-12), $CClF_3$ (R-13) and $CHClF_2$ (R-22). As can be seen from Fig. 4, each improved curve is in better agreement with the experimental values than the original theoretical ones in the experimental conditions. The force constants of the three hydrocarbon halides presented in Table 3 were determined by use of Chapman-Enskog's theory and by applying the experimental viscosity data at atmospheric

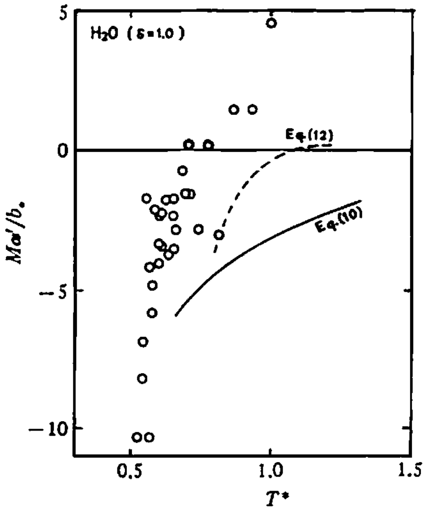

Fig. 5 Comparison of experimental and calculated value of $M\alpha'/b_*$ for water vapor

pressure in the same way as described in Refs. 2 and 3.

Figure 5 shows the comparison of the calculated and experimental values of $M\alpha'/b_0$ of warer vapor. As shown in the figure, the improved values calculated from Eq. (12) (denoted by a dashed line) are almost parallel to the experimental α'-values and do not agree with the most part of them. However, any discussion about α' of water vapor can not be adequately made beceuse the number of the measured points of water vapor in low density region was less than that of the other polar gases and the scattering of the viscosity data was large. Moreover, it can be considered that the force constants of water vapor come into question.

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