

On the Relation between Total and Partial Pressure of Gas Mixture

By

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It may be inadequate, in general, to apply Dalton's law to an imperfect gas mixture. In this paper, the total pressure of an imperfect gas mixture is considered from the view point of statistical mechanics. Namely, by expressing the equation of state in the virial expansion form, the total pressure of a gas mixture was determined analytically by using the second and the third virial coefficients calculated from the values for each component gas. For one example, the second and third virial coefficients of air were calculated by two methods and compared with each other as well as with other values which were determined directly from experimental data with air. From the numerical calculations for air and also for water-mercury vapour mixture, it has been made clear that there is some difference between the total pressure calculated by our methods and the value calculated from Dalton's law, and this difference increases with the degree of imperfectness of the gas mixture.

Introduction

Though Dalton's law is used for the purpose of determining the total pressure of gas mixtures from the partial pressures of the component gases, it may be not appropriate to apply this law to an imperfect gas mixture, since this law was established for a perfect gas. It is believed that statistical mechanics can be used for determining the total pressure of a mixture of imperfect gases from the properties of its component gases. In this paper, some considerations of this approach are described. The total pressure of a gas mixture may be calculated from the equation of state of the mixture. The equation of state of an imperfect gas mixture is expressed in the virial expansion form, and the virial coefficients are determined from those of each component gas under certain assumptions. Two methods are proposed here for relating the virial coefficients of a gas mixture to those of its component gases.

In order to compare those two methods with other results, the virial coefficients and the total pressure of air, which may be considered as a mixture of nitrogen,

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oxygen and argon, are calculated numerically. In addition, the results of numerical calculations for water-mercury vapour mixtures are given, and the difference between them and the results from Dalton's law are discussed.

Analytical Considerations

In order to know the pressure of a gas mixture and its component gases in any arbitrary condition, the equation of state of the gas mixture and its component gases ought to be determined. From statistical thermodynamics, we know that the equation of state of a ν -component gas mixture and of its component gases may be written in the virial expansion form as follows,

$$\frac{pv}{RT} = 1 + \frac{B(T)}{v} + \frac{C(T)}{v^2} + \dots \quad (1)$$

$$\frac{p_1 v_1}{R_1 T} = 1 + \frac{B_1(T)}{v_1} + \frac{C_1(T)}{v_1^2} + \dots$$

$$\frac{p_2 v_2}{R_2 T} = 1 + \frac{B_2(T)}{v_2} + \frac{C_2(T)}{v_2^2} + \dots \quad (2)$$

.....

$$\frac{p_\nu v_\nu}{R_\nu T} = 1 + \frac{B_\nu(T)}{v_\nu} + \frac{C_\nu(T)}{v_\nu^2} + \dots$$

where, p : absolute pressure R : gas constant
 T : absolute temperature v : specific volume
 B & C : 2nd & 3rd virial coefficient (function of temperature only)
 $1, 2, \dots, \nu$ (subscript): values for each component gas.

By means of statistical mechanics, these virial coefficients may be expressed in terms of the intermolecular potential energy function. As the intermolecular potential energy function, the Lennard-Jones potential function has been available for spherical nonpolar molecules, and the Stockmayer potential function for simple polar molecules. These functions φ are as follows,

Lennard-Jones potential function

$$\varphi(r) = 4\varepsilon\{(\sigma/r)^{12} - (\sigma/r)^6\}. \quad (3)$$

Stockmayer potential function

$$\varphi(r, \theta_1, \theta_2, \phi_2 - \phi_1) = 4\varepsilon\{(\sigma/r)^{12} - (\sigma/r)^6\} - (\mu/r)^3 g(\theta_1, \theta_2, \phi_2 - \phi_1) \quad (4)$$

where, σ & ε : force constants
 r : intermolecular distance
 μ : dipole moment of the molecule
 $g = 2 \cos \theta_1 \cos \theta_2 - \sin \theta_1 \sin \theta_2 \cos(\phi_2 - \phi_1)$
 θ & ϕ : cylindrical angular coordinates of dipole vector.

For the Lennard-Jones potential function the second and the third virial coefficients are given as follows,

$$B^*(T^*) = \sum_{j=0}^{\infty} b^{(j)} T^{*-(2j+1)/4} \quad (5)$$

$$C^*(T^*) = \sum_{j=0}^{\infty} c^{(j)} T^{*-(j+1)/2} \quad (6)$$

$$T^* = kT/\varepsilon, \quad (7)$$

$$B^* = B/\frac{3}{8}\pi\tilde{N}\sigma^3, \quad C^* = C/(\frac{3}{8}\pi\tilde{N}\sigma^3)^2$$

where, k : Boltzmann constant

\tilde{N} : Avogadro number.

The coefficients $b^{(j)}$ are given by $b^{(j)} = -(2^{j+1/2}/4j!) \Gamma\left(\frac{2j-1}{4}\right)^{11}$, and the values $c^{(j)}$ have been calculated by Kihara². For the Stockmayer potential function, the second and third virial coefficients have been calculated by Rowlinson and others³ and expressed in the following form as a function of T^* and t^* .

$$B(T) = \frac{3}{8}\pi N\sigma^3 B^*(T^* \cdot t^*) \quad (8)$$

$$C(T) = (\frac{3}{8}\pi N\sigma^3)^2 C^*(T^* \cdot t^*) \quad (9)$$

$$t^* = \mu^{*2}/\sqrt{8}, \quad \mu^* = \mu/\sqrt{\varepsilon\sigma^3}. \quad (10)$$

On the other hand, the virial coefficients for a gas mixture depend not only upon the interaction between like molecules but also upon the interaction between unlike molecules. Therefore, in order to calculate the virial coefficients for a gas mixture, it is necessary to know the potential energy function expressing the interaction between pairs of unlike molecules. For this purpose, the following empirical combining laws are used, which relate the force constants between unlike molecules with those between like molecules according to Hirschfelder's method⁴.

$$\sigma_{\alpha\beta} = \frac{\sigma_{\alpha} + \sigma_{\beta}}{2} \quad (11)$$

$$\varepsilon_{\alpha\beta} = (\varepsilon_{\alpha} \cdot \varepsilon_{\beta})^{1/2}. \quad (12)$$

Thus, the second virial coefficient $B_{\alpha\beta}(T)$ is calculated by substituting these force constants into equation (5).

For the third virial coefficient of a gas mixture, in order to relate the third virial coefficient $C_{\alpha\beta\gamma}$ between unlike molecules with $C_{\alpha\alpha\alpha}$ of the pure species, the following equivalent force constants are assumed in the same manner.

$$\sigma_{\alpha\beta\gamma} = \frac{\sigma_{\alpha} + \sigma_{\beta} + \sigma_{\gamma}}{3} \quad (13)$$

$$\varepsilon_{\alpha\beta\gamma} = (\varepsilon_{\alpha} \cdot \varepsilon_{\beta} \cdot \varepsilon_{\gamma})^{1/3}. \quad (14)$$

The third virial coefficient between unlike molecules is calculated by introducing

these equivalent force constants into equation (6) for the third virial coefficient for a pure species.

Though the virial coefficients between different species can be calculated by using the above mentioned method, it is considered more practical to determine the virial coefficients of the gas mixture directly from the experimental data of the p - v - t relations of the component gases. So, in this paper as the second method, the virial coefficients of the gas mixture are determined by using the virial coefficients between different species calculated under the assumption that the virial coefficients between different species are given as the arithmetical mean of the virial coefficients for the pure species as follows,

$$B_{\alpha\beta} = (B_{\alpha} + B_{\beta})/2 \quad (15)$$

$$C_{\alpha\beta\gamma} = (C_{\alpha} + C_{\beta} + C_{\gamma})/3. \quad (16)$$

From these virial coefficients in both methods, the second and third virial coefficients of the gas mixture are given as follows⁵⁾,

$$B(T) = \sum_{\alpha=1}^{\nu} \sum_{\beta=1}^{\nu} B_{\alpha\beta}(T) x_{\alpha} \cdot x_{\beta} \quad (17)$$

$$C(T) = \sum_{\alpha=1}^{\nu} \sum_{\beta=1}^{\nu} \sum_{\gamma=1}^{\nu} C_{\alpha\beta\gamma}(T) x_{\alpha} \cdot x_{\beta} \cdot x_{\gamma} \quad (18)$$

where x_{α} , x_{β} and x_{γ} are the mole fractions of α -, β -, and γ -species in the mixture, and $B_{\alpha\alpha}$ is the second virial coefficient for the pure substance α , and $B_{\alpha\beta}$ is the second virial coefficient calculated for the different species α and β . The $B_{\alpha\beta}$ and $C_{\alpha\beta\gamma}$ have the same meaning for any permutation of the indices, and $C_{\alpha\alpha\alpha}$ is the third virial coefficient for pure substance α .

From the second and third virial coefficients of the gas mixture obtained as above mentioned, the pressures of the component gases and the total pressure of the gas mixture can be calculated by using these virial coefficients. The gas constant and the specific volume v for a gas mixture with ν -components are written as follows,

$$R = \sum_{i=1}^{\nu} \frac{R_i}{v_i} / \sum_{i=1}^{\nu} \frac{1}{v_i}, \quad v = 1 / \sum_{i=1}^{\nu} \frac{1}{v_i}. \quad (19)$$

Thus, the total pressure of a gas mixture can be calculated by the following equation,

$$p = T \left(\sum_{i=1}^{\nu} \frac{R_i}{v_i} \right) \left\{ 1 + \left(\sum_{i=1}^{\nu} \frac{1}{v_i} \right) \sum_{\alpha=1}^{\nu} \sum_{\beta=1}^{\nu} B_{\alpha\beta} x_{\alpha} \cdot x_{\beta} + \left(\sum_{i=1}^{\nu} \frac{1}{v_i} \right)^2 \sum_{\alpha=1}^{\nu} \sum_{\beta=1}^{\nu} \sum_{\gamma=1}^{\nu} C_{\alpha\beta\gamma} x_{\alpha} \cdot x_{\beta} \cdot x_{\gamma} \right\} \quad (20)$$

where,

$$x_i = \left(\frac{1}{M_i v_i} \right) / \sum_{i=1}^{\nu} \frac{1}{M_i v_i}, \quad i = \alpha, \beta \text{ and } \gamma. \quad (21)$$

M : molecular weight.

Numerical Example

1. For air:—

For air which is considered a mixture with three components (78 mole per cent nitrogen, 21 mole per cent oxygen and 1 mole per cent argon), by neglecting all coefficients beyond the third virial coefficient, the total pressure is as follows,

$$p = T \left(\frac{R_N}{v_N} + \frac{R_O}{v_O} + \frac{R_A}{v_A} \right) \left\{ 1 + \left(\frac{1}{v_N} + \frac{1}{v_O} + \frac{1}{v_A} \right) B + \left(\frac{1}{v_N} + \frac{1}{v_O} + \frac{1}{v_A} \right)^2 C \right\}$$

$$B = B_N x_N^2 + B_O x_O^2 + B_A x_A^2 + 2B_{NO} x_N x_O + 2B_{NA} x_N x_A + 2B_{OA} x_A x_O$$

$$C = C_N x_N^3 + C_O x_O^3 + C_A x_A^3 + 3C_{NAA} x_N x_A^2 + 3C_{NOO} x_N x_O^2 + 3C_{NNO} x_N^2 x_O + 3C_{NNA} x_N^2 x_A + 3C_{OOA} x_A x_O^2 + 3C_{OAA} x_O x_A^2 + 6C_{NOA} x_N x_O x_A$$

where the subscripts *N*, *O* and *A* are the values for nitrogen, oxygen and argon, respectively. On the other hand, the partial pressures of the component gases are given by the following three equations,

$$p_N = T \left(\frac{R_N}{v_N} \right) \left\{ 1 + \frac{1}{v_N} B_N + \frac{1}{v_N^2} C_N \right\}$$

$$p_O = T \left(\frac{R_O}{v_O} \right) \left\{ 1 + \frac{1}{v_O} B_O + \frac{1}{v_O^2} C_O \right\}$$

$$p_A = T \left(\frac{R_A}{v_A} \right) \left\{ 1 + \frac{1}{v_A} B_A + \frac{1}{v_A} C_A \right\},$$

where the second and third virial coefficients of nitrogen, oxygen and argon $B_N, C_N; B_O, C_O$ and B_A, C_A are calculated by using the Lennard-Jones potential energy function with the force constants $\sigma_N = 3.698 \text{ \AA}$, $\epsilon_N/k = 95.05^\circ\text{K}$; $\sigma_O = 3.560 \text{ \AA}$, $\epsilon_O/k = 117.5^\circ\text{K}$; $\sigma_A = 3.405 \text{ \AA}$, $\epsilon_A/k = 119.8^\circ\text{K}$ that have been given by Bird and others from the experimental data of Horborn⁶⁾, Newitt⁷⁾ and Michels⁸⁾. Figures 1 & 2 show the second and the third virial coefficients of nitrogen, oxygen and argon, respectively. In Figs. 3 & 4, $B_{NO}, B_{NA}, B_{AO}; C_{NOO}, C_{NNO}$ computed from the empirical combining law and the equivalent force constants mentioned above, are shown with the full lines, and those values calculated by using the arithmetical combining law for the virial coefficients are shown with the chain lines. Figs. 5 & 6 show the second and the third virial coefficients of air computed by the two methods described above, and it is apparent that there is good agreement among them. In Fig. 7, for comparison, the second and third virial coefficients of air given by Mori⁹⁾ from the experimental values of air, are shown with broken lines, and the chain lines show those calculated by using the force constants of air given by Bird and others derived from the experimental data for air of Horborn and others¹⁰⁾.

From this example, it is obvious that the three values for the second virial coefficient agree very well with each other and the agreement between the values for

the third virial coefficient is moderately good. And therefore, it may be adequate to determine the second and third virial coefficients of air from the data for its component gases, in such a way.

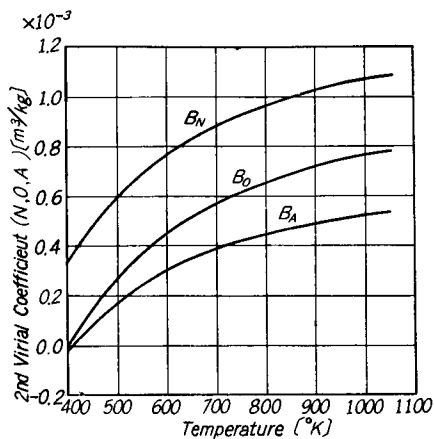


Fig. 1. The second virial coefficient for nitrogen, oxygen and argon.

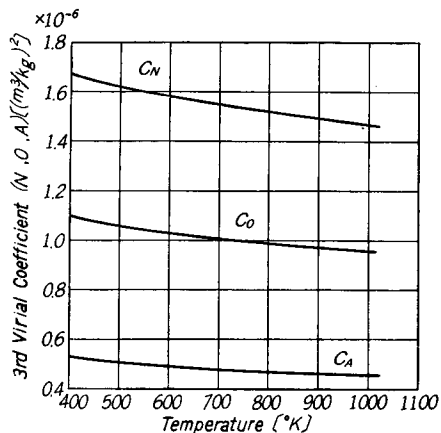


Fig. 2. The third virial coefficient for nitrogen, oxygen and argon.

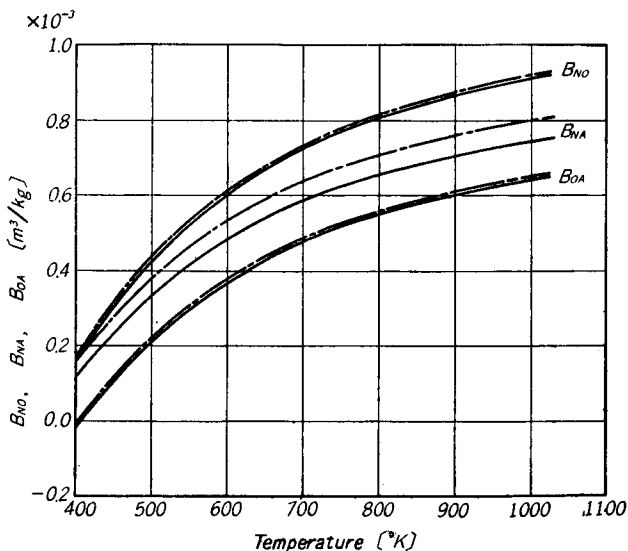


Fig. 3. The second virial coefficient (B_{NO} , B_{NO_2} and B_{O_2}).
 — 1st method,
 - - - 2nd method

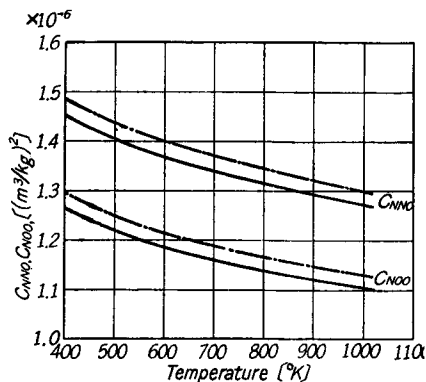


Fig. 4. The third virial coefficient (C_{NO} and C_{NO_2}).
 — 1st method,
 - - - 2nd method

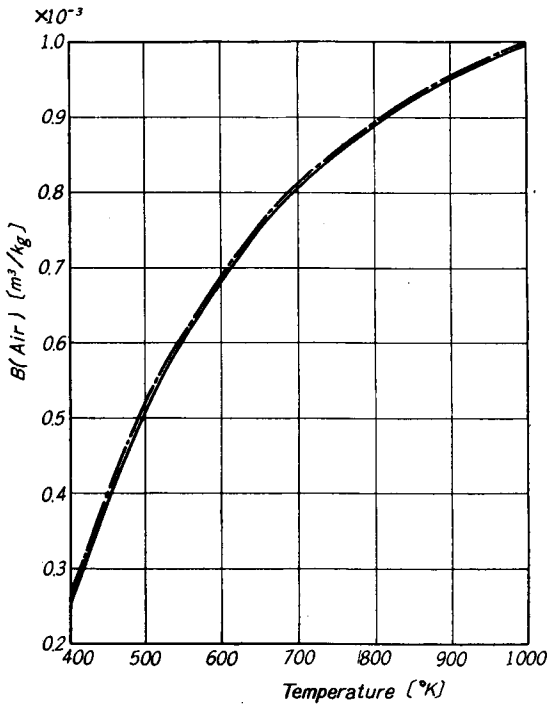


Fig. 5. The second virial coefficient for air.

— 1st method,
 - - - 2nd method

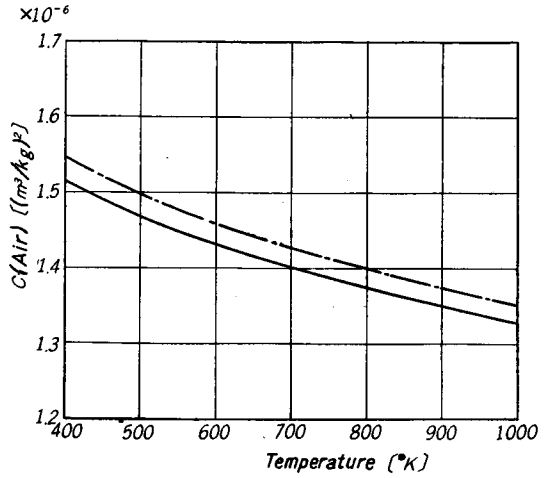


Fig. 6. The third virial coefficient for air.

— 1st method,
 - - - 2nd method

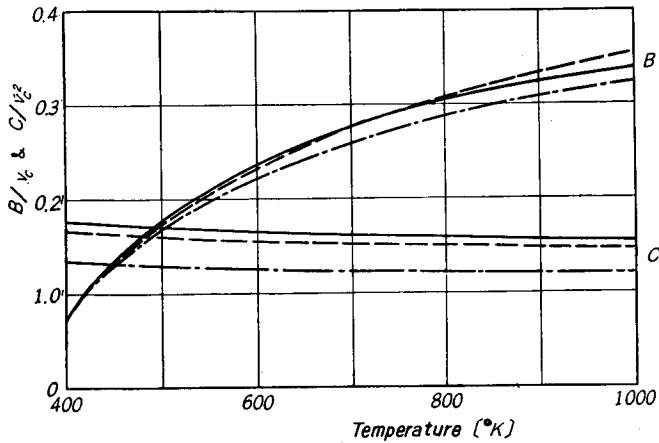


Fig. 7. Relation between the second and the third virial coefficient and temperature. ($1/v_c=340.1 \text{ kg/m}^3$)

— by 1st method,
 - - - by Y. Mori,
 - · - · by R. B. Bird

Next, the total pressure of air computed by using the above described virial coefficients are presented in Table 1, and the ratio of each those values p' —by Mori, Bird and others from the experimental data of air—to the values p calculated by our method from the experimental values for each component gas, are shown in Figs. 8 & 9. As shown in these figures, the values of Mori, Bird and others coincide well with the values by our method, but the values calculated by Dalton's law deviate from our values. This deviation increases at constant temperature as the specific volume of the air decreases, and it decreases as the temperature increases at a constant specific volume of air, in the range of our calculations.

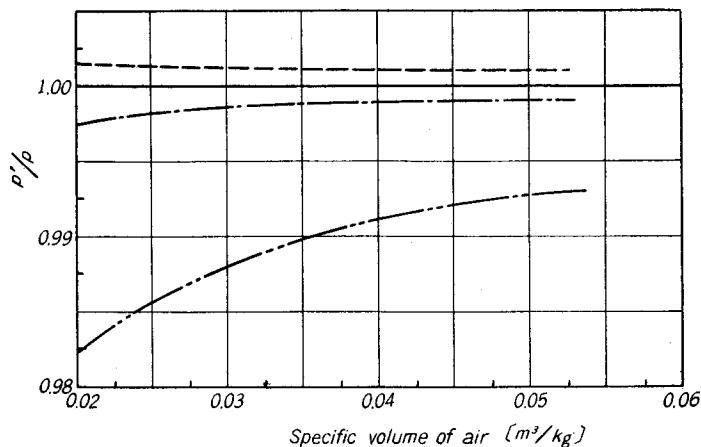


Fig. 8. Relation between p'/p and the specific volume of air. ($T=1000^{\circ}\text{K}$)

— by 1st method, - - - by R. B. Bird,
 - · - · by Y. Mori, ····· by Dalton's law

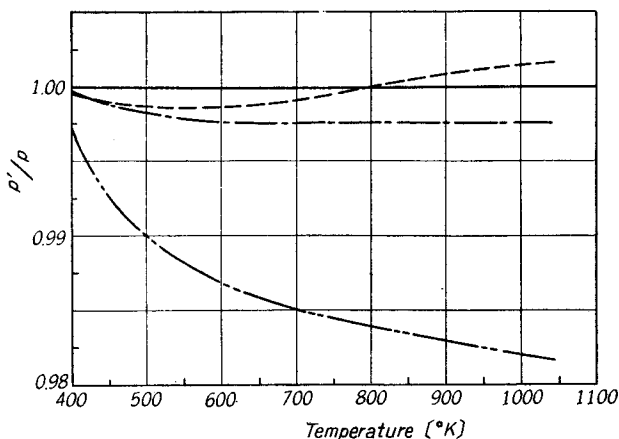


Fig. 9. Relation between p'/p and temperature of air. ($v=0.02\text{ m}^3/\text{kg}$)

— by 1st method, - - - by R. B. Bird,
 - · - · by Y. Mori, ····· by Dalton's law

Table 1. ($v=0.02 \text{ m}^3/\text{kg}$)

	Temperature °K			
	400	600	800	1000
$B_N \times 10^3 \text{ m}^3/\text{kg}$	0.3367	0.7731	0.9672	1.0735
B_O " "	-0.0059	0.4555	0.6618	0.7729
B_A " "	-0.0243	0.3042	0.4475	0.5272
$B \times 10^3 \text{ m}^3/\text{kg}$ (1st method)	0.2580	0.6973	0.8943	0.9998
B_O " " (2nd method)	0.2613	0.7020	0.8981	1.0052
$C_N \times 10^6 (\text{m}^3/\text{kg})^2$	1.6778	1.5887	1.5246	1.4702
C_O " "	1.1085	1.0283	0.9907	0.9604
C_A " "	0.5296	0.4897	0.4719	0.4576
$C \times 10^6 (\text{m}^3/\text{kg})^2$ (1st method)	1.5171	1.4301	1.3758	1.3283
C " " (2nd method)	1.5473	1.4582	1.4024	1.3535
$p_N \text{ kg/cm}^2$	46.38	70.66	94.92	111.09
p_O " "	12.27	18.50	24.73	30.95
p_A " "	0.55	0.83	1.10	1.38
$p = p_N + p_O + p_A$ "	59.20	89.99	120.75	151.42
p " " (1st method)	59.52	91.21	122.75	154.20
p " " (2nd method)	59.53	91.19	122.72	154.15

2. For water-mercury vapour mixture :—

For water-mercury vapour mixture, the total pressure is given as follows,

$$p = T \left(\frac{R_S}{v_S} + \frac{R_M}{v_M} \right) \left\{ 1 + \left(\frac{1}{v_S} + \frac{1}{v_M} \right) B + \left(\frac{1}{v_S} + \frac{1}{v_M} \right)^2 C \right\}$$

$$B = B_S x_S^2 + 2B_{SM} x_S x_M + B_M x_M^2$$

$$C = C_S x_S^3 + 3C_{SSM} x_S^2 x_M + 3C_{SMM} x_S x_M^2 + C_M x_M^3$$

where the subscripts S and M are the values for steam and mercury vapour respectively. And the partial pressures of the component gases are given by the following two equations,

$$p_S = T \left(\frac{R_S}{v_S} \right) \left\{ 1 + \frac{1}{v_S} B_S + \frac{1}{v_S^2} C_S \right\}$$

$$p_M = T \left(\frac{R_M}{v_M} \right) \left\{ 1 + \frac{1}{v_M} B_M + \frac{1}{v_M^2} C_M \right\}$$

where B_S and C_S are calculated by using the Stockmayer potential energy function with the force constants $\sigma_S = 2.65 \text{ \AA}$, $\epsilon_S/k = 380^\circ\text{K}$, $\mu_S = 1.83 \times 10^{-18} \text{ e.s.u.}$, obtained from the experimental data of Keyes and others¹¹⁾. B_M and C_M are calculated by using the Lennard-Jones potential energy function with the force constants $\sigma_M = 2.898 \text{ \AA}$, $\epsilon_M/k = 851^\circ\text{K}$, obtained from the viscosity data. And B_{SM} , C_{SSM} and C_{SMM} are calculated by the two methods mentioned above.

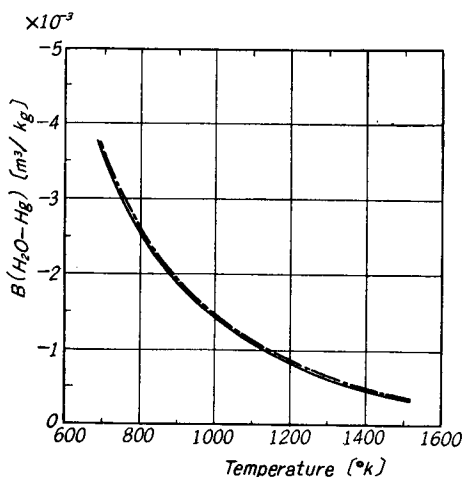


Fig. 10. The second virial coefficient for water-mercury vapour mixture.

— 1st method,
- - - 2nd method

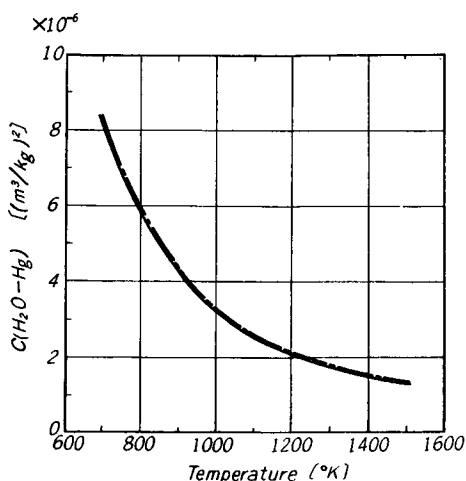


Fig. 11. The third virial coefficient for water-mercury vapour mixture.

— 1st method,
- - - 2nd method

The full lines in Figs. 10 & 11 are the second and third virial coefficients for water-mercury vapour mixture ($v_S=0.03 \text{ m}^3/\text{kg}$, $v_M=0.26 \text{ m}^3/\text{kg}$) by the first method, and the chain lines show those by the second method. At constant specific volume ($v_S=0.03 \text{ m}^3/\text{kg}$, $v_M=0.02 \text{ m}^3/\text{kg}$; $v_S=0.03 \text{ m}^3/\text{kg}$, $v_M=0.06 \text{ m}^3/\text{kg}$; $v_S=0.03 \text{ m}^3/\text{kg}$, $v_M=0.26 \text{ m}^3/\text{kg}$), the total pressure calculated by our method and the arithmetical sum of the two partial pressures are compared in Figs. 12, 13 & 14 and presented in Table 2. In these figures, the full lines represent the former, and the chain lines the latter, and Δp is the difference between the latter and the former. Furthermore, the broken lines show the relation of $p = T(R_S/v_S + R_M/v_M)$. Fig. 15 shows the relation between the ratio p_D/p and the mole fraction of mercury x_M , where p_D is the total pressure from Dalton's law and p is the value calculated by our method.

In the range of our calculations, the difference between p_D and p increases with the mole fraction of mercury. In the region of larger mole fractions of mercury, it may reach a maximum value and after that point, decrease with the further increase of the mole fraction of mercury.

In general, it is not appropriate to apply Dalton's law to a mixture of imperfect gases as shown in the above examples. On the other hand, since a gas mixture is treated as an imperfect gas throughout our method, the deviation of the total pressure calculated by our method from the true value is considered to be much smaller than that from Dalton's law, especially in the case where the imperfectness of the gas mixture is large.

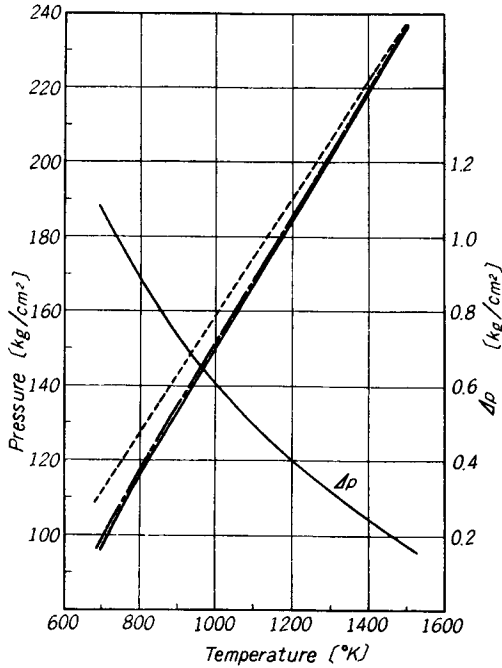


Fig. 12. Relation between the total pressure and temperature of water-mercury vapour mixture. ($v_S=0.03 \text{ m}^3/\text{kg}$, $v_M=0.26 \text{ m}^3/\text{kg}$)

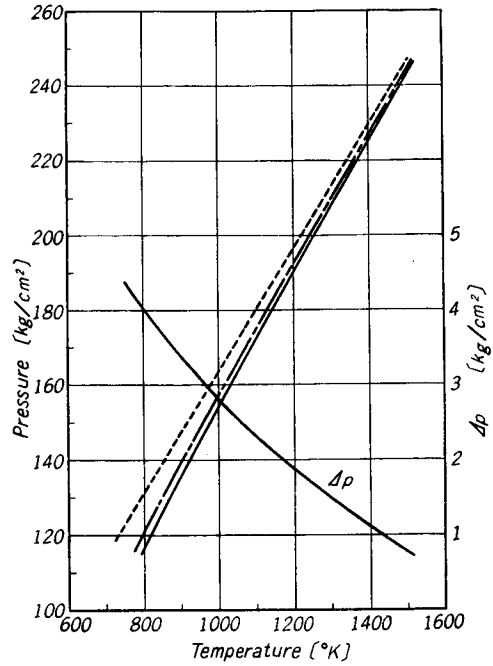


Fig. 13. Relation between the total pressure and temperature of water-mercury vapour mixture. ($v_S=0.03 \text{ m}^3/\text{kg}$, $v_M=0.06 \text{ m}^3/\text{kg}$)

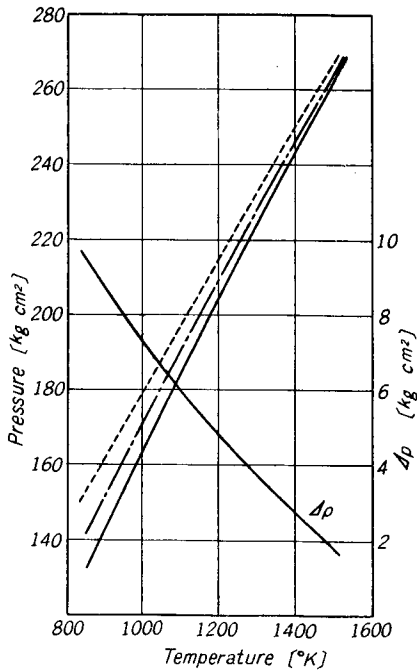


Fig. 14. Relation between the total pressure and temperature of water-mercury vapour mixture. ($v_S=0.03 \text{ m}^3/\text{kg}$, $v_M=0.02 \text{ m}^3/\text{kg}$)

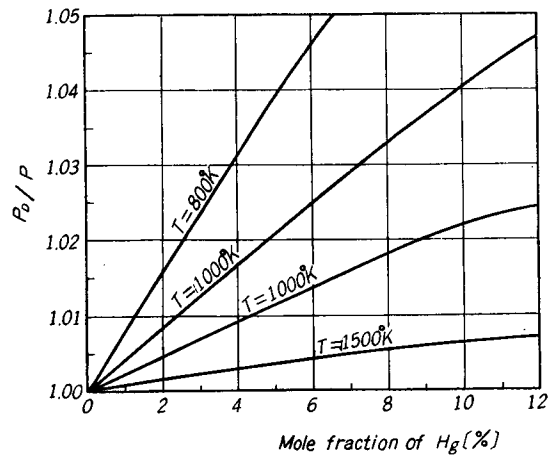


Fig. 15. Relation between p_D/p and the mole fraction of mercury in water-mercury vapour mixture. ($v_S=0.03 \text{ m}^3/\text{kg}$)

Table 2. ($v_S=0.03 \text{ m}^3/\text{kg}$, $v_M=0.26 \text{ m}^3/\text{kg}$)

	Temperature °K					
	700	800	900	1000	1200	1500
$B_M \times 10^4 \text{ m}^3/\text{kg}$	-5.459	-4.330	-3.524	-2.922	-2.078	-1.305
$B_S \times 10^3 \text{ m}^3/\text{kg}$	-3.689	-2.636	-1.939	-1.457	-0.839	-0.326
$B \times 10^3 \text{ m}^3/\text{kg}$ (1st method)	-3.586	-2.563	-1.886	-1.418	-0.816	-0.318
B " " (2nd method)	-3.621	-2.588	-1.904	-1.431	-0.824	-0.321
$C_M \times 10^8 (\text{m}^3/\text{kg})^2$	-1.384	0.590	1.237	1.376	1.325	1.123
$C_S \times 10^6 (\text{m}^3/\text{kg})^2$	8.401	6.145	4.527	3.366	2.214	1.389
$C \times 10^6 (\text{m}^3/\text{kg})^2$ (1st method)	8.065	5.898	4.347	3.232	2.126	1.334
C " " (2nd method)	8.235	6.024	4.438	3.300	2.170	1.362
$p_M \text{ kg/cm}^2$	1.14	1.30	1.46	1.62	1.95	2.44
$p_S \text{ kg/cm}^2$	97.34	115.33	132.77	149.84	183.45	235.12
$p_D = p_M + p_S$ kg/cm^2	98.48	116.63	134.23	151.46	185.40	235.56
$p \text{ kg/cm}^2$ (1st method)	97.40	115.75	133.51	150.86	184.99	235.38
p " " (2nd method)	97.27	115.65	133.43	150.79	184.95	235.36

Conclusion

It has been shown that it is possible to determine the total pressure of a gas mixture by using the force constants of each component gas or the experimental p - v - t data of each component gas. And also it has been demonstrated that it is not appropriate to apply Dalton's law to a mixture of imperfect gases in cases where the degree of imperfectness of the gas mixture is large. In such cases, it may be adequate to use our methods, i.e. the first method in which the empirical combining law for force constants and the equivalent force constants are used, and the second method in which the arithmetical combining law for the virial coefficients is assumed. The results from those two methods agree very well with each other. However, since our methods are still approximate, our results may contain some small deviation from the true value. However, in order to certify the degree of that deviation, more accurate experimental data of pure components and their mixtures are desired.

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