Studies on the Flow of Gaseous Mixtures through Capillaries. I. The Visçosity of Binary Gaseous Mixtures.

By Hiroshi ADZUMI.

(Received April 24th, 1937.)

Introduction.

When a gas flows through capillaries, the flowing formula is not unique over a wide range of pressure but depends on the proportions of the mean free path and the diameters of the capillaries, and three following cases can be distinguished: (1) If the mean free path is very small in comparison with the diameter the flowing quantity is inversely proportional to the viscosity coefficient of the gas. Such flow is called Poiseuille's or the viscous flow, and the rate of flow can be used as a means of determining the viscosity. (2) If the mean free path is large in comparison with the diameter the flowing quantity is independent of the viscosity but inversely proportional to the square root of the molecular weight of the gas, and such flow is called Knudsen's or the molecular flow. (3) When the mean free path is comparable with the diameter the mode of flow is a mixture of the above two types.

The object of the author's study is to know how gaseous mixtures flow through capillaries under various conditions and in this paper the case where the pure viscous flow takes place is treated.

Viscosities of gaseous mixtures have been measured by many authors. Viscosity—composition curves are in general not straight and some mixtures show maximum values. Several formulæ to express the viscosity of gaseous mixtures, have been given by Maxwell(1), Puluj(2), Sutherland(3), Thiessen(4), Enskog(5), and others, but these formulæ do not represent satisfactorily the results of observations. The conditions for the occurrence of a maximum point were also discussed by many authors in special cases but not considered generally.

The present author measured the viscosities of some gaseous mixtures of organic compounds, and considered the conditions for the occurrence of a maximum point for a general case. The results will be reported below.

---

(1) Maxwell, *Phil. Mag.*, (IV), **35** (1868), 212.
(2) Puluj, *Sitzber. Akad. Wiss. Wien, Math.-naturw. Klasse*, Abt. IIa, **79** (1879), 97, 745.
(3) W. Sutherland, *Phil. Mag.*, (V), **40** (1895), 421.
(4) M. Thiessen, *Verh. deut. phys. Ges.*, **4** (1902), 348.
(5) Enskog, *Inaug. Diss. Upsala*, (1917).

Experimental.

Viscosimeter. The method of measuring the viscosity is a transpiration type devised by T. Titani(6) and the capillary of the viscosimeter has the following dimensions: internal diameter, about 0.19 mm.; length, about 79 cm.

By measuring the time of flow of a definite volume of the gas (ca. 0.85 c.c. of about 1 atm. pressure) and taking air as a standard substance the viscosity for other gases can be determined relatively. As the separation of gaseous mixtures into components caused by flowing through capillaries is negligible under the experimental conditions, mixtures are treated by the same method as in the case of a simple gas. The temperatures of measurements are between 20° and 100°C.

Apparatus for Mixing Gases. The apparatus is shown schematically in Fig. 1. Cylindrical vessels A and B, each having a capacity of about 200 c.c., are connected with a capillary tube, about 2 mm. in diameter,

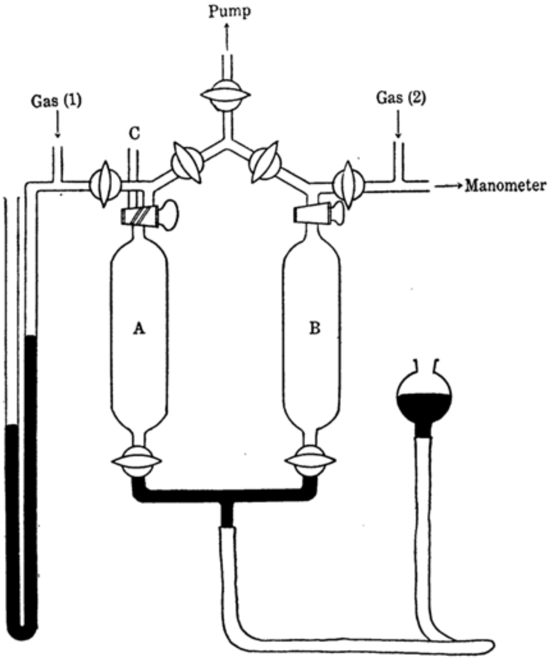

Fig. 1. Apparatus for Mixing Gases.

---

(6) T. Titani, this Bulletin, **4** (1929), 277.

provided with stopcocks. After evacuation, A and B are filled with the required quantity (read with pressure) of gases to be mixed and then by means of a mercury reservoir the gas in B is driven into A. By the equation of state of gas the molar composition of the mixture is calculated. After standing overnight, the homogeneous mixture is taken out from D.

Preparation of Gases. The preparation of gases used for experiments is as follows: *Hydrogen*: Obtained by the electrolysis of water from 30% sodium hydroxide solution, washed with conc. sulphuric acid, neutral potassium permanganate solution, basic sodium hydrosulphite sloution and then dried with calcium chloride and phosphorous pentoxide, the trace of oxygen being thoroughly removed by passing the gas over red-heated copper wire netting beforehand.

Methane and ethane: Prepared with great care by Dr. J. Horiuti. Methane was obtained from aluminium carbide and water, and ethane by the electrolysis of potassium acetate.

Acetylene: Prepared from calcium carbide and water, passed through sodium hydroxide, washed with solutions of ferric nitrate, copper sulphate, mercuric nitrate, and nitric acid, and finally acidic mercury chloride solution, then dried with calcium chloride. The sample thus purified was condensed with liquid air and carefully fractionated several times.

Propane: Obtained by the action of dilute alcoholic solution of iso-propyl alcohol on Zn-Cu couple(7), washed with fuming sulphuric acid, 2% of potassium permanganate solution, potassium hydroxide solution (1:1), then condensed and fractionated.

Propylene: Obtained by dehydration of iso-propyl alcohol with hot-concentrated phosphoric acid, washed with dilute sodium hydroxide solution and water, then condensed and fractionated.

Results of Measurements. (I) *Simple gases.* Viscosities of six following gases are shown in Table 1: H_2, CH_4, C_2H_2, C_2H_6, C_3H_6, C_3H_8. Sutherland's formula has been found to be applicable for all gases with satisfactory results. The viscosity values calculated by this formula are given in the table.

---

(7) Glastone and Tribe, *J. Chem. Soc.*, **45** (1884), 154.

Table 1. Viscosities of Simple Gases.

	Hydrogen		Methane		Acetylene	
	$\eta = 68.4\frac{T^{\frac{3}{2}}}{T+79}\cdot 10^{-7}$		$\eta = 103.9\frac{T^{\frac{3}{2}}}{T+170}\cdot 10^{-7}$		$\eta = 104.5\frac{T^{\frac{3}{2}}}{T+220}\cdot 10^{-7}$	
t°C.	$\eta\times 10^7$		$\eta\times 10^7$		$\eta\times 10^7$	
	Obs.	Calc.	Obs.	Calc.	Obs.	Calc.
0	—	877	—	1059	—	957
20	924	923	1125	1126	1022	1022
30	942	945	1171	1159	1055	1054
40	965	967	1191	1192	1085	1086
50	989	988	1220	1224	1114	1117
60	1008	1009	1255	1256	1146	1149
70	1032	1030	1289	1287	1180	1179
80	1048	1050	1316	1318	1208	1210
90	1072	1070	1345	1348	1246	1240
100	1090	1090	1380	1379	1274	1270

	Ethane		Propane		Propylene	
	$\eta = 106.0\frac{T^{\frac{3}{2}}}{T+280}\cdot 10^{-7}$		$\eta = 99.8\frac{T^{\frac{3}{2}}}{T+318}\cdot 10^{-7}$		$\eta = 100.5\frac{T^{\frac{3}{2}}}{T+302}\cdot 10^{-7}$	
t°C.	$\eta\times 10^7$		$\eta\times 10^7$		$\eta\times 10^7$	
	Obs.	Calc.	Obs.	Calc.	Obs.	Calc.
0	—	865	—	762	—	789
20	928	923	—	820	—	848
30	960	959	845	848	876	876
40	991	990	873	876	904	905
50	1016	1021	908	904	935	934
60	1051	1051	932	932	960	962
70	1080	1081	961	959	995	990
80	1109	1111	988	987	1016	1018
90	1143	1140	1013	1014	1048	1045
100	1171	1170	1040	1041	1070	1073

(II) *Gaseous mixtures.* Viscosities of seven following mixtures are given in Tables 2–8: $H_2\sim CH_4$, $H_2\sim C_2H_2$, $H_2\sim C_2H_6$, $H_2\sim C_3H_6$, $CH_4\sim C_2H_2$, $C_2H_2\sim C_3H_6$, $C_3H_6\sim C_3H_8$. The formula used for calculation of viscosities will be discussed later.

Table 2. Viscosities of $H_2 \sim CH_4$. (Fig. 2.)

$$\text{General formula: } \eta = \frac{\eta_{H_2}}{1+\frac{n_2}{n_1}1.309\frac{T+169}{T+79}} + \frac{\eta_{CH_4}}{1+\frac{n_1}{n_2}0.478\frac{T+169}{T+170}}$$

H_2 %	CH_4 %	100°		60°		20°	
		$\eta\times10^7$		$\eta\times10^7$		$\eta\times10^7$	
		Obs.	Calc.	Obs.	Calc.	Obs.	Calc.
100.00	0.00	1090	1090	1008	1008	924	924
79.17	20.83	1271	1263	1160	1159	1052	1048
69.91	30.09	1312	1305	1190	1194	1074	1078
50.96	49.04	1359	1360	1234	1237	1110	1112
31.05	68.95	1380	1379	1254	1256	1126	1128
0.00	100.00	1380	1380	1255	1255	1125	1125

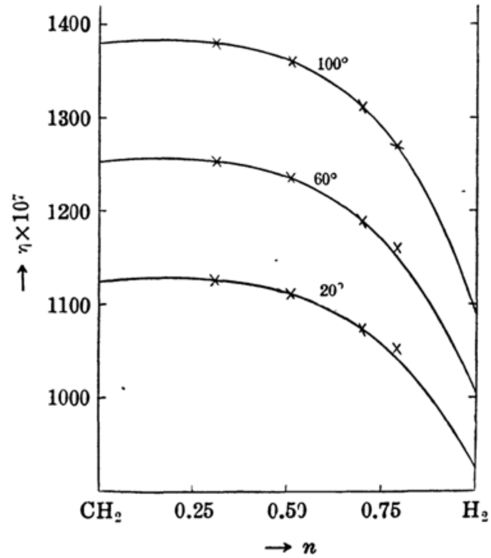

Fig. 2. $H_2 \sim CH_4$

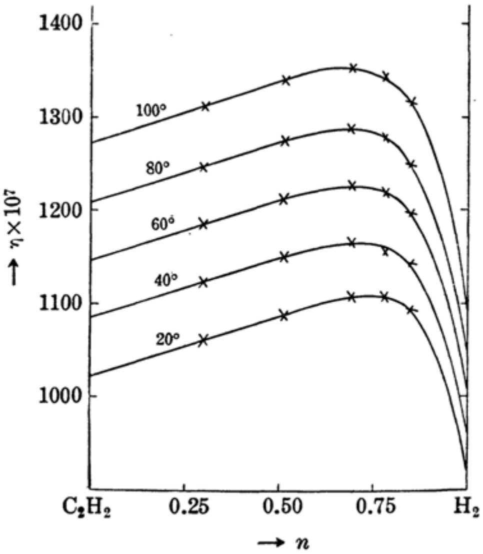

Fig. 3. $H_2 \sim C_2H_2$

Table 3. Viscosities of $H_2 \sim C_2H_2$. (Fig. 3.)

General formula: $$\eta = \frac{\eta_{H_2}}{1+\frac{n_2}{n_2}1.478\frac{T+157}{T+79}} + \frac{\eta_{C_2H_2}}{1+\frac{n_1}{n_2}0.354\frac{T+157}{T+220}}$$

H_2 %	C_2H_2 %	100°		80°		60°	
		$\eta \times 10^7$		$\eta \times 10^7$		$\eta \times 10^7$	
		Obs.	Calc.	Obs.	Calc.	Obs.	Calc.
100.00	0.00	1090	1090	1048	1048	1008	1008
84.87	15.13	1318	1293	1250	1237	1197	1183
77.88	22.12	1346	1333	1278	1274	1219	1218
69.01	30.99	1354	1360	1288	1298	1227	1239
51.28	48.72	1339	1366	1274	1301	1211	1239
29.71	70.29	1312	1335	1245	1270	1184	1206
0.00	100.00	1274	1274	1208	1208	1146	1146

H_2 %	C_2H_2 %	40°		20°	
		$\eta \times 10^7$		$\eta \times 10^7$	
		Obs.	Calc.	Obs.	Calc.
100.00	0.00	965	965	924	924
84.87	15.13	1142	1128	1094	1073
77.88	22.12	1164	1160	1106	1100
69.01	30.99	1166	1179	1106	1116
51.28	48.72	1150	1176	1086	1112
29.71	70.29	1122	1145	1059	1079
0.00	100.00	1085	1085	1022	1022

Table 4. Viscosities of $H_2 \sim C_2H_6$. (Fig. 4.)

General formula: $$\eta = \frac{\eta_{H_2}}{1+\frac{n_2}{n_1}1.516\frac{T+243}{T+79}} + \frac{\eta_{C_2H_6}}{1+\frac{n_1}{n^2}0.328\frac{T+243}{T+280}}$$

H_2 %	C_2H_6 %	100°		80°		60°	
		$\eta \times 10^7$		$\eta \times 10^7$		$\eta \times 10^7$	
		Obs.	Calc.	Obs.	Calc.	Obs.	Calc.
100.00	0.00	1090	1090	1048	1048	1008	1008
83.69	16.31	1240	1230	1183	1176	1126	1122
66.51	33.49	1255	1260	1196	1199	1138	1141
50.84	49.16	1240	1251	1181	1188	1124	1129
37.16	62.84	1224	1232	1163	1169	1104	1110
0.00	100.00	1171	1171	1109	1109	1051	1051

Table 4.—(*Concluded*)

H_2 %	C_2H_6 %	40°		20°	
		$\eta\times10^7$		$\eta\times10^7$	
		Obs.	Calc.	Obs.	Calc.
100.00	0.00	965	965	924	924
83.69	16.31	1071	1071	1013	1008
66.51	33.49	1077	1078	1013	1017
50.84	49.16	1062	1065	998	1001
37.16	62.84	1044	1046	980	981
0.00	100.00	991	991	928	928

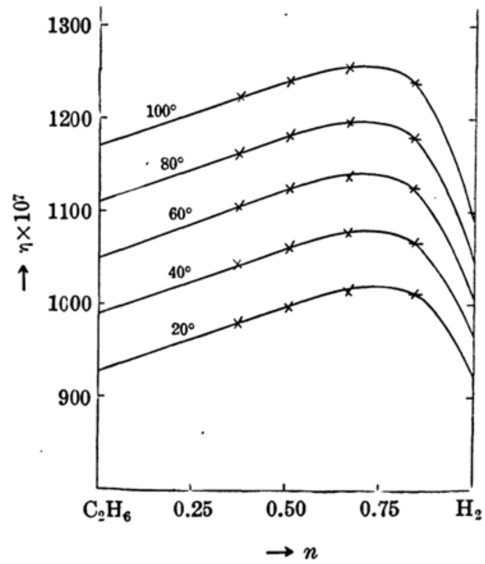

Fig. 4. H_2~C_2H_6

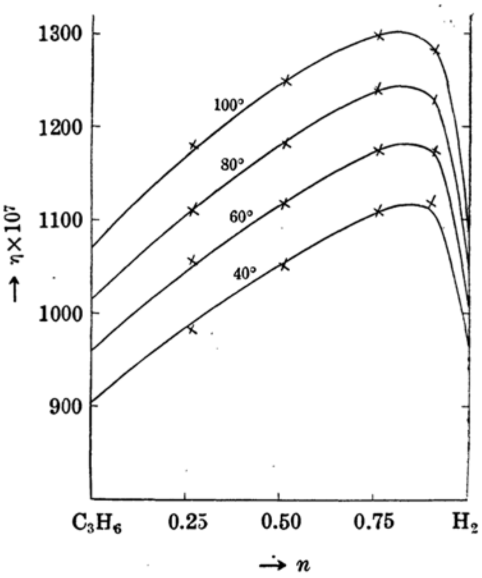

Fig. 5. H_2~C_3H_6

Table 5. Viscosities of $H_2 \sim C_3H_6$. (Fig. 5.)

General formula: $\eta = \dfrac{\eta_{H_2}}{1+\dfrac{n_2}{n_1}1.746\dfrac{T+155}{T+79}} + \dfrac{\eta_{C_3H_6}}{1+\dfrac{n_1}{n_2}0.254\dfrac{T+155}{T+302}}$

H_2 %	C_3H_6 %	100°		80°	
		$\eta \times 10^7$		$\eta \times 10^7$	
		Obs.	Calc.	Obs.	Calc.
100.00	0.00	1090	1090	1048	1048
91.10	8.90	1284	1263	1231	1221
75.88	24.12	1296	1320	1240	1263
51.02	48.98	1250	1255	1181	1197
26.76	73.24	1181	1166	1110	1107
0.00	100.00	1070	1070	1016	1016

H_2 %	C_3H_6 %	60°		40°	
		$\eta \times 10^7$		$\eta \times 10^7$	
		Obs.	Calc.	Obs.	Calc.
100.00	0.00	1008	1008	965	965
91.10	8.90	1174	1160	1118	1106
75.88	24.12	1175	1203	1110	1142
51.02	48.98	1118	1136	1050	1073
26.76	73.24	1057	1047	983	987
0.00	100.00	960	960	904	904

Table 6. Viscosities of $CH_4 \sim C_2H_2$. (Fig. 6.)

General formula: $\eta = \dfrac{\eta_{CH_4}}{1+\dfrac{n_2}{n_1}1.135\dfrac{T+172}{T+170}} + \dfrac{\eta_{C_2H_2}}{1+\dfrac{n_1}{n_2}0.854\dfrac{T+172}{T+220}}$

CH_4 %	C_2H_2 %	100°		80°	
		$\eta \times 10^7$		$\eta \times 10^7$	
		Obs.	Calc.	Obs.	Calc.
100.00	0.00	1380	1380	1316	1316
75.11	24.89	1378	1379	1310	1316
49.53	50.47	1360	1359	1300	1294
24.72	75.28	1330	1320	1264	1257
0.00	100.00	1274	1274	1208	1208

Table 6.—(*Concluded*)

CH_4 %	C_2H_2 %	60°		40°	
		$\eta\times10^7$		$\eta\times10^7$	
		Obs.	Calc.	Obs.	Calc.
100.00	0.00	1255	1255	1191	1191
75.11	24.89	1250	1254	1186	1190
49.53	50.47	1236	1231	1171	1168
24.72	75.28	1200	1193	1138	1132
0.00	100.05	1146	1146	1085	1085

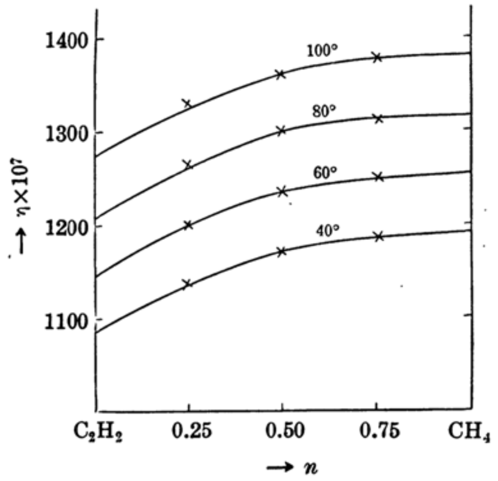

Fig. 6. $CH_4\sim C_2H_2$

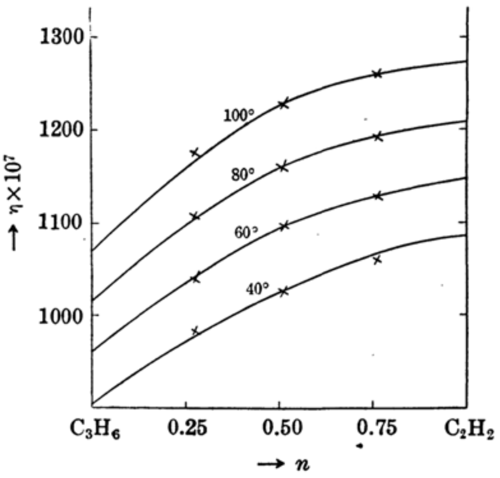

Fig. 7. $C_2H_2\sim C_3H_6$

Table 7. Viscosities of $C_2H_2\sim C_3H_6$. (Fig. 7.)

General formula:
$$\eta=\frac{\eta_{C_2H_2}}{1+\frac{n_2}{n_1}1.165\frac{T+222}{T+220}}+\frac{\eta_{C_3H_6}}{1+\frac{n_1}{n_2}0.817\frac{T+222}{T+302}}$$

C_2H_2 %	C_3H_6 %	100°		80°	
		$\eta\times10^7$		$\eta\times10^7$	
		Obs.	Calc.	Obs.	Calc.
100.00	0.00	1274	1274	1208	1208
75.99	24.01	1261	1256	1192	1194
50.79	49.21	1228	1212	1160	1151
27.39	72.61	1176	1154	1108	1095
0.00	100.00	1070	1070	1016	1016

Table 7.—(*Concluded*)

C_2H_2 %	C_3H_6 %	60°		40°	
		$\eta\times10^7$		$\eta\times10^7$	
		Obs.	Calc.	Obs.	Calc.
100.00	0.00	1146	1146	1085	1085
75.99	24.01	1130	1132	1060	1071
50.79	49.21	1094	1080	1026	1031
27.39	72.61	1040	1036	982	977
0.00	100.00	960	960	904	904

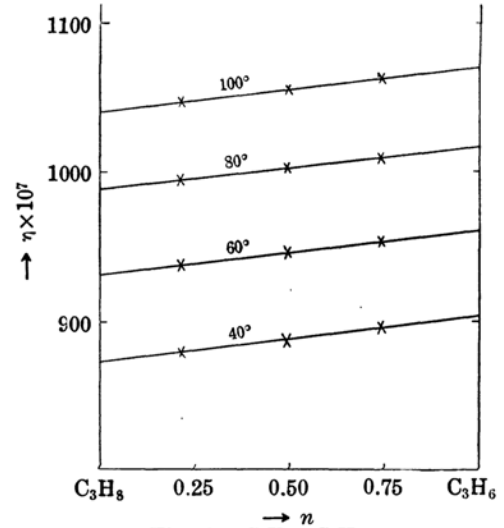

Fig. 8. $C_3H_6\sim C_3H_8$

Table 8. Viscosities of $C_3H_6\sim C_3H_8$. (Fig. 8.)

General formula: $\eta=\dfrac{\eta_{C_3H_6}}{1+\dfrac{n_2}{n_1}1.008\dfrac{T+310}{T+302}}+\dfrac{\eta_{C_3H_8}}{1+\dfrac{n_1}{n_2}0.985\dfrac{T+310}{T+318}}$

C_3H_6 %	C_3H_8 %	100°		80°	
		$\eta\times10^7$		$\eta\times10^7$	
		Obs.	Calc.	Obs.	Calc.
100.00	0.00	1070	1070	1016	1016
73.92	26.08	1062	1064	1009	1011
49.03	50.97	1056	1058	1001	1003
21.31	78.69	1046	1048	996	995
0.00	100.09	1040	1040	988	988

Table 8.—(*Concluded*)

C_3H_6 %	C_3H_8 %	60°		40°	
		$\eta\times10^7$		$\eta\times10^7$	
		Obs.	Calc.	Obs.	Calc.
100.00	0.00	960	960	904	904
73.92	26.08	952	954	896	897
49.03	50.97	946	947	886	890
21.31	78.69	940	939	878	881
0.00	100.00	932	932	873	873

As seen from the figure, the viscosities of the first four mixtures attain maximum values at definite compositions, which are about 20% of H_2 for the mixture $H_2\sim CH_4$ and about 70–80% of H_2 for the other three. The viscosity-composition curve of the mixture $C_3H_6\sim C_3H_8$ is quite linear.

Discussion.

Viscosity Formula for Binary Gaseous Mixture. The viscosity of gases depends on the diameter, the mass, and the attraction of the molecule, and further the mutual collision of the different gases must be considered in the case of a mixture and the theoretical treatment is difficult. As described above, many formulæ are proposed to express the viscosity of a gaseous mixture but they are derived almost empirically. Of those formulæ, that in which Kuenen's(8) consideration of the persistence of molecular velocity is introduced is somewhat more theoretical than the other. The experimental results will be examined first of all by this formula.

The viscosity η of a simple gas is given by

$$\eta = k m N \bar{u} \lambda f\,, \qquad (1)$$

where m is the mass, $\bar{u}$ the mean velocity, λ the mean free path of the molecule, N the number of molecules per unit volume, f the correction factor in consideration of the persistence of molecular velocity (1/0.797 in simple gas) and k a constant (0.499 × 0.797).

---

(8) J. P. Kuenen, "Die Eigenschaften der Gase," 111 (1919).

The viscosity of binary gaseous mixtures is given in a rather complicated formula. If the mean free path of the molecule in the mixture of the first type is λ_1' and the correction factor owing to the persistence in the mixture is f_1', there is obtained the following relations:

$$\eta = km_1n_1N\bar{u}_1\lambda_1'f_1' + km_2n_2N\bar{u}_2\lambda_2'f_2' \,, \qquad (2)$$

where n_1 and n_2 are mol fractions of two components. The mean free paths in a mixture are

$$\lambda_1' = \frac{1}{n_1N\pi S_1^2\sqrt{2}\left(1+\dfrac{C_1}{T}\right)+n_2N\pi S^2\sqrt{\dfrac{m_1+m_2}{m_2}}\left(1+\dfrac{C_{12}}{T}\right)} \,,$$

$$\lambda_2' = \frac{1}{n_2N\pi S_2^2\sqrt{2}\left(1+\dfrac{C_2}{T}\right)+n_1N\pi S^2\sqrt{\dfrac{m_1+m_2}{m_1}}\left(1+\dfrac{C_{12}}{T}\right)} \,,$$

where S_1 and S_2 are molecular diameters of two gases, S is a mean of these ($S=1/2(S_1+S_2)$), C_1 and C_2 are Sutherland's constants of two gases due to the attraction between the same molecules and C_{12} that between the different molecules.

According to Kuenen, the correction factors for a mixture, f', are as follows:

$$f_1' = \frac{1}{1-\dfrac{\sqrt{2}}{2}n_1N\pi S_1^2\left(1+\dfrac{C_1}{T}\right)\lambda_1'\times 0{\cdot}406-\dfrac{m_1}{m_1+m_2}n_2N\pi S^2\sqrt{\dfrac{m_1+m_2}{m_2}}\left(1+\dfrac{C_{12}}{T}\right)\lambda_1'\vartheta_1} \,,$$

where $$\vartheta_1 = \frac{m_1}{2(m_1+m_2)}+\frac{1}{4}\,\frac{m_1^2}{\sqrt{m_2}(m_1+m_2)^{\frac{3}{2}}}\ln\frac{\sqrt{m_1+m_2}+\sqrt{m_2}}{\sqrt{m_1+m_2}-\sqrt{m_2}} \,,$$

and analogous formulæ for f_2' and ϑ_2.

The viscosities of the components are also expressed by

$$\eta_1 = \frac{km_1\bar{u}_1}{\pi S_1^2\sqrt{2}\left(1+\dfrac{C_1}{T}\right)}\,\frac{1}{0{\cdot}797} \,, \qquad \eta_2 = \frac{km_2\bar{u}_2}{\pi S_2^2\sqrt{2}\left(1+\dfrac{C_2}{T}\right)}\,\frac{1}{0{\cdot}797} \,.$$

By inserting these values to equation (2), the viscosity of a mixture becomes

$$\eta = \frac{\eta_1}{1+\dfrac{n_2}{n_1}\alpha_1} + \frac{\eta_2}{1+\dfrac{n_1}{n_2}\alpha_2}\,, \qquad (3)$$

where

$$\alpha_1 = \left(\frac{S}{S_1}\right)^2 \sqrt{\frac{m_1+m_2}{2m_2}}\,\frac{1-\dfrac{m_1}{m_1+m_2}\vartheta_1}{0{\cdot}797}\,\frac{T+C_{12}}{T+C_1}\,,$$

$$\alpha_2 = \left(\frac{S}{S_2}\right)^2 \sqrt{\frac{m_1+m_2}{2m_1}}\,\frac{1-\dfrac{m_2}{m_1+m_2}\vartheta_2}{0.797}\,\frac{T+C_{12}}{T+C_2}\,.$$

Sutherland's constant C_{12} is proportional to the potential energy of two different molecules, and the only coefficient in equation (3) that can not be determined directly, and is often considered to be a geometrical mean of C_1 and C_2. For almost all mixtures, whose viscosities have been measured by many authors, such calculations of this coefficient is inadequate to adapt the experiments. However, if we take a proper value for C_{12}, the results of observation can be expressed satisfactorily by equation (3). By using such values, viscosity formulæ for all mixtures can be obtained and these are given in Tables 3–8. The viscosity curve III in Fig. 9 is obtained by equation (3) by using C_{12} calculated as a geometrical mean and curve II by observed C_{12} computed empirically from the results of observation. The comparison of the calculated and the observed values of C_{12} are shown below in Table 9.

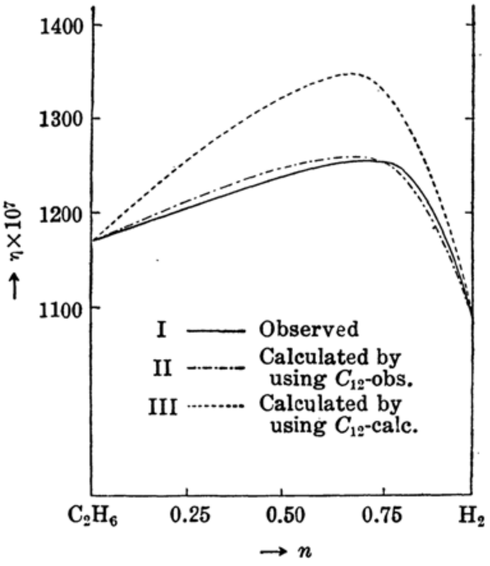

Fig. 9. H_2～C_2H_6 (100°C.)

Sutherland's Constant between Different Molecules. As described above, according to the classical theory, Sutherland's constant C_{12} has been considered to be a geometrical mean of C_1 and C_2. In 1928, Schmick⁽⁹⁾ reported that, when a dipole and a quadrupole molecules collide, the constant becomes smaller than the geometrical mean.

The potential energies between two dipole molecules, $E_{d_1d_2}$, between two quadrupole molecules, $E_{q_1q_2}$, and between a dipole and a quadrupole molecules, E_{dq}, are expressed as follows:

$$E_{d_1d_2} = -\frac{2}{3\sigma^6}\frac{\mu_1^2\mu_2^2}{kT}\,,\quad E_{q_1q_2} = -\frac{14}{5\sigma^{10}}\frac{\nu_1^2\nu_2^2}{kT}\,,\quad E_{dq} = -\frac{1}{\sigma^8}\frac{\mu^2\nu^2}{kT}\,,$$

where μ and ν are the dipole and the quadrupole moments respectively, and σ the distance between two molecules.

Then for the same sort of molecules $E_{dd} = -\frac{2}{3\sigma^6}\frac{\mu^4}{kT}$ and $E_{qq} = -\frac{14}{5\sigma^{10}}\frac{\nu^4}{kT}$, hence $E_{dq} = 0.733\sqrt{E_{dd}\times E_{qq}}$. As Sutherland's constant is proportional to the potential energy between two molecules, this becomes

$$C_{12} = 0.733\sqrt{C_1\times C_2}\,.$$

According to London's consideration⁽¹⁰⁾ from the standpoint of wave mechanics, C_{12} is always smaller than the geometrical mean, and if it be larger, we can say that some forces other than van der Waals's, for example, valency force etc. must have acted between the molecules.

There is no method to determine C_{12} directly, but as described above, we can compute it inversely from viscosities of a mixture by using equation (3).

The values of C_{12} calculated for several paires by using viscosities obtained by the present author and other authors are given in Table 9. For nine mixtures out of thirty-two the values of C_{12} are equal to, and for thirteen larger than, the geometrical means. As the examples of a dipole and a quadrupole mixtures, C_{12}'s of $NH_3\sim O_2$ and $NH_3\sim N_2$ agree very well with Schmick's theory, but those of $NH_3\sim C_2H_4$ and $NH_3\sim H_2$ do not.

It is doubtful whether the values of C_{12} obtained by such a method are exactly proportional to the potential energies or not, and therefore, such calculations serve only to the estimation of the approximate values of the constant and it is dangerous to interpret the values of C_{12} larger than the geometrical mean by the consideration of London.

---

(9) H. Schmick, *Physik. Z.*, **29** (1928), 638.

(10) F. London, *Z. physik, Chem.*, B, **11** (1931), 222.

Table 9. Calculated Values of C_{12} and the Comparison with the Geometrical Means of C_1 and C_2.

Mixture	C_1	C_2	C_{12}	$\frac{C_{12}}{\sqrt{C_1C_2}}$
$NH_3\sim N_2$	370	114	151	0.733
$NH_3\sim O_2$	370	137	165	0.733
$He\sim Ne$	76	56	52	0.80
$C_2H_4\sim O_2$	259	137	160	0.85
$C_2H_4\sim C_3H_6$	259	302	222	0.86
$NH_3\sim C_2H_4$	370	259	270	0.88
$CH_4\sim C_2H_2$	170	220	172	0.89
$CO_2\sim C_3H_8$	257	318	257	0.90
$O_2\sim CO_2$	137	257	170	0.90
$N_2\sim C_2H_4$	114	259	160	0.93
$CO\sim N_2$	102	114	105	0.97
$H_2\sim C_3H_6$	79	302	155	1.00
$C_3H_6\sim C_3H_8$	302	318	310	1.00
$N_2O\sim C_3H_8$	314	318	316	1.00
$N_2\sim O_2$	114	137	128	1.03
$C_2H_6\sim C_3H_8$	280	318	312	1.04

Mixture	C_1	C_2	C_{12}	$\frac{C_{12}}{\sqrt{C_1C_2}}$
$CH_4\sim C_2H_6$	170	280	230	1.05
$H_2\sim He$	79	76	82	1.05
$N_2\sim NO$	114	195	156	1.05
$H_2\sim NH_3$	79	370	195	1.14
$C_2H_4\sim CO_2$	259	257	290	1.14
$H_2\sim O_2$	79	137	120	1.15
$Ne\sim A$	56	163	110	1.15
$H_2\sim C_2H_2$	79	220	159	1.19
$H_2\sim Ne$	79	56	81	1.23
$CH_4\sim C_3H_8$	170	318	305	1.31
$H_2\sim C_2H_4$	79	259	200	1.40
$H_2\sim A$	79	163	163	1.44
$H_2\sim CH_4$	79	170	169	1.46
$H_2\sim CO_2$	79	257	210	1.47
$He\sim A$	76	163	163	1.47
$H_2\sim C_2H_6$	79	280	243	1.63

The Conditions for the Occurrence of a Maximum Point. The conditions for the occurrence of a maximum point have been discussed by many authors in special cases. For example, Thomsen⁽¹¹⁾ reported that in the case of mixtures of two gases of nearly equal viscosities, maximum point occurs when the ratio of molecular weights is great; and Schmick⁽¹²⁾ stated that, of mixtures of two gases, whose viscosities and molecular weights are nearly equal, those of a polar and a nonpolar molecules show maximum points.

We can not find any papers dealing with the conditions for the occurrence of a maximum point for a general case.

The general formula expressing the viscosity of a gaseous mixture is given by

$$\eta = \frac{\eta_1}{1+\frac{n_2}{n_1}a_1} + \frac{\eta_2}{1+\frac{n_1}{n_2}a_2}\,, \tag{4}$$

---

(11) E. Thomsen, *Ann. Physik*, **36** (1911), 815.

(12) G. Jung and H. Schmick, *Z. physik. Chem.*, B, **7** (1930), 130.

and by introducing Kuenen's consideration into α_1 and α_2, we can derive the special form of this equation as given in equation (3). If we write $n_1 = n$, $n_2 = 1 - n$, then

$$\eta = \frac{\eta_1}{1+\dfrac{1-n}{n}\alpha_1} + \frac{\eta_2}{1+\dfrac{n}{1-n}\alpha_2} = \frac{n\eta_1}{n+(1-n)\alpha_1} + \frac{(1-n)\eta_2}{1-n+n\alpha_2} .$$

By differentiation we have

$$\frac{d\eta}{dn} = \frac{\alpha_1\eta_1\left\{(\alpha_2-1)n+1\right\}^2 - \alpha_2\eta_2\left\{(\alpha_1-1)n-\alpha_1\right\}^2}{(n+\alpha_1-n\alpha_1)^2(1-n+n\alpha_2)^2}$$

$$= A\Big[\sqrt{\alpha_1\eta_1}\left\{\alpha_2+(1-n)\right\} - \sqrt{\alpha_2\eta_2}\left\{\alpha_1(1-n)+n\right\}\Big] , \qquad (5)$$

where
$$A \equiv \frac{\sqrt{\alpha_1\eta_1}\left\{\alpha_2+(1-n)\right\} + \sqrt{\alpha_2\eta_2}\left\{\alpha_1(1-n)+n\right\}}{(n+\alpha_1-n\alpha_1)^2(1-n+n\alpha_2)^2} .$$

If we equate $d\eta/dn$ to zero, we have the following equation, A being positive and not equal to zero.

$$\sqrt{\alpha_1\eta_1}\left\{\alpha_2+(1-n)\right\} - \sqrt{\alpha_2\eta_2}\left\{\alpha_1(1-n)+n\right\} = 0 .$$

Then the composition of the maximum or the minimum point, n_0, becomes

$$n_0 = \frac{\sqrt{\alpha_1}\,(\sqrt{\alpha_1\alpha_2\beta}-1)}{\sqrt{\alpha_1}\,(\alpha_2-1) + \sqrt{\alpha_2\beta}(\alpha_1-1)} = \frac{\sqrt{\alpha_1\alpha_2\beta}-1}{\sqrt{\alpha_1\alpha_2\beta}-1+\sqrt{\dfrac{\alpha_2}{\alpha_1}}(\sqrt{\alpha_1\alpha_2}-\sqrt{\beta}\,)} , \qquad (6)$$

where $\beta \equiv \eta_2/\eta_1$.

For the existence of a maximum or a minimum point, n_0 must be greater than nought and smaller than unity or as seen from equation (6), following conditions must be fulfilled:

$$\alpha_1\alpha_2 > \beta > \frac{1}{\alpha_1\alpha_2} \qquad \text{or} \qquad \alpha_1\alpha_2 < \beta < \frac{1}{\alpha_1\alpha_2} .$$

By inserting the values of n_0 given by (6), equation (5) becomes

$$\frac{d\eta}{dn} = A\sqrt{\alpha_1}\,\frac{n-n_0}{\sqrt{\alpha_1\alpha_2\beta}-1+\sqrt{\dfrac{\alpha_2}{\alpha_1}}(\sqrt{\alpha_2\alpha_1}-\sqrt{\beta}\,)} . \qquad (7)$$

where $A\sqrt{\alpha_1}$ is positive if $0 < n_0 < 1$.

Then if $\alpha_1\alpha_2 > \beta > \frac{1}{\alpha_1\alpha_2}$, the demoninator of equation (7) becomes positive and

$$\frac{d\eta}{dn} < 0 \qquad \text{when} \quad n < n_0 ,$$

$$\frac{d\eta}{dn} > 0 \qquad \text{when} \quad n > n_0 ,$$

hence η becomes minimum when $n = n_0$.

Next, if $\alpha_1\alpha_2 < \beta < \frac{1}{\alpha_1\alpha_2}$, the denominator of equation (7) becomes negative and

$$\frac{d\eta}{dn} > 0 \qquad \text{when} \quad n < n_0 ,$$

$$\frac{d\eta}{dn} < 0 \qquad \text{when} \quad n > n_0 ,$$

hence η becomes maximum when $n = n_0$.

By summarising,

$$\left.\begin{array}{lll} \text{when} & \alpha_1\alpha_2 > \beta > \frac{1}{\alpha_1\alpha_2} & \text{viscosity shows a minimum,} \\ & \alpha_1\alpha_2 < \beta < \frac{1}{\alpha_1\alpha_2} & \quad ,, \qquad ,, \quad \text{a maximum,} \\ & \left.\begin{array}{l} \frac{1}{\beta} < \alpha_1\alpha_2 < \beta \\ \text{or} \quad \beta < \alpha_1\alpha_2 < \frac{1}{\beta} \end{array}\right\} & \quad ,, \qquad ,, \quad \text{neither.} \end{array}\right\} \qquad (8)$$

This relations hold in the general formula of binary mixtures whatever the values of α_1 and α_2 may be, but in order to examine numerically by examples, let us insert the values given in (3) to α_1 and α_2. If we write $m_2/m_1 \equiv \mu$, $S_2/S_1 \equiv a$, $\frac{1+\frac{C_2}{T}}{1+\frac{C_1}{T}} \equiv \nu$ and assume $\frac{(T+C_{12})^2}{(T+C_1)(T+C_2)} = 1$, then

$$\left.\begin{aligned} \alpha_1\alpha_2\beta &= \left(\frac{1+a}{2a}\right)^4 \frac{B}{\nu} , \\ \frac{\alpha_1\alpha_2}{\beta} &= \left(\frac{1+a}{2}\right)^4 \frac{B\nu}{\mu} , \end{aligned}\right\} \qquad (9)$$

where $$B = \frac{1+\mu}{1.27}\left(1-\frac{\vartheta_1}{1+\mu}\right)\left(1-\frac{\vartheta_2}{1+\mu}\mu\right).$$

Above assumption is not rigorous, but, if C_{12} lies between C_1 and C_2, $(T + C_{12})^2/(T + C_1)(T + C_2)$ is not very far from unity.

The results of calculations of $\alpha_1\alpha_2\beta$ and $\alpha_1\alpha_2/\beta$ at 0°C. by formula (8) for fifty-five kinds of gaseous mixtures are tabulated in Table 10, the components of mixtures being selected as $m_2 > m_1$.

Table 10.

(1) Mixtures showing a maximum.

Mixture	$\alpha_1\alpha_2\beta$	$\frac{\alpha_1\alpha_2}{\beta}$	Observers
$H_2 \sim CH_4$	0.740	0.526	Adzumi, Graham(13), Trautz and Sorg(28).
$H_2 \sim C_2H_2$	0.574	0.485	Adzumi.
$H_2 \sim C_2H_4$	0.537	0.498	Thomsen(11), Trautz and Stauf(22).
$H_2 \sim C_2H_6$	0.499	0.517	Adzumi.
$H_2 \sim C_3H_6$	0.416	0.514	Adzumi.
$H_2 \sim C_3H_8$	0.385	0.510	
$H_2 \sim NH_3$	0.622	0.574	Thomsen(11), Trautz and Heberling(29).
$H_2 \sim CO_2$	0.675	0.242	Graham(13), Puluj(2) Thomsen(11), Breitenbach(14), Trautz and Kurz(27).
$H_2 \sim SO_2$	0.433	0.265	Trautz and Weizel(18).
$H_2 \sim NO$	0.890	0.245	Graham(13).
$H_2 \sim N_2O$	0.646	0.249	Graham(13), Trautz and Kurz(27).
$H_2 \sim HCl$	0.662	0.279	Trautz and Narath(19).
$H_2 \sim Cl_2$	0.479	0.243	Thomsen(11).
$He \sim A$	0.718	0.460	Tänzler(16), Trautz and Kipphan(23), Trautz and Binkele(25).
$He \sim Kr$	0.552	0.374	Nasini and Rossi(20).
* $CH_4 \sim NH_3$	0.852	1.120	Jung and Schmick(12).
* $NH_3 \sim C_2H_4$	0.990	1.004	Thomsen(11), Trautz and Heberling(29).

---

(13) Graham, *Trans. Roy. Soc.* (London), A, **136** (1846), 662.

(14) Breitenbach, *Wied. Ann.*, **67** (1899), 803.

(15) K. Kleint, *Verh. deut. phys. Ges.*, **7** (1905), 146.

(16) P. Tänzler, *Verh. deut. phys. Ges.*, **8** (1906), 221.

(17) A. Gille, *Ann. Physik*, **48** (1915), 799.

(18) M. Trautz and W. Weizel, *Ann. Physik*, **78** (1925), 305.

(19) M. Trautz and A. Narath, *ibid.*, **79** (1926), 637.

(20) Nasini and Rossi, *Gazz. chim. ital.*, **58** (1928), 433.

(21) M. Trautz and P. B. Baumann, *Ann. Physik*, (V), **2** (1929), 733.

Table 10.—(*Continued*)

(2) Mixtures showing no maximum.

Mixture	$\alpha_1\alpha_2\beta$	$\frac{\alpha_1\alpha_2}{\beta}$	Observers
* $H_2 \sim CO$	0.928	0.260	Graham(13), Tratz and Baumann(21).
$H_2 \sim O_2$	1.018	0.205	Kleint(15), Graham(13), Trautz and Melster(26).
* $H_2 \sim N_2$	0.960	0.254	Graham(13), Kleint(15), Trautz and Baumann(21).
* $H_2 \sim$ Air	0.954	0.247	Graham(13).
$H_2 \sim He$	2.110	0.308	Gille(17), Trauz and Kipphan(23), Trauz and Baumann(21).
$H_2 \sim A$	1.110	0.152	Trautz and Ludewigs(24), Trauz and Binkele(25).
$H_2 \sim Ne$	1.901	0.157	Trautz and Binkele(25).
$He \sim Ne$	1.20	0.455	Trautz and Kipphan(23).
$Ne \sim A$	0.744	1.260	Trautz and Kipphan(23), Trauz and Binkele(15).
$NH_3 \sim N_2$	1.694	0.521	Thomsen(11), Trautz and Heberling(29).
$NH_3 \sim O_2$	1.435	0.566	Thomsen(11), Trautz and Heberling(29).
$NH_3 \sim$ Air	1.82	0.520	Jung and Schmick(12).
$NH_3 \sim CO_2$	1.435	0.566	Thomsen(11).
$NH_3 \sim Cl_2$	1.08	0.459	Thomsen(11).
Air $\sim CO_2$	0.837	1.160	Jung and Schmick(12).
Air $\sim HCl$	0.770	1.212	
Air $\sim H_2S$	0.657	1.456	
$CO \sim N_2$	1.032	0.97	Trautz and Melster(26).
$CO_2 \sim N_2O$	0.963	1.039	Trautz and Kurz(27).
$CO_2 \sim SO_2$	0.773	1.266	Jung and Schmick(12).
$O_2 \sim CO_2$	0.745	1.333	Graham(13).
$CO \sim O_2$	1.20	0.837	Graham(13), Trautz and Melster(26).
$N_2 \sim O_2$	1.16	0.817	Graham(13), Kleint(15), Trautz and Melster(26).
$N_2 \sim NO$	0.978	1.055	Trautz and Gabriel(30).

---

(22) M. Trautz and F. W. Stauf, *Ann. Physik*, (V), **2** (1929), 737.
(23) M. Trautz and K. F. Kipphan, *ibid.*, (V), **2** (1929), 743.
(24) M. Trautz and W. Ludewigs, *ibid.*, (V), **3** (1929), 409.
(25) M. Trautz and H. E. Binkele, *ibid.*, (V), **5** (1930), 561.
(26) M. Trautz and A. Melster, *ibid.*, (V), **7** (1930), 409.
(27) M. Trautz and F. Kurz, *ibid.*, (V), **9** (1931), 981.
(28) M. Trautz and K. G. Sorg, *ibid.*, (V), **10** (1931), 81.
(29) M. Trautz and R. Heberling, *ibid.*, (V), **10** (1931), 155.
(30) M. Trautz and E. Gabriel, *ibid.*, (V), **11** (1931), 606.

Table 10.—(*Concluded*)

Mixture	$\alpha_1\alpha_2\beta$	$\frac{\alpha_1\alpha_2}{\beta}$	Observers
$CH_4 \sim O_2$	1.84	0.463	Graham(15).
$CH_4 \sim C_2H_2$	0.900	1.074	Adzumi.
$CH_4 \sim C_2H_6$	0.793	1.180	Trautz and Sorg(28).
$CH_4 \sim C_3H_8$	0.656	1.258	Trautz and Sorg(28).
$C_2H_2 \sim C_3H_6$	0.808	1.20	Adzumi.
$N_2 \sim C_2H_4$	0.539	1.895	Trautz and Melster(26).
$CO \sim C_2H_4$	0.556	1.833	Trautz and Melster(26).
$C_2H_4 \sim O_2$	2.27	0.496	Trautz and Melster(26).
$C_2H_4 \sim$ Air	1.92	0.524	Breitenbach(14).
$C_2H_4 \sim CO_2$	1.565	0.603	
$C_2H_6 \sim C_3H_8$	0.920	1.10	Trautz and Sorg(28).
$C_3H_6 \sim C_3H_8$	0.945	1.02	Adzumi.
$C_3H_8 \sim N_2O$	1.945	0.564	Trautz and Kurz(27).
$C_3H_8 \sim CO_2$	2.01	0.538	Trauz and Kurz(27).

As seen from Table 10, all mixtures showing a maximum have the values of $\alpha_1\alpha_2\beta$ and $\alpha_1\alpha_2/\beta$ always smaller than unity and in the case of mixtures showing no maximum the values are either $\left.\begin{matrix}\alpha_1\alpha_2\beta < 1\\ \alpha_1\alpha_2 > \beta\end{matrix}\right\}$ or $\left.\begin{matrix}\alpha_1\alpha_2\beta < 1\\ \alpha_1\alpha_2 < \beta\end{matrix}\right\}$, thus the conditions (8) are satisfied excepting a few mixtures prefixed by an asterisk which have values only a little different from unity, but by inserting the correct values to C_{12}, all of these will perhaps satisfy the conditions (8).

Fig. 10 is a graphical interpretation of the conditions (8). The boundary curves between the region in which a maximum point occurs and that in which a maximum point does not occur are

$$\left.\begin{aligned} &\alpha_1\alpha_2\beta = 1 \quad \text{or} \quad a = \frac{1}{2\sqrt[4]{\frac{\nu}{B}}-1}, \\ \text{and} \quad &\alpha_1\alpha_2 = \beta \quad \text{or} \quad a = 2\sqrt[4]{\frac{\mu}{B\nu}}-1\,. \end{aligned}\right\} \tag{10}$$

In Fig. 10 μ is taken as ordinate and a as abscissa when ν equals to 0.5, 1.0, and 2.0, and the shaded part between two curves of $\alpha_1\alpha_2\beta = 1$ and $\alpha_1\alpha_2 = \beta$ is the region of a maximum or a minimum point.

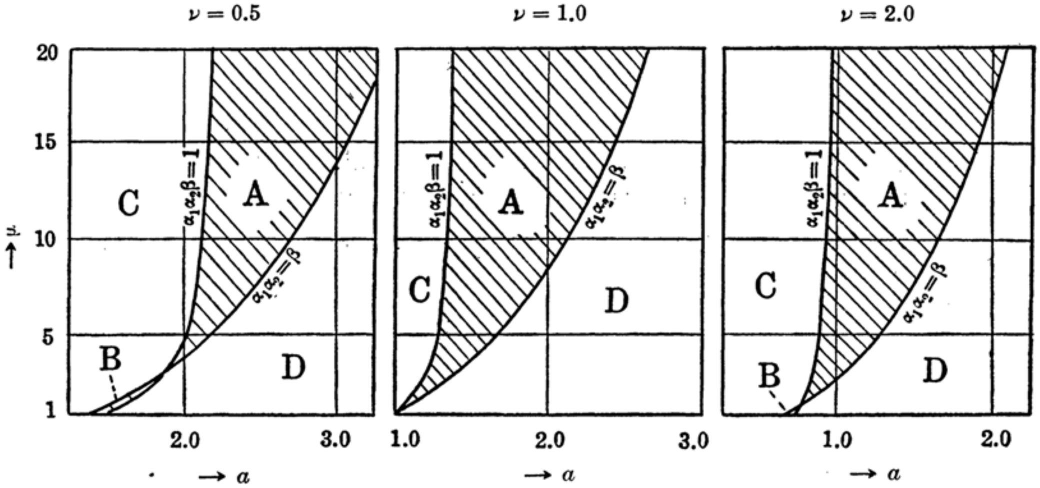

Fig. 10. Graphical Interpretation of the Conditions for the Occurrence of a Max. or a Min. Point. $\left(\mu = \frac{m_2}{m_1},\ a = \frac{S_2}{S_1},\ \nu = \frac{T+C_2}{T+C_1}\right)$.

(A) is a region of $\left.\begin{matrix}\alpha_1\alpha_2\beta < 1 \\ \alpha_1\alpha_2 < \beta\end{matrix}\right\}$ and in which a maximum point occurs,

(B) ,, $\left.\begin{matrix}\alpha_1\alpha_2\beta > 1 \\ \alpha_1\alpha_2 > \beta\end{matrix}\right\}$,, a minimum ,, ,

(C) ,, $\left.\begin{matrix}\alpha_1\alpha_2\beta > 1 \\ \alpha_1\alpha_2 < \beta\end{matrix}\right\}$,, neither max. nor min. occurs,

(D) ,, $\left.\begin{matrix}\alpha_1\alpha_2\beta < 1 \\ \alpha_1\alpha_2 > \beta\end{matrix}\right\}$,, ,, ,, ,

The mean free path of a molecule in a mixture is different from that of a simple molecule on account of the mutual collision between the different molecules, sometimes greater and sometimes smaller depending on the other molecule, and consequently the viscosity of the mixture is much complicated. It is a special case that the viscosity of a mixture follows the additive law, that is a case when $\alpha_1 = \alpha_2 = 1$. As seen from Fig. 8, for example, the viscosity of $C_3H_6 \sim C_3H_8$ is almost additive, because the molecular weights, the molecular diameters and Sutherland's constants of C_3H_6 and C_3H_8 are nearly equal respectively and then $\alpha_1 \doteqdot \alpha_2 \doteqdot 1$. In general, the viscosity curve deviates from a straight line, and by the mutual combination between α, μ, and ν, sometimes a maximum point occurs as easily seen from Fig. 10.

The existence of a minimum point is not improbable though any example has not been found. The area of (B) in Fig. 10 is very small, so that the probability of the occurrence of such a case by the combination of a few constants is very small, and when $C_1 = C_2$ a minimum point does not exist.

In order to know the relation between the conditions for the occurrence of a maximum point and the ratios of viscosities of two gases, β, we insert $a = \frac{\sqrt[4]{\mu}}{\sqrt{\beta\nu}}$ to equation (10), then we have as the formulæ of the boundary curves:

$$\left.\begin{aligned} \beta &= \frac{\sqrt{\mu}}{\nu}\left(2\sqrt[4]{\frac{\nu}{B}}-1\right)^2, \\ \text{and} \qquad \beta &= \frac{\sqrt{\mu}}{\left(2\sqrt[4]{\frac{\mu}{B\nu}}-1\right)^2\nu}. \end{aligned}\right\} \qquad (11)$$

As similarly as equation (10), this is represented graphically in Fig. 11, β being taken as abscissa. The shaded part indicates also the region of a maximum point and the greater the ratio μ is the wider this region becomes, and therefore the maximum point is very apt to occur if the ratio of two molecular weights is great. But if the ratio of two viscosities is great, the maximum point occurs only when the ratio of molecular weights is very great.

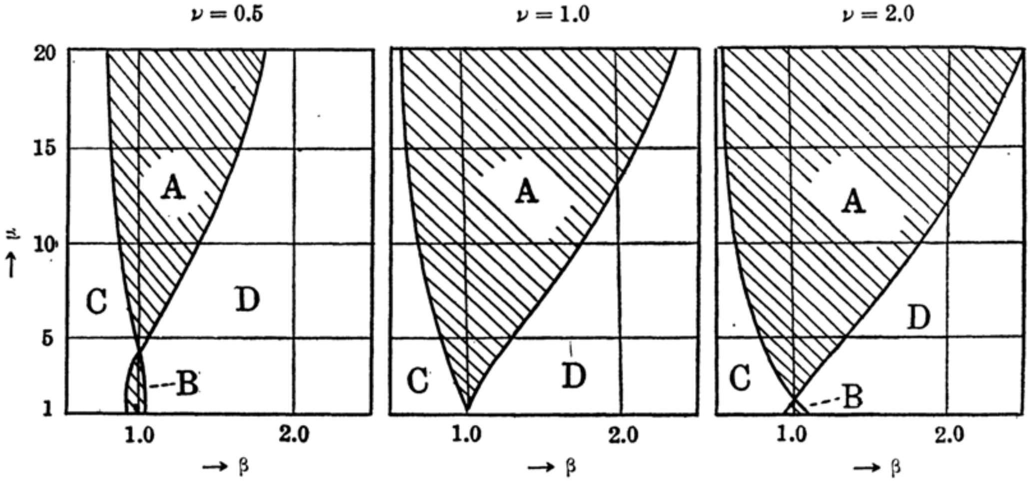

Fig. 11. $\left(\beta = \frac{\eta_2}{\eta_1}\right)$.

The above discussions are based on equation (3), in which the factor of the persistence of molecular velocity is introduced. Next, let us consider the case when the persistence of velocity is neglected, then a_1 and a_2 become:

$$a_1 = \left(\frac{S}{S_1}\right)^2 \sqrt{\frac{m_1+m_2}{2m_2}} \frac{T+C_{12}}{T+C_1},$$

$$a_2 = \left(\frac{S}{S_2}\right)^2 \sqrt{\frac{m_1+m_2}{2m_1}} \frac{T+C_{12}}{T+C_2}.$$

By inserting these values, $a_1a_2\beta$ and a_1a_2/β become the same formulæ as (9), excepting that B has the following value:

$$B = \frac{1+\mu}{2}.$$

As similarly as equation (10), this relation is represented graphically in Fig. 12. As seen from the figure, not only the shape of the shaped part is different from Fig. 10, but also the relative position of two curves of $a_1a_2\beta = 1$ and $a_1a_2 = \beta$ is inverted, and consequently the shaded part in Fig. 12 corresponds to the region of a minimum point and that of a maximum point does not exist. This is of course contrary to the fact. Namely, the consideration of the persistence of velocity is absolutely necessary to explain the occurrence of a maximum point.

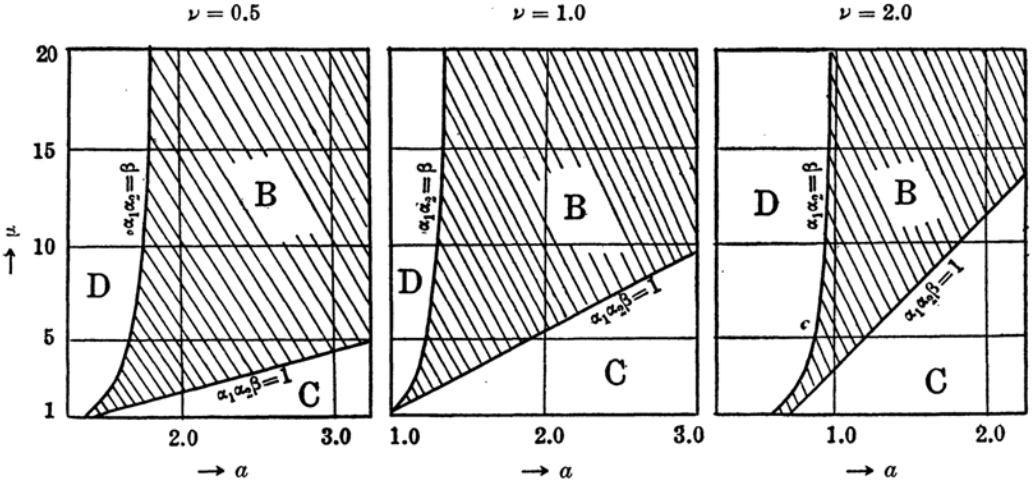

Fig. 12.

On the Displacement of the Composition of the Maximum Point. The composition of the maximum point displaces with the change of temperature. The displacements of four mixtures are given in Table 11. As seen from the table, the maximum point displaces to the lower concentration of hydrogen with the rise of temperature.

Table 11. The Compositions (H_2 %) of Maximum Points.

t°C	n_0			
	H_2~CH_4	H_2~C_2H_2	H_2~C_2H_6	H_2~C_3H_6
100	0.146	0.692	0.691	0.810
80	—	0.703	0.703	0.822
60	0.180	0.720	0.718	0.840
40	—	0.731	0.730	0.853
20	0.214	0.746	0.741	—

The composition of the maximum point is by (6)

$$n_0 = \frac{\sqrt{\alpha_1\alpha_2\beta}-1}{\sqrt{\alpha_1\alpha_2\beta}-1+\sqrt{\frac{\alpha_2}{\alpha_1}}\left(\sqrt{\alpha_1\alpha_2}-\sqrt{\beta}\right)}\,.$$

This formula contains a, μ, ν, and $\frac{(T+C_{12})^2}{(T+C_1)(T+C_2)}$ and as the first three is independent of temperature, the displacement of n_0 is due to the change of the term of Sutherland's constant. If $C_1 = C_2$, perhaps C_{12} being also the same, this term vanishes and n_0 becomes independent of temperature. Therefore, the displacement of n_0 is due to the difference of Sutherland's constants of two gases.

The Mean Free Paths of Gas Molecules in a Mixture. The mean free paths of a molecule in a simple gas and in a mixture are expressed by the following formulæ, the persistence of molecular velocity being taken into consideration.

In a simple gas:

$$\overset{(31)}{\lambda_{01}} = \frac{1}{N\pi S_1^2\sqrt{2}\left(1+\frac{C_1}{T}\right)\times 0{\cdot}797}$$

---

(31) The suffix o is used to indicate that the persistence of velocity is considered and to distinguish from the mean free path without consideration of it used in equation (3).

or $$\lambda_{02} = \frac{1}{N\pi S_2^2\sqrt{2}\left(1+\dfrac{C_2}{T}\right)\times 0.797}$$

and in a mixture: $$\left.\begin{aligned} \lambda'_{01} &= \frac{\lambda_{01}}{n_1+n_2\alpha_1}\,, \\ \lambda'_{02} &= \frac{\lambda_{02}}{n_2+n_1\alpha_2}\,, \end{aligned}\right\} \qquad (12)$$

where α_1 and α_2 have the same values as given in (3).

λ'_0's at 0°C. and 1 atm. pressure calculated by (12) for seven mixtures, whose viscosities have been measured by the present author, are given in Table 12, in which the value in brackets is the path when extremely small quantities of a gas are in another.

Table 12.

$H_2\sim CH_4$

H_2 (n_1)	$\lambda'_{01}\times 10^8$	CH_4 (n_2)	$\lambda'_{02}\times 10^8$
1.00	1445	1.00	615
0.75	1244	0.75	708
0.50	1093	0.50	834
0.25	977	0.25	1010
0.00	(880)	0.00	(1288)

$H_2\sim C_2H_2$

H_2 (n_1)	$\lambda'_{01}\times 10^8$	C_2H_2 (n_2)	$\lambda'_{02}\times 10^8$
1.00	1445	1.00	439
0.75	1202	0.75	530
0.50	1030	0.50	670
0.25	902	0.25	910
0.00	(801)	0.00	(1420)

$H_2\sim C_2H_6$

H_2 (n_1)	$\lambda'_{01}\times 10^8$	C_2H_6 (n_2)	$\lambda'_{02}\times 10^8$
1.00	1445	1.00	370
0.75	1106	0.75	447
0.50	898	0.50	567
0.25	755	0.25	772
0.00	(651)	0.00	(1209)

$H_2\sim C_3H_6$

H_2 (n_1)	$\lambda'_{01}\times 10^8$	C_3H_6 (n_2)	$\lambda'_{02}\times 10^8$
1.00	1445	1.00	285
0.75	1123	0.75	358
0.50	927	0.50	479
0.25	786	0.25	728
0.00	(682)	0.00	(1506)

$CH_4\sim C_2H_2$

CH_4 (n_1)	$\lambda'_{01}\times 10^8$	C_2H_2 (n_2)	$\lambda'_{02}\times 10^8$
1.00	615	1.00	439
0.75	594	0.75	466
0.50	574	0.50	496
0.25	556	0.25	530
0.00	(540)	0.00	(570)

$C_2H_2\sim C_3H_6$

C_2H_2 (n_1)	$\lambda'_{01}\times 10^8$	C_3H_6 (n_2)	$\lambda'_{02}\times 10^8$
1.00	439	1.00	285
0.75	422	0.75	308
0.50	405	0.50	335
0.25	389	0.25	367
0.00	(375)	0.00	(406)

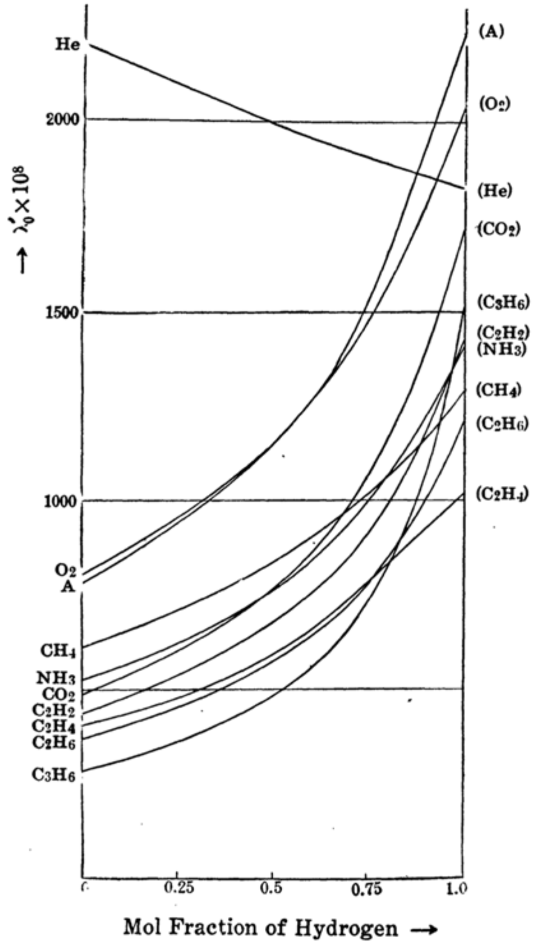

Fig. 13. The Change of the Mean Free Paths of Molecules in Hydrogen.

Table 12.—(*Concluded*)

$C_3H_6 \sim C_3H_8$

$C_3H_6\ (n_1)$	$\lambda'_{01}\times10^8$	$C_3H_8\ (n_2)$	$\lambda'_{02}\times10^8$
1.00	285	1.00	270
0.75	283.5	0.75	272
0.50	282	0.50	274
0.25	280	0.25	276
0.00	(279)	0.00	(278)

$NH_3 \sim N_2$

$NH_3\ (n_1)$	$\lambda'_{01}\times10^8$	$N_2\ (n_2)$	$\lambda'_{02}\times10^8$
1.00	522	1.00	751
0.75	558	0.75	754
0.50	600	0.50	756
0.25	648	0.25	759
0.00	(704)	0.00	(762)

$C_2H_6 \sim C_3H_8$

$C_2H_6\ (n_1)$	$\lambda'_{01}\times10^8$	$C_3H_8\ (n_2)$	$\lambda'_{01}\times10^8$
1.00	370	1.00	270
0.75	380	0.75	260
0.50	389	0.50	250
0.25	400	0.25	241
0.00	(412)	0.00	(233)

The change of the mean free paths of many gases due to the mixing of hydrogen are represented graphically in Fig. 13.

Generally the smaller mean free paths increases in the presence of molecules with the greater mean free paths and vice versa, excepting the case of $N_2 \sim NH_3$ and $C_2H_6 \sim C_3H_8$ whose change of paths are also given in Fig. 12.

Summary.

(1) Viscosities of six following simple gases and seven binary gaseous mixtures have been measured by the transpiration method over the temperature range between 20° and 100°C.

H_2, CH_4, C_2H_2, C_3H_6, C_3H_8, $H_2\sim CH_4$, $H_2\sim C_2H_2$,
$H_2\sim C_2H_6$, $H_2\sim C_3H_6$, $CH_4\sim C_2H_2$, $C_2H_2\sim C_3H_6$, $C_3H_6\sim C_3H_8$.

(2) The viscosities of the first four mixtures attain maximum values at definite compositions, which are about 20% of H_2 for the mixture $H_2\sim CH_4$ and about 70–80% of H_2 for the other three.

(3) Of several formulæ proposed to express the viscosity of gaseous mixtures, that in which Kuenen's consideration of the persistence of molecular velocity is introduced seems to be the most appropriate. The results of observation can be expressed satisfactorily by that formula, if we take a proper value for one of Sutherland's constants which is due to the attraction between the different molecules and can not be determined directly.

(4) The theoretical consideration of this Sutherland's constant by Schmick and London have been examined by thirty-two examples.

(5) The conditions for the occurrence of a maximum and a minimum points have been obtained from the discussion of the viscosity formula and these have been examined numerically for fifty-five mixtures, and found to be always correct except for only five cases.

The viscosity-composition curve deviates in general from a straight line. The deviation depends on the ratios of molecular weights, molecular diameters, and Sutherland's constants of two component gases. Especially the maximum point is very liable to occur if the ratio of two molecular weights is great.

(6) It has been shown that, if the persistence of molecular velocity be neglected, the condition for the occurrence of a maximum point cannot be fulfilled and the consideration of the persistence is absolutely necessary to explain this point.

(7) The composition of the maximum point displaces with the change of temperature. This is found to be due to the difference of Sutherland's constants of two component gases.

(8) The mean free path of each component gas has been calculated. Generally the smaller mean free paths increases in the presence of

molecules with the greater mean free paths and vice versa, excepting a few cases.

In conclusion, the author wishes to express his sincere thanks to Prof. M. Katayama for his kind guidance throughout the present study.

Chemical Institute, Faculty of Science,
Imperial University of Tokyo.