THE VISCOSITY OF NITROGEN-AMMONIA SYSTEM UNDER PRESSURES*

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The viscosities of nitrogen, ammonia and their mixtures have been measured by the use of an improved oscillating disk type viscometer at the temperatures of 25, 50, 75 and 100°C, and in the pressure range, up to about 120×10^5 Pa for nitrogen and up to around the saturated vapor pressure of ammonia for both ammonia and nitrogen-ammonia gaseous mixtures. The accuracy of the measurements is estimated to be within ±0.3%.

The initial pressure (density) dependence of the viscosity of ammonia and nitrogen-ammonia mixtures, that is, $\eta_0^{-1}(\partial\eta/\partial P)_{P\to0}$ (or $\eta_0^{-1}(\partial\eta/\partial\rho)_{\rho\to0}$) where η is the viscosity of gases, η_0 is the viscosity extrapolated to zero pressure (or density), P is pressure and ρ is density, changes its sign from negative to positive with an increase in not only temperature but also the mole fraction of nitrogen.

Introduction

Viscosity of gases that is one of transport coefficients, alike diffusion coefficient and thermal conductivity, has been required not only as a clue to clarify molecular interactions but also as an important physical property in chemical engineering.

Though fairly accurate experimental data at high temperature or at high pressure have been reported, most of them are the data about nonpolar molecules at low pressure1) and at high pressure2) and about their mixtures at low pressure3), and some of them are the data about polar molecules under pressures4,5,6) and about binary gas mixtures containing polar molecules at low pressure7). There is only one investigation about nonpolar-polar gas mixture under pressures8).

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The viscosity of pure gases and gas mixtures at low pressure has theoretically been treated by Chapman and Enskog9), and that of nonpolar dense gases was theoretically studied by Enskog10). But the viscosity of polar gases and gas mixtures under high pressure has not theoretically been treated yet. In order to develop the theory for the viscosity of polar gases and gas mixtures under pressures, the precise experimental data of the viscosity are more needed. So, the viscosity of nitrogen-ammonia gas mixture, as an example of the nonpolar-polar system, has been measured at 25, 50, 75 and 100°C using an improved oscillating disk type viscometer. For nitrogen, the measurements have been performed up to about 120×10^5 Pa, and for ammonia and nitrogen-ammonia mixtures, up to around the saturated vapor pressure of ammonia at each temperature, *i. e.*, about 10×10^6 Pa at 25°C, 20×10^5 Pa at 50°C, 35×10^5 Pa at 75°C and 60×10^5 Pa at 100°C.

Experimental apparatus

The principle of the measurements of gas viscosity was the same as that of Iwasaki *et al.*11) and Kestin *et al.*12) The modifications were undertaken in order to increase the precision of the measurements.

The suspension system of the viscometer is shown in Fig. 1. An oscillating disk ① was made of nonmagnetic stainless steel (SUS-316), and two fixed plates ② and three spacers ③ were made of quartz. They were polished to make their working surfaces which faced each other optically flat and parallel. The top and bottom ends of a fused quartz wire ④ of about 0.002cm in dia. were gripped in the center of the two small conical jaws ⑤ and ⑧, and assembled to the cylinder ⑦ and the mirror holder ⑩ by the tightening nuts ⑥ and ⑨. The thicker part of truncated cone of the conical jaws ⑧ was about 0.07cm in dia. In order to fix the wire at the center of the jaw chucks, a leading groove was set on the center of the longitudinal section. The mirror ⑬ (0.4×0.3×0.07cm) was made of quartz with aluminum backing. Vertical adjustment was obtained with the aid of the screw thread between the cylinder ⑦ and the nut ⑭, and angular adjustment by turning the nut ⑭. After these adjustments, the cylinder was fixed to the plate ⑰ by the bolt ⑮ and the bushing ⑯. The parts shown in numbers, ⑤–⑫ and ⑭, were made of super-invar, *i. e.*, iron-nickel-cobalt alloy, whose thermal-expansion coefficient was comparable to that of quartz13). The rest of the suspension system was the same as that used by Iwasaki *et al.*11) The characteristics of the suspension system are given in Table 1. This oscillating system was assembled so that the separation between b_1 and b_2 was equal using a thickness gage accurate to 0.0365cm. All these preparations were performed before this system was fixed in the high pressure vessel which was separated into the upper part

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10) S. Chapman and T. G. Cowling, "The Mathematical Theory of Non-Uniform Gases", Cambridge University Press (1961)

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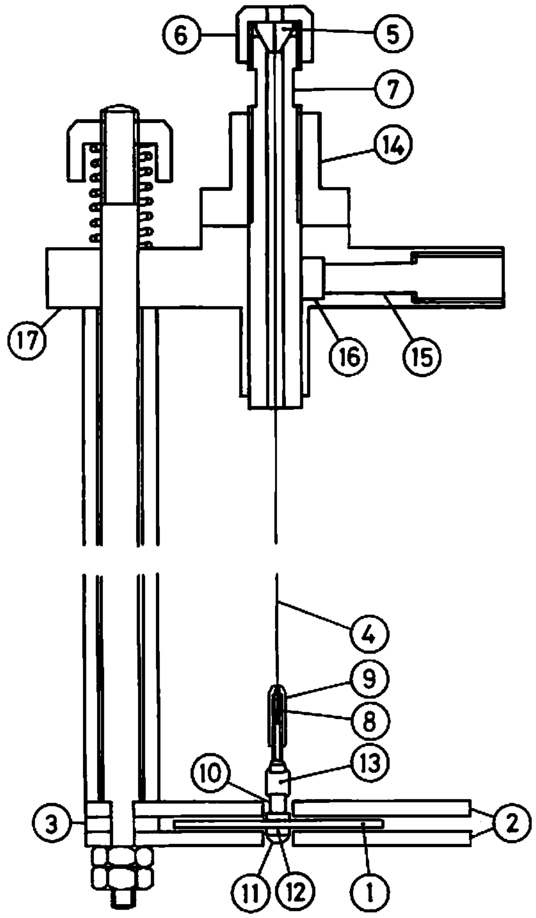

Fig. 1 Suspension system

①: Oscillating disk
②: Fixed plate
③: Spacer
④: Quartz wire
⑤: Conical jaws
⑥: Tightening nut
⑦: Cylinder
⑧: Conical jaws
⑨: Tightening nut
⑩: Mirror holder
⑪: Screw
⑫: Washer
⑬: Mirror
⑭: Nut
⑮: Bolt
⑯: Bushing
⑰: Plate

Table 1 Characteristics of suspension system (25°C)

Total separation between fixed plates		$D=0.1755\pm0.0002$cm
Upper separation		$b_1=0.0372\pm0.0005$cm
Lower separation		$b_2=0.0372\pm0.0005$cm
Radius of disk		$R=1.3997\pm0.0001$cm
Thickness of disk		$d=0.1012\pm0.0001$cm
Moment of inertia of suspension system		$I=4.8109\pm0.0017$g · cm^2
Natural period of oscillation and damping decrement	25°C	$T_0=22.598$ s $\Delta_0=5.0\times10^{-5}$
	50°C	$T_0=22.577$ s $\Delta_0=5.2\times10^{-5}$
	75°C	$T_0=22.556$ s $\Delta_0=5.4\times10^{-5}$
	100°C	$T_0=22.536$ s $\Delta_0=5.6\times10^{-5}$

and the lower one equipped with a quartz window. By turning two sets of three fine adjustment screws, the working surfaces of the oscillating disk and the fixed plates were controlled to be horizontal and parallel. The pressure vessel was mounted at the center of a double-walled oil thermostat. In the instruments of Iwasaki *et al.*[16] and Kestin *et al.*,[12] the rotating motion of the oscillating disk was given by applying a magnet just at the start of the motion, but in this apparatus, in order to remove a possible magnetic influence upon the viscometer and experimental fluids, the rotating motion was started by hand with a slight and careful twist of the whole thermostat on a ball bearing which was fixed on the base.

Pressure above 25×10^5 Pa was measured by the pressure balance of free piston type[14], and up to 25×10^5 Pa by the precision Bourdon gage. The Bourdon gage was calibrated by the use of a different pressure balance designed for moderate pressure. The accuracies of the pressure balance and the Bourdon gage were both within $\pm0.01\times10^5$ Pa. In the pressure measuring system, a U-tube type detector containing mercury for balancing of pressure was set up between the gas line and the oil line. One side of the U-tube was made of Pyrex glass capillary and the movement of the mercury level was detected by a phototransistor attached to the capillary. The other side served as a mercury reservoir, and connected with the capillary by a stainless steel pipe. The sensitivity of the detector was 0.1 cmHg.

The temperature of the pressure vessel and oil in the thermostat was measured by copper-constantan thermocouples which were calibrated against a platinum resistance thermometer provided with a calibration certificate by the National Bureau of Standards. The accuracy of the temperature measurement was ±0.01°C.

Nitrogen was supplied from the Nippon Oxygen Co., Ltd., and its purity was 99.99%. Commercial ammonia was dried with metallic sodium and purified by redistillation four times. The composition of the mixtures was determined by absorbing ammonia into a normal sulfuric acid solution.

Calculation of viscosity

The evaluation of the absolute viscosity of gases for the oscillating disk type viscometer was detailed in Newell's theory[15], but the calculating method used in the present study is the same as the modified method used by Iwasaki and Kestin[16].

In the calibration measurements to determine an empirical value of the constant C_N in Eq. (11) in Ref. 16, the empirical value C_N' was obtained using nitrogen, whose viscosity was accurately measured, at 25°C and at pressure up to 40×10^5 Pa. The viscosity value of nitrogen at 25°C was calculated from the following equation as standard.[17]

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$$\eta_{N_2, 25^\circ C} = [177.86 + 107.19\rho + 749.6\rho^2 - 434.0\rho^3] \times 10^{-7} \text{ Pa. s} \qquad (1)$$

where ρ is density in g · cm^{-3}. The mean value of C_N' was $C_N' = 1.1315$.

The average of deviations from the mean value was ±0.03%, and maximum deviation was 0.09%. Theoretical value obtained from Eq. (13) in Ref. 16 was $C_N = 1.1304$.

The densities required for the calculation were obtained from F. Din[18] for nitrogen and from K. Date[19] for ammonia. The densities of gas mixtures were calculated from Vri chart (Vri: ideal reduced molar volume) prepared by K. Tezuka[20]. For nitrogen-ammonia mixtures, the measured values at 50°C obtained by K. Date[21] were in agreement with the calculated ones within ±0.5%. It was the sufficient accuracy to calculate the viscosity of gases[4]. C_N' was checked before and after the series of measurements using nitrogen at 25°C, as it was slightly varied with the change of temperature etc. The experimental accuracy was estimated to be within ±0.3%.

Results

The experimental results obtained for nitrogen, ammonia and their mixtures are given in Tables 2 to 4. Figures 2 and 3 show the results for nitrogen and for ammonia, respectively, along with comparison with the data obtained by other investigators. The viscosity of nitrogen at the conditions of this work was measured by Micheles *et al.*[22] (25 to 75°C), Flynn *et al.*[23] (25 and 100°C), Kestin *et al.*[24] (25°C) and Iwasaki[25] (25 and 100°C). The data obtained in this work agree well with those obtained in the above-mentioned investigations within the experimental error at 25, 50 and 75°C, but are about 0.5% higher than those of Iwasaki at 100°C. Among the available viscosity data for ammonia under pressures in the literature[5,6,8], the data obtained by Iwasaki *et al.*[6] can be compared with those obtained in this work at each temperature. As can be seen from Fig. 3, the data of Iwasaki *et al.* agree well at 25 and 50°C, but are about 0.3% higher at 75°C and about 0.7% higher at 100°C than those of this work. The viscosity of ammonia shows a steady decrease with increasing pressure at 25, 50 and 75°C, but takes a minimum in the neighborhood of 30×10^5 Pa at 100°C.

Figures 4 and 5 show the results obtained for the gas mixtures at 50 and 100°C. It is evident that the initial slope of the viscosity, *i. e.*, $(\partial\eta/\partial P)_{P\to 0}$ where η is viscosity of gases and P is pressure, changes its sign from negative to positive as the mole fraction of nitrogen increases. Figure 6 shows the isobaric behavior of the viscosity of gas mixtures at 100°C. At other temperatures (25 to 75°C), the change of viscosity with pressure was smaller than that at 100°C. There are no other

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[18] F. Din, "Thermodynamic Functions of Gases" Vol. 3, Butterworths, London (1960)

[19] K. Date, *This Journal*, **43**, 1 (1973)

[20] K. Satoh, "Bussei Josu Suisanho", Maruzen, Japan (1954)

[21] K. Date, *This Journal*, **43**, 63 (1973)

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Table 2 Viscosiy of nitrogen

Pressure 10^5 Pa	Density $g \cdot cm^{-3}$	Viscosity 10^{-7} Pa·s	Pressure 10^5 Pa	Density $g \cdot cm^{-3}$	Viscosity 10^{-7} Pa·s
	25 ℃			75 ℃ (continued)	
1.22	0.00138	178.0	2.48	0.00240	199.9
1.32	0.00149	178.2	2.48	0.00240	199.8
1.87	0.00212	178.1	4.15	0.00402	200.1
2.60	0.00294	178.2	5.79	0.00560	200.2
3.14	0.00355	178.2	9.84	0.00951	200.7
4.99	0.00564	178.4	15.22	0.01469	201.3
7.14	0.00808	178.9	20.24	0.01953	201.9
10.19	0.01153	179.3	36.38	0.03497	204.3
15.18	0.01719	180.0	51.44	0.04928	206.6
20.36	0.02314	180.8	61.29	0.05857	208.4
31.28	0.03549	182.6	77.07	0.07328	211.1
40.93	0.04643	184.5	92.17	0.08716	214.5
51.18	0.05803	186.8	102.34	0.096363	216.5
61.16	0.06931	189.1	102.37	0.096401	216.7
71.96	0.08143	191.7	112.14	0.10512	218.8
82.02	0.09265	194.5	121.68	0.11357	221.0
91.98	0.1037	197.1	124.46	0.11600	221.6
102.76	0.11548	200.4		100 ℃	
112.17	0.12564	203.1			
120.63	0.13476	205.9	1.23	0.00111	211.1
120.68	0.13482	206.0	1.46	0.00132	211.4
	50 ℃		2.07	0.00187	211.5
			2.40	0.00217	211.7
1.27	0.00132	189.3	2.92	0.00264	211.5
1.95	0.00203	189.4	3.26	0.00295	211.6
3.02	0.00315	189.5	3.74	0.00337	211.6
5.06	0.00527	189.7	4.79	0.00432	211.8
6.71	0.00699	189.9	5.70	0.00515	211.6
6.71	0.00699	189.7	9.19	0.00829	212.2
10.13	0.01056	190.4	15.16	0.01364	212.9
15.40	0.01605	191.0	19.90	0.01789	213.4
20.31	0.02116	191.7	31.00	0.02779	214.7
36.29	0.03777	194.3	40.84	0.03649	216.3
51.86	0.05384	197.1	51.81	0.04615	218.0
61.76	0.06396	298.9	61.50	0.05460	219.6
76.88	0.07931	202.4	72.60	0.06418	221.6
91.94	0.09443	205.9	81.95	0.07216	223.3
102.40	0.10476	208.4	91.49	0.08022	225.0
112.13	0.11424	211.1	102.50	0.089208	227.0
122.32	0.12413	213.7	111.32	0.096743	229.2
	75 ℃		121.27	0.10499	231.5
			121.32	0.10499	230.9
1.32	0.00127	199.8			

Table 3 Viscosiy of ammonia

Pressure 10^5 Pa	Density g·cm^{-3}	Viscosity 10^{-7} Pa·s	Pressure 10^5 Pa	Density g·cm^{-3}	Viscosity 10^{-7} Pa·s
	25 ℃			75 ℃	
1.17	0.000819	100.6	1.27	0.000755	119.7
1.19	0.000832	100.8	1.40	0.000832	119.5
1.28	0.000893	100.6	2.63	0.00157	119.5
1.31	0.000918	100.7	5.12	0.00315	119.1
1.37	0.000961	100.6	10.18	0.006365	118.4
1.71	0.00120	100.6	15.31	0.009859	117.7
2.11	0.00148	100.5	20.31	0.01371	117.3
2.31	0.00163	100.4	25.26	0.01779	117.1
2.62	0.00186	100.2	28.52	0.02075	117.0
3.22	0.00230	100.2	30.51	0.02269	116.7
4.10	0.00295	99.8	32.41	0.02465	116.8
5.16	0.00376	99.4	33.68	0.02602	116.6
6.23	0.00461	99.1	34.61	0.02709	116.6
7.37	0.00555	98.7	35.45	0.02803	117.1
8.20	0.00625	98.4		100 ℃	
8.97	0.00693	98.1			
9.66	0.00760	97.9	1.27	0.000708	128.9
9.66	0.00760	97.9	1.65	0.000921	128.8
	50 ℃		4.15	0.00232	128.7
			6.98	0.00395	128.5
1.22	0.000783	110.2	12.69	0.007380	128.0
1.37	0.000882	110.1	20.33	0.01229	127.8
2.15	0.00139	110.0	25.26	0.01578	127.8
3.25	0.00211	109.9	30.48	0.01970	127.5
5.29	0.00349	109.4	35.47	0.02376	127.8
7.16	0.00480	108.9	40.41	0.02810	128.3
10.21	0.007053	108.3	45.68	0.03328	128.6
13.17	0.009393	107.7	50.81	0.03893	129.3
16.41	0.01213	107.1	53.89	0.04271	129.8
16.43	0.01215	107.2	55.74	0.04527	130.2
18.40	0.01393	106.7	57.84	0.04840	130.7
19.68	0.01527	106.5	59.85	0.05157	131.8
19.97	0.01569	106.6	60.62	0.05283	132.3

viscosity data for nitrogen-ammonia mixtures above atmospheric pressure.

The isothermal viscosity values at atmospheric pressure (1.013×10^5 Pa) are shown in viscosity-composition plot in Fig. 7. These values were obtained by extrapolating the relations between the viscosity of gas mixtures and pressure. The values obtained by Trautz *et al.*[26] agree with those obtained in this work, but the difference between the values obtained by Pal *et al.*[27] and those of this work is comparatively large.

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[27] A. K. Pal and A. K. Barua, *Indian J. Phys.*, **41**, 713 (1967)

Table 4 Viscosiy of nitrogen-ammonia mixtures

Pressure 10^5 Pa	Density $g \cdot cm^{-3}$	Viscosity 10^{-7} Pa·s
25 ℃		
N_2(0.198)-NH_3(0.802)		
1.16	0.000895	119.2
3.08	0.00239	118.8
5.05	0.00391	118.5
7.15	0.00564	118.4
9.14	0.00741	118.1
9.81	0.00802	118.1
N_2(0.403)-NH_3(0.597)		
1.23	0.00106	136.8
3.07	0.00266	136.8
5.10	0.00444	137.0
7.08	0.00625	137.1
9.10	0.00812	137.1
9.81	0.00879	136.9
N_2(0.550)-NH_3(0.450)		
1.29	0.00120	149.5
3.04	0.00283	149.6
5.06	0.00476	149.8
7.05	0.00674	149.8
9.13	0.00886	150.0
9.79	0.00955	150.2
N_2(0.801)-NH_3(0.199)		
1.27	0.00132	167.5
3.09	0.00322	167.7
5.05	0.00526	168.0
7.14	0.00750	168.1
9.07	0.00964	168.6
9.81	0.0105	168.6
50 ℃		
N_2(0.201)-NH_3(0.799)		
1.26	0.000900	127.8
3.07	0.00220	127.7
6.19	0.00455	127.5
10.06	0.007595	127.0
14.25	0.01103	127.1
17.25	0.01366	126.8
19.09	0.01521	126.9
20.06	0.01608	126.8
N_2(0.407)-NH_3(0.593)		
1.27	0.00101	146.9
3.09	0.00247	146.9
5.84	0.00467	146.9
10.13	0.008265	147.0
14.27	0.01190	147.6
17.23	0.01459	147.6
19.23	0.01642	147.6
20.04	0.01719	147.7
N_2(0.596)-NH_3(0.404)		
1.27	0.00111	163.7
3.07	0.00269	163.7
6.19	0.00543	164.1
10.05	0.009025	164.3
14.20	0.01280	164.8
18.29	0.01668	165.2
20.05	0.01837	165.4
50 ℃		
N_2(0.801)-NH_3(0.199)		
1.26	0.00121	178.2
3.10	0.00298	178.4
6.19	0.00595	178.8
10.13	0.009805	179.2
14.25	0.01389	179.8
18.23	0.01788	180.3
20.06	0.01972	180.4
75 ℃		
N_2(0.229)-NH_3(0.771)		
1.30	0.000376	139.7
1.90	0.00129	139.7
5.21	0.00352	139.5
10.23	0.007005	139.5
15.19	0.01054	139.6
20.39	0.01472	140.0
25.23	0.01872	140.0
30.26	0.02304	140.0
33.61	0.02608	140.3
35.68	0.02798	140.4
N_2(0.407)-NH_3(0.593)		
1.19	0.000880	155.7
5.15	0.00382	156.0
10.21	0.007725	156.2
14.98	0.01154	156.5
20.17	0.01592	157.0
25.30	0.02037	157.5
30.37	0.02488	158.0
33.48	0.02771	158.3
35.59	0.02963	158.4
N_2(0.570)-NH_3(0.430)		
1.26	0.00101	172.1
5.15	0.00414	172.4
10.24	0.008300	173.0
15.10	0.01252	173.3
20.41	0.01680	173.9
24.67	0.02050	174.5
30.41	0.02569	175.2
33.05	0.02810	175.5
35.41	0.03032	176.0
N_2(0.799)-NH_3(0.201)		
1.28	0.00114	188.5
4.80	0.00428	188.6
10.22	0.009145	189.1
15.04	0.01349	189.9
20.24	0.01821	190.4
25.14	0.02272	190.8
30.16	0.02742	191.6
33.31	0.03041	192.5
35.36	0.03240	192.8
100 ℃		
N_2(0.203)-NH_3(0.797)		
1.32	0.000818	145.4
5.07	0.00315	145.5
10.12	0.006493	145.3
17.66	0.01159	145.6
25.35	0.01706	145.9
32.63	0.02250	146.3
40.44	0.02876	147.2
45.20	0.03272	147.9
50.77	0.03763	148.6
53.85	0.04038	149.6
56.54	0.04286	150.0
58.70	0.04490	150.1
60.71	0.04685	150.6
N_2(0.399)-NH_3(0.601)		
1.32	0.000909	163.9
5.13	0.00351	164.2
10.04	0.006990	164.6
20.32	0.01459	165.4
30.64	0.02261	166.1
40.53	0.03051	167.3
48.13	0.03682	168.3
52.59	0.04062	169.0
55.61	0.04329	169.9
58.18	0.04559	169.6
60.62	0.04778	170.0
N_2(0.597)-NH_3(0.403)		
1.24	0.000940	182.0
5.25	0.00399	182.5
10.15	0.007810	182.8
19.42	0.01511	183.9
30.31	0.02387	184.9
40.39	0.03222	186.2
50.60	0.04076	187.9
55.58	0.04501	188.5
58.26	0.04732	189.2
60.33	0.04913	189.4
N_2(0.789)-NH_3(0.211)		
1.28	0.00106	197.4
5.17	0.00428	198.0
10.31	0.008565	198.3
20.29	0.01696	199.2
30.05	0.02503	200.5
39.68	0.03311	201.8
50.60	0.04265	203.1
54.79	0.04641	204.0
57.65	0.04906	203.9
60.42	0.05167	205.0

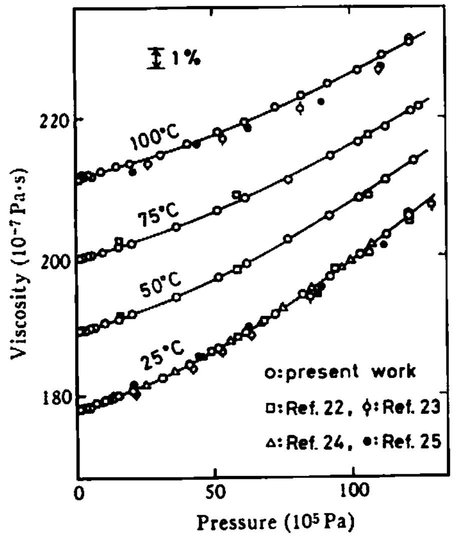

Fig. 2 The viscosity of nitrogen

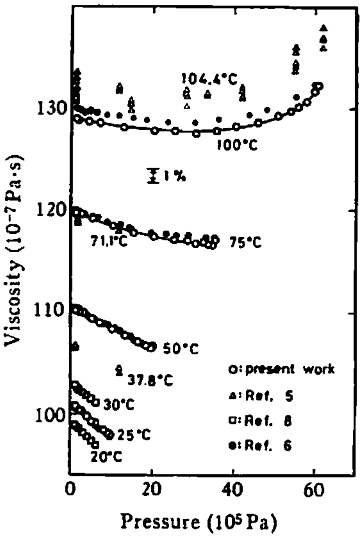

Fig. 3 The viscosity of ammonia

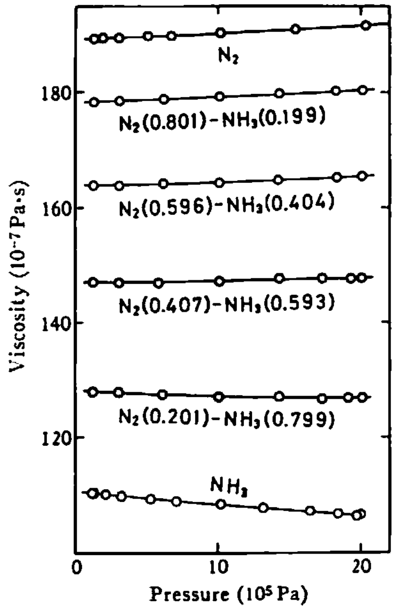

Fig. 4 The viscosity of nitrogen-ammonia system at 50°C

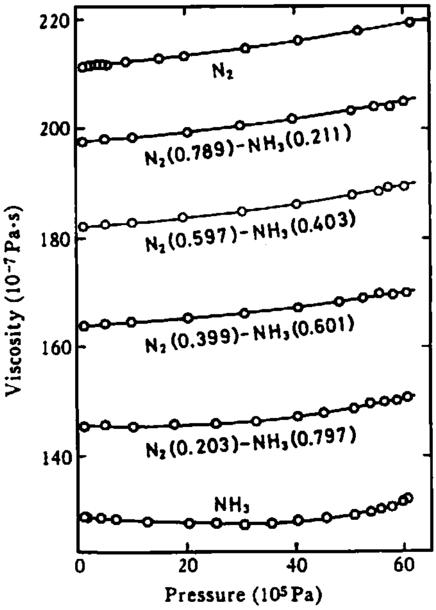

Fig. 5 The viscosity of nitrogen-ammonia system at 100°C

Discussion

It is known that the viscosity of gases depends on pressure and can be approximately expressed

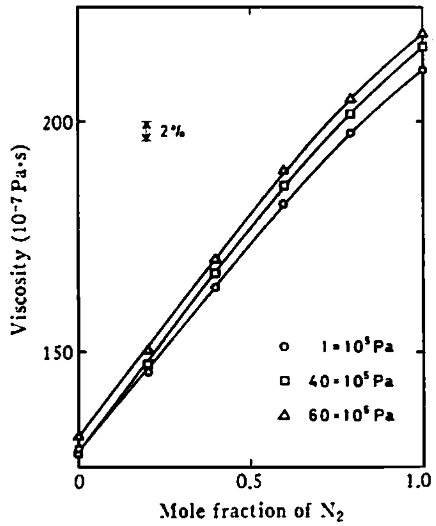

Fig. 6 The isodaric behavior of the viscosity of nitrogen-ammonia mixtures at 100°C

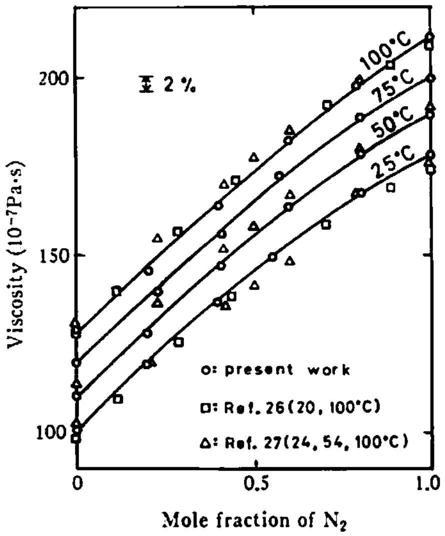

Fig. 7 The viscosity of nitrogen-ammonia system at atmospheric pressure

in terms of a power series expansion in pressure P as follows[28]:

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

where η_0 is the viscosity extrapolated to zero pressure, and $\alpha=\eta_0^{-1}(\partial\eta/\partial P)_{P\to0}$ is called the initial pressure dependence of viscosity in this study. The viscosity of polar gases such as ammonia[5,6], sulfur dioxide[4] and water vapor[29,30] shows a steady decrease with increasing pressure in lower temperature region, and after taking a minimum shows a steady increase with pressure above a certain temperature, *i. e.*, the sign of α is reversed from negative to positive at a certain temperature as temperature increases. As can be seen from Figs. 4 and 5, the sign of α is likewise reversed with increasing mole fraction of nitrogen in this experimental temperature range.

Expressing the viscosity of gases as a function of density ρ instead of pressure P, Eq. (2) can be rewritten as

$$\eta=\eta_0(1+\alpha'\rho+\cdots\cdots) \qquad (3)$$

where η_0 is the viscosity extrapolated to zero density, and $\alpha'=\eta_0^{-1}(\partial\eta/\partial\rho)_{\rho\to0}$ is the initial density dependence of viscosity. The experimental viscosities of nitrogen, ammonia and their mixtures were represented by the polynomial of density as follows:

$$\eta=\eta_0(1+\alpha'\rho+\beta'\rho^2+\gamma'\rho^3) \qquad (4)$$

Constants in Eq. (4) were obtained by means of least squares method and are listed in Table 5.

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28) F. Danon and I. Amdur, *J. Chem. Phys.*, **54**, 4841 (1971)

29) J. Kestin and H. E. Wang, *Physica*, **26**, 575 (1960)

30) S. L. Rivkin, A. S. Levin and L. B. Israilevsky, "The VII th International Conference on the Properties of Water Substance" held at Tokyo on 1968

Figure 8 shows the relation between α' and the mole fraction of nitrogen x_{N_2} at each temperature. Each isotherm was approximated by straight line, and then the mole fraction of nitrogen at $\alpha'=0$, $x_{N_2}^0$, was obtained from the diagram. The relation between temperature and $x_{N_2}^0$, is linear as shown in Fig. 9.

The initial pressure dependence of viscosity of gases, α, was theoretically treated by Stogryn *et al.*,31) Singh *et al.*32) and Danon *et al.*28) In the theoretical approximation for α, it was considered that gas was a mixture of monomers and dimers and that α depended on a dimerization effect and a collisional transfer effect31). Especially, since polar gas such as water vapor33) was apt to form dimers, it was reasonable to consider that α for polar gas depended on the effect of dimerization more than for nonpolar gas32). Therefore, it was possible to account for the decrease of the viscosity

Table 5 Values of constants in Eq. (4)a)

Temp. (°C)	x_{N_2}	η_0	α'	β'	$\gamma' \cdot 10^2$
25	0	100.99	−3.58	−169	143
	0.198	119.54	−3.68	555	−363
	0.403	137.03	−2.07	655	−490
	0.550	149.26	1.84	−347	228
	0.801	167.39	0.656	− 27.0	29.6
	1.000	177.88	0.600	4.54	− 0.0237
50	0	110.44	−2.80	− 8.53	26.8
	0.200	127.94	−1.06	30.5	0.994
	0.406	147.02	−0.739	139	− 46.9
	0.596	163.51	0.648	− 12.6	6.35
	0.801	178.08	0.462	32.4	− 11.2
	1.000	189,13	0.549	4.41	− 0.0318
75	0	119.81	−2.20	53.6	− 2.50
	0.229	139.71	−0.370	35.9	− 6.03
	0.407	155.73	0.189	26.9	− 4.61
	0.570	171.92	0.717	− 4.99	2.24
	0.799	188.29	0.577	− 6.46	3.57
	1.000	199.61	0.531	3.65	− 0.00203
100	0	128.91	−1.03	27.3	0.171
	0.203	145.54	−0.348	31.0	− 1.63
	0.399	163.90	0.434	10.6	− 0.621
	0.597	181.95	0.628	1.25	0.637
	0.789	197.43	0.411	10.2	− 0.774
	1.000	211.23	0.507	4.18	− 0.0413

a) $\eta=\eta_0(1+\alpha'\rho+\beta'\rho^2+\gamma'\rho^3)$ η: 10^{-7}Pa · s ρ: g/cm^3

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31) D. E. Stogryn and J. O. Hirschfelder, *J. Chem. Phys.*, **31**, 1545 (1959)
32) Y. Singh, S. K. Deb and A. K. Barua, *ibid.*, **46**. 4036 (1967)
33) J. P. O' Connel and J. M. Prausnitz, *IEC Fundamentals*, **8**, 453 (1969)

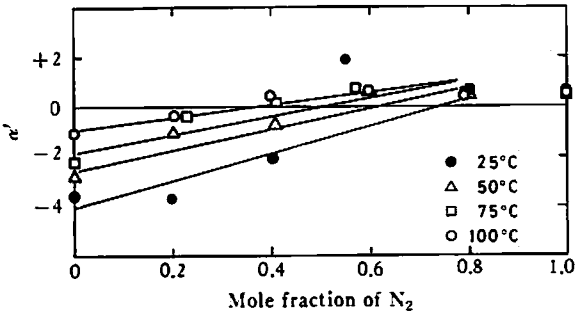

Fig. 8 The relation between α' and the mole fraction of nitrogen

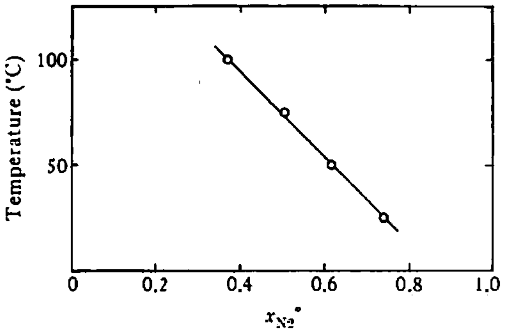

Fig. 9 The relation between temperature and $x_{N_2}{}^*$

of the polar gases with increasing pressure. The reason mentioned above and the diagram of Fig. 8 suggest that the rise of temperature and the presence of nonpolar gas disturb the formation of dimers of polar gas. In order to clarify this phenomenon, it is necessary to carry further on the research of the initial density dependence about other nonpolar-polar gas mixtures.

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