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The Viscosity of Gaseous Sulfur Dioxide

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The viscosity of sulfur dioxide was measured in the range from 25 to 160 °C over the pressure range from 1 atm to near the saturation pressure at 25—150 °C and up to 80 atm at 160 °C by means of the oscillating-disk method. The accuracy of the measurement is, at worst, ±0.25%, except that the accuracy is ±0.3% for the range above 66 atm at 160 °C. It is found that, at temperatures from 25 to 75 °C, the viscosity of sulfur dioxide shows a steady decrease with the pressure (or density) and that at 100 and 125 °C it passes through a minimum. On the contrary, the isotherms at 150 and 160 °C show a steady increase with the pressure (or density). The data were fitted to the polynomials of the density at each temperature investigated. The coefficient of the first-order density term in the equation, which indicates the degree of the initial density dependence, changes its sign from negative to positive at a temperature of about 135 °C.

The present paper continues the work on the experimental determination of the viscosity of polar gases and constitutes a sequel to our paper on ammonia.[1)] Investigations of the effect of the pressure on the viscosity of sulfur dioxide have been made by several investigators,[2—4)] but the results have not indicated any definite relation between the viscosity of sulfur dioxide and the pressure (or density). Therefore, the measurements of the viscosity of sulfur dioxide were undertaken in the range from 25 to 160 °C over the pressure range from 1 atm to near the saturation pressure at 25—150 °C and up to 80 atm at 160 °C.

TABLE 1. CHARACTERISTICS OF SUSPENSION SYSTEM (25 °C)

Total separation between plates		$D=0.1732_4$ cm
Upper separation		$b_1=0.0386_6$ cm
Lower separation		$b_2=0.0386_6$ cm
Radius of disk		$R=1.3955_7$ cm
Thickness of disk		$d=0.0959_1$ cm
Moment of inertia of suspension system		$I=4.5119$ g·cm^2
Damping decrement and natural period of oscillation		
25 °C	$\Delta_0=2.73\times10^{-5}$	$T_0=30.00$ s
50 °C[a)]	$\Delta_0=2.52\times10^{-5}$	$T_0=26.20$ s
75 °C	$\Delta_0=4.54\times10^{-5}$	$T_0=30.09$ s
100 °C	$\Delta_0=7.61\times10^{-5}$	$T_0=30.13$ s
125 °C	$\Delta_0=12.74\times10^{-5}$	$T_0=30.13$ s
150 °C	$\Delta_0=20.92\times10^{-5}$	$T_0=30.15$ s
160 °C	$\Delta_0=24.92\times10^{-5}$	$T_0=30.03$ s

a) Suspension wire used for 50 °C is different from that used for other temperatures.

Experimental

Measurements were made by the Maxwell-type oscillating-disk method. The apparatus as well as the experimental procedures was identical with those described in Ref. 1 and so need not be repeated here.

The characteristics of the suspension system are recorded in Table 1.

Materials. The sulfur dioxide was obtained from the Showa Denko Co., which reported a purity of about 99.9%. It was purified and dried by several distillations, with a generous rejection of the head and tail fractions, and finally by distillation through phosphorus pentoxide tubes.

Evaluation of Results

The results of measurements were evaluated with the aid of Newell's theory in a manner similar to, but not quite identical with, that used in Ref. 1. In this experiment, the evaluations were made by means of Eqs. (1) and (2), not by Eq. (4) as in Ref. 1. This was because the neglect of the terms containing a, f, and h in Eq. (2) in Ref. 1 was found to cause a discrepancy over 0.3% in the results evaluated by Eqs. (1) and (2) in Ref. 1 in the high-density region at temperatures over 125 °C.

It was confirmed that the present measurements satisfactorily fulfill the conditions under which Eqs. (1) and (2) in Ref. 1 will yield the required accuracy

TABLE 2. EXPERIMENTAL RESULTS AT 150 °C

Pressure P atm	Density ρ g/cm^3	Period T s	Decrement $\Delta\cdot10^2$	Viscosity η μpoise	Boundary layer thickness δ, cm	δ/b
1.00	0.00185	30.235	3.4264	182.55	0.6876	17.764
3.30	0.00619	30.202	3.4262	182.79	0.3766	9.729
6.53	0.01244	30.242	3.4328	182.95	0.2657	6.864
9.683	0.018758	30.181	3.4320	183.33	0.2166	5.594
14.036	0.027854	30.188	3.4420	183.88	0.1780	4.598
19.759	0.040551	30.194	3.4570	184.71	0.1478	3.819
24.917	0.052812	30.200	3.4803	186.00	0.1300	3.358
34.909	0.079639	30.342	3.5492	188.95	0.1067	2.756
43.707	0.10884	30.298	3.6268	193.53	0.0924	2.386
53.640	0.14859	30.384	3.7875	201.77	0.0807	2.085
60.677	0.18891	30.468	4.0019	212.86	0.0735	1.900
67.291	0.25354	30.533	4.4360	235.93	0.0668	1.726

of 0.1%. For example, the experimental results at 150 °C are shown in Table 2. All of the δ/b values were larger than 1.25.

The apparatus constant, C_N, was determined experimentally using air and nitrogen at 1 atm and 25 °C respectively. The values of C_N obtained by the two methods showed a good agreement, the discrepancy being only 0.04%.

The mean value of

$$C_N = 1.13124$$

was adopted for further calculation. However, for the measurement at 50 °C the suspension wire was different from that for the other temperatures and the check measurements of the instrument constant, C_N, were made following the method described above. The following value was obtained, showing a very slight discrepancy from the above value, and was used for the calculation at 50 °C:

$$C_N = 1.12881.$$

The accuracy of the present experiments is at worst ±0.25%, considering the uncertainty in the determination of Δ and T, and other sources of error, except that the accuracy is ±0.3% for the range above 66 atm at 160 °C for a reason to be described later.

Results and Discussion

The experimental results are presented in Table 3 and in Figs. 1 and 2. The density of sulfur dioxide was calculated using the compressibility data given by Kang *et al.*[5]

The densities marked * in Table 3 have an uncertainty of 10% at most, because of the difficulty of the estimation by the interpolation of the P–V–T data given by Kang *et al.*, the region being near the critical state. Therefore, the viscosity data in this range were

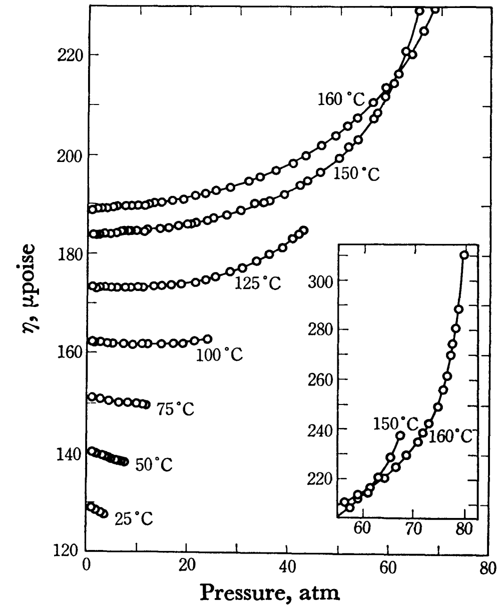

Fig. 1. Effect of pressure on viscosity of sulfur dioxide.

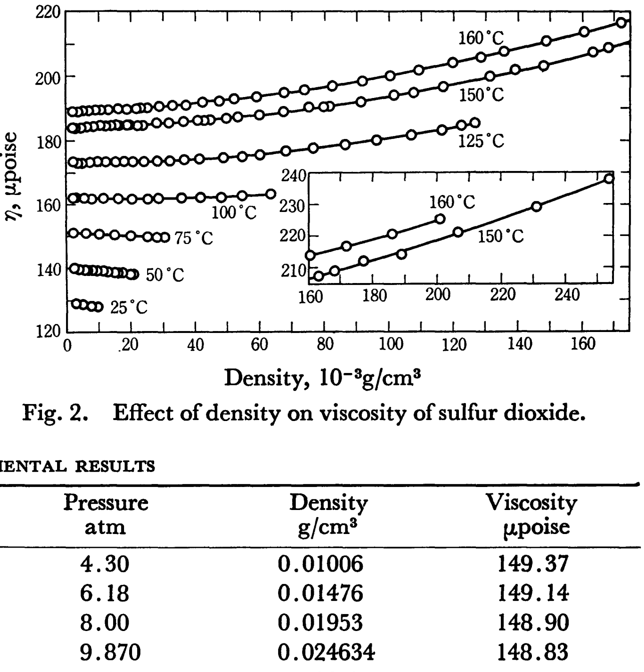

Fig. 2. Effect of density on viscosity of sulfur dioxide.

TABLE 3. EXPERIMENTAL RESULTS

Pressure atm	Density g/cm^3	Viscosity μpoise
	25 °C	
1.00	0.00267	127.93
1.74	0.00471	127.51
2.67	0.00738	127.12
3.36	0.00941	126.79
	50 °C	
1.00	0.00245	139.26
1.67	0.00411	138.94
2.12	0.00526	138.89
3.08	0.00775	138.74
3.74	0.00950	138.41
4.40	0.01128	138.35
4.90	0.01265	138.18
5.59	0.01457	137.79
6.09	0.01601	137.72
6.57	0.01740	137.55
7.32	0.01960	137.43
7.68	0.02069	137.26
	75 °C	
1.00	0.00226	150.03
2.67	0.00591	149.82
4.30	0.01006	149.37
6.18	0.01476	149.14
8.00	0.01953	148.90
9.870	0.024634	148.83
11.107	0.028170	148.52
11.841	0.030340	148.55
	100 °C	
1.00	0.00211	161.02
1.50	0.00317	160.92
2.61	0.00557	160.96
3.86	0.00833	160.76
5.18	0.01127	160.67
7.13	0.01579	160.58
8.94	0.02011	160.49
10.933	0.025029	160.56
12.227	0.028336	160.57
14.824	0.035273	160.72
16.969	0.041331	160.88
19.222	0.048078	160.92
21.196	0.054347	161.34
23.867	0.063443	161.87

TABLE 3. (Continued)

Pressure atm	Density g/cm^3	Viscosity μpoise
	125 °C	
1.00	0.00197	172.03
1.55	0.00307	171.85
2.47	0.00492	171.92
3.52	0.00704	171.97
4.61	0.00930	171.95
5.87	0.01194	171.91
7.36	0.01512	172.06
8.55	0.01770	172.03
9.820	0.020506	172.06
11.217	0.023650	172.16
13.082	0.027952	172.20
14.740	0.031884	172.45
16.470	0.036517	172.62
18.463	0.041127	172.88
21.252	0.048469	173.39
23.421	0.054442	173.78
25.378	0.060059	174.30
28.073	0.068225	175.28
30.627	0.076528	176.07
33.507	0.086641	177.42
35.998	0.096125	178.75
38.574	0.10694	180.24
40.567	0.11633	182.01
41.760	0.12231	183.11
42.611	0.12691	183.79
	150 °C	
1.00	0.00185	182.55
1.55	0.00288	182.44
1.69	0.00315	182.74
2.31	0.00430	182.56
3.30	0.00619	182.79
4.46	0.00842	182.70
5.47	0.01036	182.99
5.53	0.01049	182.95
6.53	0.01244	182.95
7.18	0.01372	183.24
8.05	0.01546	183.19
8.35	0.01607	183.28
9.683	0.018758	183.33
9.990	0.019389	183.28
11.217	0.021911	183.10
12.068	0.023684	183.49
14.036	0.027854	183.88
15.938	0.031742	184.00
16.052	0.031987	183.81
17.821	0.036153	184.18
19.759	0.040551	184.71
20.621	0.042543	184.73
21.509	0.044615	185.00
23.554	0.049491	185.58
24.917	0.052812	186.00
27.577	0.059539	186.68
30.461	0.067133	187.61
33.224	0.074786	188.85
34.909	0.079639	188.95
35.582	0.081619	189.34
38.763	0.091347	190.75
41.877	0.10153	192.54
43.707	0.10884	193.53
46.143	0.11667	195.14
49.797	0.13118	198.29
51.686	0.13938	200.30
53.640	0.14859	201.77
56.505	0.16332	205.82
57.348	0.16796	207.07
58.870	0.17706	210.37
60.677	0.18891	212.86
62.986	0.20637	219.37
65.526	0.23090	227.27
67.291	0.25354	235.93
	160 °C	
1.00	0.00181	187.50
2.11	0.00384	187.63
2.72	0.00497	187.68
3.78	0.00692	187.73
4.90	0.00904	188.03
5.63	0.01042	188.06
6.98	0.01299	188.11
8.40	0.01575	188.29
9.862	0.018622	188.39
11.406	0.021702	188.46
11.917	0.022731	188.56
13.006	0.024941	188.83
14.725	0.028489	189.07
16.693	0.032619	189.42
18.766	0.037087	189.82
21.143	0.042328	190.46
23.273	0.047164	190.95
25.317	0.051925	191.48
28.166	0.058765	192.08
31.663	0.067523	193.41
34.051	0.073768	194.18
37.161	0.082266	195.46
40.478	0.091866	197.03
43.207	0.10026	198.50
46.207	0.10994	200.63
49.011	0.11957	202.54
51.445	0.12847	204.32
53.299	0.13564	206.08
56.391	0.14845	209.28
59.097	0.16062	212.18
61.396	0.17171	214.82
64.123	0.18622	218.68
66.537	0.20089	223.38
68.600	0.21539*	227.87
70.824	0.23379*	233.31
71.807	0.24313*	236.91
73.068	0.25666*	241.05
74.769	0.27895*	247.40
75.717	0.29330*	254.49
76.522	0.30704*	259.97
77.094	0.31776*	264.50
77.521	0.32677*	272.68
78.278	0.34404*	278.11
78.609	0.35220*	286.09
79.792	0.38631*	308.04

TABLE 4. EFFECT OF ERROR IN DENSITY UPON THE CALCULATION OF VISCOSITY (160 °C)

Pressure P atm	Viscosity η μpoise Density from Table 3	Viscosity η' μpoise Density increased by 5%	% difference in viscosity $\frac{\eta'-\eta}{\eta}\times 100\%$	Viscosity η'' μpoise Density increased by 10%	% difference in viscosity $\frac{\eta''-\eta}{\eta}\times 100\%$
68.600	227.87	227.91	0.019	227.95	0.038
70.824	233.31	233.36	0.021	233.40	0.040
71.807	236.91	236.96	0.021	237.01	0.041
73.068	241.05	241.10	0.022	241.15	0.042
74.769	247.40	247.45	0.023	247.51	0.045
75.717	254.49	254.55	0.024	254.61	0.047
76.522	259.97	260.03	0.025	260.09	0.049
77.094	264.50	264.57	0.026	264.63	0.051
77.521	272.68	272.75	0.028	272.83	0.054
78.278	278.11	278.20	0.029	278.27	0.056
78.609	286.09	286.18	0.031	286.26	0.060
79.792	308.04	308.15	0.035	308.26	0.070

excluded from the viscosity *vs.* density curves (Fig. 2).

In order to assess the extent of the deviations in viscosity values resulting from the uncertainty in the density, it will be necessary to refer to the effect of the density on the calculation of the viscosity using Eqs. (1) and (2) in Ref. 1. The density of the gas appears in the final, working equation (1) in Ref. 1. However, for this purpose the value of the density need not be known extremely accurately, because it affects the value of β^2 taken from Eq. (2) in Ref. 1 approximately proportionately, thus nearly cancelling the direct effect upon the viscosity, η, in Eq. (1). A test calculation was performed for the range of pressures higher than 66 atm at 160 °C in the case of the densities increased by 5% and then by 10% of those listed in Table 3. The results are recorded in Table 4. Thus, considering the uncertainty of the density, the accuracy of the viscosity in the range discussed above is assumed to be better than ±0.3%.

The diagram in Fig. 1 represents the variation in the viscosity of sulfur dioxide with the pressure for the given temperatures. The viscosity of sulfur dioxide shows a steady decrease with the pressure at the temperatures from 25 to 75 °C. The viscosities at 100 and 125 °C pass through a minimum. On the contrary, the isotherms at 150 and 160 °C show a steady increase with the pressure; $(\delta\eta/\delta T)_P>0$ at low pressures and $(\delta\eta/\delta T)_P<0$ at high pressures.

The diagram in Fig. 2 representing the variation in the viscosity of sulfur dioxide with the density is similar to that in Fig. 1, but $(\delta\eta/\delta T)_\rho>0$ under the present experimental conditions.

Figures 1 and 2 show that the initial pressure (or density) dependency of the viscosity of sulfur dioxide changes its sign from negative to positive as the temperature increases. This relation can be clarified by representing the viscosity of sulfur dioxide in the polynomials of density as follows:

$$\eta = \eta_0 + b\rho + c\rho^2 + d\rho^3. \qquad (1)$$

The constants of Eq. (1), η_0, b, c, and d, were evaluated using the measured values of the viscosity at each temperature; they are recorded in Table 5. These equations reproduce the measured values with a deviation not exceeding ±0.1% except for the several measured values in the high-pressure region at 150 °C, which were also reproduced within the limits of experimental error. The initial density dependence of the viscosity of sulfur dioxide, b, and the viscosity at zero density, η_0, are both given against the temperature in Fig. 3. This figure shows that the relation between the

TABLE 5. VALUES OF CONSTANTS IN Eq. (1)[a)]

Temp. °C	η_0	b	$c\cdot 10^3$	$d\cdot 10^6$
25	128.38	−0.169		
50	139.51	−0.112	0.18	
75	150.24	−0.090	1.12	
100	161.09	−0.044	0.885	
125	172.03	−0.020	1.053	−1.26
150	182.54	0.0243	0.720	0.067
160[b)]	187.52	0.0304	0.877	−0.717

a) $\eta=\eta_0+b\rho+c\rho^2+d\rho^3$. η: poise, ρ: 10^{-3}g/cm^3

b) Density up to 0.20089 g/cm^3.

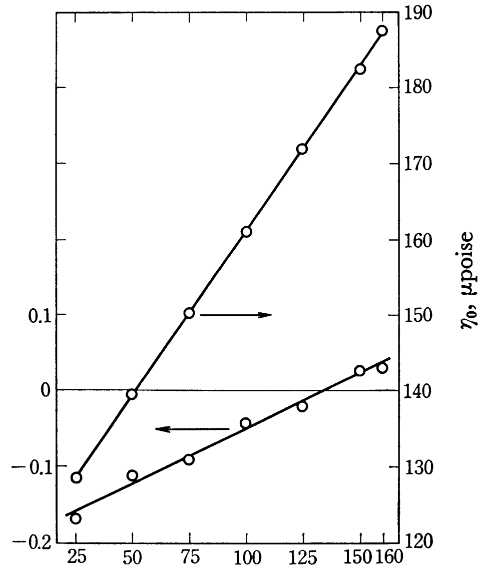

Fig. 3. Effect of temperature on η_0 and b in Eq. (1).

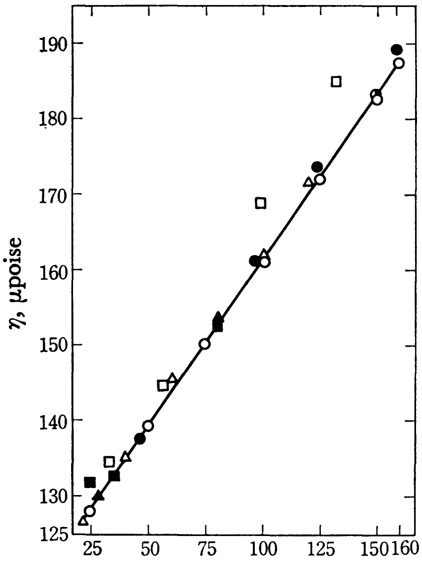

Fig. 4. Comparison of results for sulfur dioxide at 1 atm. ●, Trautz and Weizel⁶⁾; △, Titani⁷⁾; ◑, Trautz and Winterkorn⁸⁾; ■, Chakraborti and Gray⁹⁾; □, Pal and Barua¹⁰⁾; ▲, Stewart and Maass¹¹⁾; ○, present work.

initial density dependence and the temperature is linear and changes its sign from negative to positive at about 135 °C, and that the relation between the viscosity at zero density and the temperature is also linear.

The measurements of the viscosity of sulfur dioxide at high densities were performed by Awbery and Griffiths(2) for the liquid region, by Shimotake and Thodos(3) at 200 °C, and by Stakelbeck.(4) The measurements by Stakelbeck were performed not only for the liquid region but also for the gaseous region up to 40 °C, using the falling-body method, and so can be compared directly. However, his data indicate a rapid increase in the viscosity with an increase in the pressure, in disagreement with the present measurements.

In Fig. 4, the relation between the viscosity of sulfur dioxide at 1 atm and the temperature is given. Here, the data given by other investigators(6–11) are limited to those obtained in the same temperature range as that of the present measurement, thus making it possible to compare directly.

The data plotted show a good agreement within 1% among all the investigators, except that the value at 25 °C by Chakraborti and Gray(9) and those by Pal and Barua(10) show a large discrepancy.

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