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Thermal Conductivity of Gases. III. Some Values of the Thermal Conductivities of Argon, Helium, and Nitrogen from 0 °C to 75 °C at Pressures of 1×10^5 to 2.5×10^7 Pascals*

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Accurate measurements of the thermal conductivities of Ar and He agree with the theoretical value of 2.5 $\phi \eta c_v$ (η = viscosity, c_v = specific heat capacity at constant volume ϕ is a number slightly greater than 1 depending upon the intermolecular potential). Measurements of the thermal conductivities of N_2 at 9.6 and 75 °C as a function of pressure up to 2.53×10^7 Pa help to appraise the validity of other measurements of the thermal conductivities of dense gases. The excess conductivity of nitrogen (the additional conductivity resulting from pressure) is shown to be a function of only the density of the nitrogen from 0 to 700 °C and pressures up to 1.3×10^8 Pa.

Key words: Excess thermal conductivity of N_2; thermal conductivity of Ar; thermal conductivity of He; thermal conductivity of N_2.

1. Introduction

The values of thermal conductivities being reported in this paper were obtained by the author at the Massachusetts Institute of Technology. They were to have been part of a more extensive set of measurements, but further work was not completed with this equipment because the author came to the National Bureau of Standards. Even though the number of these results is small, the measurements are thought to be sufficiently accurate to be useful in three respects:

(1) Some measurements of the thermal conductivities of helium allow the factor $f = K/(\eta c_v)$ to be more exactly evaluated. It had appeared to be anomalously low. In this equation K is the thermal conductivity, η the viscosity, and c_v the specific heat capacity at constant volume.

(2) Some measurements of the thermal conductivities of argon, in combination with the values for helium, provide high and low values for calibration of other devices for relative determinations of the thermal conductivities of gases.

(3) Some measurements of the thermal conductivities of nitrogen, up to 2.53×10^7 Pa of pressure, help to provide a guide for evaluating the accuracy of other determinations of the thermal conductivities of dense gases.

2. Experimental Procedure

Measurements were made on the equipment and by the procedures described in previous publications [1, 2].[1] Briefly, the "thermal conductivity cell" was a concentric cylinder type, used with its axis vertical. It was made of silver, and consisted of an "emitter," "guard," "receiver," and "receiver extension." The emitter was a cylinder about 2.2 cm in diameter and 11.4 cm long. It was surrounded by the receiver except at its upper end, and was held in place by spacing pins so that a uniform "conductivity gap" of 0.068 cm was attained. The guard and receiver extension comprised an extension of the emitter and receiver at the upper end of the cell and extended the cell geometry adjacent to them.

The power input was determined by potentiometric measurements of the emf's across two voltage dividers, one to evaluate the potential drop across the heater and the other to evaluate the potential drop across a standard resistor. The temperature difference between the emitter and the receiver was determined from the measured emf's of two Chromel–P Alumel difference couples, one pair of junctions near the top and one pair about midway of the length of the emitter.

Electrical power was supplied to the emitter heater, usually in such amounts as to produce differential thermocouple emf's of 50, 100, and 200 μV. However, whenever the temperature rise was so large that turbulent convection was detected, the measurements were made with less power. For helium, the available power was only sufficient to heat the emitter to a temperature that produced an emf of 50 μV on the difference thermocouples.

The observed quantities were the ambient temperature, measured by a calibrated platinum resistance

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[1] Figures in brackets indicate the literature references at the end of this paper.

thermometer, the pressure measured by a deadweight gage and emf's, as follows: (1) a value of the emf of a voltage divider for a standard resistor, V_{std}, (2) a value of the emf of a voltage divider for the emitter heater, V_{em}, (3) a value of the emf of the upper emitter-receiver Chromel–P Alumel difference couple and (4) a value of the emf of the middle emitter-receiver Chromel–P Alumel difference couple $\Delta V_{\Delta t}$. The two difference couple readings were averaged to give $\Delta V_{\overline{\Delta t}}$, and the "apparent conductivity" observed is then

$$K = q/(C\Delta t) = (V_{std}V_{em}/\Delta V_{\overline{\Delta t}}) \times (D\, dE/dt) = \psi\, (D\, dE/dt). \qquad (1)$$

The quantity D is a product of calibration constants and the thermal expansion of silver and dE/dt is the thermoelectric power of the thermocouples. In general for each level of power input, two groups of emf's were measured, each group consisting of three complete sets. The temperature of the bath was adjusted according to the power input so that the average gas temperature in the conductivity gap was constant. The values of the ratio $\psi = V_{std} \times V_{em}/\Delta V_{\overline{\Delta t}}$, plotted versus the average thermocouple emf's, $(\Delta V_{\overline{\Delta t}})$, were extrapolated so the intercept, ψ_0, represented a ratio free of heat transfer by convection [1].

2.1. Argon

Argon gas was introduced into the cell directly from a tank of "lamp grade" gas (stated to be more than 99.99% pure). The six sets of emf's at a power level sufficient to produce about a 200 μV thermocouple emf are presented in the following table as a typical example:

TABLE 1. *Sample data for* $Ar_{(g)}$ *at* 0 °C *and* 1×10^5 *Pa pressure*

		Emitter–receiver			
(1)	(2)	(3)	(4)		
V_{std} (μV)	V_{em} (μV)	Upper $\Delta V_{\Delta t}$ (μV)	Middle $\Delta V_{\Delta t}$ (μV)	Average $\Delta V_{\overline{\Delta t}}$ (μV)	ψ (μV)
3467.2	4217.9	197.19	197.61	197.40	7.4085
3460.6	4209.8	196.17	196.32	196.25	7.4236
3458.3	4206.1	195.85	195.98	195.92	7.4244
3502.6	4261.6	200.44	200.66	200.55	7.4430
3503.2	4262.3	200.60	200.87	200.73	7.4387
3503.3	4262.4	200.74	200.99	200.87	7.4339

The comparison of ψ for Ar at 0 °C at each power level showed no dependence on temperature rise (i.e., no detectable transfer of heat by convection), so that ψ_0 was obtained by averaging the results.

$$10^{-4}\psi_0 = 1/3(7.4285 + 7.4247 + 7.4340) = 7.4291\ \mu\text{V}$$

with an estimate of the standard deviation of ±0.15 percent.

The extrapolated values of ψ for argon at 25 °C and 1 atm pressure were obtained from the equation

$$\psi = \psi_0 + A\ \Delta V_{\overline{\Delta t}}$$

with the following values of constants resulting from a least squares solution:

$$\psi_0 = 7.78717 \times 10^4\ \mu\text{V}$$

$$A = 23.96.$$

The estimate of the standard deviation from the straight line is $S = 70\ \mu$V.

2.2. Helium

Helium gas, stated to be more than 99.99 percent pure, was used directly from the tank. The measurements of helium were carried out with one-fourth the temperature rises used for Ar, and showed no significant dependence on the size of the temperature gradient. The thermal conductivity of helium was measured in four states: At 2.39×10^5 Pa and 0.00 °C, at 1.01×10^7 Pa and 0.00 °C, at 2.39×10^5 Pa and 75 °C, and at 1.01×10^7 Pa and 75 °C. The measurements at 2.39×10^5 Pa pressure and average gas temperature of 75.00 °C gave clear evidence of progressive contamination of the helium by desorption from the apparatus, and it never became possible to repeat them. Hence there are only three valid values of ψ_0. It was expected that the absolute uncertainty in the emitter-receiver difference couples remained the same for all measurements, and was of marginal significance for the usual range. However, the temperature rise possible was only one-fourth that used for Ar and N_2, and so the higher $\Delta V_{\Delta t}$ values should have the higher relative accuracy. The averages were therefore weighted according to the temperature rise.

At 2.39×10^5 Pa pressure and 0.00 °C,

$$10^{-5}\psi_0 = 1/7\{4\ (6.30376) + 2\ (6.30357) + 6.28868\} = 6.30151$$

with an estimate of the standard deviation of 0.15 percent.

At 1.01×10^7 Pa pressure and 0.00 °C,

$$10^{-5}\psi_0 = 1/7\{4\ (6.48407) + 2\ (6.48556) + 6.52213\} = 6.48993$$

with an estimate of the standard deviation of 0.15 percent.

At 1.01×10^7 Pa pressure and 75.00 °C,

$$10^{-5}\psi_0 = 1/7\{4\ (7.28658) + 2\ (7.28520) + 7.25989\} = 7.28794$$

with an estimate of the standard deviation of 0.15 percent.

2.3 Nitrogen

Nitrogen gas was introduced from the tank directly into the thermal conductivity apparatus. The thermal conductivities of nitrogen gas were determined for 1×10^5 and $1.01=10^7$ Pa pressure at 9.60 °C, and for 1×10^5, 1.01×10^7 and 2.53×10^7 Pa pressure at 75 °C. The temperature rise of the emitter was large enough to produce emf's of the difference thermocouples as large as 200 μV (about 5 °C), and some dependence on temperature rise was observed at all pressures. The extrapolated values of χ were calculated by the method of least squares for the equation

$$\psi=\psi_0+A\ \Delta V_{\overline{\Delta t}}$$

with the following results:

TABLE 2. *Constants for nitrogen equation by least-squares determination*

t	10^5 Pa	ψ_0	A	S
9.60	1	109,567	2.5775	159
9.60	101	134,307	13.452	99
75.00	1	126,009	0.5888	119
75.00	101	143,527	6.9634	61
75.00	253	176,387	18.983	182

The quantity S is the estimate of the standard deviation from the straight line.

The thermal conductivities were determined by calculations, first of "apparent thermal conductivities," and then of "net conductivities." The apparent conductivity is given by eq (1) where

$$D=X/(C_{23}\cdot\phi(t)\cdot R_{\mathrm{std}})=5.534_7\times 10^{-9}/\phi\ (t)\ \mathrm{w\ m^{-1}K^{-1}},$$

with X the product of the voltage divider factors, C_{23} the cell constant at 23 °C and R_{std} the value of the standard resistance. The relative thermal expansion of silver is

$$\phi(t)=1/l_{23}\ \Delta l/\Delta t$$

$$=1+0.1862\times 10^{-4}\ (t-23\ {}^\circ\mathrm{C})+7.4\times 10^{-9}\ (t-23\ {}^\circ\mathrm{C})^2.$$

The thermoelectric power, dE/dt, is the calibrated emf for the Chromel–P Alumel thermocouples in μV/K, and has been reported previously [2].

The net or true conductivity is $K=K_{\mathrm{app}}-K_{\mathrm{vac}}+\Sigma\delta K$, where K_{vac} accounts for the power transferred in a vacuum and $\Sigma\delta K$ consists of 3 relatively small corrections:

(1) fK_{app}, to account for the assymetric temperature distribution of the emitter.

(2) $-7\times 10^{-4}\ K_{\mathrm{app}}$, to account for the change of heat flow through the pins with increasing conductivity of the medium in the conduction gap.

(3) $0.02556\ (K_{\mathrm{app}})^2$, to account for the temperature differences arising in the metal body of the cell between the thermocouple junctions and the conduction gap, for both the emitter and the receiver.

The values of the factors and the net conductivities are given in table 3.

TABLE 3. *Summary of thermal conductivity calculations* K *in units of w m^{-1} K^{-1}*

t °C	10^5 P (Pa)	ψ_0 (μV)	10^9 D	dE/dt	10^2 K_{app}	10^2 K_{vac}	f	$0.02556\times K_{\mathrm{app}}$	$\frac{\Sigma\delta K}{K_{\mathrm{app}}}$	10^2 K	kgρ/m^3
					Argon						
0.00	1.01	74291	5.537	41.51	1.707_4	0.0578	0.0010	0.0004	0.0007	1.651	1.78
25.00	1.01	77872	5.535	42.43	1.828_7	.0645	.0010	.0004	.0007	1.765	1.64
					Helium						
0.00	2.39	630151	5.537	41.51	14.48_6	0.0578	0.0081	0.0037	0.0111	14.59*	0.421
0.00	101.3	648993	5.537	41.51	14.91_7	.0578	.0083	.0038	.0114	15.03	17.86
75.00	101.3	728794	5.529	43.22	17.41_7	.0850	.0095	.0044	.0132	17.56	14.01
					Nitrogen						
9.60	1.01	109567	5.536	41.96	2.545_2	0.0611	0.0016	0.0006	0.0015	2.488	1.2
9.60	101.3	134307	5.536	41.96	3.119_9	.0611	.0019	.0008	.0020	3.065	121.5
75.00	1.01	126009	5.529	43.22	3.011_4	.0850	.0018	.0007	.0018	2.932	0.98
75.00	101.3	143527	5.529	43.22	3.430_0	.0850	.0021	.0009	.0023	3.353	95.4
75.00	253.3	176387	5.529	43.22	4.215_3	.0850	.0025	.0011	.0129	4.143	217.0

*Corrected for temperature jump, $K=14.61\times 10^{-2}$ w m^{-1} K^{-1}.

The uncertainty of the results at the 99 percent confidence level is estimated to be ±0.7 percent and is derived as the root of the sum of the squares of the estimates of the 3 σ errors of the following:

Estimate of the error in the value of the

Cell constant	$3\ \sigma=\pm 0.1\ \%$
Emitter power	$3\ \sigma=\pm 0.1\ \%$
Extrapolation to ψ_0	$3\ \sigma=\pm 0.45\%$
Temperature rise	$3\ \sigma=\pm 0.45\%$
"Vacuum conductivity"	$3\ \sigma=+0.004\times 10^{-2}$ w m^{-1} K^{-1}

3. Discussion

According to the kinetic theory of gases, the ratio $f=K/(\eta c_v)$, should be [3]

$$f=2.5f_\lambda^{(3)}/f_\eta^{(3)}$$

for a monatomic gas. The value of $f_\lambda^{(3)}/f_\eta^{(3)}$ depends upon the potential function of the particular atom and is only about 3×10^{-3} greater than unity for helium. For the value of helium reported for 0 °C and 2.39×10^5 Pa, adjusted to zero pressure, and for the heat capacity at constant volume of 3/2 R per mol, the value of f is

$$f=14.60\times10^{-2}\times0.0040026/(1.86885\times10^{-5}\times8.3143\times1.5)=2.5073.$$

The value of the viscosity was derived from publications by Kestin et al. [5, 6, 7], and the value of the gas constant was taken from an evaluation of the fundamental constants [8]. The calculated value of the ratio, where the values of $f_\lambda^{(3)}$ and $f_\eta^{(3)}$ are derived for the same potential parameter given for the viscosity, is 2.5085. Thus the values from experiment and theory are in agreement within 0.05 percent, much less than the experimental uncertainties.

For many gases the increase of the thermal conductivity with pressure is principally a function of density. The increase found for helium at 0 °C is $\partial K/\partial\rho=2.41\times10^{-4}$ in the units of table 3. When the pressure effect is deducted from the thermal conductivity value found for helium at 75.00 °C and 1.01×10^7 Pa pressure, the value of K for the same temperature and zero pressure is 17.22×10^{-2} w m^{-1} K^{-1}. Kestin's experimental value for the viscosity of helium at 75.00 °C is 22.07 kg m^{-1} s^{-1}, so that $f=2.5042$, for the derived value of helium, an agreement with the theoretical value of 2.5085 that must be somewhat fortuitous.

The author suggests that most thermal conductivity values for helium are low because of contamination by degassing of the apparatus. Such an effect would diminish with pressure, and might be confused with "temperature jump" effects. The length of the temperature jump is [9]

$$g=\frac{2-a}{a}\frac{4c}{\gamma+1}\frac{K}{\eta c_v}L$$

where a is the thermal accommodation coefficient, $0.491\leqslant c\leqslant0.499$, $\gamma=c_p/c_v$, and L is the mean free path. When a is taken to be 0.37 [10], the value of g is about 8 L. At 10^5 Pa pressure and 0 °C, $L=12\times10^{-6}$ cm, so that the relative effect of the temperature jump on the heat transfer in the M. I. T. cell amounts to 2.8×10^{-3}. Probably the uncertainty of the theory of the temperature jump should be assumed to be 20 or 25 percent. A correction to the conductivity of helium calculated for the effects of temperature jump at 1 atm involves too much uncertainty, hence the choice of 2.39×10^5 Pa, at which pressure the reduction of heat transfer from temperature jump in the cell is calculated to be 1.2 parts per thousand. On the other hand, for both nitrogen and argon the effects of temperature jump at one atmosphere pressure and 0 °C are calculated to be less than 0.5 part per thousand.

A similar calculation of $f=K/(\eta c_v)$ for argon gave the value of 2.506 at 0 and 2.500 at 25 °C, when the values of the viscosities were derived from the work of DiPippo and Kestin [7]. The viscosity values used were 21.10 kg m^{-1} s^{-1} at 0 °C and 22.21 kg m^{-1} s^{-1} at 25 °C. The value of $f_\lambda^{(3)}/f_\eta^{(3)}$ for argon is about 1.001 in this temperature range, so that the theoretical value of f is 2.503. The viscosity results published by Kestin et al. are likely to be so accurate that the discrepancies between experimentally and theoretically derived values of f will come predominantly from errors in the measurement of the thermal conductivities. The agreement of the experimentally derived values of f with the theoretically derived values is within 0.12 percent, distinctly less than the uncertainties to be expected for the most accurate measurements of thermal conductivities.

The determination of the thermal conductivities of gases at high densities is apt to be erroneously high because of heat transfer by convection. It has been expected that the parallel plate cell, with the conductivity gap horizontal and the emitter plate above the receiver, would be free of the effects of convection. The accurate performance of a thermal conductivity cell in this configuration has been attained by Michels and Sengers [11]. Heat transfer by convection was expected for our coaxial cylinder cell. The measurements were made as a function of temperature rise so that the resulting data could be extrapolated to zero temperature rise; the effects of convection, so long as it was entirely laminar, could thereby be eliminated. Heat transfer by laminar convection in a vertical coaxial cylinder cell will not significantly involve the emitter if the conductivity gap is sufficiently extended in each direction by long guards. Such equipment has been used by Ziebland [12], and Johannin, Le Neindre and other workers at the Laboratoire des Hautes Pressions of the CNRS at Bellevue, France [13, 14]. A large fraction of thermal conductivity measurements at high density, however, have been reported for equipment for which neither adequate experimental precautions nor appropriate treatment of data were used.

At low densities, the thermal conductivity of nitrogen has been accurately measured also by Nuttall and Ginnings [15], and Michels and Botzen [16]. Within the stated experimental errors, their values and the low density values reported in this paper are in agreement.

The additional thermal conductivity of nitrogen due to pressure, the "excess thermal conductivity", appears to be a function of density only. The data for figure 1 are given in table 4, which covers measurements from 0 to 700 °C, and pressures up to 1.3×10^8 Pa (1284 atm).

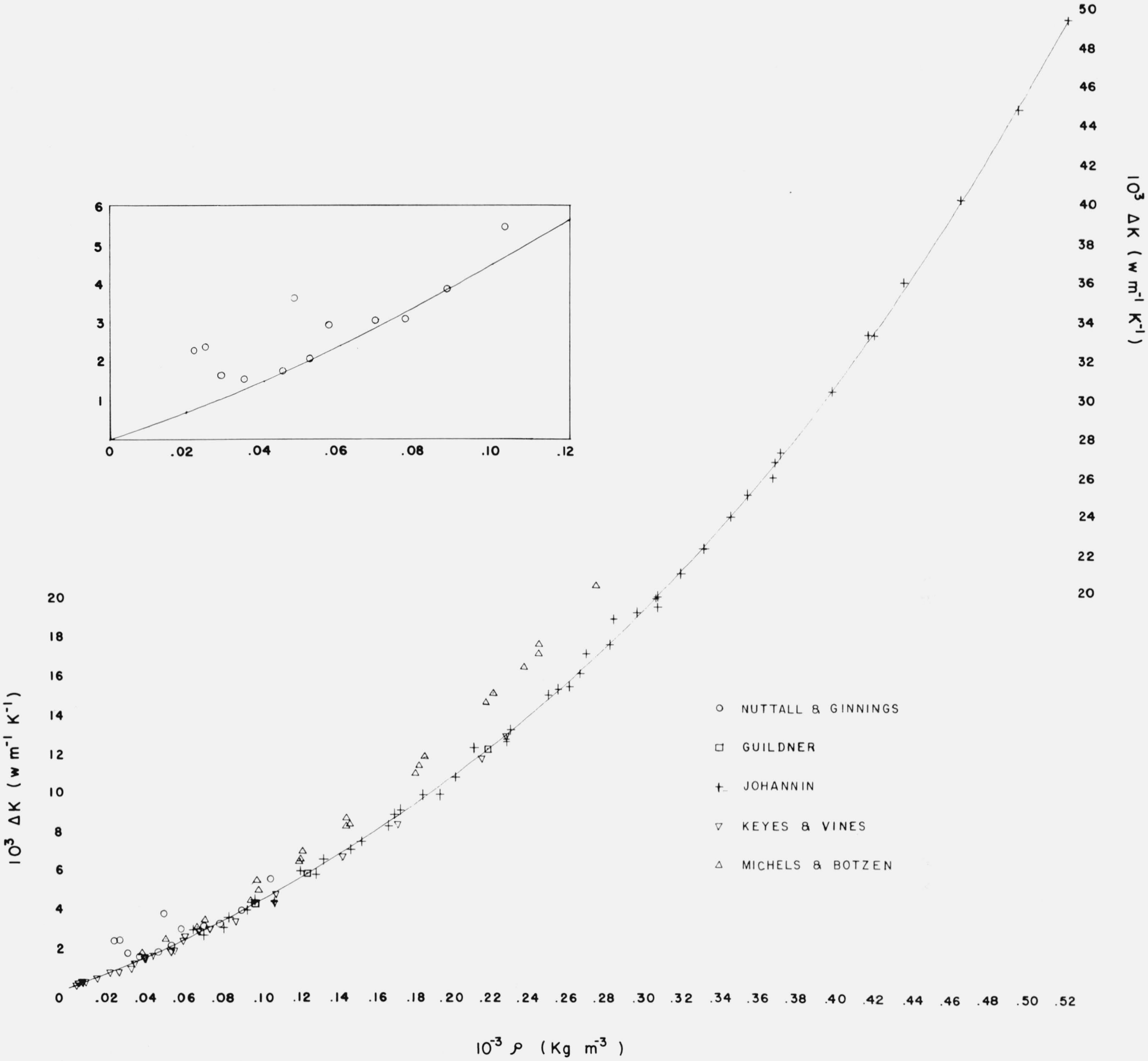

FIGURE 1. *The excess thermal conductivity of nitrogen versus its density.*

The data are obtained from various papers reporting measurements made at temperatures from 0 to 700 °C, and at pressures from 1×10^5 to 1.3×10^8 Pa.

TABLE 4. *The excess thermal conductivity of nitrogen at higher pressures*

Guildner

10^{-5} P (Pa)	10^{-3} ρ (kg/m^3)	10^3 ΔK (w m^{-1} K^{-1})
9.6 °C		
101.3	0.122	5.77
75 °C		
101.3	0.0954	4.21
253.3	.217	12.11

Michels and Botzen

25 °C			50 °C		
P	ρ	ΔK	P	ρ	ΔK
1	0.00114	0	1	0.00106	0
10.5	.0119	0.5	12.4	.0129	.6
21.8	.0217	0.9	34.2	.0356	1.5
33.0	.0367	1.7	63.4	.0656	3.0
43.3	.0491	2.4	94.3	.0969	4.9
61.6	.0697	3.4	118.0	.120	6.9
85.0	.0960	5.4	142.6	.143	8.6
106.1	.119	6.5	188.0	.184	11.8
128.6	.143	8.2	231.1	.220	15.0
163.2	.179	10.9	262.1	.244	17.5
202.7	.216	14.5			
224.1	.236	16.3			
270.0	.274	20.5			

75 °C		
1	0.00098	0
12.6	.0121	.4
37.1	.0356	1.3
74.2	.0705	3.0
99.1	.0934	4.4
127.1	.118	6.4
157.8	.145	8.3
202.5	.181	11.3
244.4	.216	14.5
258.4	.244	17.0

Nuttall and Ginnings

	10^{-5} P = 50.6 Pa		10^{-5} P = 101.3 Pa	
t °C	ρ	ΔK	ρ	ΔK
50	0.052	2.09	0.103	5.44
100	.045	1.76	.088	3.85
150			.077	3.10
200	.035	1.55	.069	3.06
300	.029	1.67	.057	2.97
400	.025	2.39	.048	3.64
500	.022	2.30		

Values of the densities were derived from numerous *PVT* data, and are probably not in error more than ±1 percent. Most of the values of ΔK are those published by Keyes and Vines [17].

P. Johannin

75 °C			125 °C		
P	ρ	ΔK	P	ρ	ΔK
102	0.095	4.4	100	0.081	3.5
181	.171	9.0	196	.151	7.4
390	.305	19.8	390	.266	16.0
588	.397	30.4	588	.353	25.1
782	.464	40.2	782	.419	33.3
977	.520	49.4	1039	.494	44.8
200 °C			300 °C		
101	0.069	2.6	197	0.105	4.3
198	.127	5.7	396	.192	9.8
395	.227	12.6	588	.260	15.3
593	.306	19.4	782	.318	21.1
782	.366	26.0	977	.367	26.8
977	.416	33.3	1301	.434	36.0
400 °C			500 °C		
198	0.091	3.9	196	0.079	3.0
391	.165	8.2	390	.145	7.0
584	.227	12.6	577	.200	10.7
780	.281	17.5	787	.254	15.2
984	.330	22.4	1024	.306	19.9
1172	.370	27.3	1217	.344	24.0
600 °C			700 °C		
197	0.070	3.0	197	0.063	2.9
392	.131	6.5	395	.119	5.9
588	.183	9.8	590	.168	8.8
782	.229	13.1	783	.210	12.2
973	.269	17.0	978	.249	14.9
1111	.295	19.1	1173	.283	18.8

Keyes and Vines [17]

139.70 °C			249.40 °C		
P	ρ	ΔK	P	ρ	ΔK
4.48	0.0036	0.17	3.45	0.0022	0.04
6.89	.0056	0.29	20.68	.0132	0.42
37.57	.0303	1.21	49.49	.0312	0.84
47.57	.0382	1.55	83.42	.0518	1.76
51.36	.0412	1.59	86.18	.0534	1.84
73.77	.0585	2.39	139.9	.0846	3.26
75.15	.0596	2.51	177.5	.105	4.27
139.2	.106	4.73	303.4	.170	8.29
190.3	.141	6.62	434.4	.227	12.81
317.1	.214	11.64			
196.75 °C			300.90 °C		
7.58	0.0055	0.13	12.07	0.007	0.21
34.14	.0245	0.67	35.16	.020	0.71
			68.94	.039	1.55
			131.0	.072	2.89
347.60 °C					
13.45	0.007	0.29			
34.45	.018	0.75			
69.63	.037	1.55			
129.3	.066	2.93			

The values reported for higher densities in [15] and [16] are inconsistent with other data. The satisfactory correlation of the Keyes and Vines data tends to substantiate that the work and treatment of data may have been satisfactory, but adequate information is lacking in the paper itself.

Good measurements of the thermal conductivity of argon were made by Michels et al [18]. The values at 0 and 25 °C for a pressure of 1×10^5 Pa atmosphere are 1.633×10^{-2} w m^{-1} K^{-1} and 1.758×10^{-2} w m^{-1} K^{-1}, differing from the results in this paper by -1.1 percent and -0.4 percent, respectively. The increase of thermal conductivity at 0, 25, 50, and 75 °C for pressure increases up to 2.46×10^8 Pa (2424 atm) can be satisfactorily correlated as a function of density only.

4. Conclusions

When careful work is combined with well-designed equipment, the thermal conductivities of gases can be determined with an uncertainty of less than 1 percent even at high pressures. For the noble gases, the thermal conductivities measured for He and Ar at low density are entirely consistent with the predictions of the kinetic theory, and similar agreement can be expected for Ne, Kr, and Xe. The increase of the thermal conductivity as the gas density increases is found to be a function of density only for nitrogen in the range of 0–700 °C and up to 1.301×10^8 Pa (1284 atm) pressure.

Note added in proof:

A trade name is used in this manuscript. In no case does such identification imply recommendation or endorsement by the National Bureau of Standards.

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