THERMAL CONDUCTIVITY OF COMMON NON-POLAR POLYATOMIC GASES

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The data of pure non-polar polyatomic gases as a function of temperature has been reviewed to provide the best available values of thermal conductivity to experimentalists and theoreticians. The gases considered in detail are O_2, N_2, H_2, CO_2, CO, NO, N_2O, CH_4 and D_2 for which several sets of measured values are available. Smooth average values are tabulated at an interval of 25°C to permit linear interpolation.

In many practical design problems of great importance such as energy control in nuclear reactors, controlled combustion and explosion problems, design of outer space exploration vehicles, the knowledge of thermal conductivity, λ, of pure gases and their multi-component mixtures is essential. It is also an important piece of information in a variety of very simple problems dealing with the transfer and exchange of thermal energy as well as in many theoretical problems relating to the understanding of molecular structure of complex molecules as regards the energy equilibration between various internal and external modes. The availability of experimental data of monoatomic gases and their mixtures was reported in a comprehensive article by Gandhi and Saxena[1]. The lack of enough experimental data have encouraged to the development of many approximate, empirical and semi-empirical methods of prediction, and these are compared and contrasted recently by Saxena and Gandhi[2, 3]. A number of workers[4–10] have also given the methods for calculating the thermal conductivity of polyatomic gases and their mixtures. These approaches fundamentally suffer for want of an adequate physical picture of the process involved in the energy balance of the various modes of the polyatomic molecules. To aid theoreticians to achieve this goal, it is imperative and to some extent obligatory for the experimentalists to evolve adequate and reliable experimental data of the required type. This section is intended in this general direction with a limited and less ambitious aim and plans to review the situation of the experimental data on the thermal conductivity of pure non-polar polyatomic gases. As will be seen in certain cases a number of workers have reported the experimental data and thus this study willl throw light on their relative appropriateness and will tend to evolve the best values available at the moment. This work will also bring to lime-light the obvious contradictions in the available data and at a few places their complete absence, which will beacon the plan for new measurements. We have a programme of measurement of thermal conductivity of gases and gaseous mixtures as a function of temperature, T, going on in this laboratory and we hope to bridge at least some of these deficiencies in due course.

EXPERIMENTAL DATA

Elaborate thermal conductivity data available for many pure polyatomic gases such as O_2, N_2, H_2, CO_2, CO, NO, N_2O, CH_4 and D_2 is considered in detail here as a function of temperature.

Oxygen

The thermal conductivity data of this gas reported by a number of workers[11-31] in the temperature range -200 to 1100°C is plotted in Fig. 1. Some data could not be shown in this figure because of overlapping. The data of Geier & Schäfer[29] and Westenberg and de Hass[30] for temperature higher than 550°C are also not shown in this figure. However, these data agree well with each other and with the values of other measurements in the overlapping temperature range. The high temperature values smoothly join the low temperature curve. The data of Westenberg and de Hass[30] were obtained according to a line-source technique[32] which falls in the broad category of the dynamic methods. Ibbs & Hirst[15] measurements are according to a Katharometer of the type first given by Daynes & Shakespear[33]. The familiar parallel plate method was exploited by Todd[11] but this value is apparently about 16 per cent smaller than the smooth value of the other workers. No weight has been given to this value while recommending the smooth values of thermal conductivity as a function of temperature in Table 1. Further, the λ values in the temperature range 550 to 1100°C are based on the measurements of Geier & Schäfer[29], and Westenberg & de Hass[30]. The co-axial cylinder type of thermal conductivity cell for measurements was used by Keyes[22, 23], Waelbroeck & Zuckerbrodt[25], and Cheung *et al*[27]. Kannuluik & Martin[17], and Srivastava *et al.*[26], [28] have employed the thick-wire variant of the hot-wire cell, while Weber[13], Johnston & Grilly[20], and Pereira & Raw[31] have used the potential-lead method very often referred to as the thin-wire variant of the hot-wire cell. A group of workers[14, 16], on the other hand, have preferred to use the compensating-cell method first introduced by Gregory & Archer[34] who used two conductivity cells of different lengths in the two arms of a wheatstone bridge and it was hoped that the measurements so obtained related to the central portion of the longer cell where radial conditions of heat flow existed.

Fig. 1 clearly bears out a marked degree of consistency between the data of different workers except Todd[11]. It does not seem possible to discover anything regarding the relative accuracies of the different methods ; the values agree among themselves within a

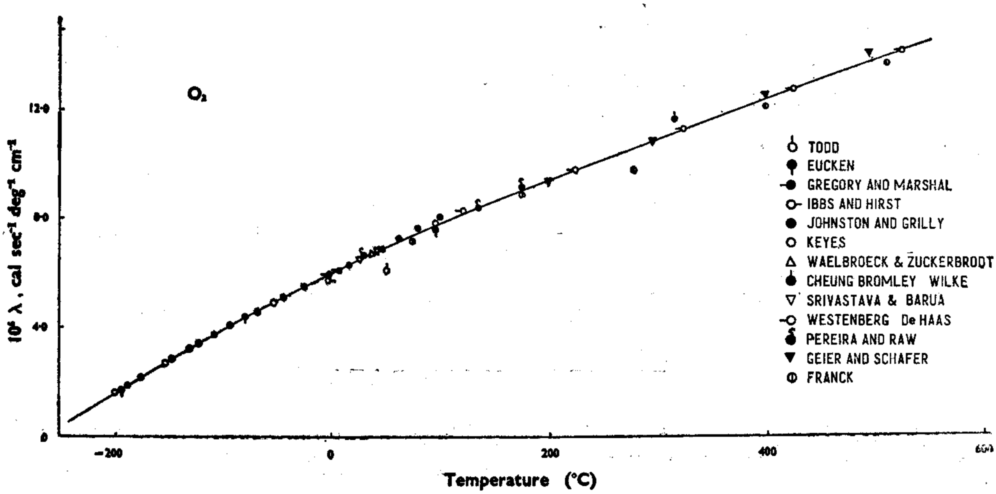

Fig. 1—Thermal conductivity of O_2 as a function of temperature

maximum deviation of about 4 per cent except Todd's[11] single value and Franck's[21] value at 280°C. Mostly the deviation is much less than this and is, thus, within the precision of the individual methods. We suggest that λ measurements above 200°C should be repeated employing various methods to have a check on the values of Franck[21], Geier & Schäfer[29], and Westenberg & de Haas[30].

Nitrogen

It is one of those gases which have been studied most extensively and the data of a large number of workers[11, 14-16, 18,21-23, 26, 27, 29, 31, 35-56] is plotted in Fig. 2 in the temperature range —200 to 900°C. Certain comments in this connection are important. Earlier values of Keyes & Sandell[40] as revised by Keyes[22, 23, 41] are in good agreement with other values. Todd's[11] value is again less by about 16 per cent as in O_2 but the measurements of Michels & Botzen[45], and Nuttal and Ginnings[49], obtained also by using parallel plate apparatus, are in good agreement with the other measurements. This indicates that something was particularly wrong in the measurements made by Todd[11] although his technique seems to be alright in general. Discrepancies in the thermal conductivity values at few temperatures are quite pronounced. The only Ibbs & Hirst[15] values at 0°C are left which differ from others by 4-5 per cent, seem to have poor precision as evidenced by their two measurements. Further, even the katharometer method used by them is open to objection[57]. However, all the measurements viewed in this context can be regarded as consistent with each other upto 300°C within experimental accuracy. At and above this temperature there are some interesting points to note. Measurements of Johannin & Vodar[50], Vines[54], Keyes [22, 23, 41] and Rothman & Bromley[48] are all on the co-axial cylinders type cell and though these individual measurements have a high degree of self consistency, they offer very systematic discrepancies and trends among themselves and differ by 3-4 per cent in value. Vines[54] values, like those of Rothman & Bromley[48], are also over an equally extensive temperature range but curiously enough these are systematically higher by 3-4 per cent. This is important when we recall that both groups have used similar conductivity cells. Westenberg & de Hass[56] values have poor precision. Schäfer &

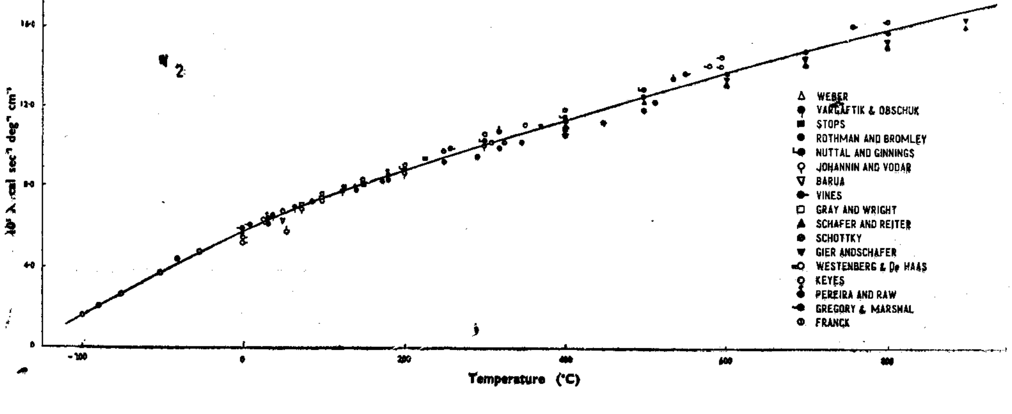

FIG. 2—Thermal conductivity of N_2 as a function of temperature

Reiter[51], and Geier & Schäfer[29] have measured thermal conductivity from 0 to 1100°C. At lower temperatures their values are consistent with other workers but at higher temperatures these are consistently lower. Values of Schottky[43] and Frank[21] are consistently smaller in the temperature range 300 to 500°C. Thus the need for more precise measurements for this gas in this temperature range is paramount; and further, it will be interesting to exploit the hot-wire type of cell as well. We plan to report such measurements from our laboratory. The values upto 900°C reported in the Table 1 are used on the compromise of the existing data; while at the two higher temperatures these are of Geier & Schäfer[29].

Hydrogen

The thermal conductivity of hydrogen is measured by a number of workers[11-13, 15-20, 22, 25, 26, 29, 34, 35, 39, 42, 55, 58-70] and all these data are plotted in Fig. 3. Unfortunately, the various sets of values are not in good agreement with each other specially above 0°C. The data of Gregory[63] obtained by the compensating-cell method seem to be systematically greater than those of Keyes[22, 68] who employed a co-axial cylinder type of cell. The latter values are, however, consistent to a good extent with the values of the other workers obtained with different types of cells, viz. parallel plates, co-axial tubes, and hot-wire. Dickens[16], and Gregory & Dock[64], who also employed the compensating type of cells, found the thermal conductivity values smaller than those of Gregory[63]. It is strongly felt that Gregory's[63] values are unreliable for this gas and hence no weight is given to this data while reporting average values in Table 1.

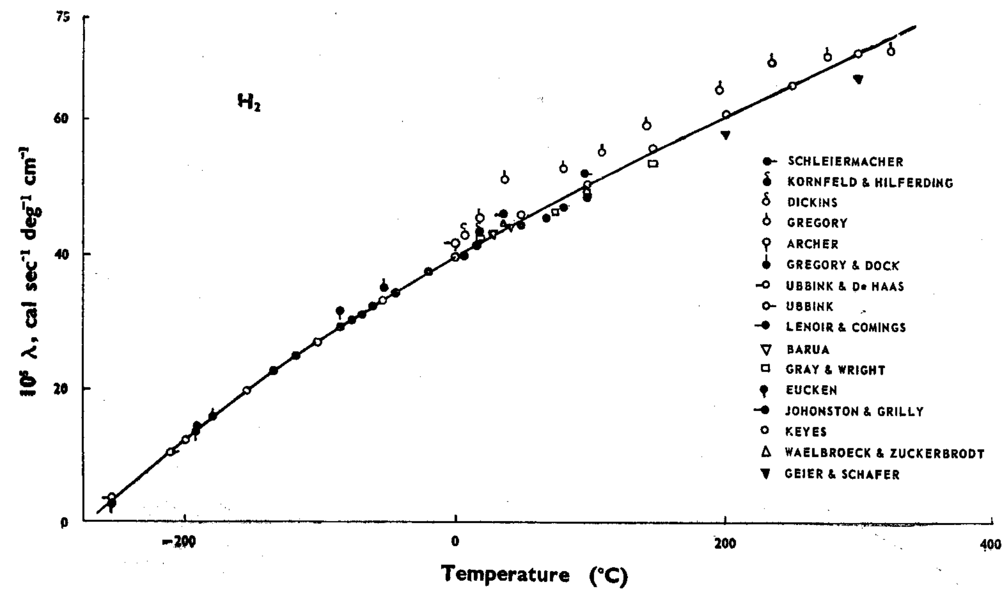

FIG. 3—Thermal conductivity of H_2 as a function of temperature

TABLE 1

SMOOTHED VALUES OF THERMAL CONDUCTIVITY, λ(CAL. SEC.$^{-1}$ DEG.$^{-1}$ CM^{-1}), OF COMMON NON-POLAR POLYATOMIC GASES AS A FUNCTION OF TEMPERATURE

Temp. °C	$10^5 \lambda$								
	H_2	N_2	O_2	CO_2	CO_2	CH_4	NO	N_2O	D_2
—250	3·50								3·62
—225	7·55								7·26
—200	11·5	1·62	1·57		1·48	1·93			10·6
—175	15·6	2·16	2·13		2·03	2·53			13·7
—150	19·5	2·71	2·69		2·59	3·14	2·59	1·21	16·4
—125	23·0	3·24	3·26		3·13	3·78	3·14	1·56	18·9
—100	26·5	3·73	3·83	1·96	3·61	4·45	3·66	1·91	21·4
—75	30·0	4·20	4·35	2·30	4·11	5·13	4·21	2·29	23·7
—50	33·2	4·68	4·86	2·66	4·60	5·79	4·74	2·69	26·0
—25	36·5	5·14	5·39	3·05	5·08	6·53	5·23	3·12	28·1
0	39·5	5·62	5·87	3·46	5·53	7·34	5·70	3·60	30·2
25	42·6	6·08	6·33	3·88	5·98	8·14	6·16	4·11	
50	45·5	6·50	6·81	4·28	6·42	9·03	6·62	4·60	
75	48·2	6·91	7·27	4·72	6·86	9·98	7·08	5·15	
100	50·8	7·31	7·68	5·18	7·28	10·9	7·52	5·70	
125	53·5	7·64	8·12	5·67		11·7	7·92		
150	56·1	8·03	8·51	6·14		12·7	8·33		
175	58·7	8·37	8·90	6·62		13·6	8·76		
200	61·1	8·74	9·24	7·12	8·72	14·6	9·16		
225	63·5	9·10	9·67	7·64		15·6	9·53		
250	65·9	9·44	10·1	8·18		16·6	9·91		
275	68·3	9·78	10·5	8·72		17·7	10·3		
300	70·5	10·1	10·8	9·26	10·1	18·8	10·7		
325		10·4	11·2	9·78		19·8			
350		10·8	11·6	10·3		20·9			
375		11·1	12·0	10·7		22·0			
400	75·6	11·4	12·3	11·2	11·5	23·0			
425		11·7	12·6	11·6		24·0			
450		12·0	13·0	12·0		25·1			
475		12·3	13·4	12·5		26·1			
500	83·7	12·6	13·7	12·9	12·8	27·1			
525		12·9	14·0	13·3					
550		13·2	14·4	13·7					
575		13·5	15·0	14·1					
600	92·0	13·8	15·3	14·4	13·9	33·2			
625		14·1	15·7	14·8					
650		14·4	16·0	15·2					
675		14·6	16·4	15·6					
700	100·7	14·9	16·7	15·9	15·1	39·0			
725		15·2	17·1	16·3					
750		15·4	17·4	16·7					
775		15·7	17·7	17·0					
800	108·2	15·9	18·1	17·4	16·3				
825		16·1	18·4	17·8					
850		16·4	18·7	18·2					
875		16·6	19·0	18·5					

TABLE 1—*contd.*

Temp °C	$10^5\lambda$								
	H_2	N_2	O_2	CO_2	CO	CH_4	NO	N_2O	D_2
900	116·5 (140·0)*	16·9	19·3	18·9	17·4				
925			19·6						
950			19·9						
975			20·2						
1000	124·0 (152·6)	17·2	20·5	19·2					
1025			20·8						
1050			21·1						
1075			21·4						
1100	131·5 (165·2)	17·9	21·7	20·3					
1200	138·5 (177·7)								

*Values within braces are those of Blais & Mann[71].

As already stated, discrepancies above 0°C are rather pronounced; and following trends are observed :

(*a*) Johnston & Grilly[20] values, obtained on the hot-wire cell, seem to yield somewhat smaller values than those of other workers though they are in good agreement with the values of Gray & Wright[55].

(*b*) The values of Geier & Schäfer[29] at 0 and 100°C are in good agreement with the data of other workers but at 200 and 300°C these are smaller. As agreement is obtained in other gases, atleast in this temperature range, nothing definite can be concluded about this trend. In Table 1, values reported above 300°C are of Geier & Schäfer[29].

(*c*) Blais & Mann[71] have used a hot-wire type of thermal diffusion column to measure thermal conductivity and have reported data between 1200 and 2000 °K. In the temperature range 1200-1473°K the only data of Geier & Schäfer[29] are available for comparison and are systematically low. Blais & Mann[71] values are as high as 20 per cent at 900°C and 29 per cent at 1200°C. As the data of Geier & Schäfer[29] for other gases in this temperature range agree well with the other available data, this comparison raises the possibility of Blais & Mann[71] data being higher than the actual values. This conclusion is in conformity with the similar suggestion made by Saxena & Agrawal[72] in connection with the data of helium. Therefore, we have excluded these data from our present survey. An effort is also, being made in this laboratory to establish this technique.

We have not included in our discussion the data of Vargaftik & Parfenov[73] as it is found to be systematically higher than the other available data[71,72]. Thus, on the whole, we find that there is an urgent need for reliable measurements for this gas above 0°C.

Carbon dioxide

It seems CO_2 has been fairly understood as it has been widely investigated and a large amount of data of different workers* are available on all the methods. These data are shown in Fig. 4. Johnston & Grilly[20], on the potential-lead method, measured the values in the temperature range—87 to 106°C, which are in good agreement with the values of Keyes[22,23,41] obtained on the co-axial cylinders method in the range—50 to 350°C. There are also a number of additional measurements in this range at a few temperatures but the agreement is not very satisfactory. The scatter of data at 0°C, 125°C and 225°C is approximately 8, 7 and 13 per cent respectively. It is worth noting that Todd's[11] value at 55°C, obtained on the parallel-plate apparatus is about 5% smaller. In addition to Johnston & Grilly[20], Sherrat & Griffiths[78] have also reported data in the temperature range 66 to 292°C on a similar conductivity cell and the two sets of measurements are in good agreement. Archer[77], using compensating-tube method, reported values in the temperature range of 12 to 319°C which though consistent among themselves, are in marked disagreement with the values of Johnston & Grilly[20], and Sherrat & Griffiths[78]. The disagreement also increases with temperature. There are four more sets of valuable measurements which extend beyond 300°C and go upto 1100°C. One is of Vines[54,80] from 0 to 900°C and the second is of Rothman & Bromley[48] from 0 to 800°C. The important point to note is that though the above two sets employ the same technique of co-axial cylinders there is no reasonable agreement between the values. The two individual sets are also not very smooth but when an attempt is made to draw a smooth curve, discrepancy becomes more pronounced at high temperatures; as an example it is about 5% at 800°C. The situation is somewhat similar to that of nitrogen. The third set is of Westenberg & de Haas[56] obtained by using the line-source technique[32]. The precision of this set is very poor as can be seen from two measurements around 775°C. The fourth set is of Geier & Schäfer[29] between 0 and 1100°C and is in good agreement with the values of Rothman & Bromley[48].

* 11—13, 15—18, 20—23, 25, 27, 29, 35 37—39, 41, 42, 44, 46 48, 51, 54, 56, 60, 62, 69, 74—81.

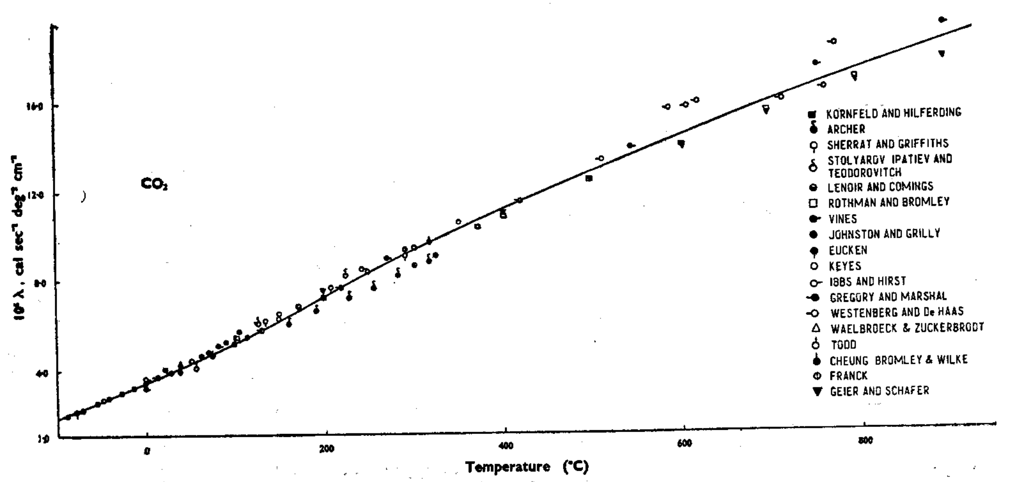

FIG. 4—Thermal conductivity of CO_2 as a function of temperature

Thus on the whole we find that at temperatures beyond 200°C the data are not satisfactory. By passing a smooth curve through the existing data we have given the compromise values upto 900°C in Table 1. In smoothing the data we have given relatively less weight to the measurements of Archer[77]. We have no particular reason to offer for this choice except the general trend of these values as regards temperature dependence though Franck's[21] values also agree with Archer's[77] measurements. The λ values at the highest two temperatures are of Geier & Schäfer[29].

This effort has brought to limelight the need for fresh measurements so that reliable and consistent values may be available. We further endeavour to suggest the use of the thick wire variant of the hot-wire method for this purpose. For information we may recall that one measurement at 0°C of Kannuluik *et al.*[17, 79] is available and this is in agreement with the average value at this temperature given by different workers.

The detailed thermal conductivity study of this gas is particularly interesting because of the internal modes of this molecule, which get readily excited at relatively lower temperatures. A critical look at λ versus T graph in Fig. 4 shows a change in its curvature at about 350°C. This, if confirmed by careful precise measurements, may lend a valuable source of information regarding the mechanism and ease of excitation of rotational modes. We feel that the rotational modes of this gas get readily excited and, therefore, after a certain temperature the rate of increase of λ with T slows down. Again at high temperatures, with the increasing participation of vibrational modes, the trend will exhibit a change.

Carbon monoxide

For carbon monoxide the relatively elaborate data are those of Johnston & Grilly[20], Keyes[22], and Geier & Schäfer[29]. The available data[11, 15–17, 20, 22, 29, 51, 60, 76, 81, 82] are plotted in Fig. 5. It is gratifying to note that these two sets [20, 22] of data, obtained on different techniques, are consistent with eacher other. Todd's[11] value at 55°C is in good agreement in

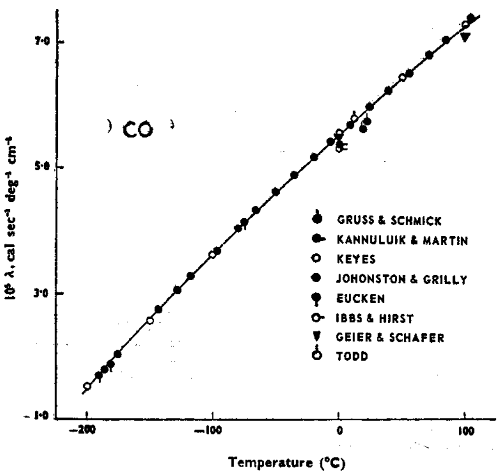

FIG. 5—Thermal conductivity of CO as a function of temperature.

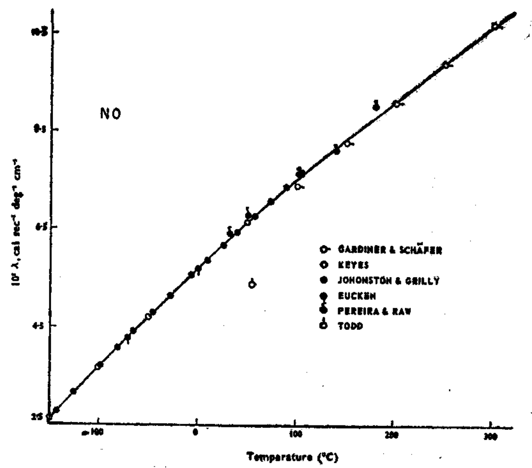

FIG. 6—Thermal conductivity of NO as a function of temperature.

this case in notable contradiction with all previous gases considered so far. At 0°C there are two more measurements[15, 17] and the maximum discrepancy assumes a magnitude as high as 5%. This, of course, is not very alarming for Ibbs & Hirst[15] values cannot given too much reliance though the other value[17] obtained on the thick-wire cell is also about 3% low. Gruss & Schmick's[83] values are also much lower. Thus, we find that firstly it will be interesting to repeat measurements in this range on the hot-wire type of cell with a view to establish the techniques and secondly to extend the temperature range of measurements beyond 100°C. Smooth values are reported in Table 1. In this Table values above 100°C are of Geier & Schäfer only.

Nitric oxide

We next consider the data on nitric oxide[11, 20, 22, 31, 60, 84] which is shown in Fig. 6. The situation is rather interesting and the main data which is due to three different groups of workers— Johnston & Grilly[20], Keyes[22], and Pereira & Raw[31] are in good agreement with each other. This lends rather good and pleasant support on the consistency in values obtained on cells of co-axial cylinder and thin-wire types. Another point worth mentioning and noting is the fact that Todd's[11] value is about 22% smaller than the smoothed value. Another value of Eucken[60] at − 71·5°C is also in good agreement. We, thus, recommend that the data on this gas be extended using a thick wire cell for reasons given in the case of discussion on CO gas. Smoothed values on the basis of Fig. 6 are given in Table 1.

Nitrous oxide

Keyes[22, 68], Johnston & Grilly[20], and Pereira & Raw[31] have reported somewhat elaborate data on N_2O and these are shown in Fig. 7 alongwith the other values[11, 13, 15–17, 51, 60, 79, 83] As in other gases, here also Keyes[22, 68] and Johnston & Grilly[20] data are in remarkable agreement though the data of Pereira & Raw[31] indicate a somewhat different trend as regards temperature dependence. Eucken's[60] value at −72°C is much higher. At 0°C, value of Kannuluik *et al.*[17, 79] agrees with the general smooth curve while the value of Ibbs & Hirst[15] is higher. We, however, do not weigh it in smoothing the data. We thus feel the necessity of extending the measurements beyond 100°C to clarify the discrepancy created by the Pereiro & Raw[31] measurements. Again, for reasons already mentioned, we feel hot-wire type of cell may be preferable to use. In reporting the smooth values in Table 1 we limit the reading to only 100°C as no weight has been given to the data of Pereira & Raw[31].

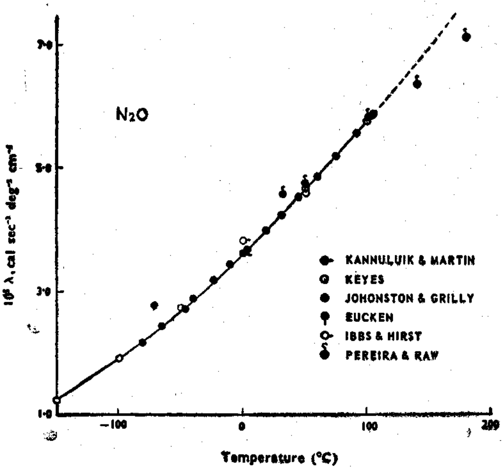

FIG. 7—Thermal conductivity of N_2O as a function of temperature

Methane

The only non-polar gas left which has been explored by a number of workers[13, 20, 22, 23, 27, 29, 39, 42, 43, 47, 60, 68, 76, 79, 82, 85] is methane. This particular polyatomic gas has always offered some incentive for investigations, both experimental and theoretical, because of its quasi-spherical shape though its inelastic nature is important. The data are consistent only upto 200°C as seen from Fig. 8. Above 200°C the differences are excessive. Schottky's[43], values go upto 500°C alongwith those of Geier & Schäfer[29]. The difference between the two sets of values increases with temperature and is maximum at 500°C, about 8%. The value of Lenoir & Comings[42], obtained by the co-axial cylinders method, is also in good agreement with the smooth values; the same holds good for the thermal conductivity value of Kannuluik & Donald[29], who have exploited the thick-wire cell method. Thus, the above discussion reveals the possibility of getting reasonably consistent values from different techniques. We suggest that an investigation extending the measurements to high temperatures beyond 200°C will be a useful piece of experimental information. Smoothed values of λ upto 500°C are reported in Table 1. The λ values above 500°C are Geier & Schäfer[29].

Deuterium

Among the gases under review deuterium has been investigated by a relatively less number of workers. The data of only five workers[65, 67, 86, 90] and that too only at a few temperatures below 0°C, are available and are shown in Fig. 9. All the data seem to be reasonably consistent within rather large margin of flexibility available as the points are at fairly distant temperatures. The λ value of Kannuluik[87] at 0°C, which he has himself discarded later on [90], is not considered here.

The comparison of the values obtained on different techniques is possible only at 0°C. Both Archer[65, 88] and Northdurft[89] have measured the thermal conductivity with the compensating-tube method and the difference between the two values is about 2%. Kannuluik[90] has exploited the thick-wire variant of the hot-wire method but after calibration like a katharometer his value can be checked with that of Northdurft[89]. The maximum difference between the existing data at 0°C[65, 86, 88-90] is about 4%. Ubbink[67] has measured the λ values employing the famous parallel plate method but his data is at very low temperatures where

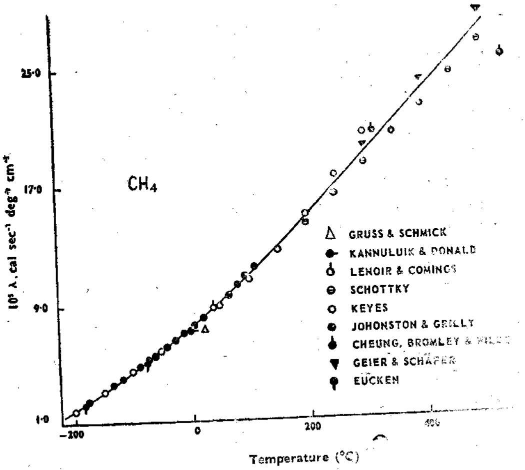

FIG. 8—Thermal conductivity of CH_4 as a functon of temperature

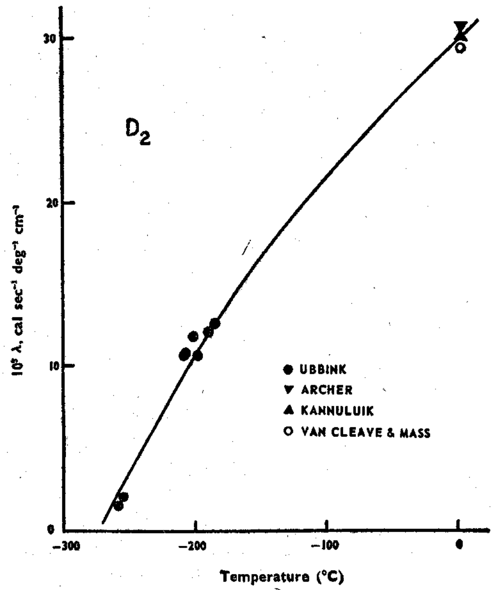

FIG. 9—Thermal conductivity of D_2 as a functiod of temperature

no check can be made as no other measurements are available. Thus the thermal conductivity of D_2 as function of temperature will be specially useful relative to other gases smooth λ values obtained from Fig. 9 are recorded in Table 1.

Very limited data are available for a large number of other non-polar polyatomic gases. Todd[11] has reported data for NO_2 ($8 \cdot 88 \times 10^{-5}$ cal-deg^{-1}– sec^{-1} at 55°C) and Franck[21] for the halogens F_2, Cl_2, Br_2, and I_2 over various temperature range—173 to 525°C. Sufficient data are also available for a large number of hydrocarbons.

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