Correlated Thermal Conductivity Data of Rare Gases and Their Binary Mixtures at Ordinary Pressures

J. M. GANDHI

Department of Physics, University of Rajasthan, Jaipur, Rajasthan, India

S. C. SAXENA

Thermophysical Properties Research Center, Purdue University, Lafayette, Ind. 47906

Most of the readily available experimental data for the thermal conductivity of rare gases and their binary mixtures are considered and graphically smoothed to generate a compromised correlated set. The analysis includes the experimental data at ordinary pressures below 1 atm. on five stable rare gases and their 10 possible binary permutations. For pure gases, the thermal conductivity data are tabulated as a function of temperature while for mixtures as a function of composition at the temperature of measurement.

RELIABLE thermal conductivity data are needed for both fundamental and applied problems. However, there is a general paucity of experimental data because of the difficulties associated with the measurements. Gandhi and Saxena (11) and Gandhi (9) have made efforts to analyze the readily available experimental data on the five rare gases and their 10 binary systems. They (9, 11) presented graphs and their efforts were directed to find the actual deficiencies, so that the experimental program of measurement of thermal conductivity of gas mixtures could be suitably oriented. Measurements on rare gases and mixtures have now been completed in the temperature range 30° to 100° C. (7, 10, 24) since the exploratory studies (9, 11), and these results are included in the correlation presented here.

The highest temperature to which elaborate rare gas mixture conductivities are available is 520°C. For pure gases, however, data are available up to much higher temperatures and a critical assessment of the extensive data is available with the tabulated recommendations (38). The present study was made independent of this particular effort (38) and was completed about 2 years ago. No attempt is made to include the works which have appeared since then. The work of Touloukian (38) has been periodically updated. Since a capsule summary (51) of this work (38)is available, the authors describe this portion of their effort only in brief. The smooth values are in good agreement, within $1^{c_{\epsilon}}$, of the recommended values (38). The authors emphasize the data on mixtures for which no correlations are available so far. The results on pure gases are also included for these have been used independently and also with data on mixtures for a variety of theoretical calculations. All the data considered refer to 1 atm. or less where thermal conductivity is pressure invariant,

Smoothing of the data on pure gases to produce a best compromised set becomes difficult as different techniques have been used and the experimental uncertainties are not known in each case with equal confidence. The extensive effort of Touloukian (38) employs for neon (above 350° K.), krypton, and xenon the data of Kannuluik *et al.* (15-17) as the basis, while for helium and argon, the recommendations are based on the consideration of all the available data. Here the latter approach has been adopted for all the gases, and in addition, the knowledge of viscosity data has been used as a guide. The discussion on pure gases is intended to be brief but the salient points are mentioned. No new experimental data are presented in this paper, but it does include some results which have become available since the completion of Touloukian's work (38). The data on binary systems are relatively scarce, but the available ones are discussed here in reasonable detail.

CORRELATION OF DATA ON RARE GASES

The thermal conductivity of monatomic gases has been measured by a large number of workers at different temperatures using different methods. Helium and argon are of special interest, for the data obtained by different established techniques are available. Kannuluik and his group (15-17), Srivastava and Saxena (37), and Srivastava and Barua (36) have used the thick-wire variant of the hot-wire cell, while Weber (47, 48), Johnston and Grilly (14), and von Ubisch (43) have exploited the thin-wire variant of the hot-wire method. On the other hand, Waelbroeck and Zuckerbrodt (45), Cheung, Bromley, and Wilke (3), Lenoir and Comings (22), and Keves (18) have employed concentric cylinders; while Walker, de Haas, and Westenberg (46) have used a flow method. The data reported by Blais and Mann (2) are obtained by a thermal diffusion column method and those reported by Sengers (23, 34) are due to the plate method. Saxena and Agrawal (27) have shown that the Blais and Mann (2) values are probably consistently higher and these are, therefore, not considered in the present correlations. The data on which this analysis is based are: He (3, 4, 13-15, 17, 18, 22, 27 36, 38-40, 41, 43-46), Ne (3, 5, 12, 15, 17-20, 22, 23, 25, 32-34, 39, 40, 41, 43-45, 47-50), Ar (3, 5, 12, 15, 17-20, 22, 25, 32-34, 37, 40, 41, 43-45, 47-50), Kr (15, 19-21, 32, 33, 36, 37, 40, 41, 43), and Xe (1, 15, 17, 21, 27, 32, 36, 40, 41, 43). For each gas, the thermal conductivity values were plotted on a large graph paper as a function of temperature. A smooth curve is passed through the different points keeping in view the accuracy of the particular data, any systematic trend in the values of a particular worker, guidance from the viscosity data and kinetic theory, and further making sure that this will join smoothly with the data above 520°C. The authors' estimates of reliability of the recommended values are, therefore, sometimes better than the actual deviations of the measured values with the smooth plot. Personal judgment plays an important role, and to this extent, there are likely to be differences in the different correlations. Some confidence can also be drawn by the intercomparison of two correlations. The smooth values only are reported in Table I. The uncertainty

01				omportarer	•
Temp.,		$\lambda \times 10^\circ$, Ca	al./Cm./Se	c./Degree	
° C.	He	Ne	Ar	Kr	Xe
-200	14.6	4.20	1.12		• • • •
-180	16.6	5.20	1.46		
-160	18.8	5.81	1.76		
-140	21.0	6.40	2.07		
-120	23.2	7.10	2.36		
-100	25.0	7.81	2.64	1.21	0.80
-80	27.0	8.40	2.92	1.38	0.92
-60	28.9	9.00	3.18	1.56	1.02
-40	30.7	9.60	3.44	1.72	1.12
-20	32.6	10.2	3.69	1.88	1.20
-0	34.2	11.0	3.94	2.04	1.29
20	35.8	11.6	4.18	2.21	1.40
40	37.4	12.0	4.42	2.36	1.48
60	38.6	12.5	4.62	2.50	1.57
80	40.0	13.0	4.86	2.63	1.65
100	41.6	13.6	5.06	2.78	1.70
120	43.5	14.1	5.30	2.91	1.80
140	44.6	14.6	5.49	3.04	1.88
160	46.2	15.1	5.68	3.16	1.95
180	47.6	15.6	5.89	3.28	2.02
200	49.1	16.0	6.08	3.39	2.08
220	50.6	16.4	6.27	3.51	2.16
240	51.9	16.8	6.45	3.62	2.24
260	53.3	17.2	6.62	3.72	2.30
280	54.7	17.6	6.82	3.84	2.36
300	56.1	18.0	6.98	3.96	2.44
320	57.5	18.4	7.16	4.07	2.50
360	60.0	19.1	7.50	4.28	2.64
400	62.8	19.8	7.88	4.50	2.76
440	65.6	20.5	8.18	4.71	2.89
480	68.2	21.2	8.51	4.92	3.02
520	70.8	22.0	8.84	5.12	3.14

Table 1. Smoothed Values of Thermal Conductivity of Rare Gases as a Function of Temperature

in the data above 200°C. is in general larger than below this temperature. As these agree well with those of Touloukian (38) and necessary deviation plots are available in the latter, the authors avoid details and give below only the brief discussion for each gas.

Most of the data on helium by the various workers are confined to the temperatures between 0° and 100°C. Many workers (4, 5, 44, 47, 48) have worked only at 0°C. Kannuluik and Carman (15), Johnston and Grilly (14), and Keyes (18) have reported measurements below 0° to -200° C. Measurements above 100° C. are from Kannuluik and Carman (15), Cheung, Bromley, and Wilke (3), and von Ubisch (43). The thermal conductivity values below 0°C. obtained using either variant of the hot-wire cell are consistent with each other and also with those obtained using coaxial cylinders by Keyes (18). At 0°C., a large amount of data mostly by the hot-wire method is available. The maximum scatter of data at this temperature is about 5%. The thick hot-wire method values of Kannuluik and Carman (15) at 0° C. agree within 1%with the values of Johnston and Grilly (14), Cheung, Bromley, and Wilke (3), and Keyes (18). From 0° to 100°C. most of the data appear to be consistent with one another, except those of von Ubisch (43), which are consistently larger than the true values. Elaborate data are not available even on the thick-wire variant of two independent workers to permit any conclusion about the possible relative consistencies. Efforts directed to produce such data are highly desirable and valuable for even assessing the experimental accuracies. The correlated values of Table I are uncertain by about 2 to 3% up to 200° C. and by about 4% above this temperature. This estimation is obtained by the assessment of departure of the actual values with the smooth recommended ones, where consideration has been given to the varying experimental uncertainty of different experiments and to systematic trends in a few cases.

In the case of Ne, there is a general dearth of data. The available data are mostly in the temperature range 0° to 100° C. Below 0° C., the data are only from Kannuluik and Carman (15) and above 100° C. from these workers and von Ubisch (43). The Sengers, Bolk, and Stigter (35) values obtained by the parallel plate method are in good agreement with those of Keyes (19, 20) and Kannuluik and Carman (15). The good agreement between the values obtained by different techniques is noteworthy. The smooth values (Table I) are likely to be uncertain by about 2%. The assessment of uncertainty above 100° C. is difficult as there is not much to compare. The smooth curve was drawn on the basis of the values of Kannuluik and Carman (15) and viscosity generated values. The high temperature point of von Ubisch (43) is given little weight.

Argon represents another interesting case for comparison of data by different workers. Below 100°C., there is a general agreement between values reported by various workers, but above 100° C., comparatively fewer data are available, and there is a large scatter of the order of 4 to 6%among the different sets. Kannuluik and Carman (15) and Schäfer and co-workers (32, 33) values appear to be consistently smaller, whereas values reported by Keves (19, 20), Cheung, Bromley, and Wilke (3), Rothman and Bromley (25), and Sengers (23, 34) agree well with one another. Vargaftik and Zimina (42) have recently measured thermal conductivity, λ , of argon and tried to correct the data of Schäfer and Reiter (33) for the temperature jump effect present in their apparatus. The corrected data seem to be reasonably consistent with other measurements. These workers (42) also recommended a smooth set which agrees with the records of Table I within less than 1%. The smooth values (Table I) may have a relatively smaller uncertainty of about 1% up to 200° C. and about 3% between 200° and 500° C. In this estimate, again, larger uncertainties in some data are regarded as peculiar to the measurement and so properly accounted in the process of graphical smoothing. The authors do not proportionately blow out their estimate of reliance just because there are some unreliable measurements, and these differ sometimes considerably on a departure plot.

Comparatively very few workers have investigated Kr and Xe. The values reported by various workers in general show disagreement and by increasing amount above 100°C. Because the thermal conductivity values of Kr and Xe are much smaller, the experimental errors are relatively pronounced. The reported values in Table I for these two gases are believed to be dependable within 5%. This estimate is intuitive and so provisional in nature. The complication arises because here the experimental thermal conductivity and viscosity data are not consistent within the framework of Chapman-Enskog kinetic theory, the thermal conductivity values being greater than those expected on the basis of viscosity. The authors have given some preference to reduce this systematic trend, and to the data of Kannuluik and Carman (15) etc., while generating values reported in Table I. In connection with the authors' measurements of thermal conductivity of mixtures using the thick-wire variant of the hot-wire method (7, 8, 24), they have also determined λ of these five rare gases at 30° or 35°, 50°, 70°, and 90°C. The theory and procedure of calculation for this method is well developed (15). In the apparatus, the convection and temperature jump effects are negligible. The maximum magnitude of radiation correction is $\overline{6}^{cc}$ for Xe. The temperature difference between the hot wire and cold wall is only about 5° C. so that the thermal diffusion separation is negligible. The apparatus is specially sensitive to detect small changes in conductivity values. In general, these values (6) are in accord with previous measurements. However, the new values of He

Table II. Values of $\lambda_{mix} \times 10^5$ for Binary Gas Mixtures as a Function of Composition, as Obtained by Smoothing Experimental Data at Temperature of Measurement

			_				Temp., °	C.					
Pair	X_1^{a}	0	18	29	30	35	38	50	70	90	100	316	520
						λ_{mix} ×	10 ⁵ , Cal.,	Cm./Sec.	/Degree				
He-Xe	0.0	33.9	35.6	36.7	35.7	35.9	37.5	37.0	38.5	39.9	42.0	55.5	73.6
	0.2		18.0	19.8	20.1		20.1	21.1	22.0	22.9			41.1
	0.4		11.0	11.5	11.0		11.8	12.4	13.1	13.7			25.5
	0.6		67	6 75	7.00		6 75	7 40	7.40	8.10			14.5
	0.0	• • •	3.50	3.63	3.80		3.63	4 20	4 30	4 60		•••	7.50
	1.0	• • •	1.90	1 42	1 97		1.25	1.46	1.50	1.63			3 34
H. K.	1.0	•••	1.02	017	1.57		1.50	01.40	1.00	1.00	•••	•••	45.9
ne-Kr	0.2		22.0	21.7	• • •	21.4	• • •	22.3	20,2	24.1	• • •	• • •	40.0
	0.4	• • •	13.3	13.3	• • •	13.5	• • •	13.9	14.5	15.2	• • •	• • •	28.0
	0.6	• • •	8.20	8.20	• • •	8,30	• • •	8.60	9,10	9.60		• • •	18.5
	0.8		4.70	4.70		4.80		5.00	5.30	5.60			11.0
	1.0		2.20	2.32		2.29	2.34	2.43	2.56	2.66	• • •		5.34
He-Ar	0.2	21.1	24.0	25.0		22.9	23.7	23.8	25.0	25.8	26.5	38.5	48.5
	0.4	14.1	15.5	16.5		15.5	15.8	16.2	16.9	17.6	18.3	25.0	32.8
	0.6	9.50	10.1	11.7		10.7	10.7	11.2	11.8	12.2	12.8	16.8	22.8
	0.8	6.31	6.50	7.00		7 10	7 10	7.30	7 70	8 20	8.00	10.7	14.6
	1.0	2 90	4.16	1.00	• • •	1.10	1.10	1.50	4.79	5.01	5.02	7.25	9.14
IL N.	1.0	0.00	4.10	4.04		4.00	4.00	4.00	90.1	0.01 91 9	0.02	1.20	57.0
ne-ive	0.2	20.7	29.5	20.7	27.6	• • •		20.0	30.1	31.3	• • •		01.0
	0.4	20.7	23.7	23.1	22.3	• • •	• • •	23.1	23.9	24.6		• • •	40.0
	0.6	16.9	19.1	18.7	17.9	• • •	• • •	18.4	19.2	19.9	• • •		37.6
	0.8	13.4	14.9	15.0	14.2		• • •	15.0	15.5	16.2			30.1
	1.0	10.8	11.6	12.4	11.8		11.8	12.3	12.7	13.2			23.6
Ne-Xe	0.2		7.50	8.00	7.80			8.20	8.50	8.90			15.9
	0.4		5.00	5.15	5.15			5.45	5.65	5.95			11.0
	0.6		3.25	3.40	3.50			3.73	3.90	4.05			7.71
	0.8		2 10	2 30	2.30			2 45	2.63	2.80			5 70
	0.0		2.10	1.00	2.00		• • • •	(40°C)	(65° C)	2.00			00
No Ka	0.9		8 90	869			9 45	(40° ().)	(05° C.)	0.40			173
ING-IVL	0.2	• • •	5.20	6,02	• • •	• • •	0.40	6.00	5.11 6.70	5.45			19.0
	0.4		5.95	6.21	• • • •		6.10	6.40	6.72	7.07	• • •	• • •	13.0
	0.6		4.30	4.60		• • • •	4.48	4.71	4.96	5.26			9.81
	0.8		3.05	3.30		• • •	3.30	3.44	3.63	3.87			7.20
Ne-Ar	0.2		9.50	9.86	• • •		9.65	9.87	10.4	10.9			19.2
	0.4		7.84	8.12			7.90	8.24	8.52	8.99			16.1
	0.6		6.41	6.60			6.52	6.83	7.04	7.48			13.2
	0.8		5.25	5.42			5.35	5.57	5.80	6.18			10.7
			0.20				(38° C)			(0 3° C)			
Ar.Xo	0.2		3 14	3 4 9			3 38			4 26			7.98
111-116	0.4	• • •	0,14	9.74	• • •	• • •	2.00		• • •	2,20	•••		7.20 5.00
	0.4	• • •	2,00	2.74	• • •	• • •	2.00	• • •	• • •	0.02	• • •	• • •	0.90
	0.6		2.03	2.15	• • •	• • •	2.11	• • •	• • •	2.85	• • •		4.85
	0.8	• • •	1.63	1.73		• • •	1.69	• • •		2.25			3.97
								(50° C.)	(70° C.)	(90° C.)			
Ar Kr	0.2		2 55	3 83		2 9/	9.75	4.00	4 10	1 34			8 1 2
AI-NI	0.2	• • •	0.00	0.00	• • •	0.04	0.10	4.00	4.19	4.04	• • •	• • •	7.00
	0.4	• • • •	3.11	3.37	• • •	3.33	3.29	3.47	3.63	3.77	• • •	• • •	1.20
	0.6	• • •	2.74	2.97	• • •	2.89	2.89	3.02	3.16	3.32	• • •	• • •	6.46
	0.8	• • •	2.46	2.63	• • •	2.56	2.57	2.70	2.84	2.98		• • •	5.86
								(40° C.)	(65° C.)				
Kr-Xe	0.2		1.98	2.08				2.22	2.36	2.56			4.80
	0.4		1.77	1.86				2.01	2.14	2.33			4.28
	0.6		1.59	1.68				1.82	1.95	2.12			3.83
	0.8		1 45	1.52				1 64	1 76	1.92			3.48
	0.0		~.10	~.~~									0.10

are smaller than the previously available ones by about 4 to 5%, a conclusion confirmed by repeated measurements with long time gaps and different charges of the gas on this apparatus (7, 10), as well as by the thermal diffusion column method (31). For Ne, Ar, and Kr the agreement between the new and the old values is satisfactory. In the case of Xe the present values are about 5% greater than the meager and insufficient data available. These values have been further confirmed in this laboratory by a different group of workers (24), and thus, the present authors feel that the old measurements are likely to be smaller. The reproducibility of these measurements is invariably within 1% while the accuracy is about 2%. In

recommending the values in Table I, a little weight has been given to these new values. The values of Table I may need later revision with the availability of new elaborate data as a function of temperature.

CORRELATION OF DATA FOR BINARY RARE GAS MIXTURES

Data for binary gas mixture thermal conductivity, λ_{mix} , are important not only for direct use but also in the prediction of multicomponent mixture values. As λ_{mix} data are not available in enough detail over wide temperature ranges,

simple but reliable methods (28–30) have been developed for their prediction. The authors mention here only the λ_{mix} data for binary mixtures of rare gases at ordinary pressures.

The experimental λ_{mix} data on binary mixtures of five noble gases have been reported by Thornton (40, 41) at $18^\circ\,C.,$ von Ubisch (43) at 29° and $520^\circ\,C.,$ and Srivastava and Saxena (26, 37) at $38^\circ\,C.$ Davidson and Music (4) have measured λ_{mix} values for the He-Ne system at $0^{\circ}\,\mathrm{C}.,$ and Cheung, Bromley, and Wilke (3) for the He-Ar system at 100° and 316° C. Measurements on He-Ar system have also been reported by Wachsmuth (44) at 0°C. Gambhir and Saxena (7), Gandhi and Saxena (11), and Mathur, Tondon, and Saxena (24) have reported values for all the 10 binary systems of five stable rare gases as a function of composition and at three to four temperatures between 30° and 100° C. The authors have plotted on a suitable scale all these λ_{mix} data as a function of composition at the actual temperature of measurement. From these graphs values were read at round and properly spaced compositions in each case (Table II). In all cases, there is a good internal consistency in the observed values, as judged from the smoothness of the experimental plot of λ_{mix} vs. composition at a particular temperature, though the uncertainty is large in many cases. This is evidenced by the critical look of Table II; values do not always fall in the right order in relation to the temperatures to which they refer. For He-Xe system all the four sets are consistent except von Ubisch values do not lie in the right order with reference to temperature and are likely to be systematically high. This indeed is confirmed by the measurements at 30°C. of Gandhi and Saxena (10). In the case of He-Kr system, Thornton values are inconsistent with those of von Ubisch, and this raises the possibility of the former data also to be somewhat on the higher side. Gambhir and Saxena (7) data at 35° C. provide some confirmation to this view and also indicate disagreement with von Ubisch values for the two pure gases. On the basis of similar critical comparisons for other systems, the authors make a few general comments. The data of von Ubisch (43) at both 29° and 520° C. are probably systematically and consistently larger than the correct values by several per cent on the average. The data of Thornton (40, 41) do not seem to be accurate and may be systematically larger than the correct values. Particular systems for which discrepancies have been observed are He-Kr, He-Ar, He-Ne, and Ar-Ne; also Ne-Xe to a lesser extent. Many discrepancies posed by the data of Thornton (40, 41), von Ubisch (43), and Srivastava and Saxena (27, 37), as explicitly mentioned in the individual cases earlier (11), are resolved to a large extent by the data from this laboratory (Table II) (7, 10, 24).

From the data of Table II, it is possible to generate smooth values of λ_{mix} , as a function of temperature. This possibility seems irrelevant in most cases because most of the data (7, 10, 24) have already been smoothed on this very criterion. Another attempt taking into account the remaining data appears redundant because, except for the Ar-He system, the data above 90°C. are available only at 520°C. and are of doubtful accuracy. The unavailability of data at 0°C. and below are further discouraging to such an approach. Therefore, the results for the Ar-He system only are presented.

The data of the Ar-He system (Table II) were plotted as a function of temperature at the compositions of 20, 40, 60, and 80% of Ar. From these plots values were read at round temperatures between 0° and 500°C. at intervals of 50°C. (Table III). This interpolation procedure also enables us to assess the λ_{mix} values of Thornton (40, 41) and von Ubisch (43). In each case, the values of these workers are greater than the smoothed values. To quote von Ubisch, this discrepancy varies between the maximum

Table III. Smoothed Values of λ_{min}	$\times 10^{5}$	for
He-Ar System as a Function of Tem	iperatu	re
at Arbitrarily Chosen Compos	itions	

Temn	Ar, %						
° C.	20	40	60	80			
	λ _{mi}	$_{\rm x}$ × 10 ⁵ , Cal./0	Cm./Sec./Deg	ree			
0	20.7	14.1	9.4	6.5			
50	24.0	16.2	11.1	7.4			
100	27.2	18.2	12.4	8.2			
150	30.3	19.8	13.6	9.0			
200	33.0	21.4	14.6	9.5			
250	35.5	23.0	15.6	10.0			
300	37.8	24.6	16.6	10.6			
350	40.0	26.1	17.5	11.0			
400	41.9	27.3	18.1	11.3			
450	43.7	28.6	18.8	11.6			
500	45.6	29.8	19.5	11.8			

and minimum limits of 21.7 and 1.4%, respectively. The average deviation for the eight points is 10.3%. The Thornton values are on the average 3.4% larger than the smooth values.

The over-all reliability associated with the λ_{mix} values of Saxena and co-workers (7, 11, 24) of Table II is about 2% and is primarily limited by the inherent uncertainty of the original data. Similarly the Thornton (40, 41) and von Ubisch (43) λ_{mix} values may be uncertain by about 5%. The uncertainty of the values of Table III is about 2% up to 350° C. but may be larger beyond this temperature. The emphasis and need for new data at elevated as well as at lower temperatures (below 0° C.) is amply borne out by this work.

No effort is made to compare these experimental values with the predictions of rigorous kinetic theory as the same has appeared in the literature (6, 7, 10, 24, 30). As one would expect, theory is generally successful in explaining the data of these spherically symmetric molecules.

ACKNOWLEDGMENT

The authors are grateful to the Ministry of Defense, New Delhi, for supporting this work, and to Y. S. Touloukian and P. E. Liley for a critical review of the manuscript.

LITERATURE CITED

- (1) Barua, A.K., Indian J. Phys. 34, 169 (1960).
- (2) Blais, N.C., Mann, J.B., J. Chem. Phys. 32, 1459 (1960).
 (3) Cheung, H., Bromley, L.A., Wilke, C.R., AIChE J. 8, 221 (1962).
- (4) Davidson, J.M., Music, J.F., U.S. Atomic Energy Comm., Oak Ridge, Tenn. Rept. No. HW-29021, (July 1953).
- (5) Dickins, B.G., Proc. Roy. Soc. A143, 517 (1934).
- (6) Gambhir, R.S., Gandhi, J.M., Saxena, S.C., Indian J. Pure Appl. Phys. 5, 457 (1967).
- (7) Gambhir, R.S., Saxena, S.C., Mol. Phys. 11, 233 (1966).
- (8) Gambhir, R.S., Saxena, S.C., Trans. Faraday Soc. 60, 38 (1964).
- (9) Gandhi, J.M., Ph.D. dissertation, Rajasthan University, Jaipur, India, 1966.
- (10) Gandhi, J.M., Saxena, S.C., Mol. Phys. 12, 57 (1967).
- (11) Gandhi, J.M., Saxena, S.C., Rajasthan University Studies (Physics) I, 7 (1965).
- (12) Gardiner, W.G., Schäfer, K., Z. Elektrochem. 60, 588 (1956).
 (13) Hansen, R.S., Frost, R.R., Murphy, J.A., J. Phys. Chem.
- 68, 2028 (1964). (14) Johnston, H.L., Grilly, E.R., J. Chem. Phys. 14, 233 (1946).
- (15) Kannuluik, W.G., Carman, E.H., Proc. Phys. Soc. (London) 65, 701 (1952).
- (16) Kannuluk, W.G., Donald, H.B., Australian J. Sci. Res. 3, 417 (1950).
- (17) Kannuluik, W.G., Martin, L.H., Proc. Roy. Soc. London A144, 496 (1934).

- (18) Keyes, F.G., Project Squid, Massachusetts Institute of Technology, Cambridge, Tech. Rept. No. 37, (April 1, 1952).
- Keyes, F.G., Trans. ASME 76, 809 (1954). (19)
- (20)Ibid., 77, 1395 (1955).
- Landolt-Bornstein, Physikalisch-Chemische Tabellen, (21)
- Lenoir, J.M., Comings, E.W., Chem. Eng. Progr. 47, 223 (1951). (22)(23)
- Michels, A., Sengers, J.V., Van de Klundert, L.J.M., Physica 29, 149 (1963).
- (24)Mathur, S., Tondon, P.K., Saxena, S.C., Mol. Phys. 12, 569 (1967).
- (25)Rothman, A.J., Bromley, L.A., Ind. Eng. Chem. 47, 899 (1955).
- (26)Saxena, S.C., Indian J. Phys. 31, 597 (1957).
- (27)
- Saxena, S.C., Agrawal, J.P., *J. Chem. Phys.* **35**, 2107 (1961). Saxena, S.C., Gambhir, R.S., *Proc. Phys. Soc.* **81**, 788 (1963). (28)
- Saxena, S.C., Gandhi, J.M., Rajasthan Univ. Studies (Physics) (29)1, 1 (1965).
- (30)Saxena, S.C., Gandhi, J.M., Rev. Mod. Phys. 35, 1022 (1963).
- Saksena, M.P., Saxena, S.C., *Phys. Fluids* 9, 1595 (1966). Schäfer, K., Grundler, K.H., Z. Elektrochem. 63, 449 (1959). (31)
- (32)
- (33)Schäfer, K., Reiter, F.W., Ibid., 61, 1230 (1957)
- Sengers, J.V., Ph.D. dissertation, Amsterdam, 1962. (34)
- Sengers, J.V., Bolk, W.T., Stigter, C.J., Physica 30, 1018 (35)(1964).
- (36)Srivastava, B.N., Barua, A.K., J. Chem. Phys. 32, 427 (1960).
- Srivastava, B.N., Saxena, S.C., Proc. Phys. Soc. (London) B70, (37)369, (1957).

- (38) Touloukian, Y.S., Thermophysical Properties Research Center, Data Book, Vol. 2, Chapter 1, Purdue University, Lafayette, Ind., 1966.
- Thomas, L.B., Golike, R.C., J. Chem. Phys. 22, 300 (1954). (39)Thornton, E., Proc. Phys. Soc. (London) 76, 104 (1960); Ibid., (40)77. 1166 (1961).
- Thornton, E., Baker, W.A.D., Proc. Phys. Soc. (London) 80, (41)1171 (1962).
- Vargaftik, N.B., Zimina, N.Kh., High Temperature 2, 645 (42)(1964)
- von Ubisch, H., Arkiv Fysik 16, 93 (1959). (43)
- Wachsmuth, J. Phys. Z. 9, 235 (1908). (44)
- Waelbroeck, F.G., Zuckerbrodt, P., J. Chem. Phys. 28, 523 (45)(1958).
- (46)Walker, R.E., de Haas, N., Westenberg, A.A., Phys. Fluids 3, 482 (1960).
- Weber, S., Ann. Physik. 54, 48, 325 (1917). (47)
- Ibid., 82, 479 (1927). (48)
- (49)Zaitseva, L.S., J. Tech. Phys. USSR 29, 497 (1959).
- (50)
- Zaitseva, L.S., Soviet Phys. Tech. Phys. 4, 444 (1959). Liley, P.E., "The Thermal Conductivity of 46 Gases at (51)Atmospheric Pressure," Proc. Symp. Thermophys. Properties 4th, University of Maryland, April 1968.

RECEIVED for review December 30, 1966. Accepted February 23, 1968

Linear Equilibrium Data for Ternary Liquid Systems and **Binary Liquid-Gas Systems**

STANLEY HARTLAND and VICTOR ANG Chemical Engineering Department, The University, Nottingham, England

> Published equilibrium data for 99 aqueous-organic systems, 47 organic-organic systems, and 39 liquid-gas systems have been tested for linearity. The best straight lines which pass either through the origin or through the mean values of the observations are considered. Slopes are tabulated for 28 systems having an error of less than 1% over the concentration range studied experimentally.

 $\mathbf{M}_{\mathrm{UCH}}$ experimental work has been performed to determine equilibrium data for ternary liquid systems. Some systems are linear, some partially linear, and others nonlinear. Both mathematical expressions and graphical methods have been employed to represent the distribution data. Various laws of distribution have been proposed but these are frequently empirical and are not obeyed over wide concentration ranges (8, 22). The common assumption of a linear equilibrium relationship in many theoretical analyses of the performance of absorption and extraction equipment leads to simple expressions and avoids the cumbersome mathematics involved if nonlinear relationships are taken into account (6, 10, 12, 13). For the experimental verification of these theoretical analyses, and for most other experimental work on absorption or extraction equipment, it is clearly advantageous, if not essential, to employ systems exhibiting linear distribution relationships (24, 25). In preliminary design work, the use of linearized data enables quick answers to be obtained and can save valuable computer time for the more detailed calculations.

The purpose of compiling a set of systems with essentially

linear equilibrium data is thus to enable the engineer either to select a system which he desires to be linear, or to confirm whether a particular system is linear, and to provide some estimate of the error in the assumption of linearity.

ANALYSIS OF DATA

Equilibrium data were obtained for a total of 185 systems, being made up of 99 aqueous-organic systems, 47 organicorganic systems, and 39 liquid-gas systems. These are arranged alphabetically according to the solute, and some are listed in Table I. Each separate set of data is termed a system and given a separate number, and so there are included in the list identical physical systems with data obtained by different authors or at different temperatures. The concentration units in which the data are given are also tabulated with reference to their source. The search was not exhaustive but limited to systems containing fairly common substances in which the solvents were reasonably immiscible.

As is usual in the literature, the solute concentrations