

Thermal Conductivity of Light Hydrocarbons and Methane-Propane Mixtures at Low Pressures

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SINCE THE first measurements of the thermal conductivity of gases by Stefan (29), Kundt and Warburg (16), and Winkelmann (35), many modifications have been made to improve the experimental techniques for their measurement (5, 12-14, 25, 27, 30, 34) by: measuring heat flux and temperature differences more accurately; eliminating convection or mass movement of the gas; eliminating or compensating for heat losses from cell boundaries; reducing and/or accurately determining radiant heat losses; and eliminating or accurately determining the accommodation effect or the temperature discontinuities at the cell surfaces. Kraussold (15) showed that convective forces could be eliminated, if the product of the Prandtl numbers, N_{Pr} , and the Grashof numbers, N_G , was less than 600.

EQUIPMENT

The experimental apparatus (Figure 1) was of the concentric cylinder type which was introduced by Keyes and Sandell (14). The outer cylinder was of heavy brass which was bored to close tolerances, silver plated, and

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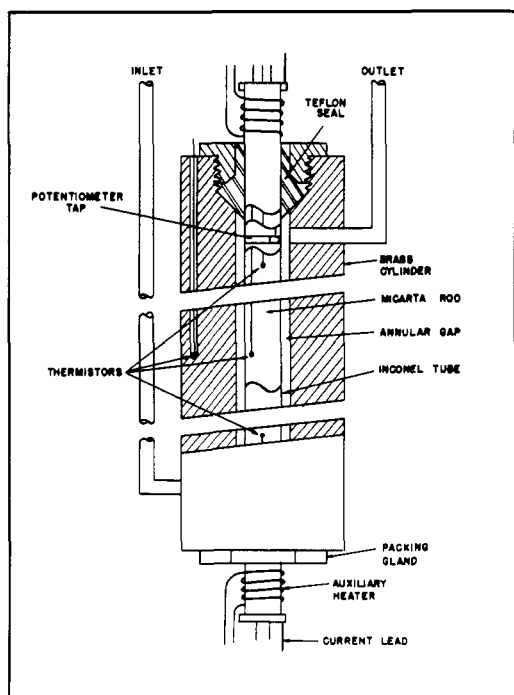


Figure 1. Thermal conductivity cell construction

polished. A polished Inconel tubing mounted concentrically with the brass cylinder served as the inner wall as well as the heating element. The cell dimensions are as follows:

Outside diameter of inner tube	1.2789 cm.
Wall thickness of inner tube	0.0835 cm.
Inside diameter of brass cylinder	1.9100 cm.
Outside diameter of brass cylinder	6.0 cm.
Effective cell length	30.718 cm.

Teflon seals held in place by packing glands made the annular cell space vacuum-tight, centered the Inconel tube in the brass cylinder, and electrically insulated the tube from the cylinder. The inner tube contained a filler rod of Micarta which had been machined to hold three thermistors and two voltage tap contacts. The voltage tap contacts were the heads of small telescoping gages to which potentiometer lead wires were soldered.

The thermistor bridge whose circuit diagram is presented in Figure 2 was used to measure the temperature drop across the annular gap containing the hydrocarbon gas. The thermistors were of the probe type (Victory Engineering Co.). A 1000-ohm resistor was placed in series with a 1.5-volt dry cell battery which served as the voltage source. The variable arms were two enclosed, switch-type resistance boxes whose ranges were 999.9 ohms in steps of 0.1 ohm. The null indicator used was a precision potentiometer or a microvolt amplifier. The thermistor whose location is shown in Figure 1 was used to detect the temperature drop across the annular layer of gas.

The temperature drops which were measured varied from 0.341° C. to 1.033° C. With the use of such low temperature drops, nonradial heat flows such as end effects were virtually absent. A second consequence of the low temperature drops was that the annular gap could be made appreciably larger and the average gap thickness measured with

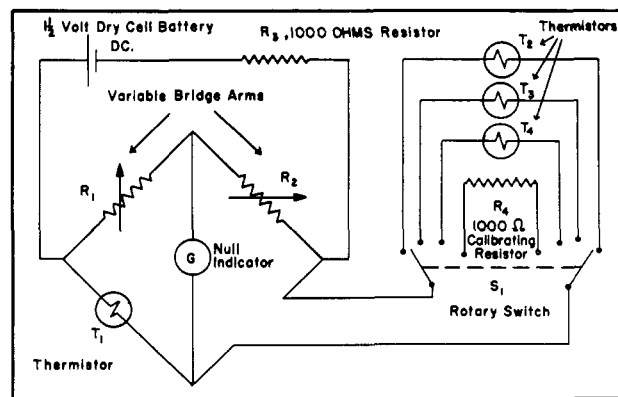


Figure 2. Thermistor bridge circuit diagram

good precision. The bath temperature was regulated with a thermistor-actuated electronic thermal regulator to less than 0.005° C.

CALIBRATIONS

Thermistor Calibration. The resistance of a thermistor may be represented by the relation:

$$R = R^0 \exp. B (T) \left[\frac{1}{T} - \frac{1}{T^0} \right] \quad (1)$$

where R is the resistance at any temperature T

R^0 is the resistance at the reference temperature, T^0

$B(T)$ is a function of the material comprising the thermistor and only slightly dependent on the temperature

Over a narrow temperature range if B is assumed constant the coefficient of resistance, $\alpha = 1/R (dR/dT)$ is related to B by the equation:

$$\alpha = -B/T^2 \quad (2)$$

For the thermistor used in the experiments, since B was about 4000° K., α varied from about -5% per ° C. at 0° C. to about -1% per ° C. at 400° C. The value of α for platinum is approximately 0.4% per ° C., so that thermistors when properly calibrated represent a sensitive resistance thermometer.

The resistance ratio of thermistor R_i at temperature T_2 to thermistor R_j at temperature T_1 may be written as:

$$\frac{R_i T_2}{R_j T_1} = \frac{R_i^0 \exp. B_i \left(\frac{1}{T_2} - \frac{1}{T^0} \right)}{R_j^0 \exp. B_j \left(\frac{1}{T_1} - \frac{1}{T^0} \right)} = \left(\frac{R_i}{R_j} \right)_{T_2}^0 \exp. B_j \left(\frac{T_1 - T_2}{T_1 T_2} \right) \quad (3)$$

where $\left(\frac{R_i}{R_j} \right)_{T_2}^0$ is the resistance ratio of the thermistors when both are at T_2 , provided that the temperature interval $(T_1 - T_2)$ is small and B_j may be assumed constant over that interval. If $T_1 T_2$ is approximated by T_2^2 , solving for $T_1 - T_2$ gives

$$T_1 - T_2 = \frac{T_2^2}{B_j} \ln \frac{(R_i/R_j)}{(R_i/R_j)_{T_2}^0} \quad (4)$$

Because the resistance ratios were obtained directly from the thermistor bridge, and the bridge arm R_1 was maintained constant at 800.0 ohms

$$\left(\frac{R_i}{R_j} \right) = \frac{R_2}{800.0}$$

and

$$\left(\frac{R_i}{R_j} \right)_{T_2}^0 = \frac{R_2^0}{800.0}$$

where R_2^0 is the resistance of bridge arm R_2 when both thermistors are at the bath temperature T_2 , and

$$\frac{R_i/R_j}{(R_i/R_j)_{T_2}^0} = \left(\frac{R_2}{R_2^0} \right)_{T_2} \quad (5)$$

so that Equation 5 may be represented by the final equation

$$T_1 - T_2 = \frac{T_2^2}{B} \ln \left(\frac{R_2}{R_2^0} \right)_{T_2} \quad (6)$$

The calibration data are listed in Table I.

Radiation Correction. The Stefan-Boltzmann radiation interchange factors (10), F_{12} , were determined by two methods. They were calculated from the emissivities of the metals as reported for highly polished states and by calculations which assumed that the discrepancies between the values of thermal conductivities of nitrogen reported by Rothman and Bromley (24) and those of several runs on

Table I. Thermistor Calibration Data

Temp., ° C.	Thermistor Resistance, Ohms	B, ° K.	R ₂ ⁰ , Ohms
49.99	3780.4	3810.9	858.04
75.00	1558.2	3898.9	858.14
100.90	698.64	3962.1	858.04
125.20	359.00	3995.6	857.52
149.79	196.41	4022.7	856.80

nitrogen by the authors were attributable to radiation effects only. The radiation interchange factors as determined by the two methods were nearly identical, yielding an average value of 0.030. The radiation corrections were calculated using this average value.

Thermometer. The thermometers used to record the bath temperature were Tagliabue precision thermometers calibrated to the nearest 0.01° C.

MEASUREMENTS

Under steady-state conditions, with the unknown gas in the annulus, the following readings were taken: voltage drop between the two voltage taps along the Inconel tube, current through the Inconel tube from a measure of the voltage drop across the standard resistor in series with the tube, thermistor resistance ratio $(R_2/R_2^0)_T$, and bath temperature.

A precision potentiometer was used to measure the voltage drop between the two taps along the Inconel tubing and the drop across the standard resistor.

Kraussold's criterion (15) requiring that the product of the Prandtl and Grashof numbers be less than 600 was fulfilled in all cases except for *n*-pentane in the neighborhood of 50° C.

MATERIALS

Table II gives the source and analysis of the pure components used.

Table II. Source and Analysis of Pure Components

Component	Source	Analysis, Mole %
Methane	Tennessee Gas Transmission Co.	99.7
Propane	Phillips Petroleum Co., instrument grade	99.5
<i>n</i> -Butane	Phillips Petroleum Co., instrument grade	99.9
<i>n</i> -Pentane	Phillips Petroleum Co., research grade	99.9
Nitrogen	Houston Oxygen Co., impurity oxygen	99.5

RESULTS

Experimental data were taken for methane, propane, *n*-butane, *n*-pentane, and four methane-propane mixtures at 50°, 75°, 100°, 125°, and 150° C. The experimental and graphically smoothed data are recorded in Tables III and IV. The average deviation of the observed data for the pure gases from the appropriate smoothed curve was 1.7% with a maximum deviation of 4.6%.

The largest uncertainties in the data are most likely to occur for methane, on the one hand, and *n*-pentane on the other. The high thermal conductivity for methane resulted in correspondingly low temperature drops across the annular gap. The convective tendencies were greatest for the low temperature runs involving *n*-pentane.

The experimental thermal conductivity data for the pure

Table III. Experimental and Calculated Thermal Conductivities for Pure Hydrocarbons

Gas	Temp., ° C.	$K \times 10^{7^a}$		$\eta \times 10^{7^b}$	$Mk/\eta^{c,d}$	$C_p^{e,e}$	$Mk/\eta^{c,f}$	% Dev.
		Obsd.	Smoothed ^g					
Methane	50	871	895	1188	12.09	8.79	12.38	2.2
	75	976	983	1263	12.49	9.06	12.73	1.9
	100	1036	1070	1334	12.87	9.36	13.11	1.8
	125	1112	1157	1402	13.24	9.68	13.53	2.1
	150	1189	1244	1468	13.59	10.01	13.95	2.6
Propane	50	477	500	876	25.17	18.78	25.26	0.4
	75	...	575	944	26.86	19.95
	100	650	650	1008	28.43	21.14	28.43	-0.4
	125	...	725	1069	29.90	22.35
	150	821	807	1130	31.49	23.55	31.49	-0.2
<i>n</i> -Butane	50	449	443	801	32.15	24.92	33.19	3.1
	75	511	511	864	34.37	26.46	35.17	2.3
	100	580	583	923	36.71	27.99	37.15	1.2
	125	657	663	979	39.36	29.50	39.09	-0.7
	150	741	740	1034	41.60	30.97	40.99	-1.5
<i>n</i> -Pentane	50	408	413	737	40.43	30.72	40.67	0.6
	75	(463) (486)	475	796	43.05	32.61	43.11	0.1
	100	540	545	850	46.26	34.50	45.54	-1.6
	125	624	622	903	49.69	36.37	47.96	-3.6
	150	701	703	955	53.11	38.24	50.37	-5.4

^aCal./sec.-cm.-° C. ^bDyne-sec./sq. cm. or poise, viscosity data (3). ^cCal./mole-° C.

^dCalculated from viscosity and thermal conductivity data. ^eHeat capacity data (1).

^fCalculated from empirical equation. ^gFrom Figure 5.

Table IV. Experimental and Calculated Mixture Thermal Conductivities^a for Methane-Propane System

$T, ^\circ \text{C.}$	% Propane	K		$K_1, (\text{F.C.})$ Rigorous	$K_2, (2),$ (F.C.)	$K_3, (22),$ (F.C.)	$K_4, (22),$ (Visc.)	$K_5, (22),$ (C_p 's)
		Obsd.	Smoothed					
50	0	871	895	(895)	(895)	(895)	(895)	(895)
	23.1	736	736	732	775	770	757	758
	43.6	655	659	638	658	680	665	667
	62.4	595	598	577	613	609	599	600
	81.2	540	545	533	553	551	545	546
	100.0	477	500	(500)	(500)	(500)	(500)	(500)
75	0	976	983	(983)	(983)	(983)	(983)	(983)
	23.1	828	825	815	861	856	842	843
	43.6	744	747	718	768	763	748	749
	62.6	683	683	654	693	690	678	679
	81.2	629	625	609	630	628	622	623
	100.0	...	575	(575)	(575)	(575)	(575)	(575)
100	0	1036	1070	(1070)	(1070)	(1070)	(1070)	(1070)
	23.1	916, 933	913	897	946	941	926	928
	43.6	872, 869	834	798	850	845	829	831
	62.6	758	769	733	773	769	757	758
	81.2	709	706	685	708	706	699	700
	100.0	650	650	(650)	(650)	(650)	(650)	(650)
125	0	1112	1157	(1157)	(1157)	(1157)	(1157)	(1157)
	23.1	1004, 991	1002	978	1031	1025	1011	1009
	43.6	923	922	876	932	926	911	909
	62.6	855	854	809	852	848	837	835
	81.2	791	785	761	784	783	776	776
	100.0	...	725	(725)	(725)	(725)	(725)	(725)
150	0	1189	1244	(1244)	(1244)	(1244)	(1244)	(1244)
	23.1	1104, 1063	1090	1064	1119	1113	1098	1099
	43.6	1013	1010	961	1019	1014	997	999
	62.6	942	939	893	938	934	921	923
	81.2	866	865	844	868	867	860	861
	100.0	821	807	(807)	(807)	(807)	(807)	(807)

^a 10^{-7} cal./sec.-cm.-° C.

hydrocarbons of this work and from other sources (17, 18, 20, 26, 31, 32) are given in Figure 3.

The data below the normal boiling points of *n*-pentane are at a pressure slightly less than 1 atm.

For the methane-propane mixtures, the average deviation of the gas mixture from the smoothed data was 1.1% with

a maximum deviation of 4.6%. The experimental data are presented in Figure 4.

TREATMENT OF DATA

The familiar Chapman-Enskog theory of nonuniform gases is accurate in predicting the coefficient of thermal

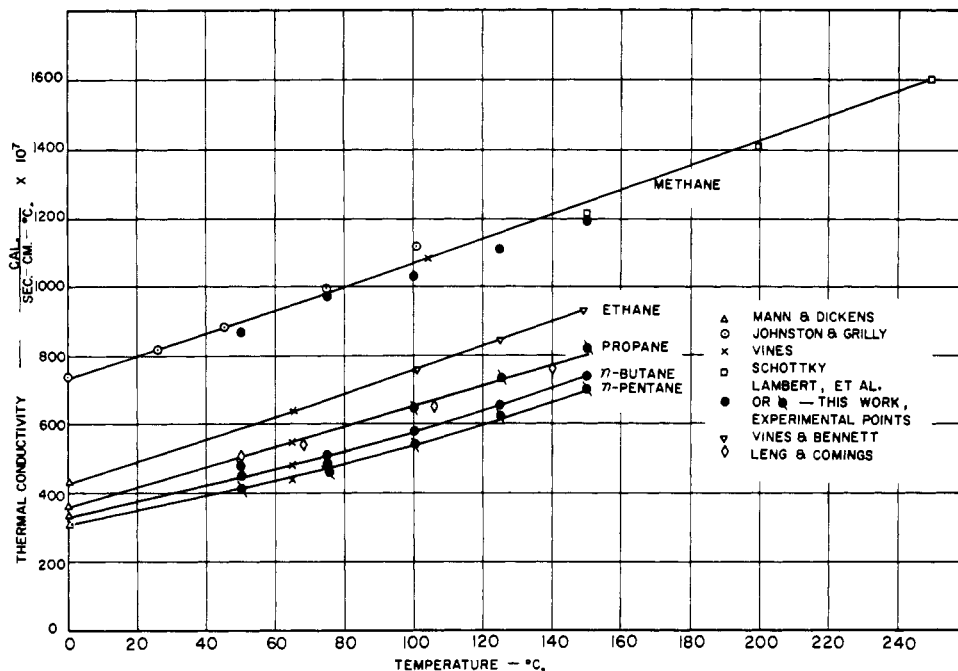


Figure 3. Thermal conductivity of light hydrocarbon gases at atmospheric pressure

conductivity for monatomic gases and gas mixtures [Chap. 8, (7)]. For polyatomic gases the Eucken type correction factor, E , for the contribution of the internal degrees of freedom to the thermal conductivity may be written as a function of the heat capacity at constant pressure, C_p :

$$E = \frac{K}{K^0} = (1 - \delta_f) + \frac{2}{5} \delta_f \frac{C_p}{R} \quad (7)$$

where K is the thermal conductivity, K^0 is the "frozen" thermal conductivity—i.e., the thermal conductivity for the gas if the internal degrees of freedom are frozen— R is the gas constant, and δ_f is a parameter to be specified. If it is assumed that the viscosity, η , is independent of the internal degrees of freedom of a polyatomic molecule, then

$$\eta = \frac{4}{15} \frac{MK^0}{R} \quad (8)$$

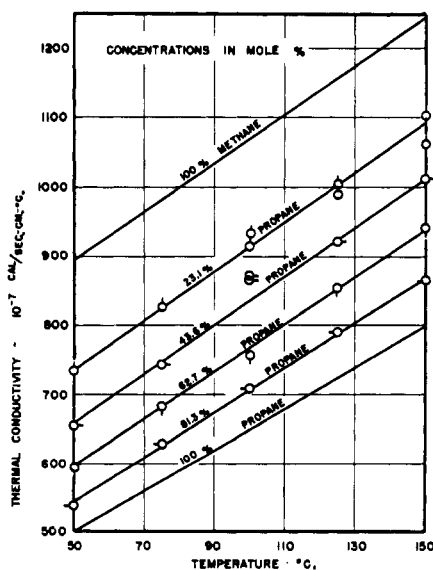


Figure 4. Thermal conductivity-temperature diagram for mixtures of methane and propane at atmospheric pressure showing experimental points

so that the Eucken-type equation becomes

$$\frac{MK}{\eta} = \frac{15}{4} (1 - \delta_f)R + \frac{3}{2} \delta_f C_p \quad (9)$$

where M is the molecular weight.

On the basis of a very simple kinetic theory argument and intuitive reasoning, Eucken (6) assigned a value of $2/3$ to δ_f . However, more rigorous interpretations by Chapman and Cowling (4) and Schaefer (28) have been shown that this factor must be related to the ratio of the self-diffusion coefficient, D_{11} , and the kinematic viscosity ratio, η/ρ , by $\delta_f = 2/3 \rho D_{11}/\eta$ inasmuch as the heat transfer effected by internal degrees of freedom is accomplished by a diffusional mechanism. On the other hand Hirschfelder (8, 9) in a formal treatment involving the consideration of each quantum state of a polyatomic molecule as a chemical species and assuming that equilibrium is maintained at each point between chemical composition and temperature requires that $\delta_f = (5/2 R) [(\rho D_{11})/(MK^0)]$. This condition is shown to be valid, if metastable states are nonexistent and the diffusion coefficients of all the quantum states are equal. The equivalence of these forms for δ_f under the required conditions follows directly from Equation 8.

The value of δ_f is a function of the molecular model chosen and temperature and may be written as $\delta_f = 4/5 (\Omega^{2.2*}/\Omega^{1.1*})$ where the collision integral ratio $A_i^* = \Omega^{2.2*}/\Omega^{1.1*}$ depends on the particular intermolecular potential function chosen to describe the gas. This ratio is unity for the rigid sphere model and as shown by Hirschfelder (8, 9) varies slightly from the value 1.106 for the more realistic Lennard-Jones (6-12) and Buckingham (6-exp.) potential models so that $\delta_f = 0.885$ to within a few per cent. Thus, for the more realistic potentials his modification of the Eucken-type factor (Equation 9) becomes

$$\frac{MK}{\eta} = 0.857 + 1.32 C_p \quad (10)$$

Values of MK/η calculated with the smoothed experimental conductivities and the viscosity values recommended by Carr, Kobayashi, and Burrows (3) are tabulated in Table III. A plot of MK/η vs. $C_i = C_p - 5/2R$ is shown in

Figure 5. The heat capacity data are those from American Petroleum Research Project 44 (1). The best straight line through the smoothed experimental points from sources cited on Figure 3 is given by Figure 5 and the relation

$$\frac{MK}{\eta} = 1.04 + 1.29 C_p \quad (11)$$

This compares favorably with Equation 10. However,

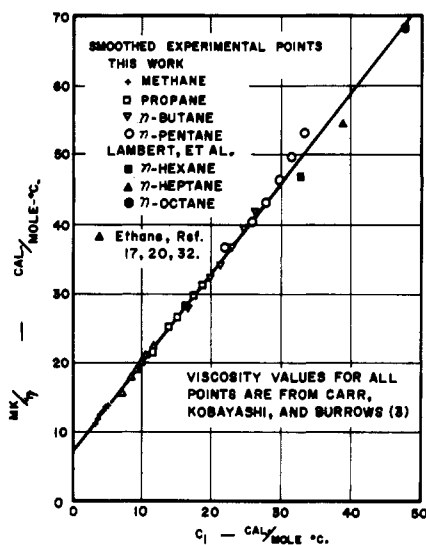


Figure 5. MK/η vs. internal heat capacity, C_i , for first eight normal paraffin hydrocarbons

Equation 11 yields an empirical value of $\delta_f = 0.86$. This lower value of δ_f is in agreement with the value of 0.85 that Hirschfelder (8, 9) obtained from an analysis of the study made by Vines and Bennett (32) of the experimental data available for a large variety of gases. With viscosity data of Carr, Kobayashi, and Burrows (3), it is seen that the experimental data of Lambert and others (17) now fall within the limits of reasonable experimental error. As shown in Table III, the observed value of MK/η has a mean deviation from the straight line of 1.5% with a maximum deviation of 5.4%.

Mixtures. Recently Hirschfelder (9) has derived an Eucken-type relation to express the coefficient of thermal conductivity of a polyatomic gas mixture $K(\text{mix})$ in terms of the "frozen" thermal conductivity of the mixture $K^0(\text{mix})$. His final expression for a mixture of polyatomic gases is

$$K(\text{mix}) = K^0(\text{mix}) + \sum_{i=1}^n \frac{K_i - K_i^0}{\sum_{j=1}^n \left(\frac{D_{ij}}{D_{ij}^0} \right) \left(\frac{x_j}{x_i} \right)} \quad (12)$$

where K_i is the experimental value of the conductivity for pure component i , x_i is the mole fraction, n is the number of components, and the first approximation to the diffusion ratio D_{ij}/D_{ij}^0 may be related to the frozen thermal conductivity ratio K_i^0/K_j^0 as $(D_{ij}/D_{ij}^0)(A_{ij}/A_{ij}^0) = (K_i^0/K_j^0)$. In most cases and over a wide temperature range $(A_{ij}^0/A_{ij}^0) \cong 1$. As K_i^0 is not directly measurable, it must be calculated from force constant data or directly from viscosities or from K_i and an Eucken factor.

$K^0(\text{mix})$ may be evaluated according to the first approximation for monatomic gas mixtures as given by Equations 8.2-36 and 8.2-43 of Hirschfelder and others (7). A much simpler, yet equally rigorous, first approximation has been recently developed (21, 23) such that

$$K^0(\text{mix}) = 4 \frac{\begin{vmatrix} L_{n1} & \dots & L_{1n} & x_1 \\ \vdots & & \vdots & \vdots \\ L_{n1} & \dots & L_{nn} & x_n \\ x_1 & \dots & x_n & 0 \end{vmatrix}}{\begin{vmatrix} L_{11} & \dots & L_{1n} \\ \vdots & & \vdots \\ L_{n1} & \dots & L_{nn} \end{vmatrix}} \quad (13)$$

and where L_{ii} and L_{ij} are given by L_{ii}^{H} and L_{ij}^{H} for Hirschfelder's Equation 8.2-43 (7). For the special case of a binary mixture this equation may be reduced to the following form

$$K^0(\text{mix}) = \frac{Q_{11} \frac{x_2}{x_1} + Q_{22} \frac{x_1}{x_2} + \left[2 Q_{12} + P \left(\frac{K_2^0 + K_1^0}{K_1^0 K_2^0} \right) \right]}{\frac{Q_{11} x_2}{K_1^0 x_1} + \frac{Q_{22} x_1}{K_2^0 x_2} + \left[\frac{P}{K_1^0 K_2^0} + \frac{1}{P} (Q_{11} Q_{22} - Q_{12}^2) \right]} \quad (14)$$

where

$$P = 2(M_1 + M_2)^2 A_{12}^* K_{12}^0$$

$$Q_{12} = [55/4 - 3B_{12}^* + 4 A_{12}^*] M_1 M_2$$

$$Q_{11} = [15/2 M_1^2 + 25/4 M_2^2 - 3M_1^2 B_{12}^* + 4 M_1 M_2 A_{12}^*]$$

and Q_{22} is of the form of Q_{11} with the subscripts interchanged. Because the quantities inside the brackets are constant for a given temperature, Equation 14 permits fairly rapid evaluation of $K^0(\text{mix})$ for a binary mixture of various concentrations at a given temperature. The "frozen" conductivities K_1^0 , K_2^0 , and K_{12}^0 may be calculated as the first approximations of the Chapman theory [Equations 8.2-31 and 8.2-34 (7)].

With Lennard-Jones (6-12) force constant (F.C.) parameters ϵ and σ as given by Table I-A (7) and the usual combining laws $\epsilon_{12} = (\epsilon_1 \epsilon_2)^{1/2}$ and $\sigma_{12} = \sigma_1 + \sigma_2/2$ Equations 12 and 14 were used to calculate heat conduction coefficients for the binary methane-propane system. These are given by K_i (Table IV) and are consistently lower than the experimental coefficients.

From rigorous kinetic theoretical considerations Brokaw (2) has developed an approximation for $K^0(\text{mix})$ of the Wassiljewa (33) form. His equation may be written as

$$K^0(\text{mix}) = \sum_{i=1}^n \frac{K_i^0}{1 + \sum_{j=1}^n G_{ij} \left(\frac{K_j^0}{K_i^0} \right) \left(\frac{x_j}{x_i} \right)} \quad (15)$$

where

$$G_{ij} = \left[1 + 2.41 \left(\frac{M_i - M_j}{M_i + M_j} \right)^2 \left(1 + 0.8585 \frac{M_j}{M_i - M_j} \right) \right]$$

Here the approximation has been made that the collision integral ratios $A_{ij}^* = B_{ij}^* = 1.1$. This equation with the Lennard-Jones force constant "frozen" conductivities and Equation 12 was used to calculate thermal conductivity coefficients for the methane-propane system. These are listed as K_2 in Table IV and are in remarkably good agreement with the experimental values.

If the G_{ij} of Equation 15 do not vary a great deal from unity, the approximation can be made that the terms with frozen conductivities in the numerators of Equation 12 tend to cancel out. This tendency is further strengthened by the counteraction of terms because if $G_{ij} > 1$, then

$G_{ji} < 1$. Thus, the following approximation to Equations 12 and 15 may be made

$$K(\text{mix}) = \frac{\sum_{i=1}^n K_i}{1 + \sum_{\substack{j=1 \\ j \neq i}}^n \left(\frac{K_j^0}{K_i^0} \right) \left(\frac{x_j}{x_i} \right)} \quad (16)$$

This very simple equation has been derived in a different manner by Mason and Saxena (22) and is similar to the successful Lindsay-Bromley empirical formula (19).

Equation 16 was employed with the Lennard-Jones frozen coefficients to calculate binary conductivities for the methane-propane system. These are tabulated under K_3 (Table V) and are only slightly lower than the coefficients K_2 calculated by the Brokaw Equation 15.

Mason and Saxena (22) have further shown that the frozen conductivity ratio K_i^0/K_j^0 , may be approximated by

$$\frac{K_i^0}{K_j^0} = \frac{1}{2(2)^{1/2}} \left[1 + \left(\frac{K_i^0}{K_j^0} \right)^{1/2} \left(\frac{M_i}{M_j} \right)^{1/4} \right]^2 \left[1 + \frac{M_i}{M_j} \right]^{-1/2} \quad (17)$$

This equation is justifiable in that certain collision integral ratios which appear in the first approximations approach unity.

With Equations 8 and 17, viscosity ratios of the pure components may be used directly for the calculation of mixture thermal conductivities by Equation 16. By this procedure and with the viscosity data of (3) (Table III) for the methane-propane system, the thermal conductivities given under K_4 (Table IV) were obtained.

The Eucken-type Equation 7 may be used to determine the frozen conductivity ratio K_i^0/K_j^0 from the measured true coefficient K_i . In this fashion Hirschfelder's Eucken factor given by a value of $\delta_i = 0.885$ in Equation 7 was used with Equations 16 and 17 to obtain the binary coefficients for the methane-propane system as listed under K_5 (Table IV). The heat capacity data (1) (Table III) were employed in these calculations.

Both K_4 and K_5 agree very well with the experimental data. With pure component experimental viscosity or conductivity data they are readily calculated. As a consequence, it is expected that they would provide a quick and easy means of approximating mixture conductivities of light hydrocarbon molecules.

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