

Diffusion Coefficients in Hydrocarbon Systems: *n*-Hexane in the Gas Phase of the Methane—, Ethane—, and Propane—*n*-Hexane Systems

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Experimental information concerning the molecular transport of the lighter hydrocarbons in the gas phase is limited. For this reason a study was made of the Maxwell diffusion coefficients of *n*-hexane in the gas phase of the methane-*n*-hexane, and propane-*n*-hexane systems.

Maxwell diffusion coefficients were determined at steady state for pressures up to 70 lb./sq. in. at temperatures between 70° and 220°F. The effects of interfacial resistance were considered and uncertainties as to the behavior at the end of the transport path were eliminated. Coefficients were reported with partial pressure and with fugacity as the potential. Fick diffusion coefficients were calculated for each component on the assumption that the gas phase was an ideal solution.

These data indicated that the Maxwell hypothesis with fugacity as the potential in an ideal solution is a fair description of the transport characteristics of the lighter hydrocarbons in the gas phase at relatively low pressures. A regular decrease in the Maxwell diffusion coefficient with an increase in the molecular weight of the stagnant component was observed.

The molecular transport of components in the gas phase has been the subject of many investigations. The work of Maxwell(12) and Stefan(25, 26, 27) laid a satisfactory foundation for the treatment of such processes in situations where the kinetic theory may be applied. Chapman and Cowling(7) extended the analysis to include the effects of changes in composition, and Enskog(8) contributed materially to an understanding of such phenomena in gases at elevated pressures. Recently a review of experimental work in this field was presented(21, 22), and summaries of the basic equations of transport are available(10). It does not therefore appear necessary to review the literature of gas-phase diffusion or to consider in detail the derivation of the basic equations of transport.

Drickamer's(16, 29) active program is an example of the current interest in the field of molecular diffusion* in the gas phase. Little experimental work is available concerning the diffusion coefficients of the lighter hydrocarbons in the gas

phase even for low pressures. Schlinger(21) measured the Maxwell diffusion coefficients of *n*-heptane and *n*-hexane in air at atmospheric pressure for temperatures from 100° to 220°F. Recently Carmichael(4) reported the coefficients of methane and *n*-heptane in the gas phase of the methane-*n*-heptane system. The work was carried out at pressures up to 70 lb./sq. in. in the temperature interval between 100° and 220°F., and the results indicated good agreement of the experimental data with the Maxwell hypothesis.

Drickamer recently reported experimental evidence(24, 30, 31) of a resistance at an interface associated with a transport process. Schrage(23) predicted that such resistances are of primary importance at low pressures where the mean free path of the molecules becomes large. Schlinger(21) and Carmichael(4) found the resistance at the gas-liquid interface of the *n*-heptane-air, the *n*-hexane-air, and the *n*-heptane-methane systems to be small.

The present work involved the measurement of the diffusion coefficients of *n*-hexane in the gas phase of the methane-, ethane-, and propane-*n*-hexane systems for

pressures up to 70 lb./sq. in. in the temperature interval between 70° and 220°F. The experimental measurements were made under steady conditions and regard was taken of the resistance at the interface and of possible variations in the effective transport length as a result of nonuniform conditions at each end of the transfer path. In addition, the Fick diffusion coefficients of each of the components were calculated as a function of pressure, temperature, and composition. Throughout this discussion and in the associated figures the pressures are reported in pounds per square inch absolute or pounds per square foot absolute.

METHODS AND APPARATUS

The details of the methods and the apparatus employed have been described(5). Figure 1 depicts schematically the steady state equipment used for the present measurements. The *n*-hexane in the liquid phase was introduced at a known rate into the chamber A and flowed upward through the porous disk B. After evaporation at the upper surface of the porous disk it diffused through the gas phase in the working section C and was withdrawn from the apparatus at D by circulating the stagnant component, which was brought to equilibrium with *n*-hexane liquid

*Molecular diffusion here refers to that component of material transport which is not due to turbulence and does not imply the very low-pressure phenomena to which this term is also applied.

at 32°F. in the condenser. The rate of introduction of the *n*-hexane into the diffusion cell as a liquid at *B* was measured with a probable error of 0.1% and was maintained by the use of a modulated electric drive for a mechanical injector. The speed of the drive was controlled by means of a quartz oscillator and a suitable electronic circuit(15). Supplemental electrical heaters were provided at *A* of Figure 1 in order to permit the temperature of the liquid evaporating at *B* to be controlled within small limits. Multijunction platinum-constantan thermocouples were employed to establish the temperature of the evaporating liquid with a probable error of less than 0.03°F. Attainment of steady state was indicated by the constancy of the level of the meniscus in the capillary manometer *E*, which also indicated constancy of the exposed surface of liquid in the porous disk. Most of the measurements were made with a capillary depression of approximately 1 in. of *n*-hexane at the evaporating surface *B*.

The mole fraction of *n*-hexane at *D*, the upper end of the transport path, was maintained at a steady small value by the circulation of the stagnant component across the upper end of the diffusion cell. The gaseous mixture of the stagnant component and *n*-hexane being circulated was in equilibrium with a liquid phase of this system at 32°F. at the pressure of measurement. To avoid large effects from convection at the upper end of the diffusion cell, the circulating gas was introduced through the interstices of a tube bundle located in the upper part of the cell. The gas was returned through the interior of the tubes of the bundle to the condenser. Circulation was maintained by means of a small centrifugal blower in the external circuit. Details of the condenser have been described(5).

The temperature of the diffusion path was known with a probable error of 0.05°F. from the international platinum scale, and variations with time and position were less than 0.02°F. The temperature of the surrounding oil bath was controlled by means of a platinum resistance thermometer which actuated a modulating circuit(15) through a galvanometer and photoelectric cell combination. The temperature was related to the international platinum scale by means of a resistance thermometer of the strain-free type(14), which was compared with the indications of a similar instrument which had been calibrated at the National Bureau of Standards.

The pressure of the system was determined by means of a multiple mercury-and-butyl phthalate manometer. The relative elevations of the six phthalate-mercury interfaces were determined by means of a cathetometer, the probable error in the

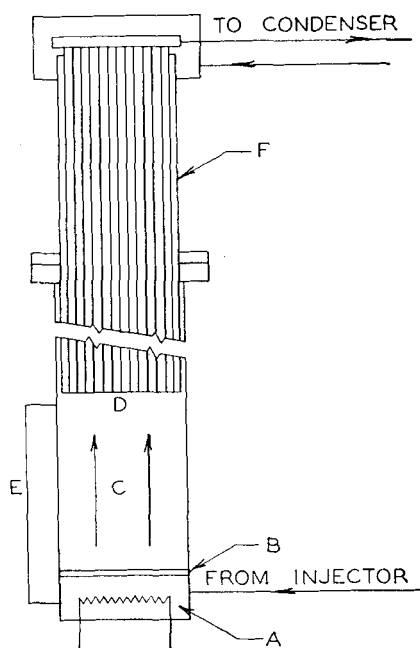


Fig. 1. Schematic arrangement of steady state equipment.

difference in elevation of the interfaces being less than 0.003 in. Corrections were made for local gravity and for the temperature of the phthalate-and-mercury column as well as for irregularities in the pitch of the screw of the cathetometer. The probable error in the measurement of pressure was less than 0.01 lb./sq.in. However, during the investigation of the ethane-*n*-hexane and the propane-*n*-hexane systems measurable decreases in pressure were observed as a result of the large negative partial volume of *n*-hexane in these systems.

The change in length of the transfer path from one set of measure-

ments to another was determined by means of a cathetometer and was known within 0.002 in. after suitable corrections for the effect of temperature upon the dimensions of the equipment(4). An additional correction to the actual length of the transfer path was made in order to take into account the end effects associated with the evaporation of the liquid at the porous plate *B* of Figure 1 and of the disturbance associated with the circulation of the gaseous mixture at *D*(5). The composition of the gas circulated at *D* was determined from the equilibrium behavior of the particular binary system at 32°F. for the pressure in question. This equilibrium information is based upon the data of Boomer and Johnson(3) for the methane-*n*-hexane system and upon ideal solutions(11) for the ethane- and propane-*n*-hexane systems. The measurements of Michels and Nederbragt(13) together with recent compilations(17,19) were used to establish the volumetric behavior of methane, ethane, and propane; whereas the Benedict equation of state with the constants recently suggested by the originator(1) was used to determine the volumetric behavior of *n*-hexane in the gas phase. The fugacities of the components were also determined from the Benedict equation of state.

The diffusion coefficients and the resistance at the interface were established by solution of each of the following equations(21) simultaneously for at least two different transport lengths:

$$D_{M,k} = \frac{\dot{m}_k b_k T (l_G - l_c)}{\ln \left(\frac{p_{j,t}}{p_{j,i} + b_k T r_{i,k} \dot{m}_k} \right)} \quad (1)$$

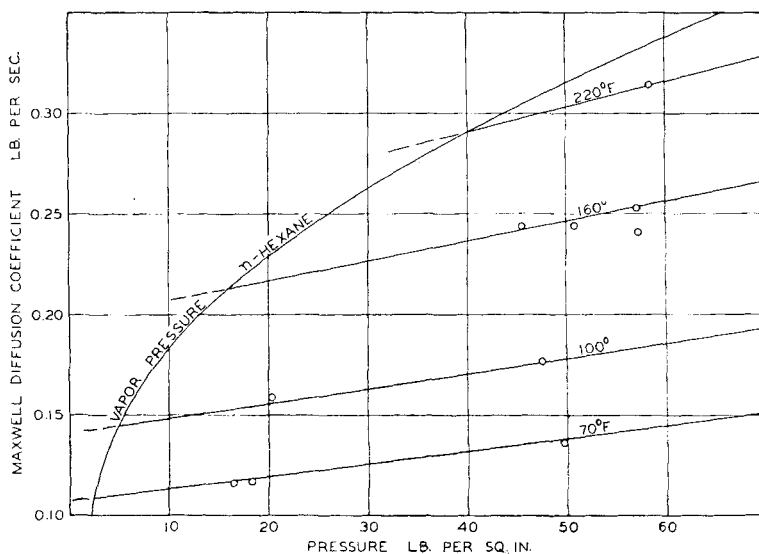


Fig. 2. Diffusion coefficients for *n*-hexane in the methane-*n*-hexane system.

TABLE 1.—MAXWELL DIFFUSION COEFFICIENTS FOR *n*-HEXANE AND PROPERTIES IN THE GAS PHASE

Pressure, lb./sq. ft.	Temperature, transport path, °F.	Compressibility factor Stagnant component	<i>n</i> -Hexane	Vapor pressure <i>n</i> -hexane, lb./sq. ft.	Fugacity of pure <i>n</i> -hexane, lb./sq. ft.	Diffusion coefficient Perfect gas, lb./sec.	Ideal solution, lb./sec.
Methane- <i>n</i> -hexane							
2,361	69.95	0.9976	0.9279	354.38	2,197	0.1149	0.1161
7,141	69.74	0.9936	0.7360	354.24	5,445	0.1345	0.1358
2,627	69.97	0.9974	0.9197	354.10	2,423	0.1164	0.1164
2,929	100.00	0.9978	0.9300	711.36	2,727	0.1557	0.1584
6,836	99.98	0.9950	0.8227	717.40	5,668	0.1730	0.1764
6,545	159.99	0.9967	0.9001	2,279.52	5,877	0.2357	0.2439
8,223	160.02	0.9960	0.8719	2,282.40	7,147	0.2438	0.2527
7,366	159.98	0.9964	0.8875	2,268.00	6,556	0.2354	0.2439
8,250	160.00	0.9960	0.8700	2,276.64	7,168	0.2323	0.2400
8,403	220.00	0.9972	0.9180	5,748.48	7,679	0.2943	0.3130
Ethane- <i>n</i> -hexane							
2,265	70.27	0.9920	0.9300	357.26	2,115	0.0902	0.0917
3,061	100.54	0.9907	0.9265	720.00	2,842	0.1080	0.1107
4,353	100.00	0.9867	0.8959	712.80	3,862	0.1145	0.1171
7,276	99.94	0.9777	0.8091	711.36	6,006	0.1132	0.1167
4,473	160.60	0.9899	0.9325	2,299.70	4,190	0.1536	0.1607
8,275	160.32	0.9813	0.8705	2,289.60	7,183	0.1461	0.1534
4,250	160.59	0.9904	0.9363	2,278.10	3,995	0.1525	0.1593
7,537	219.95	0.9873	0.9265	5,739.80	6,957	0.1856	0.1988
Propane- <i>n</i> -hexane							
2,245	70.00	0.9829	0.9342	354.38	2,093	0.0443	0.0453
2,919	100.00	0.9814	0.9308	712.80	2,694	0.0742	0.0765
3,349	100.00	0.9785	0.9195	713.52	3,089	0.0697	0.0721
4,320	160.00	0.9793	0.9353	2,278.10	4,037	0.1074	0.1128
8,887	159.68	0.9565	0.8592	2,263.70	7,614	0.0976	0.1050
4,859	160.30	0.9767	0.9270	2,288.20	4,499	0.1095	0.1148
8,773	160.23	0.9570	0.8613	2,291.00	7,531	0.0938	0.1007
8,718	220.32	0.9679	0.9149	5,771.50	7,847	0.1428	0.1532

$$D_{M,k} = \frac{\dot{m}_k b_k T (l_G - l_c)}{Z \left(\frac{f_k}{P} \right) \ln \left(\frac{f_k - f_{k,i}}{f_k - f_{k,i} + b_k T r_{i,k} \dot{m}_k} \right)} \quad (2)$$

Equation (1) assumes perfect gas behavior and employs partial pressure as the potential, whereas Equation (2) is based upon an ideal solution with fugacity as the potential. By proper choice of path lengths and transport rates it was possible to isolate to a large extent the effect upon the diffusion coefficient of uncertainties in transport length and of resistance at the interface. As was indicated earlier(5), the corrections to the transport length were found to be a single-valued function of the Reynolds number of the flow in the tube bundle F of Figure 1, which brought the circulating gas to the upper end of the transport path.

Throughout all this work the weight rate of flow of the gaseous mixtures to the upper end of the transport path was maintained at known values between 4×10^{-6} and 8×10^{-6} lb./sec. The Reynolds number was taken as a linear function of the weight rate of flow at a given state, as indicated in the following expression:

$$\frac{Re}{Re^+} = a \left(\frac{\eta}{\eta^+} \right) (\dot{m}_i' + \dot{m}_k') \quad (3)$$

In estimating the Reynolds number of the flow in the tubes at the upper end of the diffusion path, suitable

small corrections were made for the effect of temperature and pressure upon the viscosity of methane, ethane, and propane when mixed with *n*-hexane(2, 18, 20). Available calibrations(5) were employed to establish quantitatively the influence of the Reynolds number upon the effective length of the diffusion path.

MATERIALS

The sample of deaerated *n*-hexane, obtained from the Phillips Petroleum Company, had a specific weight of 40.8817 lb./cu.ft. at 77°F. and atmospheric pressure, as compared with a value of 40.87932 reported by Rossini(17) for an air-saturated sample. The index of refraction at 77°F. relative to the D lines of sodium was 1.3737, as compared with the value for an air-saturated sample of 1.37226 reported by Rossini(17) for the same temperature. It is believed that the sample employed contained only traces of air and less than 0.002 mole fraction of material other than *n*-hexane.

Methane was obtained from the San Joaquin Valley through the courtesy of The Texas Company and as received contained less than 0.002 mole fraction of impurities. Before use it was passed over calcium chloride, activated charcoal, and an-

hydrous calcium sulfate at a pressure in excess of 200 lb./sq.in. A spectroscopic analysis indicated that methane obtained from this source and subjected to the above-described treatment contained less than 0.001 mole fraction of heavier hydrocarbons, nitrogen or argon.

Ethane and propane were obtained from the Phillips Petroleum Company. The ethane was reported to contain less than 0.0006 mole fraction of impurities and was employed without further purification. There was no indication upon freezing in liquid air that the sample contained any quantities of volatile components. A spectroscopic analysis of the sample indicated that it contained less than 0.0019 mole fraction of material other than ethane. The impurity was predominantly ethene. The propane, reported to contain less than 0.002 mole fraction of impurities, was subjected to a fractionation similar to that described for *n*-hexane. The sample employed contained less than 0.0001 mole fraction of material other than propane, as indicated by spectroscopic analysis.

RESULTS

A total of twenty-six measurements of the rate of transport of *n*-hexane was made in the gas phase of the methane-*n*-hexane, ethane-*n*-hexane, and propane-*n*-hexane systems; the data therefrom are available(6). The results of the calculation of the diffusion

coefficients from these experimental data are recorded in Table 1, which includes the pressure, temperature, compressibility factor, vapor pressure, fugacity, and diffusion coefficients for each of the states investigated. The vapor pressure of *n*-hexane was based upon the critical review of Rossini(17). The fugacities were calculated from the Benedict equation of state utilizing the coefficients proposed by the originator(1). The equation of state was employed to predict the fugacity and volumetric behavior for *n*-hexane at pressures in excess of vapor pressure. In the application of Equation (1) it was assumed that the phases were perfect gases and in the use of Equation (2) that they were ideal solutions.

The Maxwell diffusion coefficients were calculated from Equations (1) and (2) by means of available information(5) concerning the small influence of Reynolds

number, described in Equation (3), upon the effective length of the diffusion path. For these relatively low pressures there is little to choose between the diffusion coefficients calculated on the basis of a perfect gas and those derived from the behavior ascribed to an ideal solution. Throughout the remainder of the discussion the Maxwell diffusion coefficients based upon fugacity as a potential in an ideal solution were employed.

Maxwell diffusion coefficients for *n*-hexane in the gas phase of the methane-*n*-hexane system are shown in Figure 2. The curves were drawn to indicate a substantial increase in the coefficients with an increase in pressure, as was found in the methane-*n*-heptane system(4). A firm evaluation of such trends with pressure must await measurements over a wider range of this variable than was covered in the present study. The data shown in Figure 2 yielded a

standard deviation of 0.00475 lb./sec. for the experimental data from the straight lines drawn through the points for each of the four temperatures. Smoothed values of this coefficient for *n*-hexane are recorded in Table 2. It should be realized that the apparent effect of pressure found in the methane-*n*-hexane system at the lower temperatures is an indication that the Maxwell hypothesis(12) does not describe the transport process with precision.

Experimental data for the Maxwell diffusion coefficients for *n*-hexane in the ethane- and in the propane-*n*-hexane systems are shown in Figures 3 and 4 respectively. In these instances there was no marked tendency for the coefficients to change with pressure; however, in the case of the propane-*n*-hexane system the data indicated a possible tendency for the Maxwell diffusion coefficient to decrease with an increase in pressure. Some difficulty was experienced in the experimental work for the propane-*n*-hexane system as a result of the diffusion of propane through the liquid *n*-hexane in the porous-glass disk *B* of Figure 1. It was necessary to eliminate some of the measurements involving the longer transfer paths since the rate of diffusion of propane through the disk under steady conditions was significant. With shorter transfer paths the gross motion of the *n*-hexane liquid in the pores of the disk more than compensated for the molecular diffusion of the propane into the *n*-hexane liquid. The standard deviation of the measurements for the ethane-*n*-hexane system was 0.003 and in the propane-*n*-hexane system 0.005 lb./sec. from the smoothed data. Table 3 records smoothed values of the Maxwell diffusion coefficient for *n*-hexane in the ethane- and propane-*n*-hexane systems. Since the data of Figures 3 and 4 indicated no significant influence of pressure upon the Maxwell diffusion coefficient, there was no need to include the latter variable in the tabulation, as was done in Table 2 for the methane-*n*-hexane system.

The influence of temperature upon the Maxwell diffusion coefficient is shown in Figure 5. The behavior of *n*-hexane in the methane-*n*-hexane system was compared at a pressure of 40 lb./sq.in. with the behavior in the ethane- and propane-*n*-hexane systems, which were found not to be influenced by pressure. The diffusion coefficient for *n*-hexane in-

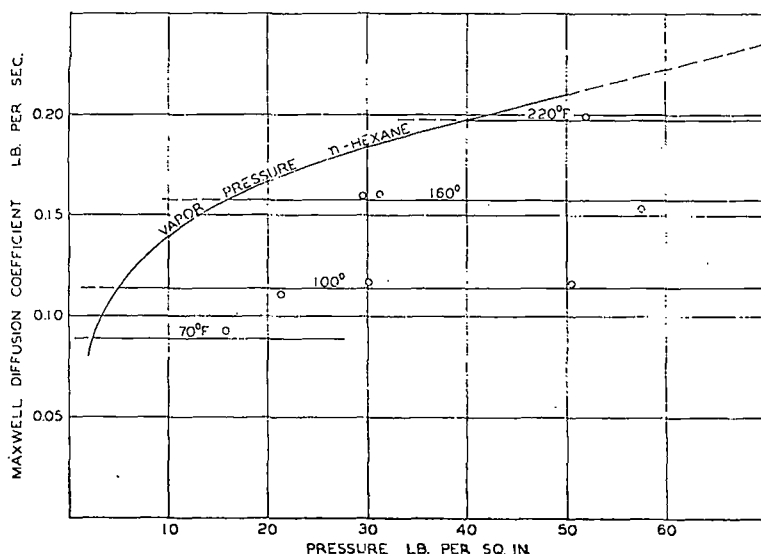


Fig. 3 Diffusion coefficients for *n*-hexane in the ethane-*n*-hexane system.

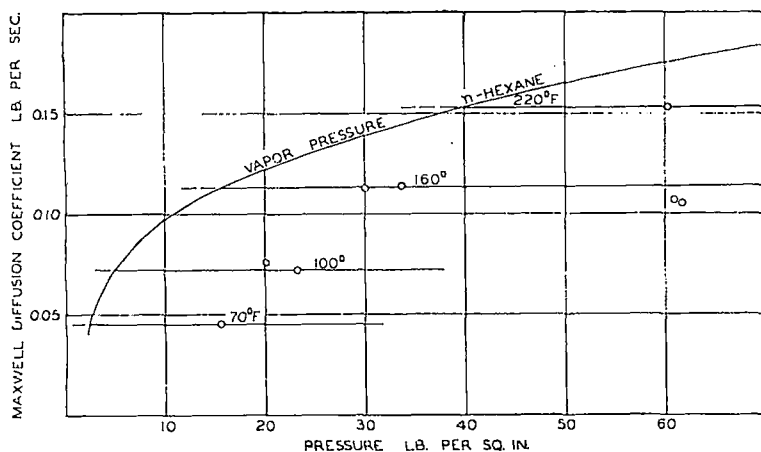


Fig. 4. Diffusion coefficients for *n*-hexane in the propane-*n*-hexane system.

TABLE 2.—MAXWELL DIFFUSION COEFFICIENTS FOR *n*-HEXANE IN THE METHANE-*n*-HEXANE SYSTEM

Temperature, °F.	Pressure, lb./sq. in.					
	14.696	20	30	40	50	60
70	0.1160*	0.1190	0.1255	0.1320	0.1380	0.1443
100	0.1520	0.1560	0.1634	0.1710	0.1765	0.1855
130	0.1811	0.1876	0.1962	0.2049	0.2128	0.2211
160	0.2168	0.2262	0.2368	0.2458	0.2555
190	0.2545	0.2645	0.2762	0.2863
220	0.2909	0.3031	0.3155

*Maxwell diffusion coefficients are expressed in pounds per second.

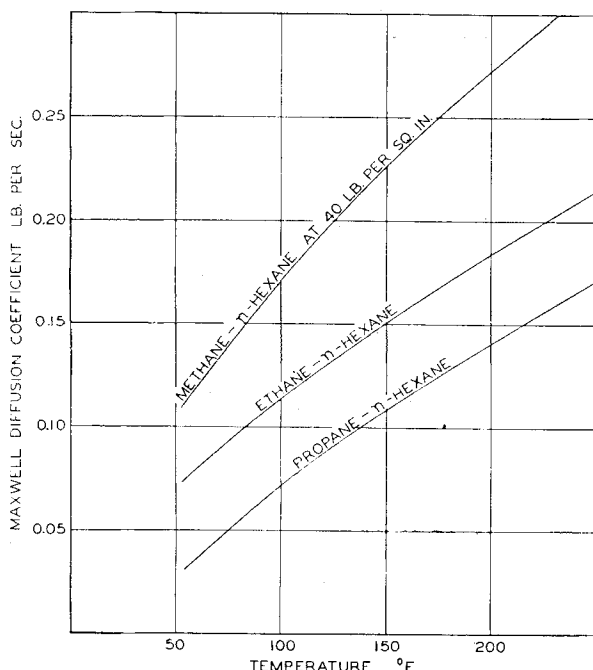


Fig. 5. Effect of temperature on diffusion coefficients for *n*-hexane.

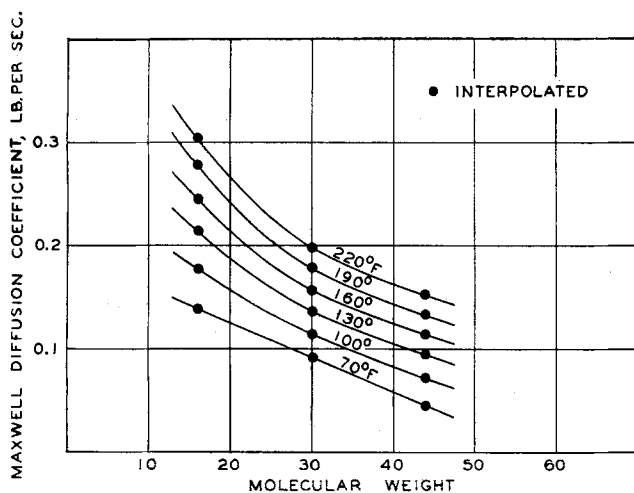


Fig. 6. Effect of molecular weight of stagnant component on diffusion coefficient for *n*-hexane.

creases more rapidly with an increase in temperature for the methane-*n*-hexane system than it does for either the ethane- or the propane-*n*-hexane systems. A pressure of 40 lb./sq. in. was chosen for the methane data in order to permit comparison throughout the temperature range investigated

and still involve as low a value as feasible.

Figure 6 depicts the effect of the molecular weight of the stagnant component upon the Maxwell diffusion coefficients for *n*-hexane at several temperatures. Again the data for methane at a pressure of 40 lb./sq.in. were compared with

TABLE 3.—MAXWELL DIFFUSION COEFFICIENTS FOR *n*-HEXANE IN THE ETHANE-*n*-HEXANE AND PROPANE-*n*-HEXANE SYSTEMS

Temperature, °F.	Ethane- <i>n</i> -hexane	Propane- <i>n</i> -hexane
70	0.0890*	0.0450
100	0.1136	0.0720
130	0.1361	0.0948
160	0.1571	0.1133
190	0.1778	0.1336
220	0.1980	0.1525

*Maxwell diffusion coefficients are expressed in pounds per second and are considered to be independent of pressure.

the diffusion coefficients in the other systems, which were independent of pressure. The experimental points correspond to those recorded in Tables 2 and 3. There is a rapid decrease in the diffusion coefficient with an increase in molecular weight, as would be predicted from the Sutherland equation(28). The regularity of decrease in the diffusion coefficient with an increase in molecular weight offers encouragement in regard to predictions of the transport behavior of hydrocarbon gases.

FICK DIFFUSION COEFFICIENTS

The Fick diffusion coefficient (9) may be defined by

$$D_{F,k} = - \frac{\dot{m}_{k,d}}{\left(\frac{\partial \sigma_k}{\partial x} \right)} \quad (4)$$

Equation (4) applies only to one dimensional transport, and the component flux is that resulting from molecular diffusion and does not include the transport associated with any gross motion of the fluid. It may be shown that the Fick coefficients are related to the Maxwell coefficients for a perfect gas in the following way:

$$D_{F,k} = \left(\frac{n_j}{n_j} \right) \frac{D_{M,k}}{P} \quad (5)$$

In case the gas phase follows the behavior of an ideal solution but may deviate from perfect gas behavior, Equation (5) assumes the form

$$D_{F,k} = \frac{\left(\frac{n_j}{n_j} \right) \left(\frac{f_k}{P} \right) \left(\frac{Z^2}{P} \right) D_{M,k}}{[1 + \sigma_k (\dot{V}_j - \dot{V}_k)]} \quad (6)$$

By utilization of Equation (6) the Fick diffusion coefficients for n -hexane were computed from the Maxwell coefficients as a function of pressure, temperature, and composition and are available(6) for the methane- n -hexane, ethane- n -hexane, and propane- n -hexane systems.

The Fick diffusion coefficients for the two components are related by the following expression:

$$D_{F,j} \frac{\partial \sigma_j}{\partial x} = -D_{F,k} \frac{\partial \sigma_k}{\partial x} \quad (7)$$

Equation (7) may be simplified to

$$D_{F,j} = \frac{\sigma_k^0}{\sigma_j^0} D_{F,k} \quad (8)$$

Equation (8) and the information, already discussed, concerning the volumetric behavior of each of the components were utilized to compute the Fick diffusion coefficients for methane, ethane, and propane, which are available(6). The Fick diffusion coefficients are markedly functions of pressure, temperature, and composition. They are discussed here because it is often more convenient to employ such coefficients in the analysis of transport processes involving hydrodynamic velocities than the Maxwell coefficients.

The results obtained in this investigation indicate that the Maxwell hypothesis(12) is a reasonable description of the transport phenomena in the gas phase of the binary systems of methane, ethane, and propane with n -hexane at low pressures. The differences in composition within the experimental transport path were significant, particularly at pressures approaching the vapor pressure of n -hexane. However, there was only a small change in the coefficient for the methane- n -hexane system with pressure and none for the other systems. Such behavior indicates that the Maxwell hypothesis took into account the effect of composition upon the transport to at least a first degree of approximation.

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NOTATION

- a = dimensional constant
- b = specific gas constant of phase $\langle R/M \rangle$, ft./°R.
- b_k = specific gas constant of component k , $\langle R/M \rangle$, ft./°R.
- $D_{F,k}$ = Fick diffusion coefficient of component k , sq.ft./sec.
- $D_{M,k}$ = Maxwell diffusion coefficient of component k , lb./sec.
- f_k = fugacity of component k in the phase, lb./sq.ft.
- f_k^0 = fugacity of component k in the pure state, lb./sq.ft.
- l_c = effective correction to gross transfer distance, ft.
- l_G = gross transfer distance, ft.
- M = average molecular weight of phase
- \dot{m}_k = weight rate of transport of component k , lb./sec. (sq. ft.)
- \dot{m}_k' = total weight rate of circulation of component k , lb./sec.
- n_k = weight fraction of component k
- n_k = mole fraction of component k
- P = pressure, lb./sq.ft.
- p_j = partial pressure of component j , (Pn_j) , lb./sq. ft.
- R = universal gas constant, ft./°R.
- r_i = resistance at interface, sec./ft.
- Re = Reynolds number
- T = thermodynamic temperature, °R.
- V_k = molal volume of component k , cu.ft./lb.mole
- x = distance in direction of diffusion path, ft.
- Z = compressibility factor, PV/bT
- η = absolute viscosity, (lb.) (sec.)/sq.ft.
- σ_k = concentration of component k , lb./cu.ft.
- σ_k^0 = molal concentration of component k , lb.mole/cu.ft.
- ∂ = partial differential operator

Superscripts

- 0 = pure state
- $+$ = reference state

Subscripts

- d = diffusion
- i = conditions at interface
- j = component j , the stagnant component
- k = component k , the diffusing component
- t = conditions at exit of transfer tube

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