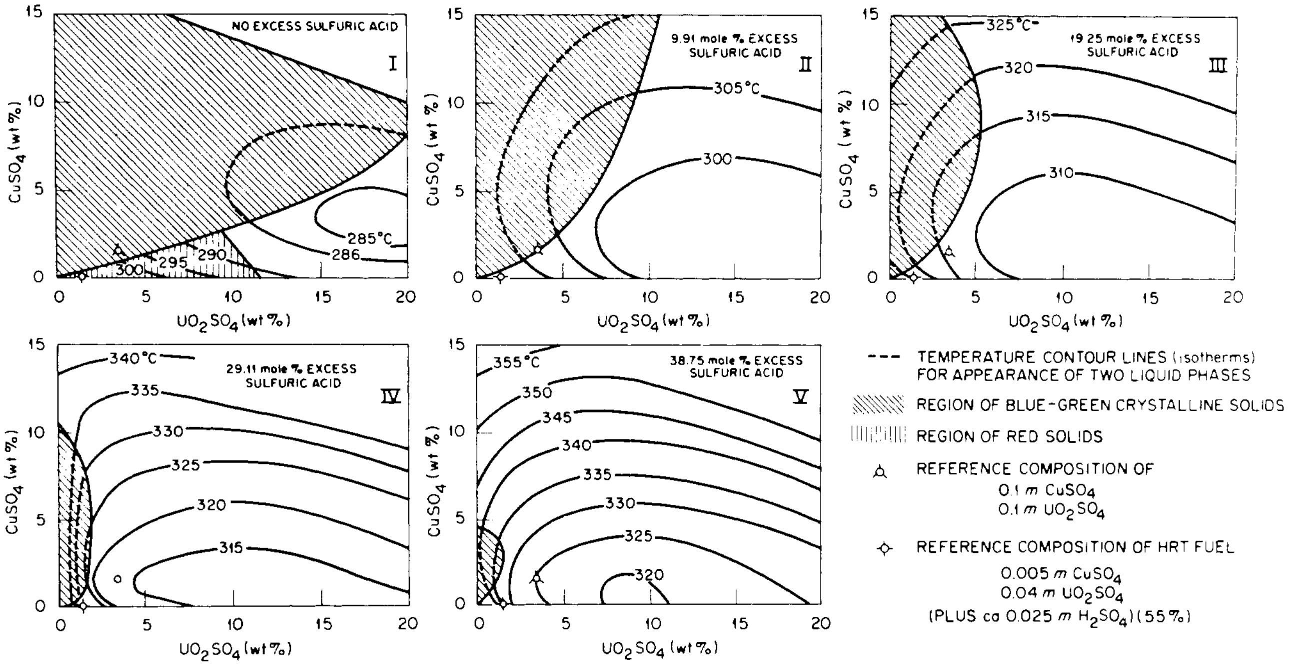

Figure 1. Phase-transition temperatures in solutions containing cupric sulfate, uranyl sulfate, and sulfuric acid

against one of the composition variables yields smooth curves from which interpolation values may be obtained. Figure 1 shows temperature contour plots obtained in this manner for each of the five acidity values. The regions in which solid phases were encountered are indicated. A comparison shows that as the amount of excess sulfuric acid is increased the scope of the precipitation regions shrinks and that the temperature at which liquid phase separation occurs for a specific concentration is elevated. If sufficient acid is present to prevent precipitation of a solid, pure copper sulfate-sulfuric acid solutions will yield a second liquid phase in a manner completely analogous to the behavior or uranyl sulfate solutions.

Solution compositions falling within the precipitation regions are probably of no interest for reactor use, because of the temperature limitation. The data in the two-liquid phase region provide an upper temperature limit for reactor use for any composition within the scope of this study. Relying on past experimental data (*4*), the two liquid phase appearance temperatures for the heavy water system are expected to be of the order of 5° to 10°C. lower than for the corresponding ordinary water system.

LITERATURE CITED

(1) McDuffie, H. F., Compere, E. L., Stone, H. H., Woo, L. F., Secoy, C. H., "Homogeneous Catalysis for Homogeneous Reactors. Catalysis of the Reaction Between Hydrogen and Oxygen," Southwide Chemical Conference, ACS, Memphis, Tenn., Dec. 6, 1956.

(2) Posnjak, E., Tunell, G., *Am. J. Sci.* **218,** 1-34 (1929).

(3) Secoy, C. H., *J. Am. Chem. Soc.* **72,** 3343 (1950).

(4) Secoy, C. H., others, "The Reactor Handbook AECD-3646," Vol. 2, 1st ed., Chap. 4.3, p. 559, Technical Information Service, U. S. Atomic Energy Commission, Oak Ridge, Tenn., May 1955.

Received for review May 24, 1958. Accepted October 9, 1958. Division of Industrial and Engineering Chemistry, Symposium on Chemistry and Reprocessing of Circulating Nuclear Reactor Fuels, 133rd meeting, ACS, San Francisco, Calif., April 1958. Based on work performed at Oak Ridge National Laboratory, operated by Union Carbide Corp. for the Atomic Energy Commission. Other articles from this symposium will be published in the February 1959 issue of *Industrial and Engineering Chemistry*.

Diffusion Coefficients in Hydrocarbon Systems. Methane in the Liquid Phase of the Methane-Santa Fe Springs Crude Oil System

H. H. REAMER and B. H. SAGE
California Institute of Technology, Pasadena, Calif.

Little experimental work is available concerning the molecular transport of methane in the liquid phase of hydrocarbons except the earlier work of Pomeroy (*10*) and of Lacey and others (*1, 4, 5, 7*). More recently, interest in this field has revived and at the present time some information is available concerning the transport of methane in binary systems made up of this hydrocarbon and the paraffin hydrocarbons from propane through *n*-decane, with the exception of octane and nonane (*11, 13-16*). Kirkwood (*6*) has set forth some of the basic relationships of molecular transport and these have been extended to a number of situations of particular interest to petroleum production (*9*).

The work of Drickamer has made a marked contribution to an understanding of transport in liquid and gas phases at elevated pressures. His studies, directed toward an understanding of the behavior at a gas-liquid interface (*24, 25*),

Table I. Analytical Distillation of Santa Fe Springs Crude Oil[a]

Fraction No.	Still Vapor Temp., °F.	Cumulative Recovery, Vol. %	°API at 60°F.	ASTM Engler Dist. Type	Distillation Temperature, °F.					Av. Boiling Point, °F.
					Initial	10%	50%	90%	Maximum	
1	186	4.90	80.0	Natural gasoline	68	92	146	244	266	156.7[b]
	186	6.91	80.5	Natural gasoline	78	95	133	176	197	134.5
	174	5.0	81.1	Natural gasoline	76	98	134	188	228	139.6
2	213	9.77	65.9	Gasoline	110	146	188	236	298	191.0
	213	10.80	60.3	Gasoline	...	...	...	...	...	...
	209	10.0	62.5	Gasoline	138	167	188	218	262	191.2
3	240	14.74	59.3	Gasoline	148	184	214	262	316	219.4
	240	14.82	55.7	Gasoline	...	...	...	...	...	...
	242	15.0	57.1	Gasoline	168	201	215	252	300	221.6
4	260	19.69	55.3	Gasoline	184	210	238	284	336	244.5
	260	18.49	...	Gasoline	...	...	...	...	...	...
	267	20.0	54.2	Gasoline	218	228	242	276	320	250.2
5	292	24.64	51.8	Gasoline	220	242	264	306	360	272.0
	292	23.16	...	Gasoline	...	...	...	...	...	...
	301	25.0	50.8	Gasoline	248	258	274	304	346	280.2
6	338	29.61	47.6	Gasoline	264	286	306	345	406	314.6
	338	29.17	...	Gasoline	...	...	...	...	...	...
	341	30.0	47.2	Gasoline	282	292	308	344	386	315.6
7	373	34.68	44.1	Gasoline	320	332	348	380	424	354.5
	373	33.22	...	Gasoline	...	...	...	...	...	...
	387	35.0	43.8	Gasoline	320	330	346	381	430	354.2
8	420	39.73	40.9	Gasoline	368	380	393	416	438	396.1
	420	37.58	...	Gasoline	...	...	...	...	...	...
	432	40.0	40.9	Gasoline	362	374	391	426	467	397.8
9	458	44.70	38.8	Kerosine	408	430	440	456	476	441.6
	458	42.44	...	Kerosine	...	...	...	...	...	...
	472	45.0	39.1	Kerosine	407	416	431	461	502	437.3
10	494	50.40	36.5	Kerosine	446	474	482	498	516	483.0
	494	47.51	...	Kerosine	...	...	...	...	...	...
	518	50.0	36.5	Kerosine	445	459	474	503	540	479.2
11	625	60.65	33.9	Gas oil	514	540	555	587	620	559.8
	625	60.28	...	Gas oil	...	...	...	...	...	...
	605	60.0	33.7	Gas oil	494	516	535	565	597	538.6
12	740	71.02	28.7	Gas oil	600	630	651	686	706	653.5
	740	70.28	...	Gas oil	...	...	...	...	...	...
	705	70.0	29.9	Gas oil	588	608	624	651	675	627.2
Residue	...	28.98	15.6							
	...	29.77	...							
	...	30.0	15.8							

[a] Composition expressed in mass per cent: methane, trace; ethane, 0.014; propane, 0.216; isobutane, 0.286; *n*-butane, 0.726; isopentane, 0.671; *n*-pentane, 0.789.

[b] (*21*).

are of particular interest. Interest in the resistance at an interface (*3*) has continued but the work of Schrage (*23*), which has been substantiated by the experimental work of Schlinger (*22*), indicated that at pressures in excess of several atmospheres the influence of the resistance at the interface is negligible.

In the present investigation measurements of the Fick diffusion coefficient of methane in the liquid phase of the methane-Sante Fe Springs crude oil system (referred to here as crude oil) at pressures up to 3600 p.s.i. in the temperature interval between 40° and 280°F. were made. The upper limit of pressure was established by the possible separation of an asphaltic phase (*2*).

METHODS AND APPARATUS

The methods employed were similar to the transient methods used by Lacey and others (*10*). In principle, they involve the measurement of the rate of introduction of methane into a quiescent, isochoric, two-phase mixture of methane and crude oil previously brought to equilibrium at a somewhat lower pressure. The pressure over the equilibrium heterogeneous mixture was increased rapidly by a predetermined increment and the quantity of methane required to maintain this variable weight system under isobaric, isothermal conditions was determined. The equipment employed has been described (*13*).

The quantity of crude oil employed was determined gravimetrically by weighing bomb techniques (*20*). The uncertainty in the weight of crude oil introduced was less than 0.05%. Pressures were measured with a probable error of 0.1 p.s.i. or 0.05%, whichever was the larger measure of uncertainty. The constancy of the pressure was established by means of a slack-diaphragm type of manostat (*17*). It is believed that the pressure was related to the initial value within 0.03 p.s.i. Temperatures were determined with a platinum resistance thermometer of the strain-free type (*8*). The temperature of the system was related to the international platinum scale within 0.02° F.

The rate of introduction of the methane was adjusted automatically (*18*) to maintain isobaric conditions, and the quantity of methane introduced was recorded in a digital fashion as a function of time. Measurements were taken at intervals of approximately 5 minutes during the diffusion process. The uncertainty in the volumetric determination of the quantity of methane introduced was less than 0.1% of the total quantity added during the period of measurement.

From the weight of methane introduced into the isochoric vessel as a function of time, the Fick diffusion coefficient may be evaluated (*13*) from the relation

$$D^*_{F,k} = \left[\frac{(1 + \bar{V}^*_{j,l}\,\sigma_j + \bar{V}^*_{k,l}\,\sigma_k)^*}{(1 + \bar{V}^*_{j,l}\,\sigma_j)^*\left(1 + \dfrac{\sigma_k}{\sigma_j}\right)^*}\right]\left[\frac{\pi\, m_k^2}{4\theta(\sigma_{k,l,e} - \sigma_{ko})^2}\right] \times \left[\frac{\bar{V}^*_{k,g}}{\bar{V}^*_{k,g} - \bar{V}^*_{k,l}}\right]^2 \qquad (1)$$

In the derivation of Equation 1 (*13*) it was assumed that local equilibrium (*6*) existed and the quantities within the first and third brackets of Equation 1 were evaluated from interpretation of available equilibrium data (*19*, *21*).

The first bracket takes into account the hydrodynamic velocity (*13*) in the liquid phase, whereas the third bracket is the ratio of the quantity of methane added to that crossing the interface. In other words, the third bracket takes into account the change in the total volume of the liquid phase.

It is possible to derive expressions similar to Equation 1 which take into account the resistance at the interface. Experimental measurements (*22*) have shown, however, that the resistance at the interface can be neglected. The negligibly small deviation from a linear relationship between the weight of methane transported across the interface and the square root of time confirms this point of view. It is not necessary to take into account any resistance in the gas phase if local equilibrium exists at the interface. The composition on the liquid-phase side of the interface is solely a function of the prevailing temperature and pressure in so far as the system can be treated as binary.

A typical set of experimental results obtained at an operating pressure of 424.8 p.s.i. and a temperature of 220° F. is shown in Figure 1. The large number of experimental points resulted from the automatic digital recording equipment described in an earlier paper (*18*). The standard deviation of the experimental points from a straight line was 1.37×10^{-6} pound, for the time period indicated between the two vertical arrows. This standard deviation is based upon the assumption that all of the error lies in the weight of methane introduced into the vessel and that there is none in the time.

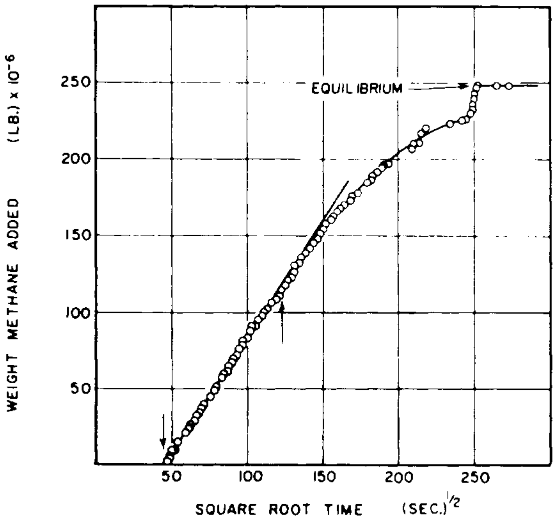

Figure 1. Typical experimental measurements at 220° F.

The total quantity of methane introduced into the isochoric vessel for the attainment of equilibrium at the higher pressure is indicated by the topmost arrow in Figure 1. It should be emphasized that Equation 1 serves as a satisfactory representation of the Fick diffusion coefficient only for situations in which the quantity of methane crossing the interface is not more than half of that required to bring the entire phase to equilibrium. In later stages in the diffusion process the influence of the finite length of the diffusion path becomes significant (*13*).

MATERIALS

The methane used in this investigation was obtained from a well in the San Joaquin Valley of California. The methane sample was passed successively over calcium chloride, freshly activated charcoal at dry ice temperature, Ascarite, and anhydrous calcium sulfate at room temperature. It was then passed through a spiral coil immersed in a mixture of dry ice and trichloroethylene. All of these processes were carried out at pressures in excess of 300 p.s.i. Special spectroscopic analysis indicated that the methane, after being subjected to the above-described treatment, contained less than 0.0003 mole fraction of material other than methane.

The crude oil was obtained in 1935 from a well in the Santa Fe Springs Field. It was stored in a sealed steel drum and a solvent extraction analysis yielded results which are available (*21*). A second, more recent analysis from a sample obtained from the steel drum after stirring is reported in Table I. The deviations are considered to be insignificant. The other pertinent physical properties of this crude oil, which has a gravity of 34.9° API at 60° F., are reported in Table I.

EQUILIBRIUM DATA

As indicated by Equation 1, detailed information concerning the partial volumetric behavior of the components of this system, which is being treated as though binary, is required to interpret the diffusion measurements. An earlier study of the methane-crude oil system was employed for this purpose. The associated partial volumetric behavior of methane and of the crude oil is available (*19*). These data were extrapolated slightly, with respect to both temperature and pressure, to obtain the necessary information. Figure 2 shows the volumetric correction factor for methane in the liquid phase of the methane-crude oil system as a function of pressure and temperature. This factor is the first bracketed term of Equation 1. Similar information for the weight correction factor for methane in this system is shown in Figure 3. This factor is the last bracketed term of Equation 1. The deviation of these correction factors from unity is much smaller for the methane-crude oil system than is encountered with such systems as methane-*n*-butane (*15*). The necessary values of concentration of the two components were obtained from the original volumetric study (*21*).

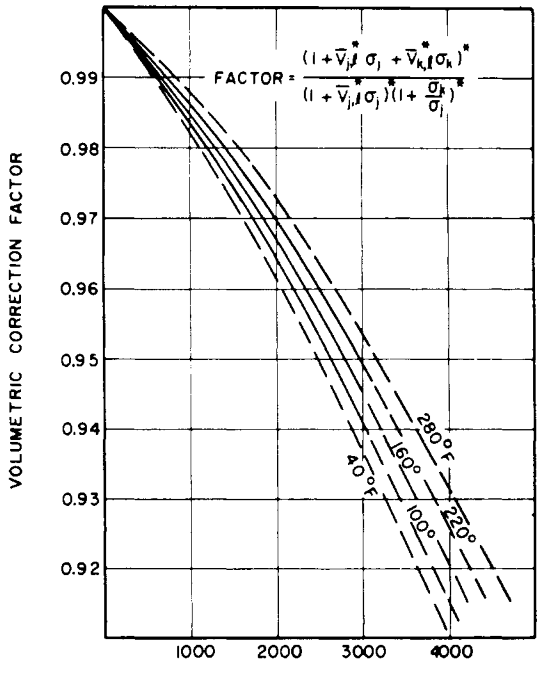

Figure 2. Volumetric correction factor for methane-Santa Fe Springs crude oil system

EXPERIMENTAL DATA

Information similar to that shown in Figure 1 was obtained at other temperatures between 40° and 280°F. A sample of the detailed experimental results obtained for the measurements covered in Figure 1 is set forth in Table II, which includes approximately 25% of the tabular information obtained.

Table III records the experimental information obtained from the application of a least squares fit to the linear relationship between the weight of methane crossing the interface and the square root of time, together with the associated experimental conditions. In addition, the values of the volumetric correction factors have been included. From these the Fick diffusion coefficient as described by Equation 1 was computed. Values both with and without consideration of the effect of hydrodynamic velocity are reported in Table III. The standard errors of estimate reported in Table III are based upon the assumption that all of the error is associated with the weight of methane crossing the interface and none in connection with the time.

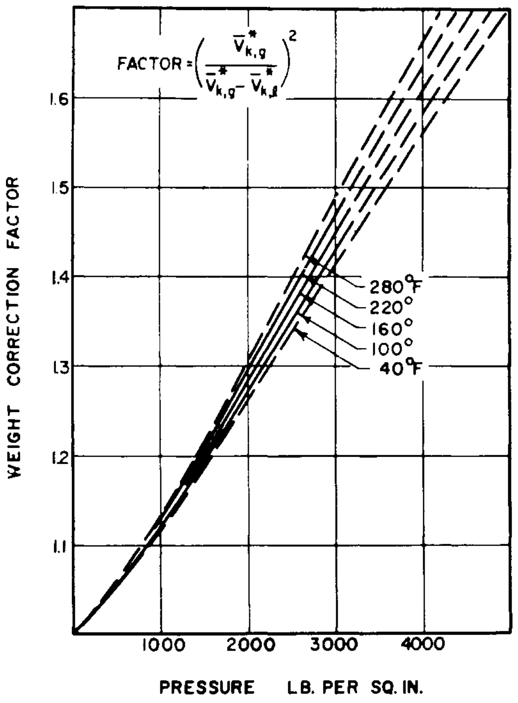

Figure 3. Weight correction factor for methane-Santa Fe Springs crude oil system

The experimental results obtained in this investigation are presented as a function of pressure in Figure 4. Lines of constant composition have been included as a matter of interest. The experimental points shown in Figure 4 were located at a linear average of the initial and final pressures used in each measurement. A substantial part of the deviation of the experimental data from the smooth curves may be ascribed to uncertainties in the equilibrium data, particularly in values of the partial volumes of methane and of the crude oil. Likewise, uncertainties in the values of the concentration required for the solution of Equation 1 add to the

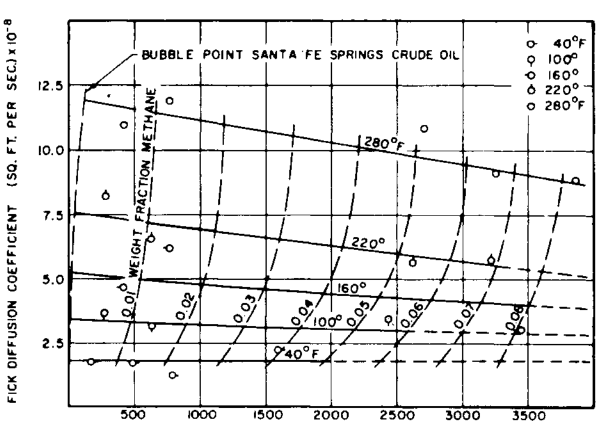

Figure 4. Effect of pressure upon Fick diffusion coefficient for methane

Table II. Sample of Experimental Measurements at 220° F.

Pressure, P.S.I.A.	Weight Fraction Methane Liquid Phase	Time, Sec.	Methane Added[a], Lb. × 10^{-6}
125.6[b]	0.0012	0	...
424.8[c]	0.0068	2170	...
		2229	1.952
		2319	3.905
		2369	5.857
		2419	7.810
		2469	9.762
		2769	11.715
		2949	15.620
		3499	21.477
		3699	23.429
		3789	25.382
		3809	27.334
		4169	29.287
		4269	31.239
		4319	33.191
		4619	35.144
		4819	37.096
		5019	39.049
		5719	44.906
		6019	48.811
		6269	50.763
		6869	56.621
		7019	58.573
		7219	60.526
		7369	62.478
		7669	64.431
		7819	66.382
		8019	68.335
		8119	70.288
		8469	72.240
		8819	74.193
		8919	76.145
		9119	78.098
		9219	80.050
		9819	82.002
		10019	83.955
		10219	85.907
		10269	87.860
		10719	89.812
		11068	91.765
		11518	95.670
		12068	97.622
		12368	99.574
		12718	101.527
		13568	107.384
		14068	109.337
		14568	111.289
		14968	115.194
		15468	117.146

[a] Weight added to heterogeneous isochoric system.

[b] Initial equilibrium pressure.

[c] Constant operating pressure during diffusion.

lack of agreement of the experimental data with the smooth curves presented.

The standard deviation of the experimental measurements shown in Figure 4 from the smooth curves was 0.6 x 10^{-8} square foot per second, when it was assumed that all of the uncertainty lay in the Fick diffusion coefficient and none in the determination of the temperature, pressure, or composition. Smoothed values of the Fick diffusion coefficient are reported in Table IV as a function of state, after correction for the hydrodynamic velocity.

The effect of temperature upon the Fick diffusion coefficient for methane in the methane-crude oil system is shown in Figure 5. As would be expected, there is a significant increase in the coefficient with an increase in temperature. The Fick diffusion coefficient is presented as a function of the molecular weight of the less volatile component for three temperatures in Figure 6. It is apparent that the coefficient in the methane-crude oil system is slightly lower than that estimated for a paraffin hydrocarbon system involving a less volatile component of the same molecular weight. The data included in Figure 6 are based upon studies of the transport of methane in the liquid phase of the methane-propane (*16*), methane-*n*-butane (*15*), methane-*n*-pentane (*11*), methane-*n*-heptane (*14*), methane-*n*-decane (*13*), and methane-white oil systems (*12*). It is not surprising that the methane-crude oil system yields slightly smaller Fick diffusion coefficients than would be obtained for systems containing no aromatic or naphthenic compounds.

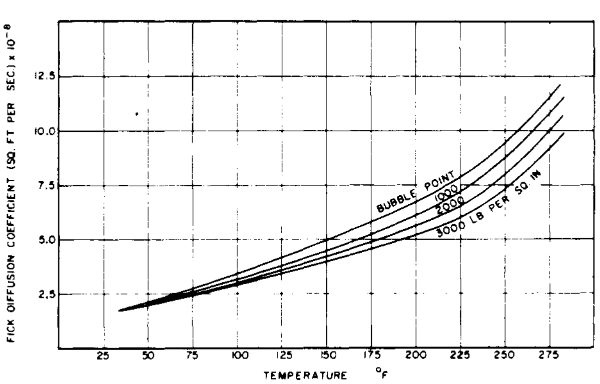

Figure 5. Influence of temperature upon Fick diffusion coefficient for methane

The accumulation of experimental data upon the diffusion coefficients of the lighter hydrocarbons has not as yet progressed to the point where a generalization of the results to permit estimation of the diffusion coefficients is worthwhile. These diffusion coefficients are somewhat smaller than those for methane in a binary hydrocarbon system involving a paraffinic, less volatile component of the same molecular weight.

ACKNOWLEDGMENT

This work constitutes a contribution from Project 37 of the American Petroleum Institute at the California Institute of Technology. The methane was supplied through the courtesy of The Texas Co. Virginia Berry prepared the data in a form suitable for publication. B. Lawson Miller assisted in the preparation of the manuscript, which was reviewed by W. N. Lacey.

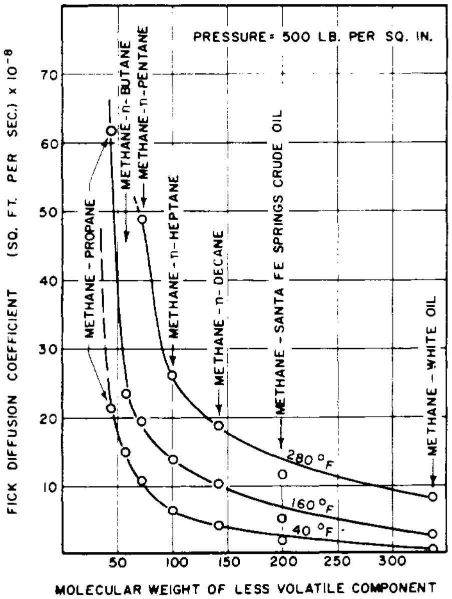

Figure 6. Effect of molecular weight of less volatile component upon Fick diffusion coefficient for methane in the liquid phase

Table III. Summary of Experimental Results[a]

Pressure, P.S.I.A.		Methane in Liquid Phase				$\frac{(\Delta m_k)^2}{\theta}$	Standard Error of Estimate, Lb.	Volumetric Correction Factor	Fick Diffusion Coefficient, Sq. Ft./Sec., $\times 10^{-8}$	
		Composition Weight Fraction		Concn., Lb./Cubic Foot						
Initial	Final	Initial	Final	Initial	Final	Lb.^{2}/Second			Uncorrected	Corrected
						40° F.[b]				
15.4	340.1	0.0005	0.0090	0.022	0.470	1.651×10^{-12}	1.43×10^{-6}	0.9941	1.795	1.785
340.4	641.4	0.0090	0.017	0.470	0.887	1.177	1.69	0.9889	1.804	1.784
641.5	945.6	0.017	0.024	0.887	1.306	0.943	1.88	0.9833	1.244	1.223
1346.3	1850.3	0.035	0.047	1.821	2.410	3.126	3.21	0.9649	2.364	2.281
						100° F.				
27.7	508.2	0.0004	0.011	0.027	0.577	5.113×10^{-12}	3.42×10^{-6}	0.9919	3.710	3.680
486.4	786.2	0.010	0.017	0.546	0.877	1.522	16.62	0.9874	3.204	3.164
2178.4	2674.4	0.050	0.062	2.479	3.012	3.546	18.20	0.9484	3.642	3.454
						160° F.				
265.4	565.6	0.0048	0.012	0.253	0.542	1.738×10^{-12}	1.56×10^{-6}	0.9918	4.688	4.649
565.1	965.8	0.012	0.019	0.542	0.941	4.226	7.96	0.9857	6.293	6.203
3240.8	3662.9	0.073[c]	0.084[c]	3.367[c]	3.812[c]	1.984	0.98	0.9288[c]	3.292	3.058
						220° F.				
125.6	424.8	0.0012	0.0068	0.096	0.341	2.246×10^{-12}	1.37×10^{-6}	0.9945	8.303	8.257
424.8	824.8	0.0068	0.014	0.340	0.688	3.454	1.63	0.9890	6.648	6.575
2315.7	2914.8	0.045	0.060	2.110	2.723	7.519	12.26	0.9506	5.964	5.670
2916.8	3508.2	0.060	0.078[c]	2.724	3.311[c]	6.384	18.06	0.9371[c]	6.090	5.707
						280° F.[d]				
266.2	570.0	0.0031	0.0085	0.182	0.415	2.653×10^{-12}	1.94×10^{-6}	0.9933	11.056	10.982
569.7	956.8	0.0085	0.016	0.415	0.722	5.083	1.76	0.9883	12.056	11.914
2459.2	2951.5	0.046	0.058	2.052	2.518	7.907	5.38	0.9540	11.406	10.881
2949.8	3556.6	0.058	0.074[c]	2.518	3.100[c]	9.891	21.74	0.9411[c]	9.692	9.121
3557.5	4158.5	0.075[c]	0.091[c]	3.102[c]	3.661[c]	8.330	52.70	0.9278[c]	9.537	8.848

[a] Effective cross-sectional area for diffusion was 0.019262 square foot.

[b] Values of composition, concentration, and volumetric correction factor extrapolated from data at higher temperatures.

[c] Extrapolated from data at lower pressures.

[d] Values of composition, concentration, and volumetric correction factor extrapolated from data at lower temperatures.

Table IV. Fick Diffusion Coefficient for Methane

Pressure, P.S.I.A.	Methane in Liquid Phase		Fick Diffusion Coefficient, Sq.Ft./Sec., $\times 10^{-8}$
	Compn. weight fraction	Concn. lb./cu.ft.	
	40° F.[a]		
500	0.013	0.687	1.8
1000	0.026	1.379	1.8
1500	0.038	2.004	1.8
2000	0.051	2.578	1.8
2500	0.063	3.123	1.8[b]
3000	0.074	3.619	1.8[b]
3500	0.084	4.069	1.8[b]
	100° F.		
500	0.011	0.568	3.3
1000	0.022	1.160	3.2
1500	0.034	1.730	3.1
2000	0.046	2.280	3.1
2500	0.058	2.828	3.0
3000	0.070[b]	2.361[b]	2.9[b]
3500	0.083[b]	3.901[b]	2.8[b]
	160° F.		
500	0.009	0.489	5.0
1000	0.020	0.982	4.7
1500	0.030	1.491	4.6
2000	0.042	2.012	4.4
2500	0.053	2.544	4.3
3000	0.066[b]	3.090[b]	4.1[b]
3500	0.080[b]	3.681[b]	4.0[b]
	220° F.		
500	0.008	0.409	7.3
1000	0.018	0.858	6.9
1500	0.028	1.319	6.6
2000	0.038	1.800	6.3
2500	0.050	2.282	6.0
3000	0.063	2.816	5.7
3500	0.077[b]	3.379[b]	5.4[b]
	280° F.[c]		
500	0.007	0.348	11.6
1000	0.017	0.759	11.2
1500	0.026	1.189	10.7
2000	0.036	1.636	10.3
2500	0.047	2.052	9.9
3000	0.059	2.560	9.5
3500	0.073	3.024	9.0

[a] Values of composition and concentration extrapolated from data at higher temperatures.

[b] Extrapolated from data at lower pressures.

[c] Values of composition and concentration extrapolated from data at lower temperatures.

NOMENCLATURE

$D_{F,k}$ = Fick diffusion coefficient of component k, square feet/second

m_k = weight of component k added per unit area of interface, pounds /square foot

$\underline{m}_k$ = total weight of component k crossing the interface, pounds

V_k = partial specific volume of component k, cubic feet per pound

Δ = difference in

θ = time, seconds

σ_k = concentration of component k, pounds per cubic foot

Superscript

$*$ = average condition

Subscripts

e = conditions at equilibrium

g = gas phase

i = conditions at interface

j = component j, stagnant component

k = component k, diffusing component

l = liquid phase

o = initial conditions

LITERATURE CITED

(1) Bertram, E. A., Lacey, W. N., *Ind. Eng. Chem.* **28**, 316 (1936).
(2) Botkin, D. F., Reamer, H. H., Sage, B. H., Lacey, W. N., "Fundamental Research on Occurrence and Recovery of Petroleum 1943," American Petroleum Inst., pp. 62-70.
(3) Emmert, R. E., Pigford, R. L., *Chem. Eng. Progr.* **50**, 87 (1954).
(4) Hill, E. S., Lacey, W. N., *Ind. Eng. Chem.* **26**, 1324 (1934).
(5) *Ibid.*, p. 1327
(6) Kirkwood, J. G., Crawford, B., Jr., *J. Phys. Chem.* **56**, 1048 (1952).
(7) Lacey, W. N., *Oil Gas J.* **30**, No. 8, 15; No. 9, 68 (1931).
(8) Meyers, C. H., *Bur. Standards J. Research* **9**, 807 (1932).
(9) Opfell, J. B., Sage, B. H., *Ind. Eng. Chem.* **47**, 918 (1955).
(10) Pomeroy, R. D., Lacey, W. N., Scudder, N. F., Stapp, F. P., *Ibid.*, **25**, 1014 (1933).
(11) Reamer, H. H., Duffy, C. H., Sage, B. H., *Ibid.*, **48**, 282 (1956).
(12) *Ibid.*, p. 285.
(13) Reamer, H. H., Opfell, J. B., Sage, B. H., *Ind. Eng. Chem.* **48**, 275 (1956).
(14) Reamer, H. H., Sage, B. H., *A.I.Ch.E. Journal* **3**, 449 (1957).
(15) Reamer, H. H., Sage, B. H., *Ind. Eng. Chem., Chem. Eng. Data Series* **1**, 71 (1956).
(16) Reamer, H. H., Sage, B. H., *Ibid.*, **3**, 54 (1958).
(17) Reamer, H. H., Sage, B. H., *Rev. Sci. Instr.* **29**, 709 (1958).
(18) Reamer, H. H., Sage, B. H., "Transport Properties in Gases. Measurement of Diffusion Coefficients in Gases and Liquids at Elevated Pressures," pp. 62-74, Northwestern University Press, Evanston, Ill., 1958.
(19) Sage, B. H., Hicks, B. L., Lacey, W. N., "Drilling and Production Practice 1938," American Petroleum Institute, pp. 402-20.
(20) Sage, B. H., Lacey, W. N., *Trans. Am. Inst. Mining Met. Engrs.* **136**, 136 (1940).
(21) Sage, B. H., Webster, D. C., Lacey, W. N., *Ind. Eng. Chem.* **28**, 984 (1936).
(22) Schlinger, W. G., Reamer, H. H., Sage, B. H., Lacey, W. N., "Fundamental Research on Occurrence and Recovery of Petroleum, 1952-1953," American Petroleum Institute, pp. 70-106.
(23) Schrage, R. W., "Theoretical Study of Interphase Mass Transfer," Columbia University Press, New York, 1953.
(24) Tung, L. H., Drickamer, H. G., *J. Chem. Phys.* **20**, 6 (1952).
(25) *Ibid.*, p. 10.

Received for review December 13, 1957. Accepted May 5, 1958.