Translational-Rotational Coupling Parameters for Mutual Diffusion in *N*-Octane

The infinite dilution diffusion coefficients of methane, ethane, *n*-propane, *n*-pentane, *n*-heptane, *n*-decane, *n*-dodecane, and *n*-tetradecane have been measured in *n*-octane using the Taylor dispersion technique in the temperature range 304-435 K at 1.72×10^6 N/m^2. Using the framework of the rough hard sphere theory, the values of the translational-rotational coupling parameter have been determined for each solute-solvent pair at each temperature. The coupling parameter was found to be independent of temperature over the entire temperature range of the experiments performed and it has the same value for solutes ranging from *n*-pentane to *n*-tetradecane. A calculation scheme is presented for accurate determination of infinite dilution diffusion coefficients for a wide range of solutes in *n*-octane at a wide temperature range.

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Introduction

Diffusion coefficients in liquids over an extended temperature and pressure range are needed for design of mass transfer processes. In addition, the mutual diffusion coefficient is most important in providing information regarding the nature of interactions between unlike molecules. It is thus a valuable quantity in developing a theory on the dynamics of fluids. The interpretation of diffusion measurements has for sometime been based on the rough hard sphere theory. However, the ambiguities involved in assigning a value to the translational-rotational coupling parameter has prevented the development of a successful predictive equation valid at a wide temperature and pressure range. The diffusion of a single solute species at very low concentration in a pure molecular fluid is the simplest case of mutual diffusion. The infinite dilution diffusion coefficients measured in this work in binary *n*-alkane systems at a wide temperature range have been used to study the nature and temperature dependence of the coupling parameter.

The interpretation of mutual diffusion measurements in dense fluids has been based on a model developed by Chandler (1975) for self-diffusion. In this model, self-diffusion coefficient of a real fluid is approximated by a rough hard sphere fluid which is related to the smooth hard sphere fluid by the expression:

$$D_{RHS} = A D_{SHS} \quad \text{where} \quad 0 < A \leq 1 \tag{1}$$

A is a roughness factor that accounts for the effect of coupling between translational and rotational momentum exchange in polyatomic fluids. The physical explanation of this phenomenon is that the coupling provides an additional mechanism for the relaxation of the velocity of a particle in the fluid. As a result, the velocity autocorrelation function will decay to zero more rapidly if the coupling exists than if it does not. The more rapid decay leads to a smaller value of the integral over the correlation function and thus a smaller value for D_{RHS}.

A similar form also holds for infinite dilution diffusion coefficients (Bertucci and Flygare, 1975):

$$D_{12,RHS} = A_{12} D_{12,SHS} \quad \text{where} \quad 0 < A_{12} \leq 1 \tag{2}$$

where A_{12} accounts for the effect of coupling between unlike molecules. Normally, $D_{12,SHS}$ is expressed as a ratio that corrects the Enskog diffusion coefficient for the effect of correlated molecular motions at high densities. The positive deviations arise from a vortex flow pattern and are expected to increase as the mass of the diffusing particle increases. On the other hand, for a lighter diffusing particle there is the probability of backscattering which leads to a negative deviation. These effects have been studied quantitatively by computer simulation studies (Alder et al., 1974) and the resulting corrections have usually been expressed as ratios. These are normally given as function of σ_1/σ_2, m_1/m_2, and V/V_o where σ_1 and σ_2 are the effective hard sphere diameters of the solute (1) and solvent (2) molecules, m_1 and m_2 are mass of the solute (1) and solvent (2) molecules. V_o is

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the close packed hard sphere volume of the solvent and is given by:

$$V_o = N\sigma_2^3/\sqrt{2} \qquad (3)$$

where N is the Avagadro's number. Therefore, Eq. 2 can be written as:

$$D_{12,RHS} = A_{12}\left(\frac{D_{12,SHS}}{D_{12,E}}\right)D_{12,E} \qquad (4)$$

$D_{12,E}$ is the Enskog (1939) expression given by:

$$D_{12,E} = \frac{3}{8\sigma_{12}^2 n}\left(\frac{kT}{\pi m_{12}}\right)^{1/2}\frac{1}{g(\sigma_{12})} \qquad (5)$$

where n is the number density, $g(\sigma_{12})$ the contact value of the pair correlation function of hard spheres, k Boltzmann's constant, T absolute temperature, σ_{12} the mean of σ_1 and σ_2, and m_{12} the reduced mass of the solute (1) and the solvent (2). The contact values of the pair correlation function is given by:

$$g(\sigma_{12}) = \frac{1}{1-\eta} + \frac{3\eta\sigma_1}{(1-\eta)^2(\sigma_1+\sigma_2)} + \frac{\eta^2\sigma_1}{2(1-\eta)^3\sigma_2} \qquad (6)$$

where $\eta = \pi n\sigma_2^3/6$ at infinite dilution. The above equation is obtained by extending the Carnahan-Starling rule to liquid mixtures (Mansoori et al., 1971) where the pair correlation function is expressed as a linear combination of the virial and compressibility results.

At present, there is no formal way to calculate A_{12} and the value can be extracted only from experimental data. The development of a theory for prediction of the translational-rotational coupling parameter has been hindered mainly due to the following three reasons.

1. Due to lack of accurate mutual diffusion data at a wide range of experimental conditions, the determination of A_{12}'s temperature and density dependency was not possible.

2. The ambiguities involved in assigning the molecules an effective hard sphere diameter have prevented the usage of a consistent set of diameters among the investigators in this area. Since a 1% change in the diameter may make a difference of several percent in the calculated translational-rotational coupling parameter, the level of uncertainty resulted in inconsistent A_{12} values.

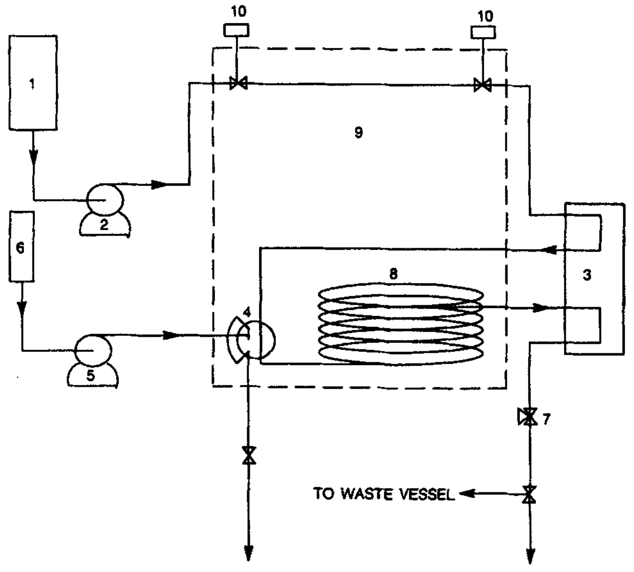
3. The ratios $(D_{12,SHS}/D_{12,E})$ have so far been obtained with interpolation of limited computer results which also caused large uncertainties in the A_{12} values.

$A_{12}$was found to be 0.70 for polyatomic solute and solvent species by fitting diffusion data of 11 solutes in 8 solvents to the RHS theory (Evans et al., 1981). However, the correction factors were obtained by interpolation of limited and inaccurate MD simulation data of Alder et al. (1974). Bertucci and Flygare (1975) obtained A_{12} values between 0.470 and 0.552 for six different polyatomic pairs, but the calculations were based on the assumption that $(D_{12,SHS}/D_{12,E})$ was independent of solution composition. Czworniak et al. (1975) studied mutual diffusion in 11 binary systems of organic liquids and extracted the A_{12} values from experimental data. However, it was assumed that the correction factors varied linearly with composition. Recently, Dymond et al. (1985) studied tracer diffusion in octamethylcyclotetrasiloxane at 323 K and at pressures up to 59 MPa and reported values for the translational-rotational coupling constant. The correction factors were obtained by MD simulation for the actual size and mass ratios of the systems studied. Surprisingly, A_{12} values were density-dependent. The hard sphere diameters were determined from PVT data using the expression (Watts and McGee, 1976):

$$10^9\sigma(\mathrm{m}) = 0.11611[10^6 V_m(\mathrm{m^3/mol})] \qquad (7)$$

where V_m is the molar volume at the freezing point. However, this expression is valid only for smooth hard spheres, i.e., for systems where A is unity. Therefore, the calculated diameters may be in considerable error.

A consistent set of V_o values for n-alkanes is obtained by simultaneously fitting self-diffusion and viscosity coefficient data to the hard sphere model (Dymond and Awan, 1988). In addition, MD simulation data of Easteal and Woolf (1988) at closely spaced mass ratios $(0.10 \leq m_1/m_2 \leq 1.67)$ and size ratios $(0.5 \leq \sigma_1/\sigma_2 \leq 1.0)$ at reduced volumes (V/V_o) between 1.5 and 2.0 enables one to determine $(D_{12,SHS}/D_{12,E})$ at a wide range with acceptable accuracy. Therefore, we have measured the infinite dilution diffusion coefficients in the homologous series n-alkanes in the reduced volume range which is covered by new MD simulation data to study the nature of the translational-rotational coupling parameter. For this purpose, n-octane was chosen as the solvent and the solutes were methane, ethane, n-propane, n-pentane, n-heptane, n-decane, n-dodecane, and n-tetradecane. The temperature range was 304–435 K which correspond to a reduced volume range of 1.52–1.99.



1. Solvent reservoir
2. Solvent pump
3. Refractive index detector
4. 6-port sample injection valve
5. Sample injection pump
6. Sample solution
7. Backpressure regulator
8. Coiled dispersion tube
9. Heated enclosure
10. Pressure transducers

Figure 1. Taylor dispersion apparatus.

Table 1. Infinite Dilution Diffusion Coefficients of *n*-Alkane Solutes in *n*-Octane at 1.72 × 10^6 N/m^2 (D_{12} × 10^9 m^2/s)

	Temperature (K)					
Solute	304	335	361	387	411	435
CH_4	6.67 ± 0.01	9.75 ± 0.01	12.71 ± 0.06	16.29 ± 0.05	19.75 ± 0.10	24.04 ± 0.21
C_2H_6	4.89 ± 0.02	7.14 ± 0.01	9.23 ± 0.11	12.00 ± 0.08	14.77 ± 0.04	17.97 ± 0.12
C_3H_8	4.00 ± 0.04	5.92 ± 0.00	7.79 ± 0.01	10.01 ± 0.03	12.32 ± 0.07	15.16 ± 0.10
C_5H_{12}	3.19 ± 0.04	4.79 ± 0.00	6.30 ± 0.04	8.15 ± 0.08	9.99 ± 0.07	12.27 ± 0.09
C_7H_{16}	2.66 ± 0.02	3.85 ± 0.05	4.98 ± 0.08	6.22 ± 0.06	8.12 ± 0.04	10.07 ± 0.10
$C_{10}H_{22}$	2.18 ± 0.01	3.28 ± 0.04	4.22 ± 0.09	5.38 ± 0.08	7.07 ± 0.03	9.17 ± 0.05
$C_{12}H_{26}$	1.92 ± 0.02	2.90 ± 0.01	3.84 ± 0.05	4.99 ± 0.09	6.31 ± 0.01	8.23 ± 0.01
$C_{14}H_{30}$	1.76 ± 0.01	2.61 ± 0.02	3.53 ± 0.01	4.46 ± 0.04	5.66 ± 0.03	7.48 ± 0.04

Experimental Procedure

We have used the Taylor dispersion technique to measure the infinite dilution diffusion coefficients of *n*-alkanes in *n*-octane. In this method, a narrow pulse of solute (1) diluted in the solvent (2) is injected into a tube in which the solvent is moving in laminar flow. The combined effects of laminar bulk flow and molecular diffusion cause the narrow pulse to disperse into a Gaussian shape. At the end of the diffusion tube the concentration $C(t)$ is recorded as a function of time data as the peak elutes through a detector. Taylor (1953) has shown that for injection of material of mass M concentrated at a point $x = 0$ and at time $= 0$, the concentration is given by:

$$C = \frac{M}{2\pi r^2 \sqrt{\pi k t}} \exp\left[\frac{-(L - \bar{u}t)^2}{4kt}\right] \tag{8}$$

where

$$k = D_{12} + \frac{r^2 \bar{u}^2}{48 D_{12}} \tag{8a}$$

and r is the tube radius, $\bar{u}$ the average solvent velocity, L the length of the dispersion tube and t the time. A standard parameter estimation technique is applied to the above equation for calculating the diffusion coefficients (Rodden, 1988).

A schematic diagram of the equipment is given in Figure 1. Solvent is sparged continuously with helium. It is pumped though a line that has a capillary restrictor and a backpressure regulator into a heated enclosure. The solvent then flows out of the enclosure to the reference side of the concentration detector (LDC/Milton Roy Refractomonitor) and back into the heated enclosure where the solute sample is injected via a six-port injection valve. The solute sample may be either a liquid solute prepared gravimetrically by mixing with the solvent or a gas solute prepared by saturating the solvent at low pressure. The dispersion of the sample peak is then detected at the sample side of the detector. A digital multimeter automated by a microcomputer is used to record the response curve on a floppy disk. The details of the experimental assembly and procedure are given elsewhere (Matthews and Akgerman, 1987).

Table 2. Comparison of Infinite Dilution Diffusion Coefficients in *n*-Octane

Solute	Temp. K	D_{12} × 10^9 m^2/s Other Work	D_{12} × 10^9 m^2/s This Work
CH_4	333	9.45*	9.52
CH_4	373	14.20*	14.25
CH_4	403	18.10*	18.41
C_7H_{16}	304	2.70**	2.63
C_7H_{16}	335	4.00**	3.84
C_7H_{16}	304	2.88†	2.63

*Chen et al. (1982)
**Alizadeh and Wakeham (1982)
†Matos Lopes and Nieto de Castro (1985)

Results and Discussion

The infinite dilution diffusion coefficients for each solute in *n*-octane at each temperature are given in Table 1. The average standard deviation is 0.8% and the accuracy of the data is estimated to be within ±1%. A comparison of our experimental values with the literature is presented in Table 2. Our methane diffusivity data were fit into a function of the Form $D_{12} = a + bT + cT^2$ to be able to provide a comparison with the experimental values of Chen et al. (1982). All of the literature mea-

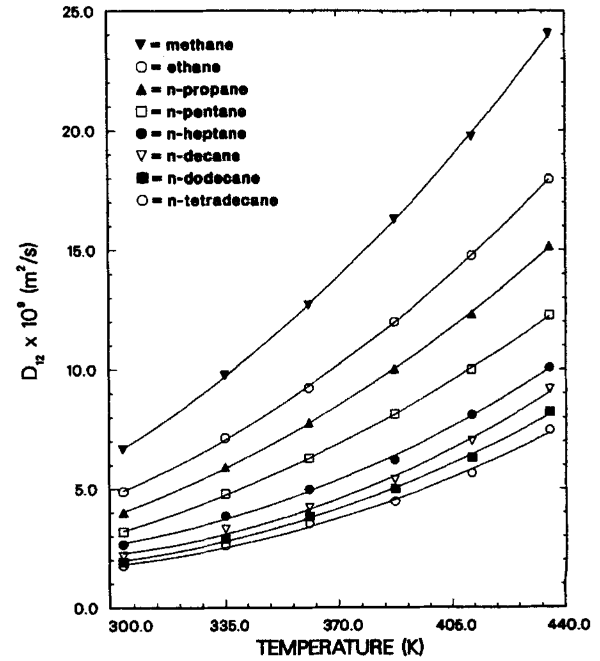


Figure 2. Temperature dependency of the infinite dilution diffusion coefficients of *n*-alkanes in *n*-octane at 1.72 × 10^6 N/m^2.

Table 3. Coefficients of the Functions $V_o = aT^b$ and $V_o = a + bT$ for n-Alkanes

Solute	a	b	Temp. Range, K	No. of Points
CH_4	57.26	−0.2045	100–300	8
C_2H_6	60.85	−0.1234	100–320	9
C_3H_8	97.29	−0.1452	90–300	6
C_4H_{10}	111.8	−0.1226	140–320	5
C_6H_{14}	181.9	−0.1429	223.2–373.2	7
C_8H_{18}	334.5	−0.1994	248.2–348.2	5
$C_{12}H_{26}$	194.4	−0.0960	298.2–373.2	4
$C_{16}H_{34}$	265.1	−0.1363	298.2–373.2	4

surements in Table 2 have been performed using the Taylor dispersion technique. We believe our values are more accurate due to the data analysis technique we have developed which is superior in accuracy to techniques used by the referenced investigators. The temperature dependency of the diffusion coefficients of n-alkanes in n-octane is shown in Figure 2.

In order to calculate the translational-rotational coupling parameter, the V, values of n-alkanes given by Dymond (1988) were accurate fitted into functions of the form $10^6\ V_o(\mathrm{m^3/mol}) = aT^b$ for methane, ethane, propane, butane, hexane, octane and $10^6\ V_o(\mathrm{m^3/mol}) = a + bT$ for dodecane and hexadecane. The coefficients of the functions for each n-alkane are given in Table 3. The same functions were used for extrapolation to higher temperatures and a set V_o values were generated at each experimental condition for the solutes and the solvent we employed. A plot of V_o vs. temperature for ethane which includes the generated V_o values at experimental conditions is presented in Figure 3. The V_o values for hexane, octane, dodecane and hexadecane were fitted into a straight line as a function of carbon number at each temperature. A plot of V_o vs. carbon number at 304 K is shown in Figure 4. V_o values for pentane, heptane, decane and tetradecane were generated by interpolation from these curves at each temperature. The density and hence molar volume of n-octane at each temperature was calculated using the correlation given in Orwell and Flory (1967). The $(D_{12,SHS}/D_{12,E})$ ratios for each solute-solvent pair were determined by using new MD simulation data of Easteal and

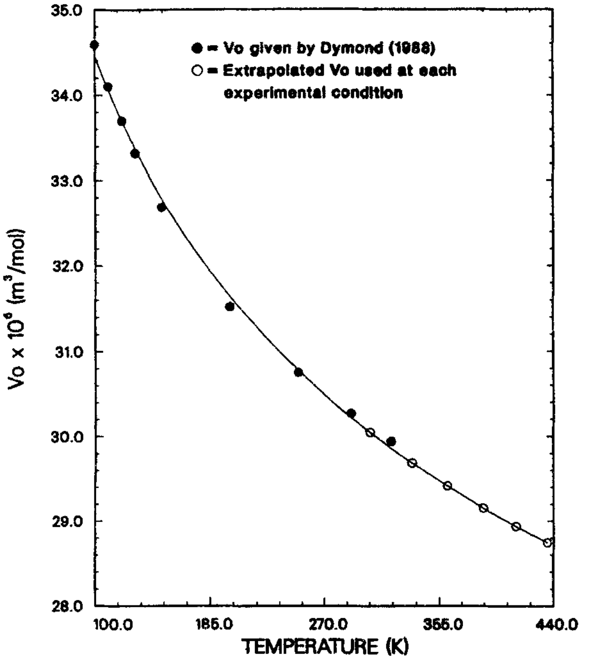


Figure 3. Temperature dependency of the close-packed hard sphere volume for ethane.

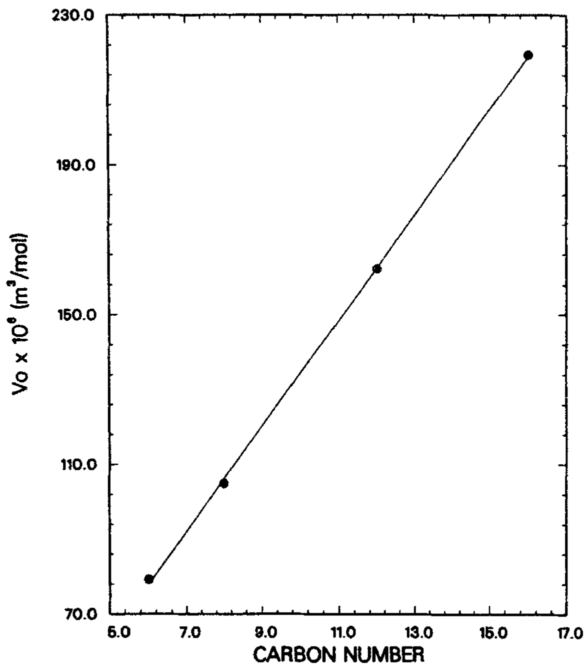


Figure 4. Variation of the close-packed hard sphere volume with carbon number at 304 K.

Table 4. Close-Packed Hard Sphere Volumes ($10^6\ V_o$, $\mathrm{m^3/mol}$) of n-Alkanes and Correction Factors for Each Solute-Solvent Pair

		Temperature (K)					
		304	335	361	387	411	435
CH_4	V_o	17.79	17.44	17.17	16.93	16.72	16.53
	R_{12}	0.53	0.61	0.68	0.74	0.77	0.82
C_2H_6	V_o	30.05	29.69	29.42	29.16	28.95	28.75
	R_{12}	0.56	0.65	0.72	0.78	0.82	0.86
C_3H_8	V_o	42.41	41.82	41.37	40.95	40.59	40.26
	R_{12}	0.57	0.67	0.75	0.82	0.86	0.90
C_5H_{12}	V_o	64.91	63.96	63.31	62.76	62.33	61.97
	R_{12}	0.62	0.74	0.83	0.89	0.93	0.99
C_7H_{16}	V_o	93.69	92.14	90.96	89.86	88.91	88.01
	R_{12}	0.66	0.78	0.87	0.95	1.01	1.04
$C_{10}H_{22}$	V_o	136.80	134.40	132.40	130.50	128.80	127.10
	R_{12}	0.69	0.83	0.91	0.99	1.09	1.12
$C_{12}H_{26}$	V_o	165.20	162.20	159.70	157.20	154.90	152.60
	R_{12}	0.72	0.86	0.93	1.03	1.11	1.12
$C_{14}H_{30}$	V_o	194.40	190.80	187.70	184.70	181.90	179.20
	R_{12}	0.74	0.87	0.94	1.03	1.14	1.14
C_8H_{18}	V_o	106.90	104.90	103.30	101.90	100.70	99.57

Table 5. Translational-Rotational Coupling Parameters for Each Solute-Solvent Pair

	Temperature (K)						
Solute	304	335	361	387	411	435	Average
CH_4	0.74	0.76	0.75	0.75	0.76	0.74	0.75 ± 0.01
C_2H_6	0.82	0.83	0.82	0.83	0.84	0.84	0.83 ± 0.01
C_3H_8	0.88	0.89	0.88	0.87	0.88	0.88	0.88 ± 0.01
C_5H_{12}	0.91	0.92	0.90	0.91	0.92	0.91	0.91 ± 0.01
C_7H_{16}	0.92	0.90	0.87	0.84	0.88	0.90	0.89 ± 0.03
$C_{10}H_{22}$	0.94	0.93	0.91	0.90	0.91	0.98*	0.92 ± 0.02
$C_{12}H_{26}$	0.90	0.91	0.92	0.90	0.90	0.99*	0.91 ± 0.01
$C_{14}H_{30}$	0.90	0.90	0.93	0.90	0.88	0.98*	0.90 ± 0.02

*Not included in the calculation of the average values due to possible errors in the extrapolated R_{12} values

Woolf (1988). The intrapolations were performed on the basis of a linear relation found between $(D_{12,SHS}/D_{12,E})$ and logarithm of (M_2/M_1) at each (σ_1/σ_2). The V_o values for the solutes and the solvent and $(D_{12,SHS}/D_{12,E})$ for each pair are given in Table 4. In this table, R_{12} represents $(D_{12,SHS}/D_{12,E})$.

The translation-rotational coupling parameters, A_{12}'s, are than extracted from the experimental data using the R_{12} values from molecular dynamics simulations and the Enskog diffusivity employing Eq. 4–6. The results are presented in Table 5. There are inherent assumptions in this approach, however, considering the accuracy of the experimental data and the MD simulations and the validity of the extrapolation of V_o to higher temperatures, the results are very encouraging (Figure 5). The coupling parameters appear to be temperature-independent for a given pair of solute and the solvent. Implications of this result are obvious, a single measurement at one temperature would enable the calculation of the diffusivities at other conditions. Furthermore, the coupling constant for solutes ranging from *n*-propane to *n*-tetradecane (C_3–C_{14}) in the solvent *n*-octane appear to have the same value of 0.91. This result enables the prediction of the diffusivities of C_3–C_{14} in *n*-octane with an average error of ±2% (slightly higher for C_3) at all conditions using a single parameter. As the molecules become more spherical, the amount of translational-rotational coupling increases resulting in a lower A_{12} value. Thus, we can conclude that the coupling parameter is dependent on the sphericity of the species involved and after a critical value for the deviation from sphericity, the parameter attains a constant value.

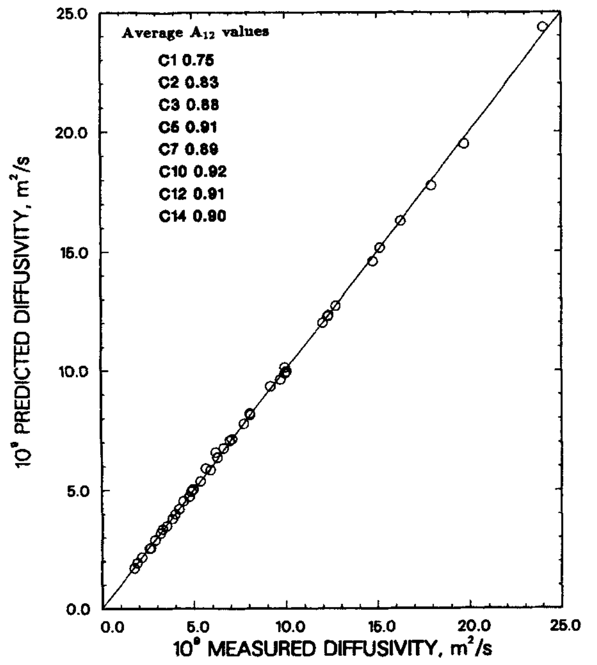


Figure 5. Comparison of the experimental diffusion coefficients with the calculated values using the average A_{12} values in Table 5 for each solute-solvent pair.

Table 6. Comparison of Calculated and Experimental Diffusion Coefficients for Spherical Solutes in *n*-Octane at 298 K with A_{12} = 0.75

			$D_{12} \times 10^9$ m²/s	
Solute	$\sigma_1 \times 10^{10}$ m	$D_{12,SHS}/D_{12,E}$	Calc.	Exp.*
Xe	3.914	0.77	4.06	3.92
CCl_4	5.254	0.73	2.56	2.49

*Chen et al. (1982)

The above observations indicate that it should be possible to predict diffusion coefficients of any solute in octane using the A_{12} values we calculated. We have calculated the diffusion coefficients of xenon and carbon tetrachloride in *n*-octane at 298 K assigning an A_{12} value of 0.75 since these are spherical molecules like methane and compared the predictions with the data of Chen et al. (1982). The results are presented in Table 6; the calculated tracer diffusion coefficients are within ±3% of experimental data. This analysis has been performed only for these two solutes at this temperature because the diameters have been calculated using high-pressure and high-temperature self-diffusion data consistent with the work performed by Dymond and Awan (1988). The effective hard sphere diameter for xenon was calculated using the correlation given by Easteal and Woolf (1984). Although the correlation is valid in the temperature range 170–289 K, the error introduced to calculate the effective hard sphere diameter at 298 K is negligible. The effective hard sphere diameter of carbon tetrachloride was determined using the equation given by Chandler (1975).

Thus, provided that consistent and accurate hard sphere diameters (V_o values) and molecular dynamics simulations at a sufficiently wide range of conditions are available, the diffusion coefficients in nonassociated liquids can be predicted with the aid of a single parameter that is temperature-independent and is dependent on the shape (sphericity) of the solute species.

Notation

A = translational-rotational coupling parameter, dimensionless
C = concentration, mol/m³
D = diffusion coefficient, m²/s
k = Boltzmann's constant 1.38×10^{-23} J/K
L = length of diffusion tube, m
m = molecular mass, kg
n = number density, m^{-3}
r = radius of diffusion tube, m
T = temperature, K
V = molar volume, m³/mol

Greek letters

σ = effective hard sphere diameter, m

Subscripts

1 = solute
2 = solvent
E = Enskog
RHS = rough hard sphere
SHS = smooth hard sphere

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