

Correlation of Dense Fluid Self-Diffusion, Shear Viscosity, and Thermal Conductivity Coefficients¹

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A general procedure has been developed for simultaneously fitting any two of the self-diffusion coefficient, the viscosity (as the fluidity), and the thermal conductivity (as its reciprocal) as Dymond reduced coefficients, (D^*, η^*, λ^*) , to a simple function of the volume and the temperature for dense fluids. For example, $D^* = \zeta_1 + \zeta_2 V_r / (1 + \zeta_3 / V_r)$, where $V_r = V[1 - \zeta_4(T - T_r) - \zeta_5(T - T_r)^2]$. T_r is any convenient temperature, here 273.15 K. As V_r is common to the two properties, only eight coefficients, ζ_j and ζ_k , are required. Such reduced transport-coefficient curves are geometrically similar for members of groups of closely related compounds. The procedure has been extended to give "family" curves for such groups by fitting a pair of transport properties for three substances from the group in a single regression. Overall, fewer coefficients are required than for other schemes in the literature, and the fitting functions used are simpler. The curves so constructed can be used for the correlation of data obtained from different sources, as well as interpolation and, to a limited extent, extrapolation. A comparison is made for a number of compound groups between simultaneous fits of the pairs $(D-\eta)$, $(D-\lambda)$, and $(\eta-\lambda)$.

KEY WORDS: correlation methods; dense liquids; diffusion coefficient; thermal conductivity; viscosity.

1. INTRODUCTION

The primary model used to describe transport properties of fluids of simple molecules is the hard-sphere model [1, 2], based on the results of computer simulations [3, 4]. Experimental results can be fitted to the model using the molecular diameter as a disposable parameter and, in the case of polyatomic molecules, a factor related to the interchange of translational and rotational energy during collisions [2, 5].

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Despite some degree of success in giving a description of the effect of molecular motion on the transport properties, the very simplicity of the hard-sphere model limits its practical application to real liquids. Most substances have molecules that lack spherical symmetry and, consequently, have liquid ranges much broader than that of the hard-sphere fluid. In addition, there are as yet no adequate theoretical methods for dealing with translational-rotational coupling or for calculating the second-order effect of the attractive forces.

Nevertheless, reduced transport property coefficients do show regularities when plotted against reduced volume [6] and it is possible to correlate data for closely related substances of similar shape [7-9]. We have developed a method to do this for the self-diffusion coefficient (D), the shear viscosity (η), and the thermal conductivity (λ). The same functional form is used for each of the three reduced properties expressed in terms of a common temperature-dependent reduced volume.

In earlier work, this approach was tested for the diffusion and viscosity coefficients of alkyl-substituted benzenes [9], where some regularity was found in the parameters used to fit these substances to the family curve. Here, the method is extended to include the thermal conductivity and is applied to the same family and the chloromethanes.

2. METHOD

In the earlier paper [9] it was shown that the temperature and volume dependence of the self-diffusion and viscosity coefficients for dense fluids can be fitted to equations of the form

$$D^* = \zeta_1 + \zeta_2 V_r / (1 + \zeta_3 / V_r) \quad (1)$$

and

$$1/\eta^* = \phi^* = \zeta_4 + \zeta_5 V_r / (1 + \zeta_6 / V_r) \quad (2)$$

where V_r is expressed in terms of molar volume V and temperature T by

$$V_r = V(1 - \xi_1(T - T_r) - \xi_2(T - T_r)^2) \quad (3)$$

T_r being any convenient reference temperature, e.g., 273.15 K. D^* and η^* are the reduced self-diffusion and viscosity coefficients introduced by Dymond [1]; they are defined by

$$\begin{aligned} D^* &= \frac{(nD)}{(nD)^x} \left(\frac{V}{V_0} \right)^{2/3} \\ &= 17.44 \cdot 10^9 \frac{D}{V^{1/3}} \left(\frac{M}{1000T} \right)^{1/2} \end{aligned} \quad (4)$$

$$\eta^* = \frac{\eta}{\eta^\infty} \left(\frac{V'}{V_0} \right)^{2/3}$$

$$= 20.929 \cdot 10^3 \eta \left(\frac{1000}{MT} \right)^{1/2} V'^{2/3} \quad (5)$$

where ∞ indicates the dilute-gas value of the transport property for the hard-sphere fluid, V_0 is the volume of random close packing, $L\sigma^3/\sqrt{2}$, D has units of $\text{m}^2 \cdot \text{s}^{-1}$, η of $\text{Pa} \cdot \text{s}$, V' of $\text{cm}^3 \cdot \text{mol}^{-1}$, M of $\text{g} \cdot \text{mol}^{-1}$, and T of K. As can be seen from the secondary expressions, the virtue of the Dymond reduced coefficients is that numerical values can be obtained without assignment of the molecular diameter, σ , implicit in the dilute-gas transport coefficients.

For the monatomic liquids and simple fluids such as methane, the Dymond reduced transport properties are essentially linear in the molar volume and it is a relatively straightforward procedure to obtain hard-sphere diameters by comparison of experimental reduced coefficients with those derived from molecular dynamics simulations. For larger, polyatomic molecules there is a definite curvature away from the hard-sphere predictions at high densities and the transport properties are nonzero well below the freezing volume of the equivalent hard-sphere fluid. The temperature dependence is, nevertheless, weak and isotherms can be moved laterally and superposed on a convenient reference isotherm. [This is equivalent to plotting against $V'/V_0(T)$.] The equations used here were introduced to take into account the curvature in a simple way, to provide a means to represent and interpolate data, and to compare results from different sources. They have been used to fit both D and η reference curves simultaneously by a nonlinear least-squares procedure, using common parameters for the lateral isotherm shift given by Eq. (3).

It was observed that the reference curves for closely related substances were generally similar and the procedure has now been extended to fit the self-diffusion coefficient and the viscosity for *three* substances simultaneously. This requires six additional parameters that shift the curves of the second and third substances along the D^* (or ϕ^*) and V_r axes. Thus for diffusion,

$$D_i^* = R_{D_i} D_1^*, \quad i = 2, 3 \quad (6)$$

for the fluidity, or reciprocal viscosity,

$$\phi_i^* = R_{\phi_i} \phi_1^*, \quad i = 2, 3 \quad (7)$$

with

$$V_{r,i} = r_i V_{r,1}, \quad i = 2, 3 \quad (8)$$

The subscript 1 indicates the primary reference fluid. The R parameters, or "inverse roughness" ratios, are analogous to Chandler's [2] translational-rotational coupling factors used in the fitting of the hard-sphere model to transport properties [5]. They take into account any differences in coupling among the three fluids. The factor r represents the ratio of the reduced molecular volumes given by Eq. (3), corresponding to the ratio of the volumes of close-packed molecules used in the hard-sphere model.

Equations (1) to (3) and (6) to (8) provide the family curves for the two properties with the same volume shift, Eq. (3) being common to both. By using data for three substances with different liquid ranges, a broad span of reduced volume can be included within the correlation. Data sets containing systematic errors that may not be obvious in a joint D - η fit for a single substance stand out in the combined fit and can then be eliminated. For the majority of substances examined, the parameters ζ_3 and ζ_6 are almost identical: this is to be expected, given the known weak density dependence of the product $D\eta$.

The thermal conductivity is treated in a similar manner as the diffusion and viscosity coefficients. The Dymond reduced reciprocal thermal conductivity is fitted jointly with either of the other two properties:

$$1/\lambda^* = \zeta_7 + \zeta_8 V_r / (1 + \zeta_9 / V_r) \quad (9)$$

$$(1/\lambda^*)_i = R_{\lambda,i} (1/\lambda^*)_1, \quad i = 2, 3 \quad (10)$$

with λ^* defined by

$$\begin{aligned} \lambda^* &= \frac{\lambda}{\lambda^x} \left(\frac{V'}{V'_0} \right)^{2/3} \\ &= 671.22 \lambda \left(\frac{M}{1000T} \right)^{1.2} V'^{2.3} \end{aligned} \quad (11)$$

where λ has units of $\text{W m}^{-1} \text{K}^{-1}$.

The family curves are based on data for up to three reference substances for which high-quality results are available, and data for other members of the group can then be matched to them.

3. RESULTS

3.1. Alkyl-Substituted Benzenes

Benzene, toluene, and mesitylene were used as the reference fluids for this class in correlating self-diffusion and viscosity coefficients and are

Table I. Coefficients for Simultaneous Transport Property Fits: Fluidity and Reciprocal Thermal Conductivity—Alkyl-benzenes^a

	(ϕ^*)	$(1/\lambda^*)$
ζ_4 or ζ_7	-0.534 460	-0.160 941
$10^2 \zeta_5$ or ζ_8	0.359 271	0.129 139
$10^{-2} \zeta_6$ or ζ_9	-0.372 936	-0.355 939
$10^3 \zeta_1$	-0.637 953	
$10^5 \zeta_2$	0.146 949	
SD (%)	1.1	1.0

	Benzene	Toluene	Mesitylene
R_λ	1	0.850 ± 0.002^b	—
R_ϕ	1	0.873 ± 0.009	$0.91_2 \pm 0.02_1$
r	1	$0.856_{08} + 0.001_2$	$0.634_{01} \pm 0.002_5$
Thermal conductivity ^c			
(rV_r) range ($\text{cm}^3 \cdot \text{mol}^{-1}$)	83.5-100.0	80.3-104.7	—
T range (K)	310-360	273-373	—
Data source	[10]	[11]	—
Viscosity			
(rV_r) range ($\text{cm}^3 \cdot \text{mol}^{-1}$)	83.3-99.2	79.4-112.3	86.8-103.0
T range (K)	298-348	218-426	298-393
Data source	[12-15]	[12, 15-17]	[18, 19]

^a T_r is 273.15 K in each case.

^b More figures than are significant are reported for computational purposes.

^c The ranges of experimental temperatures and reduced molar volumes are shown there.

again used now that the thermal conductivity is included. The correlation is weaker in that there are no high-pressure thermal conductivity data available to the author for mesitylene, but this is partly compensated by the extensive data sets available for toluene. The results and sources used are given in Tables I and II. The coefficients for D and η are very similar to those obtained earlier from the D - η correlation [9]. The values obtained do depend on the data sets: For example, slightly different values of r for mesitylene of 0.623 and 0.634 are obtained from the (D - λ) and (λ - η) correlations, respectively. Such differences need to be taken into account in fitting individual property sets to the family curves. More surprising is the closeness of the parameter ζ_9 to the value of ζ_3 and ζ_6 , as individual ($1/\lambda^*$) isotherms show very little curvature, and single-substance fits give smaller values for ζ_9 .

Table III shows the results of fits for the xylenes and ethylbenzene to the (λ - η) family curves. The fits for the viscosity are generally good and similar to those obtained with the earlier (D - η) correlation. The volume

Table II. Coefficients for Simultaneous Transport Property Fits: Self-Diffusion Coefficient and Reciprocal Thermal Conductivity—Alkyl-benzenes^a

	D^*	$(1/\lambda^*)$	
ζ_1 or ζ_7	-3.333 96	-0.155 399	
$10^2 \zeta_2$ or ζ_8	2.288 75	0.125 262	
$10^{-2} \zeta_3$ or ζ_9	-0.364 468	-0.355 941	
$10^3 \zeta_1$	-0.646 399		
$10^6 \zeta_2$	0.800 935		
SD (%)	1.9	1.3	

	Benzene	Toluene	Mesitylene
R_D	1	$0.90_6 \pm 0.01_0$	0.989 ± 0.015
R_λ	1	$0.856_4 \pm 0.006_7$	—
r	1	$0.853_{71} + 0.001_2$	$0.622_{70} \pm 0.001_3$
Self-diffusion coefficient			
(rI'_r) range ($\text{cm}^3 \cdot \text{mol}^{-1}$)	85.0–96.8	76.4–97.1	78.2–90.3
T range (K)	288–333	220–326	298–313
Data source	[20, 21]	[22]	[23]
Thermal conductivity			
(rI'_r) range ($\text{cm}^3 \cdot \text{mol}^{-1}$)	83.9–99.5	80.2–105.2	—
T range (K)	298–348	273–373	—
Data source	[10]	[11]	—

^a T_r is 273.15 K in each case.

ratios r are similar to those predicted by taking the ratios of molar volumes at atmospheric pressure. For the thermal conductivity the fits are only fair. As an example, for ethyl benzene, the thermal conductivity r value (0.86 ± 0.02) is larger than is obtained from the viscosity data (0.734 ± 0.004) or anticipated from the molar volumes ratio, 0.726. Checks were made on the toluene and benzene data used to see if the R and r values of the correlation were returned when individual properties of each substance were fitted to the family curves. The fits for toluene and the viscosity of benzene were excellent, but that for the thermal conductivity of benzene was poor, R_λ being (0.93 ± 0.02), not unity (see Table III). These results show that there is a greater latitude in the fitting of thermal conductivity, for which the isotherms show little curvature. The fits are better when the data cover a wide range of density, as is the case for toluene.

3.2. Chloromethanes

For this family the D - η correlation is based on self-diffusion and viscosity data for trichloromethane [27], supplemented by self-diffusion

Table III. Results for Fits of Literature Test Data to the Correlation Equations

	<i>o</i> -Xylene	<i>p</i> -Xylene	<i>m</i> -Xylene	Ethylbenzene	Benzene
R_2^a	$0.72_6 \pm 0.02_6$	$0.83_4 \pm 0.02_1$	$0.68_7 \pm 0.02_3$	$0.51_7 \pm 0.02_8$	$0.93_2 \pm 0.03_1^b$
r	$0.75_0 \pm 0.01_2$	$0.698_3 \pm 0.009$	$0.75_8 \pm 0.01_1$	$0.86_1 \pm 0.02_1^b$	$1.03_3 \pm 0.01_6$
r (estimated) ^c	0.7381	0.7201	0.7244	0.7263	1.000
SD (%)	0.5	0.7	0.7	0.4	1.0
Max. dev. (%)	1.1	-1.4	-1.3	1.1	-2.2
Expt. precision (%)	± 0.3	± 0.3	± 0.3	± 0.3	± 0.3
(rV_1) range (cm ³ ·mol ⁻¹)	88.3-100.4	85.3-95.9	87.2-103.6	86.6-100.9	86.2-103.2
T range (K)	308-360	305-358	305-358	305-358	310-361
Ref. No.	24	24	24	25	10
R_ϕ	0.960 ± 0.006	$0.911-0.008$	0.933 ± 0.004	0.829 ± 0.004	1.000 ± 0.006
r	$0.75_0 \pm 0.01_2$	$0.698_3 \pm 0.009$	$0.75_8 \pm 0.01_1$	0.734 ± 0.004	1.0000 ± 0.0005
r (estimated)	0.7381	0.7201	0.7244	0.7263	1.000
SD (%)	0.9	0.7	0.5	0.6	1.1
Max. dev. (%)	2.2	-1.3	1.0	-2.5	-6.5
Expt. precision (%)	± 2	± 2	$\pm 0.5, \pm 2$	± 2	Various
(rV_1) range (cm ³ ·mol ⁻¹)	83.0-95.2	87.4-98.2	85.5-98.2	85.6-99.1	83.2-99.2
T range (K)	298-348	298-348	298-348	298-348	298-348
Ref. No.	26	26	15, 26	26	12-15

^a More figures than are significant are reported for computational purposes.

^b See text regarding the test fits.

^c r can be estimated from the ratio of the molar volume of reference substance 1 (benzene) to that of the test compound.

Table IV. Coefficients for Simultaneous Transport Property Fits:
Self-Diffusion Coefficient and Viscosity—Chloromethanes^a

	D^*	$(1/\eta^*)$
ζ_1 or ζ_3	-2.605 50	-0.331 737
$10^2 \zeta_2$ or ζ_4	2.132 95	0.277 063
$10^{-2} \zeta_3$ or ζ_5	-0.307 573	-0.300 575
$10^3 \zeta_1$	-0.843 941	
$10^5 \zeta_2$	0.425 387	
SD ($^{\circ}\sigma$)	2.6	3.9

	CHCl ₃	CCl ₄
R_D	1	$1.5_{27} \pm 0.1_3$
R_η	1	$1.6_{86} \pm 0.1_6$
r	1	$0.725_{80} \pm 0.008_7$
Self-diffusion coefficient		
(rT_r) range (cm ³ ·mol ⁻¹)	70.3–88.4	66.0–76.0
T range (K)	278–348	283–328
Data source	[27]	[28]
Viscosity		
(rT_r) range (cm ³ ·mol ⁻¹)	69.2–89.7	65.9–75.4
T range (K)	278–348	303–323
Data source	[27]	[13]

^a T_r is 273.15 K in each case.

(¹⁴C tracer) [28] and viscosity coefficients [13] for tetrachloromethane. A Hayward equation of state, based on the compressibility results of Eastal and Woolf [29] and the densities at atmospheric pressure measured by Benson and Winnick [30], was used to obtain molar volumes of CCl₄ at the experimental state points. The coefficients for the correlation are presented in Table IV.

Table V shows the results of fits to the family curve for the NMR spin-echo self-diffusion results of Prielmeier and Lüdemann [31] for CHCl₃, CH₂Cl₂, and CH₃Cl. The equations of state used to calculate molar volumes for CH₂Cl₂ and CH₃Cl were based on the data of Kumagai and Iwasaki [32], Diguët *et al.* [33], and Hsu and McKetta [34].

The results for CHCl₃ give a value of r equal to unity within experimental error, but R_D is 1.07; this shows excellent agreement between the results of Lüdemann's group and our own for the relative temperature and density dependence of $D(\text{CHCl}_3)$, with the factor of 7% most likely due to differences in calibration methods. For CH₂Cl₂, the fit is generally fair, though there are large deviations at the highest pressures (> 100 MPa)

Table V. Results for Fits of Literature Test Data to the Correlation Equations

	CHCl ₃ ^a	CHCl ₃	CH ₂ Cl ₂	CH ₃ Cl	Si(CH ₃) ₄
R_D^b	0.99 ₉ ± 0.01 ₅	1.06 ₅ ± 0.08 ₇	1.3 ₂ ± 0.1 ₆	0.85 ₉ ± 0.08 ₈	1.07 ₃ ± 0.02 ₀
r	1.000 ± 0.002	1.00 ₃ ± 0.01 ₃	1.25 ₆ ± 0.02 ₂	1.88 ₇ ± 0.03 ₉	0.600 ₂ ± 0.001 ₃
r (estimated) ^c	1.000	1.000	1.250	1.80-1.88	0.5920
SD (%)	2.0	3.5	6.4	2.7	6.4
Max. dev. (%)	4.5	-6.8	16	5.0	-11
Expt. precision (%)	±2	±3	±3	±3	±0.3
(rV_r) range (cm ³ ·mol ⁻¹) ^d	70.3-88.4	75.3-88.7	73.7-87.2	84.9-90.8	64.1-91.7
T range (K)	278-348	254-370	299-406	258-313	298-373
Ref. No.	27	31	31	31	36

	Si(CH ₃) ₄
R_D^b	1.30 ₄ ± 0.02 ₄
r	0.603 ₅ ± 0.001 ₂
r (estimated)	0.5920
SD (%)	7.1
Max. dev. (%)	15.7
Expt. precision (%)	—
(rV_r) range (cm ³ ·mol ⁻¹)	65.6-88.9
T range (K)	298-373
Ref. No.	36

^a This column contains the results of a test fit for this data set against the full family curve: The agreement is excellent.

^b More figures than are significant are reported for computational purposes.

^c r can be estimated from the ratio of the molar volume of reference substance 1 (CHCl₃) to that of the test compound.

^d Only data fitting within the range of the correlation were used.

at the highest temperature, 406 K. The r value (1.26 ± 0.02), is consistent with that predicted from the ratio of the molar volumes, 1.250. For CH_3Cl , the r value is (1.89 ± 0.04), close to values estimated from Lennard-Jones potential diameters (1.84) [35] and from fits to the rough hard-sphere model (1.80) using the procedures followed in earlier work [5]. The R_D parameter is (0.86 ± 0.09). This value is less certain than those for the other chloromethanes as only a small number of points overlap the family curve. If CH_3Cl is used as the third reference fluid in forming the correlation, then this value is reduced to 0.62.

The order of the inverse coupling factors, R_D , is $\text{CH}_3\text{Cl} < \text{CHCl}_3 < \text{CH}_2\text{Cl}_2 < \text{CCl}_4$, with CH_3Cl showing the greatest translational-rotational coupling. There appears to be no obvious correlation with deviation from spherical symmetry or dipole moment.

Analysis of the same systems using the hard-sphere model [5] (except CCl_4 , for which the data lie outside the range of application of this model) yields Chandler coupling factors of 0.65, 0.68 [5], and 0.69, respectively, suggesting rather less variation of translational-rotational coupling along the series than is implied by the present correlation.

We have also attempted to fit data for the fluoromethanes and chlorofluoromethanes to the chloromethane curves but could find no good match. Curiously, the self-diffusion and viscosity coefficients of tetramethyl silane [36] do fit well (except at the highest temperature, 373 K), with an r value matching that predicted and an inverse coupling factors of 1.07 for self-diffusion coefficient and 1.30 for the viscosity.

Further analysis of other systems is in progress.

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