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Application of Corresponding States Principles for Prediction of Self-Diffusion Coefficients in Liquids

A corresponding states approach has been used successfully to correlate reduced self-diffusivity of pure liquids with reduced temperature and acentric factor. In addition, dimensional analysis techniques have been used to accurately relate viscosity, normal boiling point and temperature with self-diffusivity, as well as to relate the three transport properties.

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SCOPE

In the study of the transport properties of mixtures of liquids, considerable effort has been devoted to the coefficient of mutual diffusion from both the experimental and the theoretical points of view. On the theoretical side, a number of equations have been proposed relating the mutual diffusion coefficient to self-diffusion coefficient and activity of each component in the mixture (McCall and Douglass, 1967; Loflin and McLaughlin, 1968; Skelland, 1974). A number of relationships between self-diffusion coefficients and viscosities of liquids have been proposed in the literature (Li and Chang, 1955; Dullien, 1972).

As diffusivity and thermal conductivity have not been so related, the first purpose of this study is to propose new relationships between diffusivity and easily measurable properties such as boiling point and viscosity with reasonable accuracy. A correlation relating diffusivity to both thermal conductivity and viscosity is also desirable.

The second purpose of this study is the derivation of a corresponding states approach for the prediction of self-diffusion coefficients as has previously been done for viscosity and thermal conductivity.

CONCLUSIONS AND SIGNIFICANCE

Beginning with dimensional analysis a relationship between self-diffusivity, viscosity, boiling point and temperature has been derived which predicts self-diffusivity with an average error of 2 to 3%:

$$\frac{\mu^{2/3}}{T} D = 6.35 \times 10^{-8} (T/T_b)^{0.83}$$

By a similar approach, a relationship between the three transport properties (viscosity, thermal conductivity and diffusivity) has been derived with a mean deviation of ±3.5%:

$$\sqrt{\frac{K}{v_s^3}}\, D = 4.65 \times 10^{-4} \left(\frac{KT}{v_s^2\, \mu}\right)^{0.979}$$

It has been shown that the corresponding states principle is applicable to prediction of diffusivity. Using the generalized correlation for diffusivity developed in this work, an average error of 4.5% was obtained for twenty systems in the prediction of self-diffusion coefficients of pure liquids. The generalized correlations also can be used to predict diffusivity of a liquid at any temperature if diffusivity is known at one temperature:

$$D_r = (0.4 - \omega)\, D_r^{(1)} + (\omega - 0.2)\, D_r^{(2)}$$

where $D_r^{(1)}$ and $D_r^{(2)}$ are functions of reduced temperature.

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Relationships between self-diffusion coefficients and viscosities have been proposed in the literature. Li and Chang (1955) gave the relationship

$$\frac{D\mu}{kT} = \frac{1}{2\pi}\left(\frac{N}{V}\right)^{1/3} \tag{1}$$

which was obtained by a modification of the Stokes-Einstein equation.

Dullien (1972) obtained simple relationships which can be used to predict self-diffusion coefficients of liquids with an average error of 4%. In addition to the customary parameters, one of these equations contains the critical volume:

$$\frac{\mu VD}{RT} = 0.124 \times 10^{-16} V_c^{2/3} \tag{2}$$

The other equation uses Lennard-Jones potential parameters:

$$\frac{\mu VD}{RT} = 0.129 \times 10^{-16}\, \sigma^2(\Omega_D\, \Omega_V)^{1/2}\, T_c^{1/2} \tag{3}$$

THEORY

Dimensional Analysis

Experimental and theoretical correlations for diffusivity (Bird, Stewart and Lightfoot, 1960) show that

$$D = f(\mu, V, T, k) \tag{4}$$

Viscosity μ is related to the molar volume V and other properties:

$$\mu = g(T_b, V, T, h) \tag{5}$$

Eliminating V between Equations (4) and (5), we get

$$D = F_1(\mu_F, T_b, T, k, h) \tag{6}$$

where for simplicity μ_F is viscosity in force dimension.

Application of dimensional analysis leads to the following formulation for the diffusion coefficient:

$$D = \alpha\, \mu_F^a\, T_b^b\, T^c\, k^d\, h^e \tag{7}$$

$[D] = L^2 t^{-1}$ $\quad L$ = length

$[\mu_F] = F L^{-2} t$ $\quad t$ = time

$[T_b] = T$ $\quad F$ = force

$[T] = T$ $\quad T$ = temperature

$[K] = F L T^{-1}$

$[h] = F L t$

The dimensional analysis produces the following values for the exponents:

$$a = -2/3, \quad c = 1 - b, \quad d = 1, \quad e = -1/3$$

Using these exponents and combining variables, we get

$$(\mu_F^2\, h)^{1/3}\frac{D}{kT} = \alpha(T_b/T)^b \tag{8}$$

If we substitute $\mu_F = \mu/g_c$ and combine the three universal constants k, h and g_c, Equation (8) becomes

$$\frac{\mu^{2/3}}{T} D = a\,(T_bT)^b \tag{9}$$

where a and b are constants to be determined from data on D, μ, T_b and T. Note that although $(\mu_F^2\, h)^{1/3}\, D/kT$ is a dimensionless group, $\mu^{2/3}D/T$ is not. Thus, a is dimensional, while b is a dimensionless number.

The same procedure can be followed to produce a relationship between diffusivity and thermal conductivity. Bridgman's formula for thermal conductivity states that

$$K = f(v_s, V, k) \tag{10}$$

TABLE 1. SELF-DIFFUSIVITY AND VISCOSITY OF PURE LIQUIDS*

Substance	T_b, °K	T, °K	$10^3\mu$, N·s/m²	$10^9 D$, m²/s
Chloroform	335.05	200.0	2.20	0.50
Chloroform	335.05	298.2	0.542	2.58
Carbontetrachloride	349.5	313.2	0.739	1.82
Carbontetrachloride	349.5	333.2	0.585	2.44
n-pentane	309.05	298.2	0.215	5.535
n-pentane	309.05	308.7	0.195	6.29
n-hexane	341.9	298.2	0.2937	4.21
n-heptane	371.6	194.7	2.51	0.415
n-heptane	371.6	250.3	0.702	1.52
n-heptane	371.6	273.2	0.525	2.08
n-heptane	371.6	298.2	0.386	3.12
n-heptane	371.6	305.7	0.360	3.22
n-heptane	371.6	329.7	0.282	4.21
n-heptane	371.6	353.4	0.229	5.76
n-heptane	371.6	369.0	0.202	6.56
n-octane	398.8	298.2	0.5082	2.25
n-nonane	424.0	298.2	0.6621	1.70
2-methylbutane	301.0	298.2	0.219	5.30
3-methylpentane	336.4	298.2	0.307	3.61
2,3-dimethylbutane	331.1	298.2	0.361	3.50
2,2-dimethylbutane	322.9	298.2	0.351	3.41
2-methylpentane	333.56	298.2	0.295	3.98
Benzene	353.27	288.2	0.696	1.87
Benzene	353.27	298.2	0.599	2.22
Benzene	353.27	308.2	0.520	2.59
Benzene	353.27	318.2	0.465	3.04
Benzene	353.27	328.2	0.415	3.50
Benzene	353.27	338.2	0.375	4.03
Benzene	353.27	373.2	0.261	6.15
Benzene	353.27	423.2	0.171	10.0
Benzene	353.27	473.2	0.121	15.0
Benzene	353.27	523.2	0.082	23.9
Benzene	353.27	561.7	0.056	32.7
Methanol	337.86	308.2	0.482	2.71
Methanol	337.86	318.2	0.482	3.37
Methanol	337.86	328.2	0.378	3.88
Water	373.2	293.2	1.005	2.2
Water	373.2	298.2	0.8937	2.51
Water	373.2	313.2	0.656	3.5
Water	373.2	333.2	0.4688	5.3
Water	373.2	473.2	0.133	23.0
Water	373.2	573.2	0.0896	44.0
Water	373.2	623.2	0.0728	61.0

* μ and D from Dullien (1972).

The velocity of sound v_s is given by

$$v_s = \sqrt{g_c\gamma(\partial P/\partial\rho)_T} \tag{11}$$

The quantity $(\partial P/\partial\rho)_T$ is readily obtainable from isothermal compressibility measurements or from an equation of state or compressibility correlation. γ is the heat capacity ratio which for liquids is almost equal to unity.

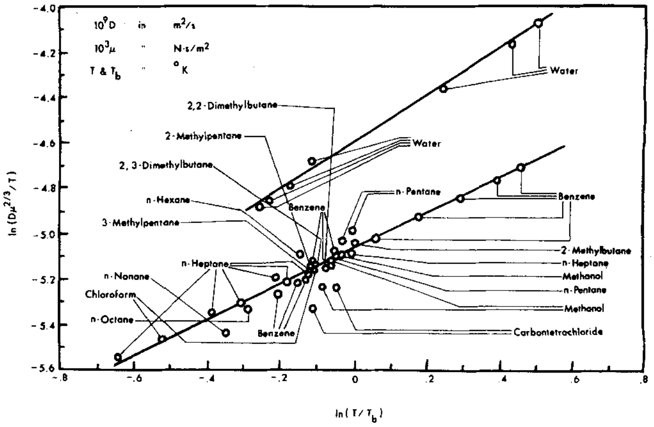

Figure 1. Dependency of diffusion coefficents on viscosity and normal boiling point.

By eliminating V between Equations (4) and (10), we get

$$D = f(\mu, v_s, K, T, k) \tag{12}$$

where dimensions of v_s and K are $L\,t^{-1}$ and $F\,t^{-1}\,T^{-1}$, respectively. Using the dimensional analysis procedure, we get

$$\sqrt{\frac{K}{k\,v_s^3}}\,D = \beta\left(\frac{KT}{v_s^2\,\mu_F}\right)^{b''} \tag{13}$$

If we substitute $\mu_F = \mu/g_c$ and take g_c and k out, Equation (13) becomes

$$\sqrt{\frac{K}{v_s^3}}\,D = a'\left(\frac{KT}{v_s^2\,\mu}\right)^{b'} \tag{14}$$

Although $\sqrt{K/k\,v_s^3}\,D$ and $KT/v_s^2\,\mu_F$ are dimensionless, $\sqrt{K/v_s^3}\,D$ and $KT/v_s^2\,\mu$ have dimensions. Thus, a' is dimensional, while b' is a dimensionless exponent.

TREATMENT OF DATA

A bank of data on self-diffusion coefficient D and viscosity μ of different substances was prepared from Dullien (1972). These data are shown in Table 1. According to Equation (9), a plot of $\ln(D\mu^{2/3}/T)$ vs. $\ln(T/T_b)$ should be linear. Such a plot is shown on Figure 1. Most of the hydrocarbons lie on a straight line. From the slope of the line, the following equation was obtained:

$$\frac{\mu^{2/3}}{T}\,D = 6.3 \times 10^{-4}\,(T/T_b)^{0.7805} \tag{15}$$

For water, the equation was derived to be

$$\frac{\mu^{2/3}}{T}\,D = 10.03 \times 10^{-14}\,(T/T_b)^{1.0245} \tag{16}$$

TABLE 2. THERMAL CONDUCTIVITY AND THE SONIC VELOCITY OF ONE-COMPONENT SYSTEMS

Substance	T °K	K*, J/m·s·°K	v_s†, m/s	$10^3\,\mu^+$, N·s/m²	$10^9\,D$,** m²/s
Chloroform	298.2	0.13942	950	0.542	2.58
Carbontetrachloride	293.2	0.10048	840	0.975	1.18
n-pentane	298.2	0.13732	1189	0.215	5.535
n-hexane	298.2	0.13942	1312	0.2937	4.21
n-heptane	298.2	0.14151	1440	0.386	3.12
n-octane	298.2	0.14570	1586	0.5082	2.25
n-decane	298.2	0.14612	1793	0.8527	1.31
Benzene	298.2	0.15994	1370	0.599	2.22
Acetone	298.2	0.17668	1442	0.3075	4.835
Water	298.2	0.60709	1520	0.8937	2.51
n-propanol	288.2	0.17751	1380	2.522	0.504
Iso-propanol	288.2	0.16328	1070	2.859	0.474

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* Values of K are taken from Perry (1963).

† Values of v_s are calculated by Equation (11) using data on compressibility coefficients from Perry (1963).

** μ and D are taken from Dullien (1972).

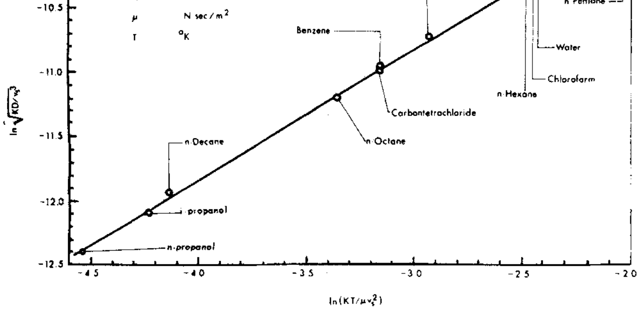

Figure 2. Dependency of diffusion coefficient on thermal conductivity.

Equation (15) can reproduce data with an average error of ±2.9%, while the average error for Equation (16) is ±2.3%. In Figure 1 most of the hydrocarbons lie on the line of Equation (15), but water, methanol and carbon tetrachloride deviate from the line.

Equation (14) indicates that the plot of $\ln(\sqrt{K/v_s^3}\,D)$ vs. $\ln(KT/\mu\,v_s^2)$ should be a straight line. The sonic velocity v_s can be calculated by Equation (11). Compressibility data were taken from Perry (1963) or were predicted from thermal conductivity data. Values of thermal conductivity K and v_s are shown in Table 2.

Figure 2 shows the plot of $\ln(\sqrt{K/v_s^3}\,D)$ vs. $\ln(KT/v_s^2\,\mu)$. From the slope and intercept of the line, constants a' and b' in Equation (14) were determined, and Equation (14) becomes

$$\sqrt{\frac{K}{v_s^3}}\,D = 4.16425 \times 10^{-13}\left(\frac{KT}{v_s^2\,\mu}\right)^{1.0126} \tag{17}$$

Equation (17) can reproduce data with a mean deviation of ±3.5%.

GENERALIZED CORRELATION FOR SELF-DIFFUSIVITY

At moderate pressure, it can be assumed that the diffusivity of liquids is independent of pressure as can be assumed for viscosity and thermal conductivity. However, diffusivity of liquids depends on temperature, as shown by all theoretical and empirical correlations. Generalized correlations have been obtained for viscosity (Graboski and Braun, 1968). In this work, similar correlations for diffusivity have been developed.

The main objective of the corresponding states approach is to predict a reduced self-diffusion coefficient from

$$D_r = \frac{D}{D_c} = D_r^0 + \omega\,D_r^1 \tag{18}$$

where:

D = absolute diffusivity of liquid
D_c = absolute diffusivity at critical temperature
D_r = reduced diffusivity, dimensionless
D_r^0 and D_r^1 = reduced diffusivity of simple and acentric fluid, respectively
ω = acentric factor

In using this approach for application to pure liquids, several facts must be kept in mind:

1. Corresponding states parameters D_r^0 and D_r^1 are not available. Thus they must be estimated from available data on pure liquid diffusivities vs. temperature.
2. Critical diffusivity data D_c are not available.

To circumvent this problem, the absolute diffusivity can be extracted from one known value of absolute diffusivity at some reference temperature.

It is assumed that in the temperature range of interest, the pressure difference between atmospheric and saturated liquid conditions would have a negligible effect on the diffusivity of the liquid. Therefore, only a saturated reduced diffusivity relationship as a function of reduced temperature is required.

The critical diffusivity D_c which is a hypothetical property may be determined by extrapolation of data to the critical temperature, or it could be predicted. If Equation (2) is applied at the critical point

$$D_c = m\,\frac{T_c}{\mu_c V_c^{1/3}} \tag{19}$$

in which m is a proportionality constant, and μ_c is the critical viscosity. Equation (2) cannot be used directly up to the critical temperature; therefore, m should be evaluated from some known value for D_c. Because values of diffusivity for benzene are available at high temperatures, D_c for benzene was determined by extrapolation. Values of μ_c are taken from Bird, Stewart and

TABLE 3. CRITICAL PROPERTIES OF ONE-COMPONENT SYSTEMS

Substance	M*	ω*	T_c,* °K	$10^3\,V_c$,* m³/kdg mole	$10^3\,\mu_c$,† N·s/m²	$10^9\,D_c$, m²/s
n-pentane	72.5	0.251	469.8	311.0	0.0238	33.8
n-hexane	86.17	0.296	507.9	368.0	0.0248	33.15
n-heptane	100.2	0.351	540.2	426.0	0.0254	32.8
n-octane	114.22	0.394	569.4	485.0	0.0259	32.46
n-decane	142.3	0.4902	617.7	603.12	0.0256	33.13
n-dodecane	170.33	0.5622	658.3	711.2	0.02589	33.0
2-methylbutane	72.15	0.2273	460.44	305.85	0.0247	32.09
3-methylpentane	86.2	0.275	504.44	366.5	0.02508	32.6
2-methylpentane	86.2	0.2791	497.5	366.5	0.0249	32.4
Benzene	78.11	0.210	562.6	260.0	0.0312	32.8
Chloroform	119.4	0.214	536.6	240.0	0.041	24.4
Acetone	58.08	0.318	508.2	209.0	0.03	40.9
Water	18.0	0.348	647.1	56.0	0.0454	56.0
Ammonia	17.03	0.25	405.6	72.5	0.0294	44.9
Methanol	32.04	0.556	512.6	118.0	0.0328	32.7
Ethanol	46.07	0.635	516.2	167.0	0.0313	26.0$^+$

* Values of M, ω, T_c and V_c were taken from Perry (1963) and Smith and Van Ness (1975).
† Values of μ_c were taken from Bird, Stewart and Lightfoot (1960).
$^+$ 21.0 extrapolated value

TABLE 4. PREDICTION OF SELF-DIFFUSION COEFFICIENT FROM CSP

Substance	ω	T, °K	$10^9\,D^*_{exp}$, m²/s	$10^9\,D_{pred}$, m²/s	Error†
n-pentane	0.2510	273.2	4.14	4.1	0.1
n-pentane	0.2510	308.7	6.29	6.38	1.4
n-hexane	0.296	298.2	4.21	4.0	−5.0
n-heptane	0.351	369.0	6.56	6.41	−2.2
n-octane	0.394	298.2	2.25	2.2	−2.2
n-octane	0.394	333.2	3.5	3.47	−2.3
n-decane	0.4902	298.2	1.31	1.3	−0.7
n-dodecane	0.5622	298.2	0.8	0.7	−2.5
N-dodecane	0.5622	333.2	1.48	1.4	−5.4
2-methylbutane	0.2273	298.2	5.3	5.6	5.6
3-methylpentane	0.275	298.2	3.61	3.9	8.0
2-methylpentane	0.2791	298.2	3.98	4.18	5.0
Benzene	0.210	338.2	4.03	3.85	−4.4
Chloroform	0.214	298.2	2.58	2.5	−3.1
Acetone	0.318	298.2	4.83	4.77	−0.6
Water	0.348	298.2	2.51	2.49	−0.8
Ammonia	0.250	213.0	3.80	3.66	−3.6
Ammonia	0.250	287.0	12.0	10.9	−9.2
Methanol	0.556	388.2	3.88	4.3	10.8
Ethanol	0.635	338.2	2.80	3.5	25.0
Ethanol (using extrapolated value for D_c = 21.0)	0.635	338.2	2.80	2.80	0.0

Average error = 4.5, bias error = 0.3, maximum error = 25.
No. of data points = 20

* Data was taken from Dullien (1972).
† Error = 100 × $(D_{calc} - D_{exp})/D_{exp}$.

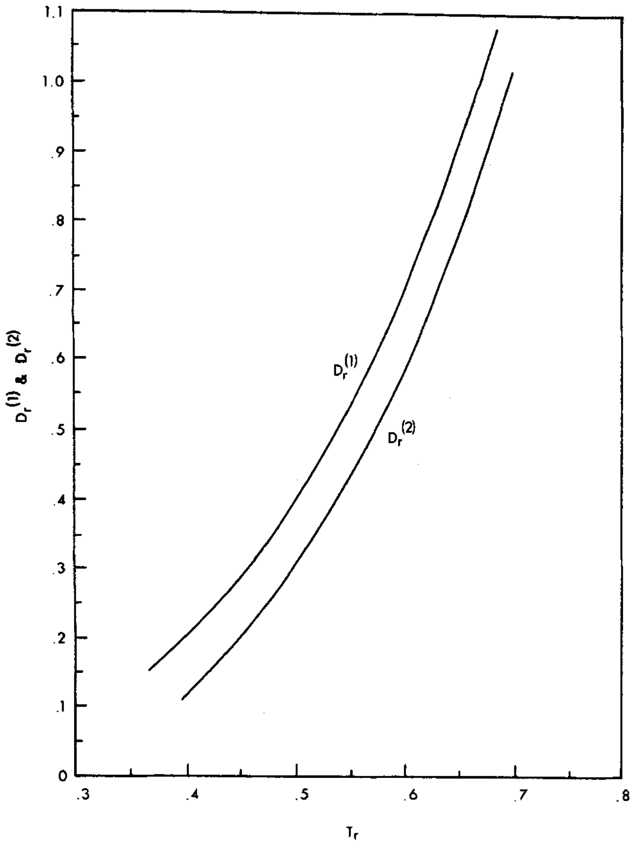

Figure 3. Generalized chart for $D_r^{(1)}$ and $D_r^{(2)}$.

Lightfoot (1960) or are calculated by

$$\mu_c = 61.6 \times 10^{-9}\,(M\,T_c)^{1/2}\,V_c^{-2/3} \qquad (20)$$

For benzene, $D_c = 32.8 \times 10^{-9}$ m²/s, $\mu_c = 3.12 \times 10^{-5}$, $V_c = 260 \times 10^{-9}$ and T_c = 497.5°K. Using these values, Equation (19) becomes

$$D_c = 1.16 \times 10^{-15}\,\frac{T_c}{\mu_c\,V_c^{1/3}} \qquad (21)$$

Equation (21) may be used to predict values of D_c relative to D_c of benzene. A tabulation of values of μ_c and D_c is given in Table 3.

Using available data (Dullien, 1972), D_c was obtained by extrapolation of data for ethanol: D_c (ethanol) = 21 × 10^{-9} m²/s, the value of D_c for ethanol from Equation (21) is 26 × 10^{-9} m²/s. This is a large deviation, but for most of the other substances, as discussed later, relatively good results were obtained.

Liquid diffusivity data were taken for *n*-pentane, *n*-heptane and benzene which have acentric factors of 0.251, 0.3506 and 0.21, respectively (Smith and Van Ness, 1975). By extrapolation of D_r to ω = 0.2 and ω = 0.4 at different reduced temperatures, the following equation was obtained:

$$D_r = (0.4 - \omega)\,D_r^{(1)} + (\omega - 0.2)\,D_r^{(2)} \qquad (22)$$

where $D_r^{(1)}$ and $D_r^{(2)}$ are plotted against T_r in Figure 3.

Table 4 shows the values of self-diffusivity predicted by Equation (22). An average error of 4.5% was observed for fourteen different substances. These results are unexpectedly good because predicted values for D_c from Equation (21) are not very accurate. Equation (22) and Figure 3 are recommended to be used for liquids having acentric factors between 0.2 and 0.4, but as it is seen from Table 4, even for some substances having $\omega >$ 0.4, the results are satisfactory. Evidently most liquids of interest have $0.2 < \omega < 0.4$

Note that in Table 4 for ethanol, if the extrapolated value of D_c is used instead of the predicted value from Equation (21), the error will be reduced from 25 to 0.0%. Therefore, extrapolated values for D_c are more preferable to the predicted values from Equation (21). Unfortunately, for many liquids, data for diffusivity in the range of critical temperature were not available. It should be emphasized that one of the reasons for having a large error for ethanol is the high value for its acentric factor.

NOTATION

a, a' = constants
b, b', b'' = constants
D = self-diffusion coefficient, m^2s
D_c = self-diffusion coefficient at critical temperature
D_r = reduced self-diffusion coefficient
g_c = gravitational constant
h = Planck's constant
K = thermal conductivity, J/m·s°K
k = Boltzman's constant
M = molecular weight
m = proportionality constant
N = Avogadro's number
P = pressure
R = gas constant
T = temperature, °K
T_b = normal boiling point, °K
T_c = critical temperature, °K
t = time
V = liquid molar volume
v_s = velocity of sound, m/s

Greek Letters

α = proportionality constant
β = proportionality constant
γ = heat capacity ratio
ω = acentric factor
μ = absolute viscosity, $N \cdot s/m^2$
μ_F = viscosity based on force unit
μ_c = viscosity at critical temperature, $N \cdot s/m^2$
ϕ = density

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Dynamics of Bubbles and Entrained Particles in the Rotating Fluidized Bed

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Trajectories of bubbles and entrained particles in the rotating fluidized bed were obtained from equations expressing a balance among inertial, centrifugal, Coriolis, gravity and drag forces. The solutions led to information on the behavior of isolated bubbles, bubble swarms and the elutriation characteristics of the rotating fluidized bed.

SCOPE

In a rotating fluidized bed, the bed particles are located in a rotating cylindrical container having a porous wall. A stable fluidized bed results from a balance of centrifugal forces arising from rotation and drag forces caused by gas entering the vessel through its porous wall. The minimum fluidization velocity increases with rotational speed and is usually large when compared with that for the familiar gravitational bed in which the earth's gravitational force opposes the gas drag force. For this reason, the rotation bed has been proposed as a high capacity gas-solid contactor for application in areas such as the processing of particulate foods and combustion of gas, oil and coal. It has also been suggested for use in propulsion in outer space, where gravity forces are small.

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