NOMENCLATURE

B, C, D, E = constants of Redlich-Kister equation
P° = vapor pressure of pure component
x = mole fraction of component in liquid phase
y = mole fraction of component in vapor phase
γ = activity coefficient
π = total pressure
ϕ = fugacity coefficient
ϕ° = fugacity coefficient of pure component
v = molar volume
B_{ij} = virial coefficients
σ^2 = variance of fit

Subscripts

1 = DMSO component
2 = n-Butanol component

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Viscosity Correlation for Mixtures of Freon-12 and Freon-22 Vapors

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Residual viscosity is a unique function of density for pure Freon-12 and Freon-22 vapors. Also, a plot of residual viscosity against density for Freon-12 and Freon-22 vapors exhibits a regular trend. These phenomena form the basis for predicting the viscosity of mixtures of Freon-12 and Freon-22 vapors.

THE RESIDUAL viscosity, defined as the difference between the viscosity, μ, at a particular temperature and density, and the viscosity, μ_0, at the same temperature but at zero density (*10*), is a unique function of density for some substances. This concept signifies that a single smooth curve represents the viscosity over a wide temperature and pressure range. The residual viscosity correlation, therefore, serves to represent the experimental viscosity measurements and to predict the viscosity at the temperature and pressure conditions of practical interest.

The correlation technique of residual thermal conductivity of gases and liquids with density, proposed by Abas-Zade, was extended on parallel lines to residual viscosity-density correlation by Shimotake and Thodos (*15*) for liquid and gaseous argon, helium, neon, krypton, and xenon, and by Brebach and Thodos (*1*) for nitrogen, hydrogen, and oxygen in the liquid and gaseous state. The residual viscosity phenomena was confirmed by Dolan *et al.* (*5*) for n-butane; by Starling *et al.* (*16*) for propane, and by Eakin *et al.* (*6*) for ethane. The concept was extended successfully to the methane-n-butane and methane-propane systems by Dolan *et al.* (*4*). It was also a useful extrapolation technique.

EMPIRICAL CORRELATIONS

There are four general methods for computing the viscosity of gas mixtures. The method of Hirschfelder *et al.* (*9*), based on rigorous kinetic theory and described by Hirschfelder *et al.* (*8*), is the most reliable, but complicated and time-consuming. Buddenberg and Wilke (*2*) proposed a method based on the Sutherland equation (*17*), eliminating the determinant, but retaining the need for unlike molecular diffusion coefficients. Later, Wilke (*19*) proposed a further development, eliminating even the need for the various unlike molecular diffusion coefficients, and presented a simple equation for gas mixtures. Another method, developed by Herning and Zipperer and successfully employed by Dolan and co-authors (*4*), is significant for its simplicity. This mixing rule is accurate for those mixtures which contain less than 25% hydrogen. Still another method was developed by Dean and Stiel (*3*) for calculation of the viscosity of nonpolar gas mixtures from the molecular weights and critical constants of the components. It claims novelty over the above three methods in enabling computation of viscosity of gas mixtures at moderate and high pressures without viscosity information on the pure components.

THEORY

For many gases, the residual viscosity can be expressed in terms of density as

$$\mu - \mu_0 = A\rho + B\rho^2 \qquad (1)$$

where

$$\mu = \mu(\rho, T)$$

and

$$\mu_0 = \mu(0, T)$$

The coefficients, A and B, which depend on the nature of the gas, are only mildly varying functions of temperature (*11*). Equation 1 signifies that if a regular trend exists in the plot of residual viscosity against density for the pure components, then the concept of residual viscosity can be extended to mixtures by relating A and B to composition by the following equations

Table I. Coefficients, μ_0, a, and b, and Reliability Information for Freon-12 and Freon-22 Vapors

Temp., °C.	Coefficient $\mu_0 \times 10^2$ Poise	$a \times 10^2$ Poise/(g./cc.)	$b \times 10^2$ Poise/(g./cc.)^2	Standard Deviation, $\sigma \times 10^6$ Poise	Greatest Error, $\times 10^6$ Poise	Per Cent Greatest Error
			Freon-12			
25	0.012467	0.0027	1.5921	0.0623	0.0973	0.074
50	0.012938	0.0393	0.1034	0.0070	0.0094	0.007
100	0.014220	0.0251	0.2156	0.7938	1.1860	0.801
150	0.015655	0.0250	0.1989	0.8653	1.2430	0.748
200	0.017197	0.0166	0.2506	0.3004	0.4898	0.278
			Freon-22			
25	0.012651	0.0677	0.6494	0.4807	0.6980	0.515
50	0.013472	0.0577	−0.0223	0.2937	0.4465	0.308
100	0.015372	0.0184	0.4710	0.2718	0.4446	0.279
150	0.016931	0.0178	0.3765	0.2939	0.4458	0.247
200	0.018588	0.0128	0.4917	0.1720	0.2892	0.149

Table II. Coefficients A and B, and Reliability Information for Freon-12 and Freon-22 Vapors

Refrigerant	Coefficient $A \times 10^2$ Poise/(g./cc.)	$B \times 10^2$ Poise/(g./cc.)^2	Standard Deviation, $\sigma \times 10^6$ Poise	Probable Error, P.E. $\times 10^6$ Poise
Freon-12	0.0252	0.2358	2.2240	1.5090
Freon-22	0.0218	0.4376	2.8080	1.8940

Table III. Coefficients A_m and B_m for Mixtures of Freon-12 and Freon-22 Vapors

Molar Fraction Freon-12 x_1	Freon-22 x_2	Coefficient $A_m \times 10^2$ Poise/(g./cc.)	$B_m \times 10^2$ Poise/(g./cc.)^2
1.0	0.0	0.0252	0.2358
0.8	0.2	0.0245	0.2762
0.6	0.4	0.0238	0.3165
0.4	0.6	0.0232	0.3569
0.2	0.8	0.0225	0.3970
0.0	1.0	0.0218	0.4376

$$A_m = \Sigma x_i A_i \tag{2}$$

$$B_m = \Sigma x_i B_i \tag{3}$$

Consequently, the residual viscosity of mixtures of gases may be predicted from A and B for pure components and density of the mixtures. Equation 1 is not valid for the dense gas and the uncompressed liquid range of substances.

The density of the mixtures may be computed from the equation found in standard texts on thermodynamics (*18*). The molar volume of the mixture, $\tilde{V}_m$, is given by the equation

$$\tilde{V}_m = \Sigma x_i \tilde{V}_i \tag{4}$$

Therefore, the density of the mixture, ρ_m, is

$$\rho_m = \frac{\Sigma x_i M_i}{\Sigma x_i \tilde{V}_i} \tag{5}$$

CALCULATIONS AND CORRELATIONS

The first step in the computational technique is to determine the zero density values of viscosity presented by Makita (*12*) at temperatures 25°, 50°, 100°, 150°, and 200° C., and covering a wide range of pressures at each temperature. The viscosity values at any temperature are plotted against density and the data are correlated by the least square method, using the expression

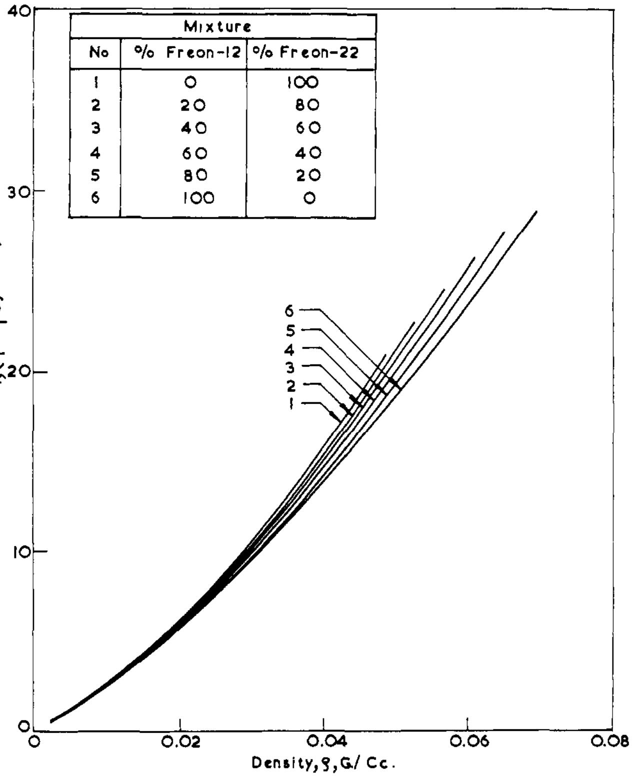
$$\mu = \mu_0 + a\rho + b\rho^2$$

The zero density values of viscosity, μ_0, so obtained, as well as the coefficients, a and b, are presented in Table I. The standard deviation, σ, defined in standard texts on statistical methods (*13*), is calculated by the following equation

$$\sigma = \left[\frac{1}{N}\sum_{i=1}^{N}(\mu_i - \bar{\mu}_i)^2\right]^{1/2} \tag{6}$$

The "greatest error" signifies the maximum difference between the experimental and correlated value of viscosity. The per cent greatest error is calculated on the basis of the experimental viscosity value.

Using the zero density values of viscosity, μ_0 given in Table I, and the experimental viscosity values of Makita (*12*), the residual viscosity, $(\mu - \mu_0)$, is plotted against density at any temperature and pressure. For Freon-12 and Freon-22 vapors, the residual viscosity is a unique



Mixture No	% Freon-12	% Freon-22
1	0	100
2	20	80
3	40	60
4	60	40
5	80	20
6	100	0

Figure 1. Variation of Residual Viscosity with Density for Mixtures of Freon-12 and Freon-22 Vapors

Table IV. Viscosity of Mixtures of Freon-12 and Freon-22 Vapors

Pressure, P.S.I.A.	Pure Components		Mixtures			
	$1.0x_1^{a}$ $0.0x_2^{b}$	$0.0x_1$ $1.0x_2$	$0.8x_1$ $0.2x_2$	$0.6x_1$ $0.4x_2$	$0.4x_1$ $0.6x_2$	$0.2x_1$ $0.8x_2$
	Viscosity, $\mu \times 10^6$, Poise					
	25° C.					
zero density	124.67^{c}	126.51^{c}	124.99^{d}	125.33^{d}	125.70^{d}	126.09^{d}
			125.17^{e}	125.62^{e}	126.00^{e}	126.31^{e}
	130.39^{f}	131.02^{f}	130.53^{f}	130.74^{f}	130.94^{f}	130.90^{f}
15	126.02^{c}	127.37^{c}	126.24^{g}	126.48^{g}	126.75^{g}	127.04^{g}
			126.42^{h}	126.77^{h}	127.05^{h}	127.26^{h}
	131.13^{f}	131.52^{f}	131.22^{f}	131.37^{f}	131.54^{f}	131.45^{f}
30	127.56^{c}	128.39^{c}	127.67^{g}	127.81^{g}	127.98^{g}	128.17^{g}
			127.85^{h}	128.10^{h}	128.28^{h}	128.39^{h}
	131.88^{f}	132.10^{f}	131.91^{f}	132.00^{f}	132.17^{f}	132.04^{f}
40	128.69^{c}	129.16^{c}	128.74^{g}	128.80^{g}	128.90^{g}	129.01^{g}
			128.92^{h}	129.09^{h}	129.20^{h}	129.23^{h}
	132.38^{f}	132.46^{f}	132.40^{f}	132.53^{f}	132.57^{f}	132.47^{f}
60	131.25^{c}	130.94^{c}	131.17^{g}	131.08^{g}	131.02^{g}	130.96^{g}
			131.35^{h}	131.37^{h}	131.32^{h}	131.18^{h}
	133.55^{f}	133.26^{f}	133.45^{f}	133.43^{f}	133.55^{f}	133.55^{f}
80	134.31^{c}	133.09^{c}	134.07^{g}	133.82^{g}	133.57^{g}	133.32^{g}
			134.25^{h}	134.11^{h}	133.87^{h}	133.54^{h}
	134.80^{f}	134.13^{f}	134.68^{f}	134.64^{f}	134.60^{f}	134.30^{f}
	50° C.					
zero density	129.38^{c}	134.72^{c}	130.31^{d}	131.30^{d}	132.37^{d}	133.50^{d}
			130.46^{e}	131.54^{e}	132.62^{e}	133.69^{e}
	140.06^{f}	140.80^{f}	140.23^{f}	140.46^{f}	140.98^{f}	140.59^{f}
15	130.62^{c}	135.51^{c}	131.45^{g}	132.35^{g}	133.33^{g}	134.37^{g}
			131.60^{h}	132.59^{h}	133.58^{h}	134.56^{h}
	140.75^{f}	141.25^{f}	140.87^{f}	141.04^{f}	141.51^{f}	141.09^{f}
50	134.09^{c}	137.85^{c}	134.71^{g}	135.39^{g}	136.15^{g}	136.95^{g}
			134.86^{h}	135.63^{h}	136.40^{h}	137.14^{h}
	142.33^{f}	142.46^{f}	142.42^{f}	142.54^{f}	142.89^{f}	142.42^{f}
80	137.81^{c}	140.51^{c}	138.26^{g}	138.74^{g}	139.23^{g}	139.85^{g}
			138.41^{h}	138.98^{h}	139.58^{h}	140.04^{h}
	144.01^{f}	143.63^{f}	143.93^{f}	143.91^{f}	144.24^{f}	143.67^{f}
120	144.24^{c}	145.13^{c}	144.42^{g}	144.58^{g}	144.74^{g}	144.90^{g}
			144.57^{h}	144.82^{h}	144.99^{h}	145.09^{h}
	146.35^{f}	145.34^{f}	146.14^{f}	146.19^{f}	146.29^{f}	145.50^{f}
160	153.13^{c}	151.31^{c}	152.81^{g}	152.47^{g}	152.11^{g}	151.72^{g}
			152.92^{h}	152.59^{h}	152.19^{h}	151.73^{h}
	149.71^{f}	147.33^{f}	149.22^{f}	148.84^{f}	148.78^{f}	147.74^{f}
	100° C.					
zero density	142.20^{c}	153.72^{c}	144.21^{d}	146.35^{d}	148.64^{d}	151.09^{d}
			144.28^{e}	146.47^{e}	148.77^{e}	151.19^{e}
	159.17^{f}	159.73^{f}	159.34^{f}	159.61^{f}	159.79^{f}	159.79^{f}
15	143.26^{c}	154.36^{c}	145.19^{g}	147.25^{g}	149.46^{g}	151.84^{g}
			145.26^{h}	147.37^{h}	149.59^{h}	151.94^{h}
	159.71^{f}	160.13^{f}	159.86^{f}	160.07^{f}	160.25^{f}	160.25^{f}
80	149.01^{c}	158.40^{c}	150.63^{g}	152.35^{g}	154.21^{g}	156.22^{g}
			150.70^{h}	152.47^{h}	154.34^{h}	156.32^{h}
	162.38^{f}	162.13^{f}	162.37^{f}	162.50^{f}	162.52^{f}	162.31^{f}
120	153.69^{c}	161.84^{c}	155.12^{g}	156.63^{g}	158.23^{g}	159.96^{g}
			155.19^{h}	156.75^{h}	158.36^{h}	160.06^{h}
	164.28^{f}	163.45^{f}	164.19^{f}	164.20^{f}	164.10^{f}	163.81^{f}
160	159.47^{c}	166.13^{c}	160.70^{g}	161.96^{g}	163.26^{g}	164.63^{g}
			160.77^{h}	162.08^{h}	163.39^{h}	164.73^{h}
	166.57^{f}	164.96^{f}	166.28^{f}	166.10^{f}	165.85^{f}	165.44^{f}
220	171.12^{c}	174.44^{c}	171.92^{g}	172.59^{g}	173.20^{g}	173.79^{g}
			171.99^{h}	172.71^{h}	173.33^{h}	173.89^{h}
	170.53^{f}	167.48^{f}	169.97^{f}	169.51^{f}	168.98^{f}	168.24^{f}
	150° C.					
zero density	156.55^{c}	169.31^{c}	158.78^{d}	161.15^{d}	163.69^{d}	166.40^{d}
			158.85^{e}	161.27^{e}	163.81^{e}	166.49^{e}
	177.99^{f}	178.73^{f}	178.13^{f}	178.57^{f}	178.76^{f}	178.61^{f}
15	157.50^{c}	169.89^{c}	159.65^{g}	161.94^{g}	164.41^{g}	167.04^{h}
			159.72^{h}	162.06^{h}	164.53^{h}	167.13^{h}
	178.46^{f}	179.08^{f}	178.59^{f}	178.98^{f}	179.17^{f}	178.96^{f}
80	162.30^{c}	173.26^{c}	164.19^{g}	166.21^{g}	168.39^{g}	170.72^{g}
			164.26^{h}	166.33^{h}	168.51^{h}	170.81^{h}
	180.80^{f}	180.81^{f}	180.77^{f}	181.08^{f}	181.09^{f}	180.81^{f}
120	166.00^{c}	175.99^{c}	167.74^{g}	169.59^{g}	171.57^{g}	173.69^{g}
			167.81^{h}	169.71^{h}	171.69^{h}	173.78^{h}
	182.29^{f}	181.90^{f}	182.23^{f}	182.47^{f}	182.42^{f}	182.06^{f}
160	170.32^{c}	179.29^{c}	171.93^{g}	173.61^{g}	175.38^{g}	177.26^{g}
			172.00^{h}	173.73^{h}	175.50^{h}	177.35^{h}
	184.09^{f}	183.11^{f}	183.90^{f}	183.96^{f}	183.78^{f}	183.31^{f}
220	178.35^{c}	185.37^{c}	179.74^{g}	181.09^{g}	182.45^{g}	183.86^{g}
			179.81^{h}	181.21^{h}	182.57^{h}	183.95^{h}
	186.98^{f}	185.15^{f}	186.57^{f}	186.48^{f}	186.16^{f}	185.53^{f}

[a] x_1 = Freon-12. [b] x_2 = Freon-22. [c] Viscosity by Equation 1 using zero density viscosity values reckoned from Makita's (*12*) experimental data. [d] Zero density viscosity values by Herning and Zipperer's (*4*) equation. [e] Zero density viscosity values by Wilke's (*19*) equation. [f] Viscosity by Dean and Stiel's (*3*) equation. [g] Viscosity by Equation 1, using zero density viscosity values by Herning and Zipperer's (*4*) equation. [h] Viscosity by Equation 1, using zero density viscosity values by Wilke's (*19*) equation.

function of density. The plot of residual viscosity against density is correlated by the least square method, using Equation 1. The values of *A* and *B* so obtained are given in Table II with the standard deviation, σ, and the probable error, P.E., calculated by the equation

$$\text{P.E.} = 0.6745\ \sigma \tag{7}$$

For evaluating the residual viscosity for mixtures, coefficients A_m and B_m are calculated, using Equations 2 and 3. Table III gives the computed values of A_m and B_m for the entire region of mixture composition. The results of residual viscosity are shown plotted against density in Figure 1.

In Table IV are given the predictions of viscosity of pure Freon-12 and Freon-22 vapors and their mixtures by the present correlation, based on the experimental data of Makita (*12*). The zero density viscosity values, μ_0, for mixtures are calculated by Herning and Zipperer's (*4*) equation and Wilke's (*19*) equation. The specific volumes of the pure components, obtained from tables found in standard texts on refrigeration (*14*), are used to evaluate the densities of the mixtures by Equation 5. The values of viscosity (the standard deviation is 14.6956×10^{-6} poise for Freon-12 vapor and 9.3036×10^{-6} poise for Freon-22 vapor) as computed by Dean and Stiel's (*3*) equations using the critical constants of the components (*7*) are presented in Table IV for comparison.

RESULTS AND DISCUSSIONS

Tables I, II, and III indicate that the present correlation satisfactorily represents the existing experimental data. A study of Table IV exhibits a large disparity in the values of viscosity given by the present correlation and the values computed by Dean and Stiel's (*3*)equations. This may be attributed to inherent errors in the method of determination of the pseudocritical constants (*3*). In Figure 1, a regular trend is noticed between the limits of the two pure components. This clearly brings out the effect of mixture composition on viscosity of the Freon-12–Freon-22 vapor mixture system.

CONCLUSION

The present correlation facilitates design of engineering systems, as viscosity information on pure Freon-12 and Freon-22 vapors and their mixtures may be obtained readily. The determination of viscosity, however, requires an accurate knowledge of the temperature-dependence of viscosity at zero density. For engineering purposes, it may be convenient to use the atmospheric viscosity values. These atmospheric viscosity values may be calculated for pure components by the correlation of Witzell and Johnson (*20*) and for mixtures by Herning and Zipperer's (*4*) equation or by Wilke's (*19*) equation.

NOMENCLATURE

a, b = coefficients in Table I
A, B = coefficients in Equation 1
A_m, B_m,
A_i, B_i = coefficients defined in Equations 2 & 3
M_i = molecular weight of the pure components
N = number of experimental points
P.E. = probable error
T = temperature
$\tilde{V}_m$ = molar volume of the mixture
$\tilde{V}_i$ = molar volume of the pure components
x_i = molar fractions of the pure components in the mixture
μ = viscosity at a given temperature and density
μ_0 = viscosity at the same temperature as μ but at zero density
$\bar{\mu}$ = viscosity as calculated from Equation 1
ρ = density
ρ_m = density of the mixture
σ = standard deviation

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Heterogeneous Phase Equilibrium in the Ethane–*n*-Dodecane System

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Vapor-liquid equilibrium compositions, pressures, and liquid molar volumes for the binary system of ethane and *n*-dodecane are presented for five isotherms between 0° and 100° C. The Flory-Huggins model of liquid solutions gave better results in fitting the experimental data than the Scatchard modification of regular solution theory.

THIS WORK was done to obtain accurate equilibrium data on the ethane–*n*-dodecane system and to test some of the simple solution models. The literature through 1967 shows no existing experimental data on this system. The ethane–*n*-octane system was reported recently by Rodrigues *et al.* (*16*).

The pure materials, ethane and *n*-dodecane, have been studied by many groups. The ethane has been studied extensively (*1–3, 13, 15, 17*) and the properties of *n*-dodecane were reported by several investigations (*8, 17*).

EXPERIMENTAL

The equipment and experimental techniques were described in detail by Kohn and Kurata (*11*) and were the same as those used in the previous studies of binary hydrocarbon systems (*9–11, 16–18, 20*). A complete schematic flow diagram of the apparatus is shown in Figure 1. The apparatus is mounted on a plywood housing with 1-inch, 90° steel flanges on the outside. The cell (*O*) temperatures were measured by a platinum resistance thermometer (*V*) and controlled by a Sargent Thermonitor proportional controller with probable error within ±0.02° C. The pressures were measured by Heise bourdon tube gages (*G*, *H*), with an accuracy of ±0.07 atm. The bourdon tube gages were checked frequently against an Ashcroft dead weight gage (*S*). The equilibrium cell (*O*) used in this study was a 10-ml. borosilicate glass cell and of the same type as those used in the earlier works.

The equilibrium cell was initially partially filled with a known amount of liquid *n*-dodecane. The weight was