Isothermal Vapor-Liquid Equilibria of Tetrachloromethane + Linear Ether or Acetal Mixtures at 298.15 K[†]

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Vapor-liquid equilibria for tetrachloromethane + a linear mono-, di-, or triether (diethyl ether, dipropyl ether, dibutyl ether, methyl butyl ether, ethyl butyl ether, 1,2-dimethoxyethane, 1,2-diethoxyethane, di(ethylene glycol) dimethyl ether) or an acetal (dimethoxymethane, diethoxymethane) were determined at 298.15 K by head-space gas chromatographic analysis of the vapor phase directly withdrawn from an equilibration apparatus. Excess molar Gibbs energies G^E for the investigated mixtures were determined by a least-squares treatment of the equilibrium data. The G^E values are generally negative and, though small, reflect effects due to (i) chain lengthening, (ii) position of the oxygen atom in the chain structure of monoethers, and (iii) distance of two -O- groups in diethers or acetals.

Introduction

In previous investigations (*1*), vapor-liquid equilibria (VLE) have been determined for binary mixtures of linear ethers or acetals with cyclohexane. In this paper, we present the results of VLE measurements for mixtures with tetrachloromethane (TCM) of the same oxygen compounds, namely, diethyl ether, dipropyl ether, dibutyl ether, methyl butyl ether, ethyl butyl ether, 1,2-dimethoxyethane, 1,2-diethoxyethane, di(ethylene glycol) dimethyl ether (diglyme), formaldehyde dimethyl acetal (dimethoxymethane), and formaldehyde diethyl acetal (diethoxymethane).

The head-space gas chromatographic (GC) analysis (*2*) was employed for measuring the vapor-phase mole fraction, y, as a function of liquid-phase mole fraction, x. This technique proved (*3*, *4*) to be simpler and faster than the conventional static or dynamic methods. The excess Gibbs energies, G^E, were calculated from the x-y data by a modified Barker method (*4*, *5*). G^E values are reported in the literature for TCM + diethyl ether (*6*), dipropyl ether (*7*), and dibutyl ether (*8*) only.

A detailed interpretation of G^E results in terms of DISQUAC, an extended quasichemical group contribution model (*9*), will be given elsewhere.

Experimental Section

An equilibration cell combined with a gas chromatograph (C. Erba 6000 Vega series) was employed for obtaining x-y data. The apparatus and procedure were described in detail in previous papers (*3*, *4*), where the reliability and suitability of the head-space GC technique for measuring VLE were checked. In most cases, the GC column was a 4 mm × 4 m stainless steel tube packed with Carbowax 20 M (C20M) on Chromosorb W 60-80. Also, columns packed with Porapak QS (PQS), Porapak N (PN), and Chromosorb 102 (C102) were employed.

Table I. Vapor Pressure p_i^*, Molar Volume V_i^*, Second Molar Virial Coefficients B_{ii}, and Cross Molar Virial Coefficients B_{ij}, at 298.15 K

compound	p_i^*/kPa	V_i^*/(cm^3 mol^{-1})	$-B_{ii}$/(dm^3 mol^{-1})	$-B_{ij}$/(dm^3 mol^{-1})
tetrachloromethane	15.23[a]	97.09[b]	1.6[f]	
diethyl ether	71.24[a]	104.72[e]	1.2[g]	1.2[i]
dipropyl ether	8.38[a]	137.68[e]	2.8[h]	1.7[i]
dibutyl ether	0.82[a]	170.43[e]	5.6[h]	2.3[i]
methyl butyl ether	18.51[a]	117.36[e]	1.9[h]	1.4[i]
ethyl butyl ether	7.51[a]	137.13[e]	2.8[h]	1.7[i]
dimethoxymethane	53.03[a]	89.12[e]	1.2[h]	1.0[i]
diethoxymethane	8.97[a]	126.35[e]	2.8[h]	1.6[i]
1,2-dimethoxyethane	9.93[b]	104.54[e]	1.9[h]	1.3[i]
1,2-diethoxyethane	1.84[c]	141.32[e]	4.0[h]	1.8[i]
diglyme	0.33[d]	142.91[e]	5.6[h]	1.9[i]
nitrogen	-	-	0.0[g]	0.1[j]

[a] Reference 11. [b] Reference 12. [c] Extrapolated from higher temperature data in ref 13. [d] Interpolated from ref 14. [e] Reference 15. [f] Extrapolated from higher temperature data in ref 16. [g] Reference 16. [h] Estimated from B_{ii} values (*16*) of related compounds. [i] Computed according to ref 10; the quoted values refer to mixtures with tetrachloromethane. [j] Reference 16; the same value applies for all mixtures of nitrogen (or oxygen) with listed substances.

All chemicals were products from Fluka or C. Erba of the best grade quality. They were used without further purification, and their purity, as checked by GC analysis, was 99.8 mass % for TCM and diethyl ether and 99.5 mass % for the other substances except dimethoxymethane, methyl butyl ether, and ethyl butyl ether, which contained about 1% impurities. Thanks to the GC technique employed in determining the vapor composition, the purity of substances is not a critical factor.

All mixtures were prepared by weighing. The liquid mole fractions x_i, after correction for buoyancy and for evaporation of constituents in the vapor space of the container, were evaluated to better than 0.0001.

The values of vapor mole fraction y_2 (the subscript 2 indicates TCM throughout this paper) were obtained from

$$y_2 = 1/\{1 + r_f/(A_2/A_1)\} \tag{1}$$

where A_2/A_1 is the measured peak area ratio of the two components in the vapor phase and r_f the response factor of the chromatograph for the mixture under examination. The overall uncertainty in y, resulting mostly from the uncertainty in A_2/A_1, was generally less than 0.005. In the dilute regions, the y precision was better than 2% of the lowest y_i.

Data Treatment

The experimental A_2/A_1 values are related to the ratio of the activity coefficients of the constituents, f_2/f_1, through the equation

$$(f_2/f_1)_{\text{exp}} = (x_1/x_2)(A_2/A_1)(w_1/w_2)(p_1^*/p_2^*)(1/r_f) \tag{2}$$

where p_i^* is the vapor pressure of the pure component i and w_i a factor relevant to vapor-phase nonideality. The latter was

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[†] Isothermal Vapor Liquid Equilibria of Mixtures Containing Organic Compounds. 4. Part 3: Reference 1. This work is a contribution to the TOM (Thermodynamics of Organic Mixtures) project (*9*).

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Table II. Experimental Vapor–Liquid Equilibrium Data (x–y) for Binary Mixtures at 298.15 K[a]

x_2	y_2	x_2	y_2	x_2	y_2
$(C_2H_5)_2O$ (1) + CCl_4 (2)					
0.0912	0.0199	0.5037	0.1831	0.8543	0.5697
0.1399	0.0333	0.5454	0.2108	0.9132	0.7091
0.2702	0.0745	0.6528	0.2968	0.9892	0.9554
0.4878	0.1313	0.7549	0.4090		
$(C_3H_7)_2O$ (1) + CCl_4 (2)					
0.0375	0.0596	0.4195	0.5568	0.7692	0.8676
0.1064	0.1642	0.5182	0.6553	0.9648	0.9822
0.2192	0.3189	0.5904	0.7340		
0.3216	0.4474	0.6670	0.7903		
$(C_4H_9)_2O$ (1) + CCl_4 (2)[b]					
0.1083	0.6548	0.5008	0.9469	0.8639	0.99274
0.2256	0.8287	0.5932	0.9643	0.8918	0.99427
0.3211	0.8880	0.7004	0.9788	0.9532	0.99784
0.4028	0.9194	0.7827	0.9867		
0.1096	0.6605	0.5144	0.9511	0.7629	0.9846
0.2349	0.8325	0.6390	0.9709	0.8174	0.9898
0.3651	0.9064	0.7022	0.9781	0.9199	0.99609
$CH_3OC_4H_9$ (1) + CCl_4 (2)[c]					
0.0966	0.0685	0.6255	0.5930	0.9448	0.9407
0.2121	0.1647	0.7525	0.7358	0.9573	0.9540
0.2783	0.2287	0.8666	0.8609	0.9680	0.9692
0.3894	0.3368	0.9261	0.9260		
0.0187	0.0126	0.4130	0.3485	0.9025	0.9007
0.0593	0.0412	0.5202	0.4691	0.9673	0.9667
0.1502	0.1179	0.6887	0.6601	0.9771	0.9753
0.1899	0.1485	0.7514	0.7394		
0.3075	0.2586	0.8241	0.8174		
$C_2H_5OC_4H_9$ (1) + CCl_4 (2)					
0.0829	0.1367	0.4584	0.6285	0.7822	0.8876
0.1725	0.2733	0.5555	0.7245	0.8359	0.9202
0.2815	0.4174	0.6464	0.7918	0.8713	0.9382
0.3691	0.5355	0.6999	0.8212	0.9452	0.9762
$(CH_3O)_2CH_2$ (1) + CCl_4 (2)					
0.0385	0.0110	0.4043	0.1673	0.7851	0.5092
0.0708	0.0226	0.5033	0.2266	0.8403	0.6032
0.1127	0.0359	0.5333	0.2511	0.9282	0.7904
0.2048	0.0698	0.6074	0.3073	0.9746	0.9203
0.2880	0.1061	0.6921	0.3924		
0.3560	0.1417	0.7428	0.4570		
$(C_2H_5O)_2CH_2$ (1) + CCl_4 (2)[d]					
0.1113	0.2845	0.3845	0.4882	0.8210	0.9039
0.1933	0.2490	0.5086	0.6264	0.9076	0.9590
0.2864	0.3720	0.5930	0.7132	0.9161	0.9619
0.3644	0.4642	0.7108	0.8212		
0.0976	0.1222	0.4844	0.6048	0.8229	0.9112
0.1145	0.1513	0.6005	0.7244	0.9436	0.9778
0.2438	0.3006	0.6376	0.7537	0.97022	0.9876
0.4094	0.5003	0.7240	0.8408		
$(CH_3OCH_2)_2$ (1) + CCl_4 (2)					
0.0547	0.0840	0.4817	0.5886	0.9088	0.9386
0.1652	0.2361	0.6071	0.6993	0.9239	0.9471
0.2663	0.3622	0.7431	0.8070	0.9669	0.9771
0.3651	0.4724	0.8382	0.8858	0.9783	0.9852
$(C_2H_5OCH_2)_2$ (1) + CCl_4 (2)					
0.0533	0.2636	0.3799	0.8236	0.7591	0.9676
0.1501	0.5404	0.4878	0.8837	0.8424	0.9816
0.2637	0.7133	0.5774	0.9191	0.9067	0.9906
0.2894	0.7510	0.6660	0.9461	0.97615	0.9979
$(CH_3OCH_2CH_2)_2O$ (1) + CCl_4 (2)[e]					
0.0499	0.7117	0.3870	0.9644	0.9076	0.99772
0.0764	0.07835	0.4866	0.9752	0.9655	0.99916
0.1644	0.8999	0.7221	0.99204		
0.2582	0.9405	0.7616	0.99321		
0.0540	0.7299	0.4000	0.9677	0.7953	0.99458
0.1228	0.8561	0.5452	0.9818	0.8625	0.99665
0.2407	0.9314	0.7476	0.99240	0.9184	0.99800

[a] The y values (which are consistent with p_i^* values in Table I) were obtained from eq 1 using the least-squares r_f's from Table III and experimental A_2/A_1 ratios. [b] The two sets of data were obtained with two different GC columns packed with C20M (upper series) and PQS, respectively. [c] The two series of data were obtained with a C102 packed GC column and different flow rates of the carrier gas. [d] The two sets of data were obtained with two different GC columns packed with PN (upper series) and PQS, respectively. [e] The upper set of measurements were carried out with a C20M GC column at 130 °C and with 0.5 mL of vapor injected; the other group of measurements were made with the same column but with programmed increase of temperature (initial isotherm at 80 °C and final isotherm at 150 °C) and 3 mL of vapor injected.

calculated by taking into account the presence of air in the vapor phase, assumed to be a three component mixture, nitrogen being the third component (*3*). The expression of w_i is given elsewhere (*3*, *10*). In Table I are collected the values of the quantities $p_i^\bullet$, $V_i^\bullet$, and the molar second virial coefficients B_{ii} and B_{ij} used for evaluating w_i. Although most of the virial coefficients B_{ii} and B_{ij} used in the calculation are estimated values and probably affected by large uncertainties, the w_i terms differ from 1 by less than 5% for all mixtures studied and partially compensate in the ratio w_1/w_2.

The activity coefficients in eq 2 were smoothed with the Redlich–Kister equation:

$$\ln f_{i,\mathrm{calc}} = \{c_1 + 3(-1)^j c_2\}x_j^2 - 4(-1)^j c_2 x_j^3 \qquad (3)$$

$$(i,j = 1,2;\ i \neq j)$$

and the coefficients c_m were obtained by a nonlinear least-squares procedure, similar to that described by Barker (*5*) for the treatment of P–x data. In our case, the objective function is

$$S = \sum_{k=1}^{n} \{\ln (f_2/f_1)_{\mathrm{exp}} - \ln (f_2/f_1)_{\mathrm{calc}}\}_k^2 \qquad (4)$$

the sum S extended over all n experimental points. The product $(p_1^*/p_2^*)(1/r_f)$, appearing in the expression for $(f_2/f_1)_{\mathrm{exp}}$, was treated as an adjustable parameter to be determined in the least-squares calculations. This is an important advantage of the present head-space GC technique for measuring VLE. In fact, neither the direct determination of the response factor r_f via calibration experiments nor the measurement of the vapor pressures $p_i^\bullet$ are necessary for obtaining activity coefficients.

For each of the systems involving dibutyl ether, methyl butyl ether, diethoxymethane, and diglyme, two different series of x–y measurements were carried out in which either different GC columns or different chromatographic variables such as oven temperature, flow of carrier gas, and volume of sampling loop were employed. In these cases, use was made of a least-squares method that was able to process data of different series altogether and to find a single set of c_m coefficients with two different r_f values.

Excess Gibbs energies G^E were calculated through the equation

$$G^E/RT = x_1x_2\{c_1 + c_2(x_1 - x_2)\} \qquad (5)$$

Results and Discussion

The experimental x–y quantities are collected in Table II. We preferred to report here the vapor mole fraction y from eq 1 instead of the more direct experimental quantity f_2/f_1 because of the wide use of y in practical applications. The c_m values are reported in Table III together with the values of the response factor r_f obtained from the fitting (and from the $p_i^\bullet$ values in Table I) and the standard deviation s for the repre-

Table III. Coefficients c_m, Equation 3, Chromatographic Response Factor r_f, Standard Deviation s for Representation of $\ln (f_2/f_1)$, and Excess Gibbs Energy G^E at $x = 0.5$ for Ether + Tetrachloromethane Binary Mixtures at 298.15 K

no.	ether	GC column	r_f	c_1	c_2	s^a	$G^E(x=0.5)^b/(\mathrm{J\ mol^{-1}})$
1	diethyl ether	PQS	1.329	−0.0548		0.015	−34 ± 5[c]
2	dipropyl ether	C20M	0.931	−0.1188		0.020	−74 ± 7[d]
3	dibutyl ether	C20M	0.622[e]	−0.2052	0.0428	0.018	−127 ± 5[f]
		PQS	0.714[e]				
4	methyl butyl ether	C102	0.776[e]	−0.1796		0.034	−111 ± 7
		C102	0.860[e]				
5	ethyl butyl ether	C20M	0.928	−0.1598		0.017	−99 ± 7
6	dimethoxymethane	PQS	1.533	0.0132		0.023	8 ± 6
7	diethoxymethane	PN	1.074[e]	−0.3420	0.0961	0.035	−212 ± 10
		PQS	1.229[e]				
8	1,2-dimethoxyethane	C20M	0.916	0.0470		0.017	29 ± 5
9	1,2-diethoxyethane	C20M	0.587	−0.2992	0.0407	0.019	−185 ± 6
10	diglyme	C20M	0.417[e]	−0.0070		0.044	−4 ± 10
		C20M	0.773[e]				

[a] $s = [\sum\{\ln (f_2/f_1)_{\mathrm{calc}} - \ln (f_2/f_1)_{\mathrm{exp}}\}^2/(n-q)]^{1/2}$, q being the number of c_m coefficients. [b] Evaluated from eq 5; the quoted uncertainties are standard deviations and were calculated from the errors on c_1 by the usual formula of error propagation. The standard deviations of c_m coefficients, not reported in the table, were obtained from s in the least-squares treatment. [c] A value of −75 J mol^{-1} was reported by Becker et al. (*6*). [d] A value of −57.8 J mol^{-1} was reported by Williamson (*7*). [e] The two r_f values of the quoted system are consistent with two different series of x–y data obtained either with different GC columns and/or different chromatographic conditions (see footnotes in Table II). Both series of data are described by a single set of c_m parameters (see text). [f] Values of −127.2 J mol^{-1} at 308.15 K and, by extrapolation, of −131.6 J mol^{-1} at 298.15 K are quoted in ref 8a. A revised value of −128.7 J mol^{-1} at 308.15 K was reported in ref 8b.

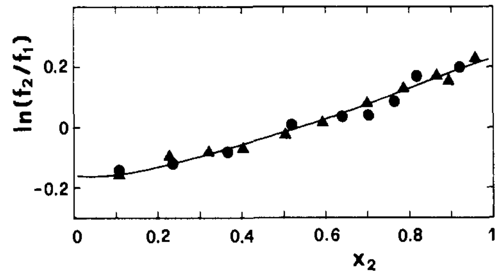

Figure 1. Plot of $\ln (f_2/f_1)$ against x_2 for dibutyl ether (1) + TCM (2) at 298.15 K: experimental results obtained with (▲) C20M or (●) PQS GC columns; (—) calculated from Redlich–Kister equation with coefficients given in ref 8a.

sentation of $\ln (f_2/f_1)$. This latter value on the average amounts to 0.02; the standard deviation of y was usually less than 0.004. In Table III, the excess Gibbs energies for equimolar mixtures, G^E ($x = 0.5$), as well as their standard deviations are also reported. These latter values were calculated from the standard deviations of the c_1 coefficient and ranged from 5 to 10 J mol^{-1}. No more than two c_m coefficients were necessary for fitting data to eq 3, which was found more appropriate than other equations such as Wilson or NRTL (nonrandom two liquids).

In Figure 1, the plot of $\ln (f_2/f_1)$ vs x_2 is shown for TCM + dibutyl ether. A fairly good agreement is observed between experimental results obtained with two different GC columns that differed in r_f. The data of Ott et al. (*8*), obtained with a conventional apparatus, lie in between.

The G^E curves, computed by eq 5, are represented in Figure 2. This shows that TCM + ether or acetal mixtures exhibit small, usually negative, deviations; in particular, for the compounds containing two methoxy groups, dimethoxymethane, dimethoxyethane, and diglyme, G^E values are very near to zero. The G^E curves are symmetrical or slightly asymmetrical, with their minima at $0.4 < x_1 < 0.5$. Our G^E values agree well with the results of Marsh and Ott (*8*) for dibutyl ether and differ by less than 40 J mol^{-1} from the results of Becker et al. (*6*) for diethyl ether and of Williamson (*7*) for dipropyl ether.

Molar excess enthalpies H^E at 298.15 K are known (*17*) for binary mixtures herein studied. From these smoothed calorimetric H^E data, which are all negative, and from G^E values given by eq 5, molar excess entropies S^E were calculated and found all negative.

When the dependence of the thermodynamic excess properties at $x = 0.5$ on the ether structure for TCM + ether systems is examined, it can be recognized that both G^E and $-TS^E$ decrease with the number of carbon atoms in the ether chain and increase with the O–O distance in the diethers, whilst the opposite happens for H^E. Moreover, there is clear evidence for the effect of position of the –O– atom in the chain structure: symmetrical monoethers have higher G^E than asymmetrical ones. A similar X^E ($X = G, H, S$) dependence on the chain length is shown by mixtures of n-alkanes in TCM (*18–20*). Similarities are also found when G^E values of the present mixtures are compared with those of mono- and polyethers + cyclohexane (*1*) and n-alkanes (*21*).

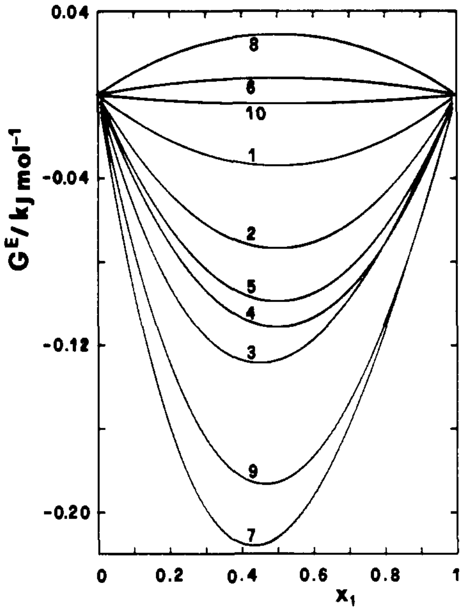

Figure 2. Calculated G^E curves for binary systems ether (1) + TCM (2) at 298.15 K. The numbers correspond to the systems as indicated in Table III.

In Table IV are collected the values of the activity coefficients at infinite dilution, f_i^∞, obtained from the parameters of eq 3, as well as the molar Gibbs energies of solvation ΔG_i° of the single compounds in the molar concentration scale. These were obtained through the equation

$$\Delta G_i^\circ = RT \ln f_i^\infty + RT \ln (p_i^* V_j^* / RT) \qquad (6)$$

where V_j^* is the molar volume of the solvent. From the trend of ΔG_i° of ethers in TCM, an average increment of −2.9 kJ mol^{-1} is calculated for the standard molar Gibbs energy of

Table IV. Activity Coefficients at Infinite Dilution f_i^∞ and Molar Gibbs Energies of Solvation ΔG_i° of the Components of Binary Ether (1) + Tetrachloromethane (2) Mixtures at 298.15 K

ether	f_1^∞	$-\Delta G_1^{\circ a}$/ (kJ mol^{-1})	f_2^∞	$-\Delta G_2^{\circ a}$/ (kJ/mol^{-1})
diethyl ether	0.947	14.72	0.947	18.35
dipropyl ether	0.888	20.18	0.888	17.83
dibutyl ether	0.780	26.26	0.850	17.41
methyl butyl ether	0.836	18.37	0.836	18.38
ethyl butyl ether	0.852	20.55	0.852	17.95
dimethoxymethane	1.013	15.28	1.013	18.59
diethoxymethane	0.645	20.80	0.782	18.36
1,2-dimethoxyethane	1.017	19.42	1.017	18.18
1,2-diethoxyethane	0.712	24.49	0.772	18.12
diglyme	0.993	27.92	0.993	17.46

[a] ΔG_i° is referred to the transfer process component(ideal gas, 1 mol dm^{-3}) = component(solute obeying Henry's law, 1 mol dm^{-3}) and evaluated through eq 6. The value for the standard process component(ideal gas, 101.325 kPa) = component(solute obeying Henry's law, at unit mole fraction) can be calculated as $\Delta G_i^\circ = RT \ln (f_i^\infty p_i^* / (101.325 \text{ kPa}))$.

solvation of a CH_2 group in TCM. This value is mostly determined by the p_i^* trend in the series, since the f_i^∞'s are near to 1 for all the compounds. The same value was found for the solvation in cyclohexane of a CH_2 group of linear ethers (*1*). From ΔG_i°'s of *n*-alkanes in TCM, collected by Abraham (*22*), an increment of -3.5 kJ mol^{-1} is obtained in a similar way.

Acknowledgment

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Glossary

A_i	peak area of component *i* in the vapor phase
B_{ij}	molar second virial coefficient (*i*, *j* = 1, 2), dm^3 mol^{-1}
c_m	coefficients in eqs 3 and 5
f_i	activity coefficient of component *i*
f_i^∞	activity coefficient of component *i* at infinite dilution
G^E	excess Gibbs energy, J mol^{-1}
ΔG_i°	Gibbs energy of solvation of component *i*, kJ mol^{-1}
p_i	partial pressure of component *i*, kPa
p_i^*	vapor pressure of pure liquid *i*, kPa
R	molar gas constant, 8.314 51 J mol^{-1} K^{-1}
r_f	response factor of gas chromatograph for a pair of components
s	standard deviation of the fit
S	sum of residuals, eq 4
T	temperature, K
V_i^*	molar volume of pure liquid *i*, cm^3 mol^{-1}
w_i	vapor-phase nonideality factor of *i*
x_i	liquid-phase mole fraction of component *i*
y_i	vapor-phase mole fraction of component *i*

Registry No. CCl_4, 56-23-5; diethyl ether, 60-29-7; dipropyl ether, 111-43-3; dibutyl ether, 142-96-1; methyl butyl ether, 628-28-4; ethyl butyl ether, 628-81-9; 1,2-dimethoxyethane, 110-71-4; 1,2-diethoxyethane, 629-14-1; diglyme, 111-96-6; dimethoxymethane, 109-87-5; diethoxymethane, 462-95-3.

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