Vapor-Liquid Equilibria of the Trifluoromethane–Trifluorochloromethane System

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Vapor-liquid equilibrium data were determined by the vapor recirculation technique for the trifluoromethane–trifluorochloromethane binary at 31.9, −0.1, −55.1, and −100.05°F over the full range of composition. The compositions of the maximum-pressure (minimum-boiling) azeotrope exhibited by this system are 0.656, 0.630, 0.579, and 0.533 mol fraction CHF_3, respectively, at pressures of 400.6, 247.1, 90.47, and 31.32 psia, respectively. The heats of vaporization of the azeotropes computed primarily from the equilibrium data, are 3639, 4723, 5845, and 6363 Btu/lb mol, respectively. Liquid-phase activity coefficients were calculated from the experimental data under the assumption that the Redlich–Kwong equation of state gave an adequate description of the vapor phase. Subsequently, the Wilson equation was fit to these activity coefficients, and a correlation was developed which permits the vapor-liquid equilibria to be computed for any temperature between 32 and −100°F.

A specified composition of trifluoromethane and trifluorochloromethane, particularly 49–51 mol % trifluoromethane, is patented (*1*) and marketed as Refrigerant 503. An investigation of Refrigerant 503 has been published (*10*) in which the equimolar mixture was treated as a pure material. In our continuing program of studies of the thermodynamic properties of mixtures of the more volatile fluorocarbons, it became of interest to determine the vapor-liquid equilibria of the trifluoromethane–trifluorochloromethane binary over the complete composition range. The temperature range covered was as wide as could be conveniently handled in our experimental apparatus. The highest temperature (32°F) was limited by the apparatus design which requires that equilibrium temperatures be lower than ambient temperature, and the lowest temperature (−100°F) was limited by the requirement that the associated equilibrium pressures be large enough to permit extraction of sufficient phase samples for analysis.

EXPERIMENTAL

An apparatus of the vapor-recirculation type, similar to one described previously (*11*), was used in this investigation. With this technique, vapor was continuously removed from the top of the vapor-liquid equilibrium cell and reintroduced at the bottom of the cell where it bubbled up through the liquid, establishing the vapor-liquid contact needed to reach equilibrium. Between removal from the top of the cell and reintroduction at the bottom, the vapor passed through a vapor-sampling coil, a pump, a variable-volume device, which permitted removal of samples from the system without reduction in system pressure, and a conditioning coil, which ensured that the vapor reintroduced at the bottom of the cell was at the equilibrium temperature. This entire vapor-circulating loop had a maximum volume of approximately 150 cc. The cell, which was usually operated at about two-thirds full of liquid, was 11 cc in volume. When equilibrium had been attained, the vapor sample was entrapped in the vapor-sampling coil between two valves and was bled off into a previously evacuated 75-cc cylinder. Liquid samples were obtained through a very small diameter probe which extended to the bottom of the equilibrium cell. The liquid sample flashed across a valve into a previously evacuated 75-cc cylinder. Details of the construction of the apparatus and of the experimental technique used have been described elsewhere (*5*).

The pressure of the system was measured with an accuracy of ±0.1 psia along the 32° and 0°F isotherms and to ±0.01 psia along the −55° and −100°F isotherms by means of Heise Bourdon-tube gauges, which had been previously calibrated against a precision deadweight gauge manufactured by the Ruska Instrument Corp. The weights of the deadweight gauge were calibrated by Ruska against class S standards, and the area of the piston was certified by Ruska on the basis of light-wave micrometer readings accurate to 5×10^{-6} in. The temperature was measured with a three-junction copper-constantan thermopile, used in conjunction with a Leeds and Northrup type K-3 potentiometer, to within ±0.02°F; the temperature scale was established by calibration against a platinum resistance thermometer which had been previously calibrated by the U.S. National Bureau of Standards.

Most of the phase samples were available at a pressure of approximately 55 psia in 75-cc stainless steel cylinders. Even those samples obtained at the lowest experimental pressures, available at about 15 psia, were quite adequate for analysis. The phase samples were analyzed with a Beckman Instruments, Inc. GC-2A gas chromatograph using a di-*n*-butyl maleate column for separation and thermal conductivity for detection. An electronic integrator was used to determine the peak areas. The composition of each phase sample was determined by direct ratio of the peak area of the unknown sample to the peak area of a known standard sample of similar composition. The standard and the unknown were always run in immediate succession. Most pairs were run in duplicate. The standard samples, representing the composition range studied, were prepared at a total pressure of two atmospheres. The composition of each standard was determined from partial pressure measurements coupled with second-virial-coefficient corrections, which were quite small for the two-atmosphere-and-less pressure range involved. The calibration procedure for these standard samples and the analytical apparatus and technique are reported elsewhere (*8*). The estimated error in each reported CHF_3 and $CClF_3$ composition is less than 0.3% of the lesser mole fraction.

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Table I. Experimental Vapor-Liquid Equilibria and Calculated Fugacity and Activity Coefficients for the CHF_3–$CClF_3$ System

Pressure, psia	Mole fraction CHF_3					
	Liquid	Vapor	ϕ_1	ϕ_2	γ_1	γ_2
	Temp 31.90°F					
285.2	0.0	0.0	...	0.767	...	1.000
309.6	0.0615	0.0979	0.881	0.747	1.588	1.010
330.5	0.1240	0.1890	0.855	0.733	1.568	1.012
351.3	0.2021	0.2783	0.829	0.720	1.454	1.027
366.7	0.2768	0.348	0.809	0.712	1.350	1.052
380.6	0.365	0.436	0.788	0.709	1.293	1.066
390.6	0.462	0.506	0.773	0.709	1.191	1.128
396.3	0.538	0.564	0.763	0.712	1.139	1.180
398.6	0.564	0.586	0.759	0.713	1.129	1.196
399.1	0.582	0.598	0.758	0.714	1.115	1.215
400.5	0.652	0.652	0.752	0.721	1.082	1.277
398.9	0.7371	0.7240	0.747	0.734	1.052	1.361
394.8	0.8062	0.7829	0.747	0.747	1.029	1.465
389.0	0.8598	0.8355	0.748	0.761	1.018	1.542
376.6	0.9378	0.9194	0.754	0.785	1.004	1.707
362.3	1.0	1.0	0.763	...	1.000	...
	Temp −0.10°F					
176.6	0.0	0.0	...	0.825	...	1.000
189.3	0.0427	0.0863	0.906	0.813	1.939	1.005
200.9	0.0922	0.1670	0.890	0.803	1.807	1.009
211.4	0.1456	0.2368	0.875	0.795	1.676	1.020
220.1	0.2003	0.2963	0.863	0.788	1.562	1.036
227.0	0.2529	0.347	0.853	0.784	1.477	1.053
235.3	0.336	0.429	0.840	0.781	1.398	1.068
239.8	0.397	0.475	0.832	0.781	1.321	1.100
243.1	0.463	0.514	0.826	0.781	1.234	1.158
245.3	0.516	0.549	0.822	0.782	1.187	1.203
246.7	0.594	0.605	0.817	0.786	1.135	1.270
246.9	0.663	0.655	0.814	0.791	1.096	1.348
245.2	0.7390	0.7121	0.812	0.798	1.061	1.455
242.5	0.7999	0.7635	0.812	0.807	1.040	1.558
237.7	0.8585	0.8209	0.814	0.818	1.025	1.660
232.8	0.9033	0.8668	0.817	0.827	1.011	1.793
228.2	0.9378	0.9092	0.819	0.837	1.006	1.886
217.3	1.0	1.0	0.827	...	1.000	...

Pressure, psia	Mole fraction CHF_3					
	Liquid	Vapor	ϕ_2	ϕ_2	γ_1	γ_2
	Temp −55.10°F					
64.70	0.0	0.0	...	0.908	...	1.000
70.70	0.0425	0.1100	0.943	0.900	2.535	1.005
75.38	0.0907	0.1958	0.935	0.894	2.232	1.012
78.76	0.1320	0.2526	0.929	0.891	2.053	1.024
82.68	0.1982	0.3229	0.922	0.887	1.820	1.048
85.45	0.2617	0.377	0.917	0.884	1.654	1.078
87.73	0.337	0.431	0.912	0.883	1.501	1.122
89.42	0.433	0.495	0.908	0.882	1.357	1.189
89.89	0.470	0.516	0.907	0.883	1.308	1.226
90.18	0.499	0.533	0.906	0.883	1.277	1.253
89.93	0.665	0.629	0.903	0.887	1.124	1.493
89.42	0.7067	0.656	0.903	0.889	1.096	1.578
88.39	0.7614	0.6928	0.903	0.892	1.063	1.716
86.59	0.8249	0.7497	0.904	0.897	1.042	1.878
84.49	0.8702	0.7975	0.905	0.902	1.027	2.012
81.46	0.9207	0.8588	0.908	0.909	1.012	2.233
79.15	0.9526	0.9071	0.910	0.914	1.006	2.404
74.41	1.0	1.0	0.915	...	1.000	...
	Temp −100.05°F					
22.38	0.0	0.0	...	0.956	...	1.000
25.24	0.0485	0.1416	0.969	0.951	3.115	1.011
26.67	0.0868	0.2074	0.966	0.948	2.685	1.025
28.33	0.1472	0.2909	0.962	0.946	2.349	1.039
29.65	0.2126	0.343	0.959	0.944	1.998	1.089
30.18	0.2507	0.376	0.958	0.943	1.890	1.105
30.51	0.2873	0.396	0.957	0.943	1.753	1.137
31.10	0.412	0.482	0.955	0.943	1.515	1.204
31.31	0.538	0.535	0.954	0.943	1.294	1.385
31.30	0.565	0.548	0.954	0.943	1.261	1.431
31.30	0.564	0.550	0.954	0.943	1.268	1.421
31.08	0.635	0.580	0.954	0.944	1.180	1.574
30.99	0.657	0.591	0.954	0.944	1.159	1.625
30.89	0.6675	0.599	0.954	0.945	1.152	1.641
30.50	0.7544	0.646	0.954	0.946	1.086	1.939
30.11	0.7839	0.6733	0.954	0.947	1.076	2.010
29.84	0.8148	0.6912	0.955	0.948	1.053	2.199
29.03	0.8538	0.7392	0.955	0.950	1.047	2.295
27.58	0.9118	0.8121	0.957	0.954	1.025	2.616
25.15	0.9747	0.9252	0.961	0.960	1.001	3.334
23.80	1.0	1.0	0.963	...	1.000	...

The CHF_3 was analyzed by the supplier, E. I. du Pont de Nemours & Co., and stated to be 99.98% pure. The purity of the $CClF_3$ was 99.90%; the major impurity was determined chromatographically, as described above, to be 0.10% CF_4.

RESULTS

Experimental vapor-liquid equilibrium data were determined at 31.90°, −0.10°, −55.10°, and −100.05°F. The data are given in Table I, and the pressure-composition diagram for 31.9°F is shown in Figure 1. A positive (minimum-boiling) azeotrope exists for this system.

Table II. Azeotropes in the CHF_3–$CClF_3$ System

Temp, °F	Pressure, psia	Mole fraction, CHF_3
31.90	400.6	0.656
−0.10	247.1	0.630
−55.10	90.47	0.579
−100.05	31.32	0.533

DISCUSSION OF RESULTS

The nature of the experimental apparatus and procedures made the attainment of the exact azeotropic conditions a matter of chance. Consequently, the azeotropic conditions were determined graphically from data on both sides of and very near to the azeotrope. These conditions are given in Table II. A graph of ln P vs. $1/T$ (absolute) of the data in Table II results in a straight line, which when extrapolated to one atmosphere yields a boiling point of −88.14°C, compared to −87.85°C cited by Sinka et al. (*10*) as the atmospheric-pressure, minimum-boiling point of Refrigerant 503. The observed change in composition of the azeotrope with pressure is in agreement with the qualitative rule credited to Kamienski (*12*), that an increase in pressure on a binary positive azeotrope produces a concentration increase of the component having a lower dT/dP value (CHF_3 in this case).

Thermodynamic consistency of the data was checked by application of the area test (*7*) to the liquid-phase activity coefficients. The experimental values of log (γ_1/γ_2) were fitted with a third-order polynomial in x_1, and the integration was performed analytically to obtain an objective evaluation of the areas. The method of calculation of the activity coefficients and the inherent assumptions in them are given in the subsequent section. The results of the area test are shown in Table III. The thermodynamic consistency is good, within the framework of the current state of the art (*7*).

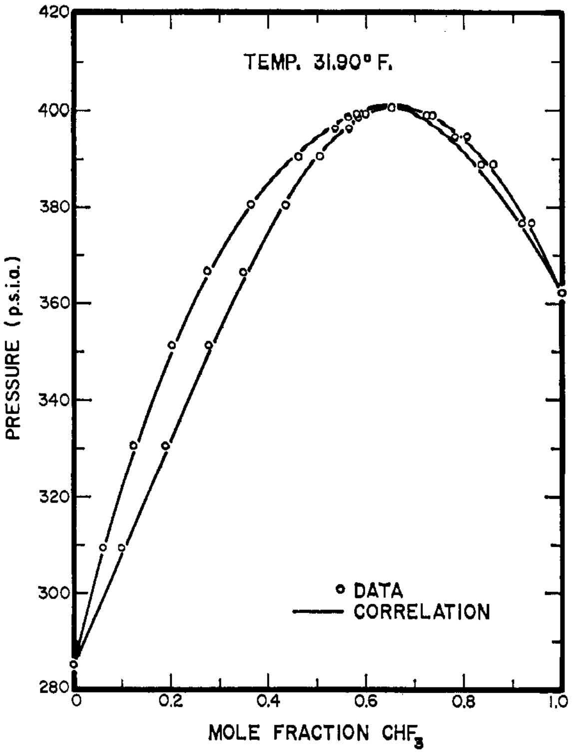

Figure 1. Pressure-composition plot for $CHF_3/CClF_3$ at 31.90° F

Table III. Thermodynamic Consistency

Temp, °F	$\left(\frac{\text{Pos. area} - \text{neg. area}}{\text{Pos. area} + \text{neg. area}}\right) \times 100$
31.90	+0.8%
−0.10	+1.7%
−55.10	+2.1%
−100.05	+0.1%

Table IV. Computed Enthalpy Quantities for the Azeotropes

Temp, °F	Heat of vaporization, Btu/lb mol	Liquid-phase integral heat of mixing, Btu/lb mol
31.90	3639	337
−0.10	4723	181
−55.10	5845	219
−100.05	6363	401

Table V. Properties of the Pure Components Used in Calculations

	CHF_3	$CClF_3$	Ref.
Critical temp, °R	538.33	543.62	*(3, 4)*
Critical press., psia	701.4	561.3	*(3, 4)*
Critical vol, ft³/lb mol	2.1356	2.8961	*(3, 4)*
Acentric factor	0.2654	0.1703	*(3, 4)*
Ω_a	0.4532	0.4400	This work
Ω_b	0.0923	0.0894	This work
Enthalpy of vaporization, Btu/lb mol			*(3, 4)*
31.90°F	4215	3969	
−0.10	5122	4795	
−55.10	6290	5865	
−100.05	7007	6537	
Liquid vol, ft³/lb mol			*(3, 4)*
31.90°F	1.089	1.495	
−0.10	0.972	1.357	
−55.10	0.855	1.208	
−100.05	0.796	1.123	

A comparison can be made between the bubble-point pressures of equimolar mixtures and the work on Refrigerant 503 by Sinka et al. (*10*) who treated 503 as a pure material and presented a vapor-pressure (bubble-point) equation for it. Bubble-point pressures were determined by interpolation of the experimental data at the equimolar composition. They were found to be 394.1, 244.5, 90.18, and 31.31 psia. Agreement with the Sinka et al. equation exists within experimental error at the two higher pressures (temperatures). The differences are 0.67 and 0.27 psia at −55° and −100°F, respectively (data are lower); −100°F is slightly outside of the range of the equation.

The general coexistence equation, derived by combining the general form of the Gibbs-Duhem equation for the vapor phase and for the liquid phase according to the requirements for phase equilibrium applied to the locus of azeotropic conditions, results in the following equation (*13*):

$$[H(\mathrm{v}) - H(\mathrm{l})] = R\left(\frac{-d\ln P}{d\frac{1}{T}}\right)[Z(\mathrm{v}) - Z(\mathrm{l})] \qquad (1)$$

Thus, the data on variation of the pressure with the temperature of the azeotrope provide the key information required to extract enthalpy quantities from the phase equilibrium data. The compressibility factor of the azeotropic vapor was computed from the Redlich-Kwong equation of state (*9*), consistent with the details given in the subsequent section. The compressibility factor of the liquid was computed by using the method and corresponding-states correlation of Chueh and Prausnitz (*7*) for calculation of the volume of the saturated liquid mixture. This part of the calculation did not introduce serious error because Z(l) was the minor term in the difference; it ranged from 0.8% (at −100°F) to 16% (at 32°F) of Z(v).

The results of the calculation of heats of vaporization of the azeotropes from Equation 1 are given in Table IV. With the heat of vaporization in hand, the integral heat of mixing of the liquid phase can be calculated by making computations about a thermodynamic cycle via the ideal-gas state. Such a cycle requires the heat of vaporization of the pure components (Table V) and an equation of state to compute the effect of pressure on the vapor-phase mixture and pure components. The Redlich-Kwong equation, having been used previously in the heat-of-vaporization calculation, was used for this calculation. The very minor effect of pressure on the enthalpy of the pure liquids between their vapor pressures and the azeotropic pressure was neglected. The results for the liquid-phase integral heat of mixing of the azeotropes are given in Table IV.

CORRELATION OF EXPERIMENTAL DATA

The correlation presented hereinafter follows closely a method demonstrated in several places by Prausnitz: See, for example, reference *7*; consequently, only a brief description featuring the numerical values, which are listed in Table V, and equations required to use the correlation is given.

Thermodynamic equilibrium between a liquid phase and a vapor phase is established when the temperature and pressure of both phases are equal and when the fugacity of each molecular species present is the same in both phases. In terms of two auxiliary functions, the fugacity coefficient and the activity coefficient, the equality of fugacity at equilibrium for any component "i" can be written

$$\phi_i y_i P = \gamma_i x_i f_i^{\circ}(\mathrm{l}) \qquad (2)$$

The pressure–volume–temperature properties of the vapor phase were described by the Redlich–Kwong equation of state (*9*). For this equation of state, the fugacity coefficient of component i can be written as follows:

$$\ln \phi_i = \ln \frac{v}{v-b} + \frac{b_i}{v-b} - \frac{2\sum_{j=1}^{N} y_j a_{ij}}{RT^{3/2}b} \ln \frac{v+b}{v} + \frac{ab_i}{RT^{3/2}b^2}\left(\ln \frac{v+b}{v} - \frac{b}{v+b}\right) - \ln \frac{Pv}{RT} \qquad (3)$$

where

$$a = \sum_{i}^{N}\sum_{j}^{N} y_i y_j a_{ij} \text{ and } b = \sum_{i=1}^{N} y_i b_i \qquad (3a)$$

The constants were obtained from

$$a_{ii} = \frac{\Omega_{a_{ii}} R^2 T_{c_{ii}}{}^{2.5}}{P_{c_{ii}}} \text{ and } a_{ij} = \frac{(\Omega_{a_{ii}} + \Omega_{a_{jj}}) R^2 T_{c_{ij}}{}^{2.5}}{2\,P_{c_{ij}}} \; (i \neq j) \qquad (3b)$$

$$b_i = \frac{\Omega_{b_i} R T_{c_i}}{P_{c_i}} \qquad (3c)$$

Equation 3b for a_{ij} and the mixing rules required follow Chueh and Prausnitz (*2*). The dimensionless constants, Ω_a and Ω_b, were evaluated by fitting the Redlich–Kwong equation to the saturated vapor volumes (*3*, *4*) for each component from the normal boiling point to the critical point. The value for these constants turned out to be the same as the ones which could have been obtained by making a correlation of the Ω's for many substances (*2*) with acentric factor. The binary interaction constant, k_{12}, which is used in the mixing rules cited, was evaluated by first calculating second-virial coefficients for Refrigerant 503 from the Martin–Hou equation-of-state constants given by Sinka et al. (*10*) and then back calculating the k_{12} required to get the same values of second-virial coefficients for the equimolar mixture from the Pitzer and Curl (*6*) correlation of second-virial coefficients. The resulting k_{12}'s are 0.091 (at 31.9°F), 0.091, 0.081, and 0.064 (at −100°F). A linear interpolation is recommended for intermediate temperatures. The resulting ϕ's are shown in Table I.

The liquid-phase standard-state fugacity, $f_i^{\circ}(\mathrm{l})$, for each component was taken to be the pure liquid component at the system temperature and at zero pressure. Both components in this study were subcritical; consequently, in the pressure correction for liquid-phase activity coefficients, the molar volumes of the pure liquids were used and were assumed to be independent of pressure. Thus, the activity coefficients, corrected to a zero reference pressure, were calculated as follows:

$$\gamma_i = \frac{\phi_i y_i P}{x_i P_i{}^s \phi_i{}^s \exp\left[\dfrac{(P - P_i{}^s) v_i(\mathrm{l})}{RT}\right]} \qquad (4)$$

The experimentally determined vapor pressures were used for $P_i{}^s$. The $\phi_i{}^s$ terms were calculated from the Redlich–Kwong equation applied to the pure, saturated vapor. The resulting γ's are shown in Table I.

The activity coefficients calculated from Equation 4 were correlated by the Wilson equation (*14*).

$$\ln \gamma_1 = -\ln(x_1 + \Lambda_{12}x_2) + x_2\left[\frac{\Lambda_{12}}{x_1 + \Lambda_{12}x_2} - \frac{\Lambda_{21}}{\Lambda_{21}x_1 + x_2}\right] \qquad (5)$$

$$\ln \gamma_2 = -\ln(x_2 + \Lambda_{21}x_1) - x_1\left[\frac{\Lambda_{12}}{x_1 + \Lambda_{12}x_2} - \frac{\Lambda_{21}}{\Lambda_{21}x_1 + x_2}\right] \qquad (6)$$

where subscript 1 refers to CHF_3 and subscript 2 to $CClF_3$.

Table VI. Optimum Parameters for the Wilson Equation

Temp, °F	Λ_{21}	Λ_{12}	Root-mean-square percentage difference between experimental and calculated γ's
31.90	0.671	0.785	0.6
−0.10	0.662	0.690	0.8
−55.10	0.598	0.558	1.1
−100.05	0.494	0.504	1.4

The parameters Λ_{21} and Λ_{12} were arbitrarily adjusted such that the root-mean-square percentage difference between calculated and experimental values of the activity coefficients was a minimum. These parameters, with the corresponding root-mean-square percentage differences, are given in Table VI for each temperature level. For the purposes of interpolation between experimental temperatures, the following correlation of Λ's with temperature is recommended:

$$\Lambda_{21} = 0.643 + 0.000872\,t \qquad (7)$$

$$\Lambda_{12} = 0.697 + 0.00254\,t \qquad (8)$$

These correlations for the fugacity coefficient and the activity coefficient allow accurate calculation of the vapor-liquid equilibria for this system at any temperature between −100° and 32°F. Once a temperature and liquid composition of the binary have been set, the system is specified, and the corresponding pressure and vapor composition can be determined. Such calculations were made for each experimental liquid composition and temperature using the correlation given in Equations 2, 3, 5–8. The overall average absolute difference between experimental and calculated pressures was 0.44%. Calculated vapor compositions had a root-mean-square difference of 0.005 mol fraction from experimental compositions. The results of the calculations for 31.9°F are superimposed on the experimental data in Figure 1.

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The donation of samples of CHF_3 and $CClF_3$ by E. I. du Pont de Nemours & Co. is gratefully acknowledged.

NOMENCLATURE

a, b = constants in the Redlich–Kwong equation of state
$f_i^{\circ}(\mathrm{l})$ = liquid-phase standard-state fugacity, psia
H = enthalpy, Btu/lb mol
k_{12} = binary interaction constant
N = number of components
P = system pressure, psia
R = gas constant
t = temperature, °F
T = temperature, °R = 459.69 + °F
v = molar volume of vapor phase, cu ft/lb mol
$v(\mathrm{l})$ = molar volume of liquid phase, cu ft/lb mol
x = liquid-phase mole fraction
y = vapor-phase mole fraction
Z = compressibility factor
$\Lambda_{21}, \Lambda_{12}$ = parameters in the Wilson equation
ϕ = vapor-phase fugacity coefficient
ϕ^s = fugacity coefficient of pure saturated vapor
Ω_a, Ω_b = dimensionless constants in the Redlich-Kwong equation of state

Subscripts

c = critical-point property
i = component i
j = component j
1 = CHF_3
2 = $CClF_3$

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Extraction of Metals from Nitrate and Sulfate Solutions by Amines

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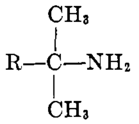
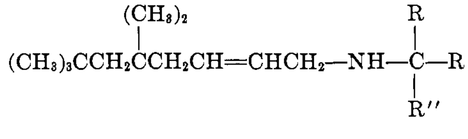
Coefficients for the extraction of 54 metals from acidified lithium nitrate and lithium sulfate solutions were determined by using representative primary, secondary, tertiary, and quaternary alkylammonium nitrates or sulfates in diethylbenzene diluent as extractants. The acid concentration of the aqueous phase was 0.2*N*, and the total nitrate and sulfate concentrations varied from 0.5–10*N* and 0.3–5*N*, respectively.

Since the solvent extraction of mineral acids with high-molecular-weight amines was first reported in 1948 (*30*), extraction characteristics of these reagents have received much study (*1–27, 29, 31, 32*). Most metal extraction surveys have been limited to acid dependence studies; furthermore, the use of a variety of diluents in these studies has made correlation of data from different sources difficult. In this work the extraction of 54 metals from acidified lithium nitrate and lithium sulfate solutions was systematically surveyed using representative primary, secondary, tertiary, and quaternary alkylam-

Table I. Description of Amines

Amine type	Compound[a]	Structure of compound
Primary	Primene JM	 $R-\overset{CH_3}{\underset{CH_3}{C}}-NH_2$ R = alkyl groups containing 15–21 carbon atoms
Secondary	Amberlite LA-1	 $(CH_3)_3CCH_2\overset{(CH_3)_2}{C}CH_2CH{=}CHCH_2{-}NH{-}\overset{R}{\underset{R''}{C}}{-}R'$ where R + R′ + R″ = different alkyl groups totaling 12–14 carbon atoms
Tertiary	Adogen 364	R_3N, where R designates mixed *n*-octyl and *n*-decyl groups
Quaternary	Adogen 464	$[R_3N\text{-}CH_3]^+Cl^-$, where R designates mixed *n*-octyl and *n*-decyl groups

[a] Primene JM and Amberlite LA-1 were supplied by the Rohm and Haas Co. and Adogen 364 and Adogen 464 by the Archer Daniels Midland Co.

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