Ternary Vapor-Liquid Equilibria for System *sec*-Butanol–Isobutanol–*n*-Butanol

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Isobaric vapor-liquid equilibrium was measured for the system *sec*-butanol–isobutanol–*n*-butanol at 760 mm Hg with a Dvorak-Boublik recirculation apparatus. Liquid activity coefficients were evaluated for the three components and correlated with the Redlich-Kister and Wilson equations. Vapor composition and boiling points were correlated from binary and ternary data and showed good agreement with the experimental information.

The butanol isomers system reported here represents an example of the prediction of multicomponent vapor-liquid equilibria of chemical species that are very similar and do not present steric effects of accommodation in solution. In addition, the binaries present exhibit opposite enthalpies of mixing effects. The vapor-liquid equilibrium data of the three binaries have already been determined (*12*). Quitszch et al. (*7*) studied the behavior of the binary *n*-butanol–*sec*-butanol at 100, 300, 500, and 700 mm Hg and found that it presented a negative deviation from ideal behavior. Similar results were obtained by Wisniak and Tamir (*12*) at 760 mm Hg. Wisniak and Tamir also correlated the boiling points of the binaries with their composition using a two-constant modified Swiestolawski equation. Murakami and Benson (*5*) found that the excess enthalpies of mixtures of *n*-butanol and isobutanol were positive, with a maximum value of 11.4 J/mol, and that those of *n*-butanol and *sec*-butanol were negative, with a maximum value of −100 J/mol at about equimolar composition.

Experimental

Purity of materials. Analytical grade reagents purchased from Merck and Fluka were used without further purification after gas chromatography analysis failed to show any significant impurities, particularly water. *n*-Butanol was at least 99.5% pure. Physical properties of the pure components appear in Table I.

Apparatus and procedure. An all-glass modified Dvorak and Boublik recirculation still (*1*) was used in the equilibrium determinations. A vacuum cartesian manostat connected to an ebulliometer allowed the pressure to be controlled to within 0.1 mm Hg. Temperature determinations were made to within ±0.01°C with a Hewlett-Packard quartz thermometer Model 2851A. The experimental features have been described previously (*11*). All analyses were carried out by gas chromatography on a Packard-Becker Model 417 apparatus provided with a thermal conductivity detector and an Autolab Model 6300 electronic integrator. the column was 200 cm long and 0.2 cm diameter and was packed with 10% Hallcomid M-18-01 on 80-100 Supelcoport and operated isothermally at 70°C. Injector temperature was 170°C, and the detector operated at 150 mA and 180°C. Calibration analyses were carried on to convert the peak area ratio to the weight composition of the mixture. Concentration measurements were generally accurate to ±0.3%.

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Results

Sixty-four constant pressure equilibria determinations were made at 760 mm Hg, and the experimental results appear in Table II.

Activity coefficients were calculated from the equation

$$\ln \gamma_i = \ln (Py_i/P_i^\circ x_i) + (B_i - V_i^\circ)(P - P_i^\circ)/RT + (P/RT)[(1 - y_i)(y_j\delta_{ij} + y_k\delta_{ik}) - y_jy_k\delta_{jk}] \quad (1)$$

where

$$\delta_{ij} = 2B_{ij} - B_i - B_j \quad (2)$$

and *i, j, k* is any permutation of 1, 2, 3.

Vapor pressures P_i° and second virial coefficients B_i were calculated according to the general equations (*2, 9*)

$$\log P_i^\circ = A - B/(C + t) \quad (3)$$

$$\log (-B_i) = \alpha_i - \beta_i \log T \quad (4)$$

where the coefficients appear in Table II.

The mixed virial coefficients B_{ij} were estimated by the correlation suggested by O'Connell and Prausnitz (6), using the molecular parameters given by the same authors. In general, the contribution of the last two terms accounted for less than 4% of the activity coefficients.

The thermodynamic consistency test was performed with the McDermott-Ellis (*4*) point-to-point test and by an overall correlation of the data with the Redlich-Kister and Wilson equations. A pair of points is considered thermodynamically consistent if the deviation value D_1 defined as

$$D_1 = \frac{1}{2}\sum_1^3 (x_{ia} + x_{ib})(\ln \gamma_{ib} - \ln \gamma_{ia}) \quad (5)$$

is less than 0.01 if the accuracy of measurement of vapor and liquid mole fraction is within ±0.001. The heat of mixing is very small so that there is no need to correct Equation 5 to account for it.

Table I. Physical Properties of Pure Compounds

Index	Compound	Refractive index at 25°C	Bp, °C 760 mm Hg
1	*sec*-Butanol	1.3949	99.7
		1.3950 (*9*)	99.6 (*9*)
2	Isobutanol	1.3938	107.7
		1.3939 (*9*)	107.7 (*9*)
3	*n*-Butanol	1.3975	117.6
		1.3973 (*9*)	117.66 (*9*)

Table II. Virial and Vapor-Pressure Constants

Compound	α_i	β_i	A	B	C
sec-Butanol	14.678	4.5	7.47431	1314.19	186.55
Isobutanol	14.711	4.5	7.22014	1190.38	166.67
n-Butanol	14.711	4.5	7.47680	1362.39	178.77

Table III. Ternary Vapor-Liquid Equilibrium Data for *sec*-Butanol(1)–*n*-Butanol(2)–Isobutanol(3)

Temp, °C		Liquid compn			Obs vapor compn			Calc vapor compn			Obs activity coeff			Calc activity coeff		
Obs	Calc	x_1	x_2	x_3	y_1	y_2	y_3	y_1	y_2	y_3	γ_1	γ_2	γ_3	γ_1	γ_2	γ_3
100.80	100.99	0.833	0.115	0.052	0.887	0.088	0.025	0.875	0.086	0.030	1.01687	0.98850	0.91015	1.00308	0.96325	1.09234
101.40	101.56	0.771	0.156	0.073	0.839	0.124	0.037	0.829	0.120	0.042	1.01663	0.90352	0.93674	1.00469	0.97056	1.05491
101.50	101.70	0.734	0.231	0.035	0.797	0.183	0.020	0.791	0.181	0.020	1.01073	1.09635	1.05188	1.00274	0.98707	1.07703
101.60	101.75	0.785	0.088	0.127	0.867	0.067	0.066	0.860	0.067	0.071	1.01689	0.94669	0.94525	1.00847	0.95370	1.01639
102.30	102.20	0.703	0.207	0.090	0.785	0.166	0.049	0.782	0.166	0.052	1.00960	0.97838	0.97078	1.00555	0.98008	1.03079
102.40	102.39	0.742	0.085	0.173	0.833	0.070	0.097	0.833	0.067	0.096	1.01135	1.00094	0.99580	1.01102	0.95849	0.98739
102.40	102.70	0.749	0.031	0.220	0.848	0.026	0.126	0.843	0.024	0.120	1.01994	1.01939	1.01717	1.01425	0.95061	0.96608
102.90	102.66	0.656	0.243	0.101	0.747	0.198	0.055	0.746	0.201	0.059	1.00744	0.97181	0.94817	1.00566	0.98610	1.01972
102.90	102.92	0.710	0.079	0.211	0.811	0.065	0.124	0.812	0.064	0.118	1.01057	0.98131	1.02325	1.01214	0.96403	0.97242
103.20	103.15	0.645	0.188	0.167	0.740	0.160	0.100	0.743	0.157	0.096	1.00410	1.00363	1.03034	1.00785	0.98264	0.99135
103.60	104.04	0.479	0.461	0.060	0.568	0.394	0.038	0.557	0.398	0.036	1.02297	0.99284	1.07272	1.00299	1.00208	1.02586
103.60	103.38	0.589	0.288	0.123	0.686	0.245	0.069	0.686	0.246	0.073	1.00475	0.98823	0.95016	1.00515	0.99324	1.00739
104.00	104.19	0.455	0.499	0.046	0.534	0.438	0.028	0.537	0.437	0.028	0.99803	1.00450	1.01491	1.00287	1.00270	1.02938
104.10	104.16	0.512	0.355	0.133	0.604	0.312	0.084	0.606	0.311	0.080	0.99960	1.00202	1.04894	1.00303	1.00024	1.00518
104.10	103.85	0.547	0.316	0.137	0.647	0.272	0.081	0.648	0.276	0.083	1.00225	0.98137	0.98195	1.00416	0.99730	1.00317
104.10	104.07	0.639	0.078	0.283	0.767	0.064	0.169	0.762	0.067	0.164	1.01708	0.93548	0.99180	1.01011	0.98098	0.96148
104.20	103.97	0.575	0.228	0.197	0.693	0.196	0.111	0.685	0.199	0.117	1.01758	0.97645	0.93214	1.00557	0.99370	0.98630
104.40	104.32	0.505	0.345	0.150	0.604	0.303	0.093	0.604	0.306	0.092	1.00263	0.99017	1.01769	1.00263	1.00094	1.00140
104.60	104.44	0.494	0.353	0.153	0.597	0.310	0.093	0.595	0.316	0.094	1.00587	0.98273	0.98996	1.00215	1.00180	1.00132
104.80	105.01	0.578	0.091	0.331	0.704	0.081	0.215	0.703	0.081	0.198	1.00656	0.99305	1.04839	1.00443	0.99553	0.96536
105.00	105.04	0.492	0.271	0.237	0.595	0.246	0.159	0.600	0.247	0.146	0.99232	1.00081	1.07572	1.00041	1.00463	0.98754
105.10	105.01	0.433	0.416	0.151	0.531	0.374	0.095	0.529	0.381	0.095	1.00268	0.98754	1.00486	0.99946	1.00509	1.00548
105.40	105.53	0.401	0.416	0.183	0.502	0.384	0.114	0.494	0.386	0.116	1.01271	1.00273	0.98343	0.99665	1.00770	1.00354
105.40	105.64	0.563	0.052	0.385	0.699	0.048	0.253	0.696	0.048	0.235	1.00437	1.00273	1.03741	1.00068	1.00007	0.96525
105.60	105.90	0.554	0.042	0.404	0.695	0.038	0.267	0.689	0.039	0.249	1.00767	0.97558	1.03526	0.99858	1.00236	0.96665
105.80	105.83	0.349	0.496	0.155	0.442	0.462	0.096	0.435	0.467	0.101	1.01010	0.99697	0.96270	0.99458	1.00737	1.01134
105.80	105.83	0.403	0.370	0.227	0.504	0.345	0.151	0.503	0.349	0.146	0.99745	0.99802	1.03396	0.99502	1.00996	0.99880
105.90	105.83	0.432	0.302	0.266	0.545	0.284	0.173	0.541	0.286	0.170	1.00263	0.99577	1.00701	0.99530	1.01064	0.99218
106.40	106.28	0.316	0.509	0.175	0.406	0.483	0.111	0.401	0.490	0.117	1.00332	0.99346	0.95331	0.99139	1.00872	1.01224
106.60	106.66	0.203	0.710	0.087	0.262	0.684	0.054	0.258	0.686	0.059	1.00070	1.00122	0.93542	0.98520	1.00349	1.02843
106.80	106.94	0.159	0.768	0.073	0.206	0.745	0.049	0.202	0.746	0.050	0.99750	1.00079	1.00383	0.98001	1.00243	1.03173
106.90	106.98	0.441	0.145	0.414	0.566	0.144	0.290	0.567	0.143	0.274	0.98468	1.02082	1.04356	0.98676	1.01698	0.98681
107.00	107.02	0.136	0.806	0.058	0.170	0.790	0.040	0.174	0.788	0.040	0.95565	1.00382	1.02348	0.97712	1.00167	1.03378
107.20	107.23	0.289	0.445	0.266	0.384	0.439	0.177	0.375	0.444	0.182	1.00873	1.00298	0.97996	0.98519	1.01402	1.00961
107.50	107.49	0.266	0.462	0.272	0.359	0.461	0.180	0.348	0.466	0.189	1.01389	1.00344	0.96344	0.98325	1.01418	1.01191
107.60	107.66	0.206	0.572	0.222	0.272	0.573	0.155	0.270	0.577	0.156	0.98846	1.00371	1.01260	0.98009	1.01051	1.01955
107.70	107.54	0.239	0.516	0.245	0.317	0.515	0.168	0.315	0.523	0.172	0.98947	0.99638	0.99070	0.98222	1.01233	1.01562
107.80	107.94	0.068	0.822	0.110	0.086	0.835	0.079	0.088	0.829	0.079	0.94018	1.01042	1.03365	0.96249	1.00270	1.03210
107.90	107.93	0.375	0.175	0.450	0.502	0.177	0.321	0.496	0.180	0.313	0.99170	1.00240	1.02276	0.97894	1.02123	0.99734
107.90	107.95	0.075	0.807	0.118	0.097	0.816	0.087	0.098	0.817	0.085	0.95811	1.00213	1.05711	0.96383	1.00309	1.03159
108.00	108.12	0.110	0.715	0.175	0.139	0.738	0.123	0.144	0.729	0.126	0.93285	1.01925	1.00390	0.96950	1.00615	1.02788
108.10	108.04	0.090	0.765	0.145	0.113	0.783	0.104	0.118	0.781	0.105	0.92366	1.00705	1.02055	0.96641	1.00443	1.02994
108.60	108.86	0.235	0.367	0.398	0.321	0.383	0.296	0.318	0.387	0.289	0.98755	1.00836	1.03828	0.97702	1.01926	1.01300
109.00	108.91	0.046	0.745	0.209	0.061	0.785	0.154	0.062	0.787	0.156	0.94551	1.00351	1.01319	0.96013	1.00630	1.02842
109.00	108.94	0.149	0.538	0.313	0.209	0.567	0.224	0.204	0.573	0.232	1.00013	1.00371	0.98405	0.97440	1.01368	1.02124
109.10	109.10	0.138	0.543	0.319	0.188	0.576	0.236	0.189	0.580	0.238	0.96799	1.00662	1.01343	0.97383	1.01362	1.02180
109.40	109.45	0.254	0.260	0.486	0.368	0.279	0.353	0.352	0.283	0.363	1.01882	1.00735	0.98374	0.97396	1.02136	1.01139
109.51	109.43	0.293	0.182	0.525	0.408	0.198	0.394	0.406	0.199	0.393	0.97550	1.01725	1.01222	0.97099	1.02205	1.00855
109.56	109.54	0.344	0.069	0.587	0.473	0.092	0.435	0.475	0.075	0.439	0.96159	1.04449	0.99763	0.96466	1.02114	1.00602

109.71	109.66	0.264	0.216	0.520	0.383	0.233	0.384	0.369	0.238	0.393	1.00933	1.00142	0.98853	0.97213	1.02156	1.01091
109.80	109.87	0.209	0.310	0.481	0.294	0.335	0.371	0.294	0.342	0.366	0.97565	0.99999	1.02901	0.97558	1.02017	1.01480
110.00	109.90	0.030	0.663	0.307	0.043	0.726	0.231	0.042	0.729	0.238	0.97752	1.00606	0.99728	0.96574	1.01032	1.02550
110.10	109.89	0.096	0.538	0.366	0.139	0.589	0.272	0.136	0.596	0.284	0.99392	1.00226	0.98034	0.97420	1.01429	1.02270
110.25	110.29	0.233	0.214	0.553	0.346	0.234	0.420	0.332	0.240	0.427	1.01412	0.99568	0.99624	0.97282	1.02041	1.01335
110.40	110.33	0.030	0.618	0.352	0.043	0.685	0.043	0.043	0.691	0.276	0.97382	1.00390	1.00791	0.97124	1.01230	1.02415
110.48	110.43	0.174	0.323	0.503	0.256	0.351	0.393	0.251	0.365	0.393	0.99685	0.98142	1.01605	0.97806	1.01930	1.01666
110.80	110.79	0.269	0.090	0.641	0.383	0.119	0.498	0.387	0.103	0.505	0.95417	1.08059	0.99829	0.96528	1.01791	1.01272
111.55	111.85	0.177	0.171	0.652	0.251	0.195	0.554	0.266	0.200	0.530	0.92631	0.99145	1.06168	0.98343	1.01608	1.01479
111.56	111.49	0.172	0.217	0.611	0.252	0.248	0.500	0.259	0.254	0.497	0.95671	0.99327	1.02211	0.98248	1.01767	1.01540
111.73	111.69	0.147	0.247	0.606	0.225	0.286	0.489	0.224	0.291	0.496	0.99372	1.00031	1.00152	0.98787	1.01761	1.01584
111.76	111.65	0.231	0.081	0.688	0.339	0.097	0.564	0.346	0.095	0.563	0.95179	1.03345	1.01632	0.97051	1.01431	1.01427
111.85	111.95	0.231	0.052	0.717	0.345	0.062	0.593	0.347	0.061	0.589	0.96568	1.02568	1.02194	0.97019	1.01205	1.01446
114.75	114.68	0.102	0.050	0.848	0.159	0.065	0.776	0.184	0.065	0.769	0.91418	1.01047	1.01634	1.05531	1.00822	1.01759
115.01	114.76	0.094	0.059	0.847	0.143	0.077	0.780	0.172	0.077	0.775	0.98447	1.00534	1.01315	1.06194	1.00947	1.01719

Ma et al. (*3*) concluded that if a set of data can be shown to be well correlated by a multicomponent thermodynamic equation such as that of Redlich-Kister or Wilson, then the assumption that the data are inconsistent according to the point-to-point test is not justified. They set an upper limit for the rmsd of 1.5% in the vapor phase to indicate a good correlation.

All the data that appear in Table III are consistent according to the McDermott-Ellis test. They were further correlated using the following Redlich-Kister equation (*8*)

$$\begin{aligned}\ln \gamma_1 = {} & x_2x_3[(B_{12} + B_{13} - B_{23}) + C_{12}(2x_1 - x_2) + C_{13}(2x_1 - x_3) + 2C_{23}(x_3 - x_2) + \\ & D_{12}(x_1 - x_2)(3x_1 - x_2) + D_{13}(x_1 - x_3)(3x_1 - x_3) - 3D_{23}(x_3 - x_2)^2 + C_1(1 - 2x_1)] + \\ & x_2^2[B_{12} + C_{12}(3x_1 - x_2) + D_{12}(x_1 - x_2)(5x_1 - x_2)] + \\ & x_3^2[B_{13} + C_{13}(3x_1 - x_3) + D_{13}(x_1 - x_3)(5x_1 - x_3)] \end{aligned} \quad (6)$$

where B_{ij}, C_{ij}, D_{ij} are the binary constants and C_1 is a ternary constant. The equations for the other two activity coefficients were obtained by cyclic rotation of the indices. Equation 6 was used with and without the ternary constant to predict the values of the experimental activity coefficient and vapor-phase composition. For the latter, the rmsd's were 0.67 and 0.68%, respectively, values that indicate that the overall correlation is very good and that they are not statistically different. In other words the ternary vapor-liquid equilibria of the ternary system *sec*-butanol–isobutanol–*n*-butanol can be predicted from the behavior of the different binaries that compose it without the need for interaction terms like C_1. Table III reports the predicted values of the activity coefficients and vapor-phase composition, and Table IV the Redlich-Kister correlating constants for the three binaries.

The ternary system was also correlated using the Wilson equation (*10*)

$$\ln \gamma_k = -\ln\left[\sum_1^m x_j\Lambda_{kj}\right] + 1 - \sum_1^m \frac{x_i\Lambda_{ik}}{\sum_1^m x_j\Lambda_{ij}} \quad (7)$$

Equation 7 contains only parameters which can be obtained from binary data. These were calculated by a Simplex optimization technique from the following equations, applicable to binary systems

$$\ln \gamma_1 = -\ln(x_1 + \Lambda_{12}x_2) + x_2\left[\frac{\Lambda_{12}}{x_1 + \Lambda_{12}x_2} - \frac{\Lambda_{21}}{\Lambda_{21}x_1 + x_2}\right] \quad (8)$$

$$\ln \gamma_2 = -\ln(x_2 + \Lambda_{21}x_1) - x_1\left[\frac{\Lambda_{12}}{x_1 + \Lambda_{12}x_2} - \frac{\Lambda_{21}}{\Lambda_{21}x_1 + x_2}\right] \quad (9)$$

The Wilson constants for the different binaries are reported in Table V, together with their rmsd. Application of Equation 7 to the ternary system showed that the overall rmsd was 0.77% so that the predicting abilities of the Redlich-Kister and Wilson equations are similar.

The boiling points of the ternary system were then correlated using the equation suggested by Wisniak and Tamir (*12*)

$$T = x_1T_1^\circ + x_2T_1^\circ + x_3T_3^\circ + \omega + \sum^3\left[x_ix_j\sum_0^k C_K(x_i - x_j)^k\right] + x_1x_2x_3[A + B(x_1 - x_2) + C(x_1 - x_3) + \ldots] \quad (10)$$

where

$$\omega = x_1\ln(y_1/x_1) + x_2\ln(y_2/x_2) + x_3\ln(y_3/x_3) \quad (11)$$

The different constants of Equation 10 appear in Table VI, and from the value of the rmsd, correlation is very good. The

Table IV. Correlation of Binary Vapor-Liquid Data, Redlich-Kister Equation

System	B_{ij}	C_{ij}	D_{ij}
sec-Butanol–isobutanol	$-1.18005 \cdot 10^{-2}$	$2.71650 \cdot 10^{-2}$	$-1.39587 \cdot 10^{-2}$
sec-Butanol–*n*-butanol	$-2.05600 \cdot 10^{-2}$	$-2.82625 \cdot 10^{-3}$	$6.24375 \cdot 10^{-2}$
Isobutanol–*n*-butanol	$5.05079 \cdot 10^{-2}$	$2.24689 \cdot 10^{-2}$	$4.86144 \cdot 10^{-3}$
sec-Butanol–isobutanol–*n*-butanol	$C_1 = 9.52208 \cdot 10^{-3}$		

Table V. Wilson Constants, Equation 7

System	Λ_{ij}	Λ_{ji}	Rmsd, % y	Rmsd, % γ
sec-Butanol–isobutanol	0.88175	1.13388	0.78	0.53
sec-Butanol–*n*-butanol	0.66549	1.50287	1.20	2.70
Isobutanol–*n*-butanol	0.57133	1.54896	0.52	1.12

Table VI. Correlation of Boiling Points, Equation 10

System	C_0	C_1	C_2	Rmsd
sec-Butanol–isobutanol	−0.51772	−1.29401	−0.59257	0.055
sec-Butanol–*n*-butanol	−5.13436	−1.03916	−2.43468	0.070
Isobutanol–*n*-butanol	−2.30700	−1.03729	−0.33503	0.042
sec-Butanol–isobutanol–*n*-butanol	$A = -2.19106$			0.147

rmsd for the prediction of the ternary boiling points from the binary data alone is 0.162 so that for practical purposes constant A may be neglected.

Acknowledgment

Yehudit Reizner and Moshe Golden helped in the experimental and numerical determinations.

Nomenclature

α, β, A, B, C = constants
B_i = second virial coefficient, cm^3/mol
B_{ij} = mixed virial coefficient, cm^3/mol (Equation 2; constant (Equation 6)
n = number of experimental points
P = overall pressure, mm Hg
P_i° = vapor pressure of component i pure, mm Hg
R = gas constant
rmsd = root-mean-square deviation,

$$\sqrt{\Sigma\Sigma(y_{ji,exp} - y_{ji,calc})^2/3\,n}$$

t, T = temperature, °C, K
T_i° = boiling temperature of component i at pressure P, K
V_i° = molar liquid volume of component i pure, cm^3/mol
x_i, y_i = mole fraction composition of component i in the liquid and vapor phases
γ_i = activity coefficient of component i
A_{ij} = constant, Wilson equation

Subscripts

exp = experimental
calc = calculated
i = component i

Literature Cited

(1) Boublikova, L., Lu, B.C.Y., *J. Appl. Chem.*, **19**, 89 (1969).
(2) Cox, J. D., *Trans. Faraday Soc.*, **57**, 1674 (1961).
(3) Ma, K. T., McDermott, C., Ellis, S.R.M., *Inst. Chem. Eng. Symp. Ser.*, **32** (1969).
(4) McDermott, C., Ellis, S.R.M., *Chem. Eng. Sci.*, **20**, 293 (1965).
(5) Murakami, S., Benson, G. C., *Bull. Chem. Soc. Jpn.*, **46**, 74 (1973).
(6) O'Connell, J. P., Prausnitz, J. M., *Ind. Eng. Chem., Process Des. Dev.*, **6**, 245 (1967).
(7) Quitszch, K., Kochler, S., Taubert, K., Geiseler, G., *J. Prakt. Chem.*, **311**, 429 (1969).
(8) Redlich, O., Kister, A. T., *Ind. Eng. Chem.*, **40**, 345 (1948).
(9) TRC Tables "Selected Values of Properties of Chemical Compounds", Thermodynamics Research Center Data Project, College Station, Tex., 1961.
(10) Wilson, G. M., *J. Am. Chem. Soc.*, **86**, 127 (1964).
(11) Wisniak, J., Tamir, A., *J. Chem. Eng. Data*, **20**, 168 (1975).
(12) Wisniak, J., Tamir, A., *Ibid.*, p 388.

Received for review April 21, 1975. Accepted August 15, 1975.