M_i	molar mass of compound *i*
n_i	refractive index of pure compound *i* for the sodium D-line
n_{12}	refractive index of the binary mixture for the sodium D-line
δn_{12}	refractive index difference defined by eq 12
R_{12}	specific refractivity of binary mixture
V^E	excess volume on mixing
x_i	mole fraction of compound *i*
w_2	mass fraction of compound *i*

Greek Letters

ρ_i	density of pure compound *i*
ρ_{12}	density of binary mixture
σ	standard deviation
ϕ_i	volume fraction of compound *i*
$\sigma(a_k)$, $\sigma(b_k)$	standard deviation of the parameters a_k and b_k, respectively
$\sigma(n_{12})$, $\sigma(\rho_{12})$	standard deviation of the fit by eqs 10 and 11, respectively

Subscripts

exptl	experimentally observed quantity
calcd	calculated quantity

Registry No. Benzene, 71-43-2; cyclohexane, 110-82-7; acetone, 67-64-1.

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Salt Effect on Phase Equilibria by a Recirculating Still

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A new still to measure the salt effect on the vapor-liquid equilibrium at low pressures was developed. It is of the recirculating type, allowing the recirculation of both phases and the determination of its composition. The still was tested with the system ethanol-water-potassium acetate at different mole fractions of salt at 101.33 kPa.

Introduction

The most work concerning the experimental determination of the salt effect on the vapor-liquid equilibrium at low pressures uses the well-known Othmer still as standard equipment (*1-5*). Hala et al. (*6*) and Malanowski (*7*) describe one disadvantage of the Othmer still as being the superheating of the gas phase, not allowing the correct determination of the equilibrium temperature. On the other hand, only the vapor phase recirculates.

The purpose of this work was to develop, construct, and test an equilibrium still to eliminate the problems mentioned above. It differs from others which have been used for salt effect studies in the sense that both equilibrium phases recirculate with the aid of a Cottrell pump. To test the proposed apparatus, the system water-ethanol-potassium acetate was chosen. This system has been previously studied by Costa Novella and Tarrasó (*1*), Meranda and Furter (*3*), and Schmitt (*4*).

Experimental Section

Apparatus. A diagram of the apparatus is shown in Figure 1; it is a modified Naumann still (*8*) especially designed for salt effect studies in the range of 300-500 K and to 150 kPa. It is of the recirculation type, in which both liquid and vapor recirculate continuously, and allows the determination of the equilibrium composition of both phases. The essential elements are a 400 cm^3 Pyrex glass flask, 2 (equipped with a magnetic stirrer, 18, and a heating coil, 1), a Cottrell pump, 3, an equilibrium chamber, 5 (both isolated from outside by a silvered vacuum jacket, 4), a condenser, 8, for the vapor phase, a cooler, 13, for the liquid phase, two sampling ports, 10 and 14, fitted with rubber septums, and a magnetic stirred mixing chamber, 16. The volume of 400 cm^3 was chosen due to the necessity of removal of about 10 cm^3 of the liquid phase for the determination of the amount of salt in solution. Also, a large liquid volume makes possible the maintenance of a constant molar salt composition. The liquid phase return branch, 12, was built as short as possible, eliminating the occurrence of salt deposits. The equilibrium flask was involved with heating tapes to prevent heat losses.

In its operation, 350 cm^3 of solution is introduced into the still through the feed system, 20, which allows feeding also at pressures other than atmospheric. The solution is then brought partially to boil by an internal electric heater, 1. The Cottrell pump, 3, carries the mixture of liquid and vapor upward to the equilibrium chamber, 5, where the two phases, after striking directly against the thermometer stem, separate into a liquid and a vapor stream. The vapor is condensated in 8 and passed through the sampling port to the mixing chamber, 16. The liquid is cooled in 13 and passed through the sampling port, 14, to 16, where it mixes with the condensate, returning to flask 2 for recirculation.

The still can be operated at constant temperature or pressure. The steady state was usually reached after 30 min of operation. Samples of both phases are taken using syringes. Changes in composition are made by purging a known amount of solution, and replacing it by an amount of one component,

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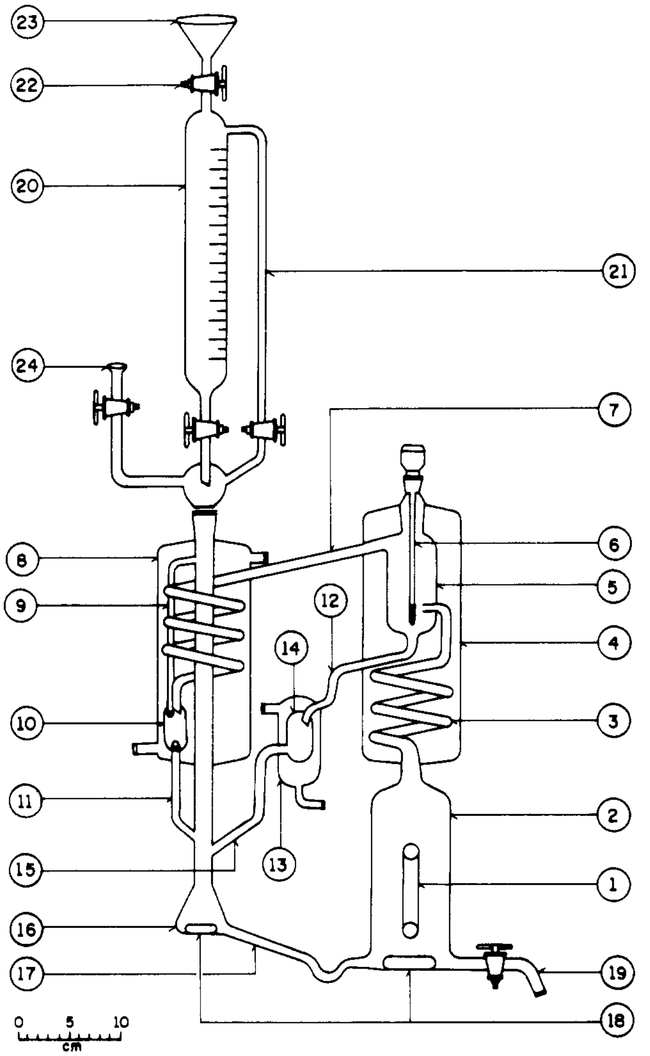

Figure 1. Diagram of the equilibrium cell: 1, heating coil; 2, Pyrex glass flask; 3, Cottrell pump; 4, vacuum jacket; 5, equilibrium chamber; 6, Pt-100 thermometer; 7, 12, return tubes for the vapor and liquid streams, respectively; 8, condenser; 9, 21, pressure equalizing tubes; 10, 14, sampling ports; 11, return tube for the condensate; 13, cooler; 15, return tube for the cooled liquid; 16, mixing chamber; 17, return tube for the mixture; 18, magnetic stirrer; 19, drain; 20, filler; 22, stopcock; 23, funnel; 24, to pressure measuring instrument, vacuum pump, and nitrogen tank.

without interruption of the operation.

The apparatus is attached with the aid of a ground-glass joint, 24, to a vacuum pump, a U-tube mercury manometer, a compressed nitrogen gas bottle, and a 200-L surge tank with nitrogen to stabilize the gas pressure in the still.

The temperature in the equilibrium chamber is measured using a platinum resistence thermometer, 6, in conjunction with a Fischer Model DT digital instrument. Pressure is determined by measuring the difference in column height of the mercury in the arms of a mercury U-tube manometer using a cathetometer. The compositions of the vapor-phase condensate samples are determined from measurements of their refractive indexes at 293.15 K with an Abbe-type refractometer. For the salt solutions a method consisting of refractometry and gravimetry was used (*9*).

The uncertainty in the pressure measurement is about ±0.02 kPa, in temperature about ±0.05 K, in refractive index about ±0.0001, and in mass about ±0.000 05 g.

The uncertainty in the final molar fraction values caused by the above uncertainties is estimated to ±0.005.

Materials. Ethanol (p.a. grade, 99.0 mol % Merck), cyclohexane (p.a. grade, 99.5 mol %, Grupo Química, Brazil), toluene (p.a. grade, 99.5 mol %, Grupo Química, Brazil), and potassium acetate (p.a. grade, 99.9 mol %, Reagen, Brazil) were used without further purification. The water used was laboratory-distilled, and its normal boiling point was 373.15 K.

Table I. Experimental Vapor Pressures P^v of Ethanol as a Function of Temperature T[a]

T/K	P^v/kPa	T/K	P^v/kPa	T/K	P^v/kPa
306.15	12.40	332.05	45.14	350.25	99.00
307.65	13.68	333.15	47.68	350.75	99.83
309.75	15.18	335.45	52.59	350.95	100.89
310.95	16.03	337.75	58.10	351.15	101.82
312.15	17.22	339.95	63.89	352.35	106.73
313.35	18.18	341.05	66.28	353.35	108.72
314.45	19.28	342.75	71.60	354.05	112.50
315.55	20.48	342.95	72.46	354.65	112.60
316.95	22.01	344.45	78.48	355.75	119.21
318.25	23.54	346.85	85.32	356.55	122.90
320.65	26.52	347.85	89.01	357.95	129.60
323.75	31.34	349.05	93.44	358.55	133.01
327.95	37.17	349.75	96.19	359.95	139.52

[a] The Antoine constants are $A = 16.171\,26$, $B = 3375.151\,304$ K, and $C = -59.341\,321$ K.

Table II. Experimental Vapor–Liquid Equilibrium Data for Cyclohexane(1)–Toluene (2) at 323.15 K

P/kPa	x_1	y_1	P/kPa	x_1	y_1
12.35	0.000	0.000	24.44	0.392	0.665
14.67	0.058	0.191	25.91	0.459	0.711
15.26	0.066	0.230	27.13	0.525	0.749
15.68	0.087	0.274	28.07	0.564	0.783
16.24	0.108	0.322	29.21	0.610	0.808
17.18	0.141	0.370	29.73	0.647	0.827
17.77	0.157	0.391	30.20	0.679	0.839
18.61	0.185	0.437	31.06	0.720	0.867
19.35	0.206	0.479	32.84	0.805	0.909
20.33	0.246	0.516	34.08	0.875	0.939
21.66	0.293	0.574	34.64	0.904	0.954
22.69	0.329	0.615	35.36	0.949	0.975
23.12	0.339	0.631	36.38	1.000	1.000

Table III. Deviations between Calculated and Measured y_1 Values for Cyclohexane (1)–Toluene (2) at 323.15 K and Ethanol (1)–Water (2) at 101.33 kPa

$$\Delta Y = \frac{1}{N}\sum_{i=1}^{N}|y_{i,\mathrm{calcd}} - y_{i,\mathrm{exptl}}|$$

system	no. of data points	ΔY
cyclohexane–toluene (T = 323.15 K)	26	0.0071
ethanol–water (P = 101.33 kPa)	26	0.0097

Results and Discussion

Performance of the New Apparatus. The performance of the apparatus was evaluated by measuring vapor pressure values for ethanol and vapor–liquid equilibrium data for the systems toluene–cyclohexane, and water–ethanol.

Vapor pressure data of pure ethanol were measured over the range 306–360 K, and the results were fitted to the Antoine equation:

$$\ln (P^v/\mathrm{kPa}) = A - B/((T/\mathrm{K}) + C)$$

The results appear in Table I. The normal boiling temperature obtained from this equation is 351.49 K, while the literature value (*10*) is 351.43 K. The average relative deviation between experimental and calculated pressures was 0.042%. The average deviation between the experimental vapor pressures and the values of Ambrose and Sprake (*11*) was 0.02 kPa.

Vapor–liquid equilibrium data have been measured for the system cyclohexane–toluene at 323.15 K and for the system water–ethanol at 101.33 kPa. The results are given in Tables

Table IV. Experimental Results for Ethanol (1)–Water (2)–Potassium Acetate at 101.33 kPa

x_1	y_1	T/K	γ_1	γ_2	x_1	y_1	T/K	γ_1	γ_2
s = 0.000									
0.000	0.000	373.15			0.536	0.669	352.95	1.184	1.529
0.023	0.200	366.55	5.094	1.036	0.611	0.710	352.35	1.126	1.639
0.042	0.289	364.15	4.373	1.026	0.718	0.761	351.75	1.050	1.912
0.073	0.399	360.45	3.952	1.031	0.773	0.798	351.65	1.027	2.019
0.138	0.493	358.05	2.812	1.027	0.816	0.831	351.55	1.016	2.096
0.147	0.509	357.55	2.775	1.025	0.869	0.882	351.45	1.016	2.069
0.164	0.521	357.05	2.593	1.041	0.897	0.897	351.25	1.009	2.317
0.209	0.566	356.25	2.274	1.029	0.924	0.921	351.35	1.002	2.402
0.238	0.562	355.75	2.020	1.099	0.936	0.930	351.35	0.998	2.528
0.295	0.587	354.95	1.753	1.157	0.951	0.943	351.35	0.996	2.691
0.377	0.614	354.15	1.478	1.264	0.975	0.972	351.45	0.998	2.584
0.437	0.627	353.65	1.326	1.379	0.992	0.984	351.45	0.993	4.618
0.490	0.640	353.25	1.225	1.493	1.000	1.000	351.45		
s = 0.025									
0.000	0.000	375.35			0.539	0.704	353.05	1.233	1.530
0.009	0.145	370.25	8.769	1.007	0.572	0.715	352.85	1.189	1.604
0.018	0.213	366.55	6.902	1.068	0.602	0.746	352.65	1.188	1.564
0.039	0.328	364.15	5.329	1.018	0.645	0.764	352.45	1.144	1.669
0.049	0.388	362.55	5.355	0.995	0.697	0.799	352.25	1.114	1.727
0.084	0.465	360.05	4.069	0.998	0.725	0.820	351.95	1.112	1.756
0.125	0.504	358.65	3.108	1.023	0.771	0.845	351.75	1.086	1.929
0.146	0.528	357.65	2.896	1.040	0.808	0.865	351.55	1.068	2.130
0.198	0.556	356.75	2.313	1.082	0.858	0.912	351.45	1.065	2.078
0.243	0.578	356.05	2.012	1.124	0.863	0.923	351.55	1.068	1.950
0.275	0.606	355.65	1.892	1.120	0.873	0.924	351.55	1.056	2.176
0.309	0.602	354.95	1.717	1.221	0.913	0.962	351.55	1.051	2.181
0.416	0.655	353.95	1.440	1.327	0.931	0.975	351.55	1.045	2.621
0.471	0.676	353.55	1.331	1.412	0.940	0.983	351.55	1.043	2.930
0.500	0.689	353.35	1.286	1.450	0.975	1.000	351.55		
s = 0.050									
0.000	0.000	374.25			0.505	0.718	353.25	1.333	1.496
0.014	0.242	369.35	9.473	0.974	0.541	0.744	353.15	1.294	1.508
0.024	0.345	366.95	8.143	0.932	0.618	0.795	352.65	1.232	1.558
0.041	0.451	362.35	7.340	0.947	0.672	0.817	352.45	1.173	1.731
0.103	0.503	360.15	3.563	1.002	0.728	0.852	352.35	1.133	1.833
0.145	0.549	358.25	2.948	1.033	0.771	0.887	352.25	1.118	1.877
0.230	0.598	356.55	2.160	1.112	0.803	0.917	352.15	1.114	1.890
0.287	0.620	355.75	1.844	1.187	0.853	0.950	352.15	1.086	2.095
0.326	0.637	354.65	1.739	1.264	0.895	0.990	352.15	1.079	2.256
0.404	0.669	354.15	1.503	1.355	0.950	1.000	352.15		
0.478	0.705	353.55	1.368	1.454					
s = 0.066									
0.034	0.427	367.55	7.029	0.833	0.489	0.740	353.95	1.382	1.405
0.048	0.495	365.75	6.141	0.798	0.533	0.762	353.55	1.323	1.466
0.103	0.541	362.25	3.534	0.894	0.540	0.768	353.45	1.323	1.461
0.120	0.557	361.35	3.240	0.912	0.585	0.781	353.35	1.246	1.603
0.132	0.568	360.65	3.083	0.924	0.642	0.824	353.25	1.202	1.613
0.178	0.593	359.45	2.494	0.969	0.697	0.881	352.95	1.196	1.450
0.193	0.607	358.65	2.420	0.985	0.744	0.895	352.75	1.147	1.758
0.273	0.658	355.65	2.070	1.101	0.777	0.924	352.75	1.133	1.677
0.289	0.659	355.35	1.978	1.138	0.788	0.929	352.65	1.129	1.760
0.337	0.671	355.15	1.740	1.027	0.818	0.953	352.55	1.120	1.824
0.381	0.693	355.05	1.592	1.234	0.846	0.987	352.45	1.124	1.031
0.394	0.694	354.75	1.563	1.276	0.934	1.000	352.35		
0.410	0.702	354.45	1.534	1.298					
s = 0.085									
0.000	0.000	375.15			0.436	0.743	353.95	1.555	1.309
0.019	0.377	372.25	9.332	0.782	0.506	0.782	353.65	1.426	1.353
0.055	0.521	366.35	5.562	0.778	0.565	0.806	353.45	1.325	1.473
0.140	0.592	360.85	3.008	0.915	0.617	0.844	353.25	1.280	1.472
0.182	0.622	358.45	2.649	0.984	0.705	0.909	352.95	1.220	1.425
0.272	0.668	356.65	2.026	1.078	0.740	0.935	352.75	1.204	1.381
0.296	0.685	356.05	1.954	1.092	0.780	0.963	352.65	1.181	1.286
0.360	0.714	355.05	1.737	1.169	0.915	1.000	352.65		
0.385	0.728	354.55	1.689	1.200					
s = 0.150									
0.000	0.000	379.45			0.271	0.735	356.65	2.238	1.048
0.024	0.473	373.35	9.239	0.734	0.411	0.799	355.15	1.694	1.199
0.086	0.608	367.45	3.998	0.741	0.433	0.811	354.05	1.700	1.263
0.126	0.653	362.25	3.492	0.849	0.511	0.844	353.65	1.524	1.414
0.206	0.704	358.35	2.652	0.962					

Table V. Critical Temperature (T_c), Critical Pressure (P_c), Critical Compressibility Factor (Z_c), Rackett's Parameter (Z_R), Acentric Factor (ω), Tsonopoulos Constants (a, b), and Parameters of the Antoine Equation (A, B, C)

	ethanol	water	cyclohexane	toluene
$T_c{}^a$/K	516.2	647.3	557.4	591.7
$P_c{}^a$/(kPa × 10^{-2})	63.8	220.5	40.7	41.1
$Z_c{}^a$	0.248	0.229	0.273	0.264
ω^a	0.635	0.344	0.218	0.257
$Z_R{}^b$	0.2520	0.2380	0.2729	0.2646
a^c	0.0878	0.0279	0	0
b^c	0.0560	0.0229	0	0
A^d	18.9119	18.3036	15.7527	16.0137
B^d/K	3803.98	3816.44	2766.63	3906.52
C^d/K	−41.68	−46.13	−50.50	−53.67

[a] Reference 21. [b] Reference 19. [c] Reference 18. [d] Reference 20.

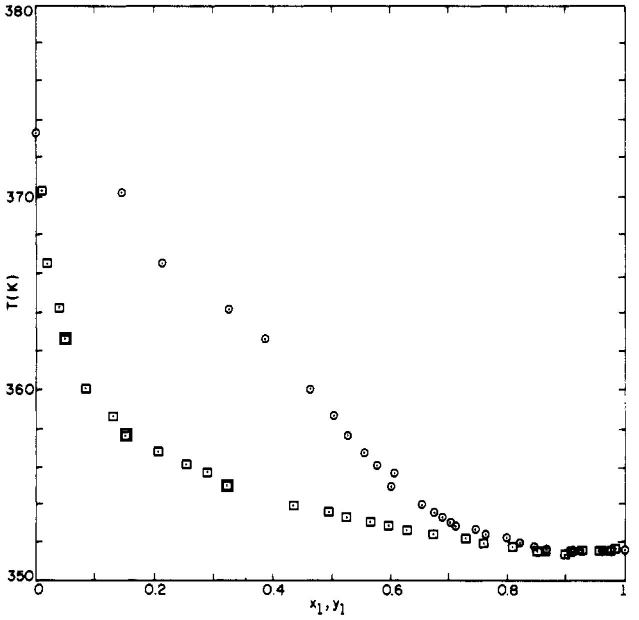

Figure 2. Temperature–mole fraction of ethanol diagram for the system water–ethanol–potassium acetate at 101.33 kPa and s = 0.025.

II and IV. Comparisons with data measured by Sieg (*12*), Myers (*13*), and Rivenq (*14*) for the system cyclohexane–toluene and with data measured by Bloom (*15*) and Jones (*16*) for the system water–ethanol show good agreement. The thermodynamic consistency of the data was tested by the Fredenslund test (*17*). The results appear in Table III. Thermodynamic quantities used in the calculations are given in Table V. The data sets were considered consistent using the criteria given in the literature.

Salt Effect Data. The effect of potassium acetate on the vapor–liquid equilibrium of ethanol–water was studied at 101.33 kPa at five different mole fractions of acetate (0.025, 0.050, 0.066, 0.085, and 0.150). The results are given in Table IV. Experimental activity coefficients for water and ethanol on a three-component basis were obtained from the equation

$$\ln \gamma_i = \ln\left(\frac{y_i P}{x_i P_i^{\mathrm{v}}}\right) + \frac{(B_{ii} - V_i)(P - P_i^{\mathrm{v}})}{RT} + \frac{(1 - y_i)^2 P \sigma}{RT}\delta$$

where $\delta = 2B_{ij} - B_{ii} - B_{jj}$. Second virial coefficients B_{ii}, B_{ij}, and B_{jj} were estimated using correlations suggested by Tsonopoulos (*18*). The pure liquid molar volumes V_i were obtained by the modified Rackett equation (*19*). For the vapor pressure P_i^{v}, literature values (*20*) were used. Pure component thermodynamic quantities used in these calculations are given in Table V. Figure 2 shows a typical curve at 101.33 kPa for the studied system at a salt mole fraction of 0.025. The azeotropic point disappears at a salt mole fraction larger than 0.066. Vapor–liquid equilibrium of the studied system have been also measured by Schmitt (*4*), Meranda and Furter (*3*), and Costa Novella and Tarrasó (*1*). All these measurements have been performed using an Othmer-type still. A comparison with Schmitt's data is possible. The data of the present work show a slight negative deviation in the vapor composition (between 0.005 and 0.02) and a negative temperature deviation between 1 and 4 K. These differences may be attributed to the Cottrell pump in the proposed still, which allows a temperature measurement without superheating.

In our opinion stills with recirculation of both phases containing a Cottrell pump seem to be an improvement of ebulliometric techniques for the measurement of the salt effect on liquid–vapor equilibria.

Nomenclature

A,B,C = parameters of the Antoine equation
a,b = Tsonopoulos constants
B_{ii},B_{jj},B_{ij} = virial coefficients, cm^3 mol^{-1}
N = number of data points
P^{v} = vapor pressure, kPa
P = pressure, kPa
R = universal gas constant, cm^3 kPa K^{-1} mol^{-1}
s = mole fraction of salt
T = absolute temperature, K
V_i = molar volume of component i, cm^3 mol^{-1}
x = mole fraction in the liquid phase
y = mole fraction in the vapor phase
z = compressibility factor
Z_R = Rackett's parameter

Greek Letters

γ_1,γ_2 = activity coefficients
ω = acentric factor

Subscripts

calcd = calculated
exptl = experimental
c = critical

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