below thermal decomposition temperature for 10–12 h during which the conductivity fell to a constant value. Measurements were made at 5–10 ° intervals in cooling and heating cycles with a Beckmann conductivity bridge (Model RC-18A) at a frequency of 1 kHz. Melt temperatures were controlled and known with a precision better than ±0.5 °C.

Results and Discussion

The impedance of a cell is generally considered as a series combination of a resistance (R_s) and the double layer capacitance (C_s). In measuring bridge, as employed, the resistances in the ratio arms were matched and the balancing arm had a parallel combination of resistance (R_s) and capacitance (C_p). From ac theory (*1*) one has,

$$R_s = R_p/[1 + (R_p C_p w)^2]$$

where $w = 2\pi f$ and f is the ac frequency in hertz.

Specific conductance (k) data at different compositions and over the accessible range of temperature (t, °C) were obtained. At a given composition k–t data could be expressed into various mathematical expressions; those in the form $k = a + bt + ct^2 + dt^3$ are presented in Table I. (Supportive data are available in supplementary material. See paragraph at end of paper.) No significance is attached to these different forms of the expressions, except that they permitted alternate evaluation of Arrhenius coefficients, E_k, at different temperatures through the derivative $\partial \ln k/\partial(1/T)$. The Arrhenius coefficient (apparent activation energies) given by

$$E_k = -R\left[\frac{\partial \ln k}{\partial(1/T)}\right]$$

was found to be temperature dependent. The range of variation of E_k (average of the values obtained from different fits) for different compositions is also included in Table I.

Conductance–composition isotherms (not shown) were smooth curves in all cases, indicating absence of structural transformations (complexation, etc.) on addition of the divalent ion. A gradual decrease in conductivity with the increase in divalent ion content, as observed, may result from increased Coulombic interactions and a shrinkage of the ionic lattice, resulting in hindered ionic migration.

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Supplementary Material Available: Supportive data for Table I (4 pages). Ordering information is given on any current masthead page.

Binary Azeotropes Containing Butyric Acids

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The formation of binary azeotropes by *n*- and isobutyric acids with 11 substances at atmospheric pressure is investigated. The choice of substances is based on consideration of polarity, association, the reduced value in the acid partial pressure, and the formation of compounds. Dimethylaniline, diethylaniline, and toluene form no azeotropes, though slight deviations from normal behavior are noticed. Other substances form positive azeotropes which may be attributed to the acid association at high acid concentration. The characteristics of each two corresponding azeotropes are very close.

Eleven substances have been selected for the investigation of the formation, at atmospheric pressure, of binary azeotropes by *n*- and isobutyric acids. The substances selected are *n*-octane, *p*-xylene, anisole, dimethylaniline, diethylaniline, acetic anhydride, toluene, and four petroleum fractions of boiling points 148.1, 143.3, 140.8, and 129.9 °C, the main constituent of the last fraction being an octane isomer, while those of the other three fractions being nonane isomers. The selection of these substances has been made according to the following.

The dielectric constant values of *n*- and isobutyric acids increase linearly with rise of temperature, due to their dissociation which favors azeotropic formation. The dielectric constant values of the *n*-acid are larger than those of the isoisomer (*6, 7*).

Quite large positive deviations from Raoult's law are obtained if one of the components is nonpolar or slightly polar, as *n*-octane, *p*-xylene, and the four petroleum fractions, while the other component is highly polar.

Negative deviations from Raoult's law may be produced when the added substance has a slightly basic character, such as dimethyl- and diethylaniline, so as to favor the formation of a compound.

To separate an azetrope of two acids, it is necessary to add a substance that reduces the partial pressure of one acid component more than that of the other. It is also desirable that the added substance can be easily removed from the distillate. According to these two basic requirements, *n*-octane, *p*-xylene, anisole, acetic anhydride, and the four petroleum fractions have been selected.

Toluene is reported to have been used in the azeotropic separation of butyric acid isomers (*9*). For this reason it was included among the chosen substances.

Experimental Section

Substances to be used in the experiments were labeled A.R. (analytical reagent). However, each substance, before use, was purified by distillation several times through a packing column. Dimethyl- and diethylaniline were distilled under reduced pres-

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Table I. Measured Values of Main Physical Properties of Used Substances

Substance	Bp, °C	Mp, °C	Density at 25 °C (g/ml)	Refractive index, 25 °C Na D-line	Difference between condensation and boiling temperature (°C)
n-Butyric acid	163.55	−5.55	0.95273	1.39581	0.03
Isobutyric acid	154.70	−46.10	0.93970	1.39340	0.05
n-Octane	125.60	−56.80	0.69878	1.39534	0.00
p-Xylene	138.20	+13.30	0.85664	1.49321	0.00
Anisole	153.90	−37.40	0.98931	1.51430	0.00
Dimethylaniline	193.40	+2.50	0.95191	1.55619	0.03
Diethylaniline	217.32	−21.30	0.93192	1.54093	0.03
Acetic anhydride	139.50	−73.00	1.07491	1.39011	0.05
Toluene	110.00	−95.00	0.86250	1.49404	0.05
Petroleum fraction	148.10				0.03
	143.30				0.03
	140.80				0.03
	129.90				0.03

Table II. Boiling Temperature-Composition Data[a]

n-Butyric acid–*n*-octane		Isobutyric acid–*n*-octane	
t, °C	wt % of acid	*t*, °C	wt % of acid
125.60	00.000	125.60	00.000
124.80	02.213	124.60	01.800
124.73	04.824	124.30	05.821
124.70	06.698	124.31	10.341
125.00	10.501	124.54	14.102
125.74	18.234	125.00	21.603
126.60	25.041	126.30	35.814
127.50	31.402	128.12	46.123
128.90	40.041	129.40	53.229
130.31	47.221	131.50	61.900
131.60	53.241	133.59	69.221
133.70	59.913	135.80	75.009
136.50	68.221	137.61	81.232
140.54	77.101	140.81	85.114
144.20	82.541	142.91	87.000
147.12	87.141	144.61	91.011
153.51	92.801	147.82	94.513
157.05	95.501	150.61	97.512
161.60	98.801	153.90	100.000
164.50	100.000		

[a] $P = 760$ mmHg.

sure to avoid decomposition by high temperature. The purity of the substance was tested by determining the refractive index, boiling point, density, and the difference between the boiling and condensation temperatures. The measured values of these properties are given in Table I (*3*).

The boiling temperature–composition (t–X) relation for any system was studied by using the ebulliometric method. A differential ebulliometer was used in measuring the boiling points of the different composition mixtures of each system at their equilibrium states with vapors (*2, 10*).

The temperature was always measured by using a Beckmann thermometer and was subject to an error, not larger than 0.05 °C (*3*). Two types of correction were applied to each measurement. The first type of correction was for the small changes of atmospheric pressure and was determined by measuring the boiling point of water using a simple ebulliometer (*2, 10*). The

Table III. Vapor–Liquid Equilibrium Data for *n*-Butyric Acid–*n*-Octane and Isobutyric Acid–*n*-Octane ($P = 760$ mmHg)

t, °C	X_1[a]	Y_1[b]	γ_1[c]	γ_2[d]	γ_1/γ_2
		n-Butyric Acid–*n*-Octane			
125.60	00.000	00.000		1.0000	
124.80	02.922	03.788	4.7751	1.0060	5.7406
124.70	06.171	05.943	3.5602	1.0191	3.4937
124.65	08.045	08.001	3.0711	1.0292	2.9895
125.00	12.642	08.140	2.3523	1.0513	2.2379
125.71	22.233	09.941	1.5902	1.1514	1.3830
127.20	37.119	13.480	1.5031	1.2841	0.9369
130.30	53.700	19.413	1.0810	1.4282	0.7571
131.60	59.648	22.011	1.0532	1.4763	0.7134
133.70	65.942	25.832	1.0341	1.5252	0.6780
136.50	73.623	31.133	1.0160	1.5811	0.6426
140.50	81.342	39.521	1.0073	1.6364	0.6155
142.70	83.900	43.012	1.0015	1.7112	0.5853
147.10	89.773	54.372	1.0021	1.6900	0.5929
153.50	94.281	70.334	1.0010	1.7180	0.5827
155.20	94.823	74.849	1.0010	1.7213	0.5782
157.20	96.654	81.190	1.0010	1.7332	0.5724
161.60	99.069	95.232	1.0000	1.7474	0.5722
164.50	100.000	100.000	1.0000		
		Isobutyric Acid–*n*-Octane			
125.60	00.000	00.000		1.0000	
124.61	03.391	05.015	3.9551	1.0060	3.9315
124.30	07.452	08.283	3.0040	1.0211	2.9423
124.15	10.505	10.401	2.6850	1.0370	2.5892
124.30	12.641	11.123	2.4024	1.0477	2.2931
124.50	17.631	12.160	1.8951	1.0761	1.7612
125.00	25.781	14.521	1.5191	1.1552	1.3152
126.31	39.113	19.122	1.2290	1.2770	0.9624
128.10	52.671	24.610	1.1002	1.4031	0.7872
129.42	59.623	28.241	1.0640	1.4622	0.7278
133.32	71.501	36.372	1.0020	1.5644	0.6407
135.41	77.121	43.052	1.0015	1.6110	0.6301
136.81	82.543	47.872	1.0008	1.6551	0.6096
137.18	83.689	48.854	1.0002	1.6642	0.6022
139.39	87.541	55.472	1.0003	1.6932	0.5925
141.52	88.299	59.691	1.0003	1.7000	0.5900
142.55	89.891	62.831	1.0002	1.7123	0.5854
143.60	89.900	64.572	1.0003	1.7060	0.5880
146.48	94.100	76.461	1.0022	1.7442	0.5745
153.90	100.000	100.000	1.0000		

[a] X_1 = weight percent of butyric acid component in liquid phase. [b] Y_1 = weight percent of butyric acid component in vapor phase. [c] γ_1 = activity coefficient of butyric acid component. [d] γ_2 = activity coefficient of other component.

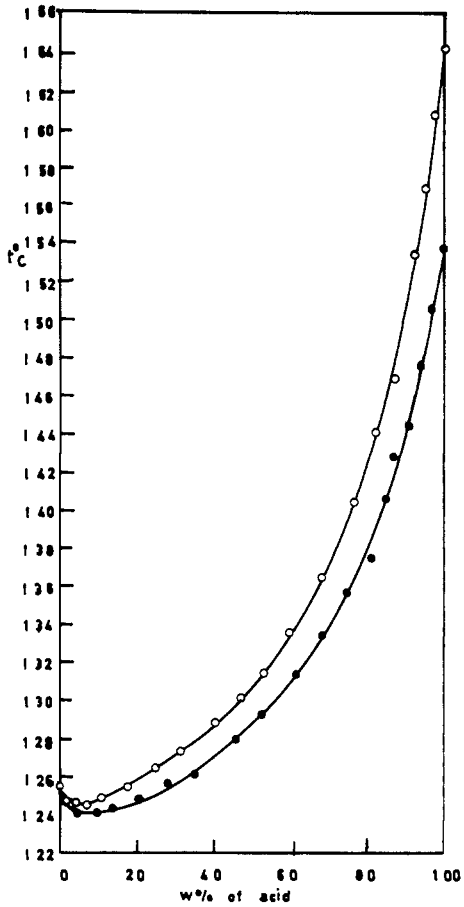
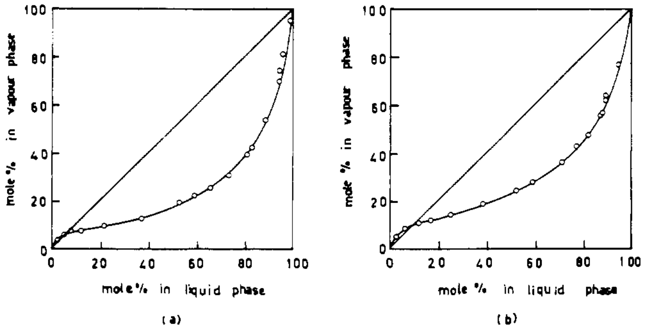


Figure 1. Temperature-composition diagram for the systems (○) *n*-butyric acid–*n*-octane and (●) isobutyric acid–*n*-octane at 760 mmHg.

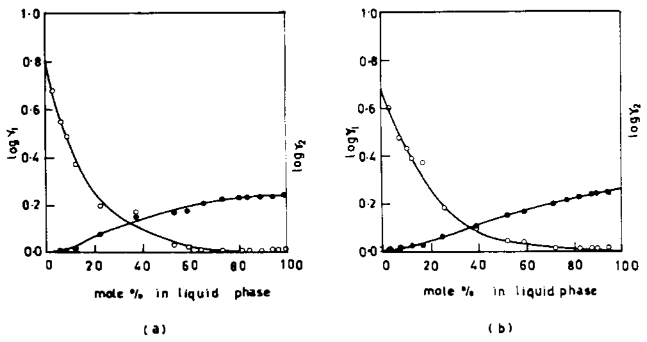


(a) (b)

Figure 2. Vapor-liquid equilibrium diagram for the systems (a) *n*-butyric acid–*n*-octane and (b) isobutyric acid–*n*-octane at 760 mmHg.

applied correction was of the order of 0.1°/atm. The second type of correction was for the exposed thermometer thread and for the secular changes of the position of the zero point on the thermometer due to the slow changes in shape of the thermometer glass bulb with time (*2*).

The vapor-liquid (V-L) equilibria were studied by the so-called flow method in which an ebulliometer was used to distill a liquid condensed from its vapors, collect the condensate in a hold-up trap, and return it to a flask without having contact with the vapors from the distillation (*2, 11*). At the equilibrium states of the liquids with their vapors, weighed samples of the liquids and vapors were analyzed by either titration with a standard sodium hydroxide solution when the system included *n*-octane or *p*-xylene, or refractive index determination if it included dimethyl- or diethylaniline. In the last analytical method a calibration curve had to be drawn for use in the determination.



(a) (b)

Figure 3. Activity coefficient-composition diagram for the systems (a) *n*-butyric acid–*n*-octane and (b) isobutyric acid–*n*-octane at 760 mmHg.

Table IV. Average Values and Standard Errors of Terms *A* and *B* of Van Laar Equations

	A		*B*	
System	Av value	Standard error[a]	Av value	Standard error[a]
n-Butyric acid–*n*-octane	0.7519	0.0309	0.2424	0.0026
Isobutyric acid–*n*-octane	0.7151	0.0358	0.2446	0.0072
n-Butyric acid–*p*-xylene	0.5173	0.0041	0.2837	0.0015
Isobutyric acid–*p*-xylene	0.3863	0.0050	0.1747	0.0025
n-Butyric acid–anisole	0.3391	0.0213	0.1186	0.0014
Isobutyric acid–anisole	0.2862	0.0045	0.2443	0.0032
n-Butyric acid–dimethylaniline	0.2879	0.0046	1.0253	0.0065
Isobutyric acid–dimethylaniline	0.3225	0.0109	0.4763	0.0062
n-Butyric acid–diethylaniline	0.3663	0.0091	0.7628	0.0121
Isobutyric acid–diethylaniline	0.5629	0.0120	0.8074	0.0056

[a] Standard error = $\sigma/\sqrt{n}$, where σ is the standard deviation of the original data, and n is the number of data.

For all systems containing, as a component, one of the five substances *n*-octane, *p*-xylene, anisole, dimethylaniline, and diethylaniline, the boiling temperature-composition (t-X) relations and the vapor-liquid equilibria (V-L) were studied. For the systems which contained acetic anhydride and toluene, only the t-X relation was studied because a positive azeotrope was formed in every case. For the four systems composed of isobutyric acid and a petroleum fraction, the t-X relation was studied for all possible points, but for the remaining four systems of the *n*-acid and a petroleum fraction, the t-X relation was studied only for a few points of low acid compositions.

Results and Discussion

t-X and V-L experimental data obtained for the two systems, *n*- or isobutyric acid and *n*-octane, are given in Tables II and III and represented in Figures 1 and 2, respectively. For each of these two systems, an azeotropic mixture was formed, boiling at a temperature close to the boiling point of *n*-octane, as indicated in Table VI. Also, as seen in Table VI, the acid percentages in the formed azeotropes are very low corresponding to 7.4 and 10.8 mol % when obtained from the V-L curves.

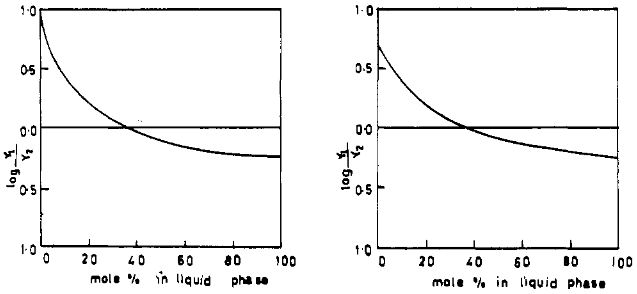


Figure 4. Redlich–Kister test for the systems (a) *n*-butyric acid–*n*-octane and (b) isobutyric acid–*n*-octane at 760 mmHg.

Table V. Results of Redlich–Kister Test

System	% of difference in area
n-Butyric acid–*n*-octane	2.1
Isobutyric acid–*n*-octane	0.0
n-Butyric acid–*p*-xylene	2.3
Isobutyric acid–*p*-xylene	5.3
n-Butyric acid–anisole	7.7
Isobutyric acid–anisole	7.8
n-Butyric acid–dimethylaniline	3.7
Isobutyric acid–dimethylaniline	4.0
n-Butyric acid–diethylaniline	5.5
Isobutyric acid–diethylaniline	0.0

Deviations from Raoult's law are positive in both systems. They are studied by first calculating, for each component in either system, the activity coefficient $\gamma = YP/XP^0$, where X and Y are the mole fractions in the liquid and vapor phases, respectively, P is the total vapor pressure (1 atm), and P^0 is the vapor pressure of the pure component at the temperature of the system, and is obtained by using the Antoine equation:

$$\log P^0 = a - b/(t + c)$$

where a, b, and c are constants obtained from published tables (*1*), and t is the temperature reading on a centigrade scale.

By plotting log γ vs. X for each component of the system, as in Figure 3, positive deviations from Raoult's law are easily preceived, according to the Gibbs–Duhem equation.

The accuracy of data of either system was tested by calculating, for each point, the terms A and B of Van Laar equations (*12*):

$$A = \log \gamma_1(1 + X_2 \log \gamma_2/X_1 \log \gamma_1)^2$$

and

$$B = \log \gamma_2(1 + X_1 \log \gamma_1/X_2 \log \gamma_2)^2$$

The average values of A and B and their standard errors, for all completely investigated systems are given in Table IV. The ratio of the standard error to the corresponding average varies between 0.0063 and 0.0625 with a median value of 0.0158.

The thermodynamic consistency was tested by applying the Redlich–Kister test (*8*) in which the curve log (γ_1/γ_2) vs. X was traced as shown in Figure 4. Because of the thermodynamic consistency of data of either system, the upper and lower subtended areas between the curves and the horizontal axis corresponding to zero ordinate were found to differ by 2.1 and 0.0% of the total areas for the *n*- and isobutyric acid systems, respectively. Table V gives the proportions of area differences for all systems that have been completely investigated. These differences are all insignificant, the largest difference being less than 8%.

For the other 20 investigated systems (composed of a butyric acid isomer and *p*-xylene, anisole, dimethylaniline, diethylaniline, acetic anhydride, toluene, or a petroleum fraction), results are given in the microfilm edition. Table VI presents the boiling points of the pure substances and the formed azeotropes and the weight percentages of the acid components of the formed azeotropes. The general conclusions which may be drawn are as follows.

The obtained experimental results have been proved to be thermodynamically consistent, as provided by Van Laar and Redlich–Kister tests.

No azeotrope mixture has been formed with the substances dimethylaniline, diethylaniline, and toluene. Slight positive deviations could be noticed with diethylaniline. With dimethylaniline, the t–X curve seems to be of the tangent zeotrope (nonazeotrope) and it is more pronounced for the systems containing *n*-butyric acid, because the difference between the boiling points of the two components is less than that of the isoacid system.

Large deviations are noticed by studying the t–X relation of the two toluene systems, the curves are almost of the type tangent zeotrope. It is worthwhile to refer here to the contradicting reports about toluene formation of azeotropes with butyric acid isomers. Schicktanz and others (*9*) state that toluene forms an azeotrope with either butyric acid isomers, while Horsley and Lecat (*4*, *5*) point out the nonformation of azeotropes.

The observed deviations from the ideal behavior are always

Table VI. Boiling Points (°C) of Used Pure Substances and Formed Azeotropes and Weight Composition of Formed Azeotropes

Substance used	Bp of pure substance	Bp of formed azeotrope with		wt % of acid in formed azeotrope	
		n-Acid	Isoacid	*n*-Acid	Isoacid
n-Butyric acid	163.55				
Isobutyric acid	154.70				
n-Octane	125.60	124.68	124.27	5.50	7.40
p-Xylene	138.20	137.60	136.75	7.50	13.50
Anisole	153.90	152.00	148.90	15.45	41.50
Dimethylaniline	193.40	No azeotrope formed			
Diethylaniline	217.32	No azetrope formed			
Acetic anhydride	139.50	137.10	135.38	24.00	35.00
Toluene	110.00	No azeotrope formed			
Petroleum fraction	148.10	144.10	142.75	27.00	35.00
	143.30	140.35	139.55	21.00	24.00
	140.80	138.80	138.40	20.00	22.00
	129.90	129.20	128.80	5.50	7.50

positive. This may be attributed to the fact that the acid component tends to associate, especially at higher concentrations.

The characteristics of any two corresponding azeotropic mixtures formed by the acid isomers are very close and in no way would lead to the separation of the two acids. This conforms with the reported acid azeotropes formed by the six substances: *m*-xylene, ethylene bromide, chlorobenzene, benzyle chloride, anisole, and water (*1*). The reported acid compositions and boiling points of anisole azeotropes are slightly different from those obtained in our investigation. Our results have been repeatedly obtained several times.

Nomenclature

X = mole fraction of component in liquid phase
Y = mole fraction of component in vapor phase
t = temperature, °C
P = total vapor pressure (= 1 atm)
P^0 = vapor pressure of pure component
a, b, c = constants in Antoine equation
A, B = terms of Van Laar equation
γ = activity coefficient

Subscripts

1 = butyric acid component
2 = nonbutyric acid component

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Supplementary Material Available: boiling temperature-composition data and vapor-equilibrium data given in tables and figures (40 pages). Ordering information is given on any current masthead page.

Vapor–Liquid Equilibrium in the System Water/1,2-Propylenediamine

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Vapor–liquid equilibrium in the system water/1,2-propylenediamine was studied under atmospheric pressure (760 mmHg) using thoroughly purified propylenediamine (bp 120.9 °C, $d^{20}{}_4$ = 0.8640, $n^{20}{}_D$ 1.44672), the refractometric method of analysis being the most reliable. The flow method and the equilibrium still of Vilim, Hala, Pick, and Fried were used in the experiments. The peculiar trend of the equilibrium curve observed is presumably associated with the presence of a half-hydrate reported in the literature. A direct proof of the absence of an azeotrope under atmospheric pressure is the very pure propylenediamine obtained on rectification of the system, starting with dilute solutions, established during the purification of propylenediamine.

Propylenediamine, $C_3H_6(NH_2)_2$, is an important intermediate product in the synthesis of preparations belonging to the dithiocarbamate series, used for pest control of plants. During the production process it must be isolated from an aqueous solution by rectification.

Data about the vapor–liquid equilibrium in the system water/propylenediamine are lacking and the data on its physicochemical properties are very scarce. The purpose of the present investigation was the study of the vapor–liquid equilibrium in the system water/propylenediamine under atmospheric pressure (760 mmHg).

In the experiments we used distilled water and propylenediamine (PDA, "Fluka", analytically pure, racemic mixture with a certified 99 wt % purity grade, bp 119–120 °C, $d^{20}{}_4$ = 0.875 and $n^{20}{}_D$ 1.4460), thoroughly purified according to the following method. It was first dried with solid KOH (analytically pure) for several days, as it had been found, by gas chromatography, to contain water. The dry propylenediamine was then purified by rectification in a heat-insulated bench-scale column (26 mm in diameter; glass-bead packing, mean diameter of beads 5.2 mm, layer depth 0.55 m). As column efficiency was relatively low, purification was carried out in two stages. The first stage involved alternating operation at total reflux (about 30 min) and a removal of the low-boiling fraction at the top of the column (3–5 min) at a reflux ratio of 2 to 3 (48 h of operation in all). The second stage starts when, in the course of the distillation, temperature of the vapor phase (measured at the top of the column) changed no more. This stage represented conventional rectification carried out at a reflux ratio of 40–45. For the experiments we used the middle fraction (about 300 ml from a total of 1500 ml) having the following physicochemical properties: boiling point 120.9 °C (120.5° (*1*) 120.9° (*4*)); density 0.8640 g/cm³ (0.8635 (*4*)); refraction coefficient 1.446 72.

The starting solutions employed were prepared by mixing propylenediamine with distilled water. They were left to stand for at least 24 h before use. The aim was to achieve chemical equilibrium in the solution, needed on account of the presence of a half-hydrate reported in the literature (*1, 3–6, 9*).

Considering the high accuracy required in the study of the vapor–liquid equilibrium of the system, gas chromatography proved inadequate as a method of analysis; it was employed (as mentioned above) only for a qualitative characterization of the