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Solubilities of Butane, Vapor Pressures, and Densities for Benzene + Cyclohexane, Benzene + Methanol, and Methanol + Cyclohexane Solutions at 298 K

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In this paper the solubilities of butane at a pressure of 101.3 kPa and a temperature of 298.15 K are presented for three mixed solvent solutions: benzene + methanol, cyclohexane + methanol, and benzene + cyclohexane. The densities and vapor pressures are also reported for these solutions at the same conditions. Whereas the benzene + methanol and cyclohexane + methanol mixtures form azeotropic solutions, they are mutually soluble for all compositions of the two solvents. On the other hand, mixtures of cyclohexane and methanol are not mutually soluble but form two immiscible liquid phases for a significant portion of the composition range, but at a higher temperature also form an azeotropic solution.

Introduction

A systematic study of gas solubilities is useful in providing design data for absorption processes, as well as indirectly useful in aiding the analysis of molecular interactions in solutions. Certain types of solvent interactions appear to reduce the solubility of nonreacting gases below that expected for simple solutions, whereas an enhancement in solubility is observed for certain other gases which tend to reversibly react with the solvent (*1*). It is expected that similar phenomena may be observed in mixed solvent solutions. For these reasons it is useful to study gas solubilities in mixed solvent solutions especially in those with strong molecular interactions and irregular solvating properties such as those used in this work.

Butane gas is highly soluble, more soluble than propane, propylene, ethane, or similar hydrocarbon gases in nonpolar, nonassociating solvents. Solubilities of nonreacting gases in nonpolar, nonassociating solvents generally follow an order of increasing solubility with increases in the normal boiling point temperature of the liquefied gas. While this approximate relationship exists, it is not sufficiently consistent to permit the prediction of gas solubility even for simple solutions. In polar and associating solvents, the effect of molecular association on solubility can be demonstrated by comparing the solubilities of a number of gases, in several solvents of varying degrees of association (*2*). In a range of associated solvents, the solubilities of most gases are observed to decrease with increases in the degree of association in the solvents. The difficulty of predicting gas solubility behavior can be illustrated with the following example: butane is highly soluble in cyclohexane at a 101.3-kPa pressure and 298.15 K, reaching an equilibrium concentration expressed as the mole fraction of butane of about 0.39. On the other hand, in water as solvent, for the same conditions the mole fraction butane solubility is about 0.0002 or approximately 1/2000 of that in cyclohexane. This example indicates that the degree of association in the solvent has a most profound effect on the solubility.

An equation for gas solubilities in ideal mixed solvent solutions based on the development of Hildebrand et al. (*3*) is

$$\ln x_{1,\mathrm{ideal}} = \phi_2 \ln x_{12} + \phi_3 \ln x_{13} \tag{1}$$

Hence, the deviation from the ideal solubility estimated by eq 1 or the excess solubility is expressed by

$$\ln x^{\mathrm{E}}_{1,\mathrm{mix}} = \ln x_{1,\mathrm{mix}} - \phi_2 \ln x_{12} - \phi_3 \ln x_{13} \tag{2}$$

In eqs 1 and 2 ϕ_2 and ϕ_3 are volume fractions of the two solvent components, x_{12} and x_{13} are the mole fraction butane solubilities for a partial pressure of 101 325 Pa in the pure solvent components, and $x^{\mathrm{E}}_{1,\mathrm{mix}}$ is the excess solubility of butane based on $x_{1,\mathrm{mix}}$ the actual solubility in the mixed solvent, and $x_{1,\mathrm{ideal}}$, the ideal solubility calculated using eq 1. It is of interest to discover the direction and the extent of the deviation of the solubility from the ideal (the excess solubility) and the relation of the deviation to the mixed solvent properties. In a former paper (*4*) it was demonstrated that the solubilities of butane in mixed solvent solutions composed of acetone and the alkanols from methanol to butanol showed a consistent behavior. A significant reduction in solubility was observed in solvent mixtures composed of acetone + methanol, and lesser reductions were observed for each of the alkanols of increasing carbon number, when compared with solubilities in nonassociating solvents. Of interest also was the fact that the excess solubility was quantitatively related to the corresponding excess volume for each of the mixed solvent solutions. Thus, it appeared that the same molecular interactions that affected the solvent densities also affected the solubility of butane in these solvent solutions. We wished to see if the effects of molecular association in the solvents on butane solubilities and solution densities were also present in the three mixed solvent solutions used in this work.

Excess volumes are based on the "ideal" specific volume:

$$V_{\mathrm{ideal}} = (\phi_2\rho_2 + \phi_3\rho_3)^{-1} \tag{3}$$

$$V^{\mathrm{E}} = V - V_{\mathrm{ideal}} \tag{4}$$

In the above equations, ϕ_2 and ϕ_3 are volume fractions of solvent components as before, ρ_2 and ρ_3 are pure solvent component densities (kg m^{-3}), and V^{E} is the excess specific volume (m^3 kg^{-1}) based on V and V_{ideal}, the actual specific

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volume and ideal specific volume as calculated using eq 3, respectively. Further, because of the highly nonideal behavior of the three mixed solvent solutions used in this work, it was apparent that a special problem existed for the calculation of the solubility data. Because accurate vapor pressure data were required for the calculation of accurate solubilities, it was necessary to measure the vapor pressures for the whole composition range of the three solvent mixtures. With a knowledge of the solution vapor pressures and densities, it was then possible to calculate accurate *n*-butane solubilities from the experimental data.

For the determination of the vapor pressure of a pure solvent, the evacuation or deaeration step is of prime importance to ensure that all the residual dissolved air is released prior to the pressure measurement. This deaeration is usually accomplished by boiling the solvent under vacuum for a short period of time. For deaeration of mixed solvents there is the possibility that the solvent composition will be changed as a result of the deaeration. In this work, however, because the three solvents had very similar vapor pressures, it could have been assumed that the composition of the evolved vapor was essentially the same as that of the liquid solution. However, to avoid any possibility of error, the solvent components were deaerated prior to mixing in the equilibration cell.

Experimental Section

The solubilities of butane in the mixed solvent solutions were measured using a method previously described (*5*) with minor modifications. The method involved charging the two deaerated solvent components from separate burets into an evacuated equilibration cell. A measured quantity of the solvent having the lowest concentration in the solution was introduced first into a cell of known mass, and the quantity was determined by mass. The cell was then cooled in an ice bath, a measured quantity of the second solvent component was introduced in the cell, and the quantity was again determined by mass. At this point in the procedure either the vapor pressure of the mixed solvent or the solubility of the butane could be measured. The procedure for solubility determinations will be summarized first. The equilibration cell, partially filled with deaerated solvent of known composition, was immersed in a constant-temperature bath and connected to a supply of butane gas at constant pressure. Next, the cell was pressurized with butane through a needle valve attached to the cell, and then agitated for equilibration. Incremental additions of butane were required until finally the cell was subjected to mechanical agitation for at least 60 min, a period of time found to be sufficient to give a maximum or equilibrium quantity of gas dissolved. The mechanical agitation of the cell was possible because the connection to the butane gas included a section of flexible rubber tubing. The quantity of gas dissolved was again determined by mass after the cell was sealed and removed from the bath and the exterior was completely dried. Because the mass of each component in the cell could be determined to 1 mg, an accurate result for the composition of the solution was obtained. Due correction for the mass of butane and solvent vapor remaining in the gas phase in the cell was made although this correction was almost negligible. The mole fraction solubility at the pressure of the experiment was given by

$$x_p = (W_g - w_g)/M_g[(W_g - w_g)/M_g + (W_1 - w_1)/M_1 + (W_2 - w_2)/M_2]^{-1} \quad (5)$$

In eq 5 W_1 and W_2 are the masses (kg) of deaerated solvent components charged into the cell, w_1 and w_2 are the masses of the solvent vapors of the two components in the gas phase of the cell, W_g is the mass of butane charged into the cell, and w_g is the amount of butane left in the gas phase at equilibrium. Also, M_1 and M_2 are the solvent molecular weights, M_g is the butane molecular weight, and x_p is the mole fraction solubility of butane at the prevailing butane partial pressure. The gas partial pressure at the pressure of the experiment was calculated from the measured mixed solvent vapor pressure, $P^\circ_{\rm mix}$:

$$p_g = P_{\rm exptl} - (1 - x_p)P^\circ_{\rm mix} \quad (6)$$

The solubility corresponding to a gas partial pressure of 101.325 kPa was determined assuming that over a small pressure range Henry's law could be applied:

$$x = 101325x_p/p_g \quad (7)$$

In eqs 6 and 7 p_g is the butane partial pressure in the gas phase (Pa), whereas $P_{\rm exptl}$ and $P^\circ_{\rm mix}$ are the pressure of the experiment and vapor pressure of the mixed solvent, respectively.

One of the modifications to the procedure from that formerly utilized was the measurement of the mixed solvent vapor pressures using the same apparatus as that used for the solubility determinations. For the determination of solvent vapor pressure, once the evacuated equilibrium cell was charged with both solvent components, the cell and the line to the quartz manometer were once again evacuated for several minutes to ensure that deaeration was complete. The vacuum line was then closed and the solution left to equilibrate. During this period it was ensured that the temperature of the quartz electronic manometer was greater than 298.15 K to prevent condensation of solvent vapor. The final equilibrium mixed solvent vapor pressure was read to 0.1 mmHg (13.33 Pa), and the accuracy was considered to be better than 0.5 mmHg (or 66.65 Pa). The procedure for the vapor pressure determinations is similar to that described by Nitta et al. (*6*).

The mixed solvent densities were measured at 298.15 K using an Anton Paar digital densitometer, model DMA 60, and densitometer cell, DMA 602W (Austria). The two accurately known densities of methanol, taken from Wilhoit and Zwolinski (*7*), and of distilled water were used for calibration of the densitometer. The solutions were prepared from the pure solvents in two burets for each solvent, a small 10 cm^3 and a large 25 cm^3 buret to permit accurate measurements of even small volumes of solvent. The solutions were prepared in 50 cm^3 volumetric flasks so that there was a check on the volumes used. There usually was a slight but noticeable change in the volume of the solution when compared with the volume of solvents charged because of the volume change on mixing. Care was also taken to keep the solvents, particularly those containing methanol, from contacting the air for any significant period of time because of possible absorption of water vapor from the air. A 3 cm^3 syringe was used to inject samples of the well-mixed solutions into the densitometer. The sample loop contained only about 0.5 cm^3 of sample so that it was possible to flush through the densitometer cell a volume of solvent before a density determination was made. The densitometer used a vibrating reed mounted in a glass capillary tube, the vibrating frequency of which was related to the density of the liquid in which it was immersed. Calibration was accomplished by determining the calibration constant, *A*, from the vibrating frequencies in the two liquids of known density as follows:

$$A = (T_1^2 - T_2^2)/(\rho_1 - \rho_2) \quad (8)$$

Subsequently the density of a solution was calculated using

Table I. Butane Solubilities x at a Partial Pressure of 101.32 kPa in the Pure Solvents as Well as Experimental Densities ρ and Vapor Pressures P° of Pure Solvents at 298.15 K

	this work	lit.	difference/%
	ρ/(kg m^3)		
methanol	786.64[a]	786.64	NA
cyclohexane	773.70	773.87[b]	−0.02
benzene	873.64	873.68[b]	−0.005
	P°/kPa		
methanol	16.92	16.97[c]	−0.25
cyclohexane	13.01	13.04[c]	−0.23
benzene	12.71	12.66[c]	+0.39
	x		
methanol	0.037 22	0.037 63[d]	−1.1
cyclohexane	0.387 1	0.396 2[d]	−2.4
benzene	0.281 3	0.285 1[d]	−1.3

[a] The methanol density from Wilhoit and Zwolinski (*7*) was used as a calibration standard for the digital densitometer. [b] ASTM Data Series Publication DS 4A (*8*). [c] Reid et al. (*9*). [d] Miyano and Hayduk (*4*).

Table II. Solubilities of Butane x_1 in Solvent Solutions at 298.15 K and a Butane Partial Pressure of 101.325 kPa

ϕ_2[a]	$x_{1,mix}$	$x^E_{1,mix}$	ϕ_2[a]	$x_{1,mix}$	$x^E_{1,mix}$
		Benzene (2) + Cyclohexane (3)			
0.0	0.3871	0.0	0.6136	0.3471	0.087
0.1004	0.3853	0.027	0.6871	0.3372	0.081
0.1948	0.3813	0.047	0.6987	0.3360	0.081
0.2974	0.3746	0.062	0.8007	0.3173	0.057
0.3020	0.3757	0.067	0.8151	0.3161	0.057
0.4015	0.3677	0.077	0.8899	0.3024	0.037
0.5299	0.3570	0.088	0.9024	0.2991	0.030
0.6059	0.3482	0.088	1.0	0.2813	0.0
		Methanol (2) + Benzene (3)			
0.0	0.2813	0.0	0.4045	0.1269	0.024
0.0079	0.2685	−0.029	0.5002	0.1051	0.026
0.0252	0.2561	−0.042	0.5073	0.1042	0.032
0.0488	0.2442	−0.043	0.5982	0.0872	0.039
0.0766	0.2330	−0.032	0.6981	0.0706	0.030
0.1148	0.2171	−0.026	0.7060	0.0693	0.027
0.1457	0.2063	−0.016	0.7981	0.0572	0.022
0.1987	0.1872	−0.005	0.9032	0.0460	0.015
0.2573	0.1681	0.006	1.0	0.0372	0.0
0.3120	0.1523	0.016			
		Methanol (2) + Cyclohexane (3)			
0.0	0.3871	0.0		immiscible region	
0.0050	0.3793	−0.009	0.7025	0.0988	0.280
0.0196	0.3636	−0.017	0.8032	0.0743	0.231
0.0259	0.3581	−0.017	0.8925	0.0551	0.140
0.0348	0.3567	0.0	0.9004	0.0538	0.135
0.0402	0.3515	−0.002	0.9039	0.0528	0.125
0.0556	0.3440	0.012	1.0	0.0372	0.0

[a] Volume fraction in solvent on a butane-free basis.

the following equation:

$$\rho_{mix} = (T_{mix}^2 - T_2^2)/A + \rho_2 \qquad (9)$$

In eqs 8 and 9 T refers to the frequency of vibration of the glass reed (s^{-1}) for the two liquids, 1 and 2, used for calibration and in the mixed solvent, T_{mix}, and the subscripted ρ refers to the corresponding density (kg m^{-3}) of components 1 and 2 and the mixed solvent. The instrument yielded replicate density results with a reproducibility of less than 0.1 kg m^{-3} when the temperature surrounding the capillary tube of the densitometer was controlled to 0.02 K.

The vapor pressure and solubility measurements were performed at Okayama University of Science and the density measurements at the University of Ottawa. For the former measurements the butane was supplied by Chugoku Kasei at a specified minimum purity of 99.0 mol %. The methanol, benzene, and cyclohexane were of HPLC grade and supplied by Aldrich Chemicals, all with a specified minimum purity of 99.9 mol %. The same grade, supplier, and purity of solvents were used for the density measurements.

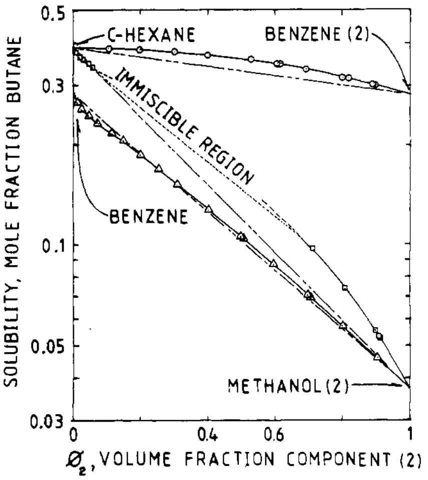

Figure 1. Solubilities of butane in the three mixed solvent solutions composed of benzene, cyclohexane, and methanol at 298.15 K and for a butane partial pressure of 101.3 kPa.

Results and Discussion

The solubilitites of butane at a temperature of 298.15 K in the pure solvents methanol, benzene, and cyclohexane as well as the densities and vapor pressures as measured in this work are compared with published values in Table I. Whereas the density of methanol as published by Wilhoit and Zwolinski (*7*) was used as a calibration standard for the digital densitometer, the deviations in densities for the other two solvents when compared with values from the ASTM Data Series publication (*8*) are small. A quartz manometer was used to measure the solvent vapor pressures, and reproducible results were attained; the deviations in vapor pressure from those published by Reid et al. for the pure solvents (*9*) are less than 0.5% so that the agreement is considered satisfactory. The explanation for the 1.1–2.4% deviation in butane solubilities in the pure solvents from those obtained earlier (*5*) is that more care was taken with the experimental procedure and the experiments were repeated in the more recent measurements.

The butane solubilities in the three mixed solvent solutions are listed in Table II and shown in Figure 1. It is of interest that for "ideal" solution behavior, the *n*-butane solubilities should be linear when plotted as in Figure 1. In spite of the large difference in solubilities for butane in benzene and in methanol, the measured solubilities are within 4% of being the ideal solubilities for the mixed solvent solutions composed of methanol and benzene. The butane solubilities are within 8% of being the ideal solubilities in benzene and cyclohexane, but deviate widely for the high concentrations of methanol in the miscible region of the methanol + cyclohexane solvent mixtures. The deviations from the ideal solubility behavior are shown as the excess solubilities in Figure 2 which also shows the excess volume for these same mixed solvent solutions. There is no strong similarity in the curves of the two excess properties as was observed for butane solubilities in acetone + alkanol solutions (*4*). There is, however, at least a qualitative similarity between the excess volume and excess solubility curves in that the excess volumes and excess solubilities are lowest for the methanol + benzene solutions, increase for benzene + cyclohexane, and are highest for the partially miscible mixed solvent solution of methanol + cyclohexane. Further, it would appear that the butane

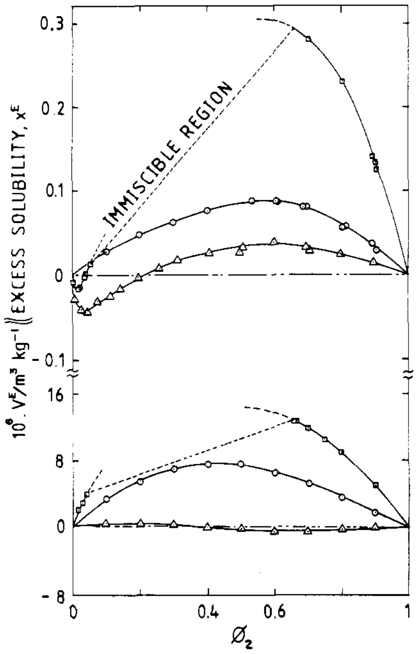

Figure 2. Excess volumes, and excess solubilities of butane, at 298.15 K for solutions of △, methanol (2) + benzene (3); ○, benzene (2) + cyclohexane (3); and □, methanol (2) + cyclohexane (3).

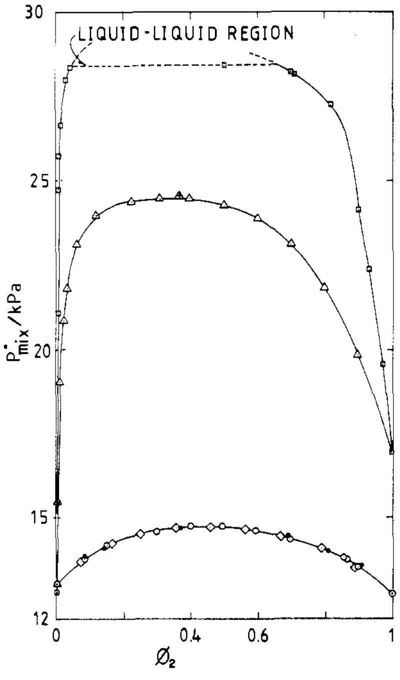

Figure 3. Vapor pressures at 298.15 K of methanol (2) + benzene (3), ●, Li and Lu (*10*); ◇, Nitta et al. (*6*); ○, this work; of benzene (2) + cyclohexane (3), ▲, extrapolated from Oracz and Kolasinska (*11*) and Tochigi et al. (*12*); △, this work; and of methanol (2) + cyclohexane (3), □, this work.

Table III. Vapor Pressures for Benzene + Cyclohexane, Methanol + Benzene, and Methanol + Cyclohexane at 298.15 K

system	x_2	$P°_{mix}$/kPa	x_2	$P°_{mix}$/kPa
benzene (2) + cyclohexane (3)	0.0	13.01	0.6375	14.59
	0.0973	13.75	0.7335	14.29
	0.1726	14.16	0.8771	13.81
	0.3407	14.54	0.8857	13.78
	0.4430	14.72	0.9146	13.50
	0.4486	14.75	1.0	12.71
	0.5426	14.73		
methanol (2) + benzene (3)	0.0	12.71	0.5910	24.43
	0.0248	19.00	0.6868	24.25
	0.0457	20.83	0.7676	23.85
	0.0663	21.78	0.8343	23.10
	0.1247	23.10	0.8981	21.81
	0.2292	23.92	0.9512	19.83
	0.3887	24.34	1.0	16.92
	0.4974	24.43		
methanol (2) + cyclohexane (3)	0.0	13.01	0.8627	28.21
	0.0081	21.68	0.8670	28.13
	0.0168	24.71	0.9233	27.25
	0.0248	25.74	0.9597	24.15
	0.0347	26.61	0.9729	22.32
	0.0770	28.06	0.9887	19.52
	0.1007	28.28	1.0	16.92
(two liquid phases)	0.7289	28.43		

Table IV. Densities ρ and Excess Volumes V^E at 298.15 K for Benzene + Cyclohexane, Benzene + Methanol, and the Miscible Solutions of Cyclohexane + Methanol

	benzene (2) + cyclohexane (3)		benzene (2) + methanol (3)	
ϕ_2	ρ/ (kg m^{-3})	$10^6 V^E$/ (m^3 kg^{-1})	ρ/ (kg m^{-3})	$10^6 V^E$/ (m^3 kg^{-1})
0.0	773.7	0	786.64	0
0.1	781.5	3.6	795.5	−0.3
0.2	790.0	5.9	804.3	−0.4
0.3	799.0	7.3	813.2	−0.7
0.4	808.4	8.0	821.9	−0.7
0.5	818.2	8.1	830.4	−0.4
0.6	828.6	7.3	838.9	−0.1
0.7	839.6	6.1	847.4	+0.2
0.8	850.4	4.4	855.9	0.4
0.9	861.5	2.8	864.7	0.3
1.0	873.6	0	873.6	0

	methanol (2) + cyclohexane (3)			methanol (2) + cyclohexane (3)	
ϕ_2	ρ/ (kg m^{-3})	$10^6 V^E$/ (m^3 kg^{-1})	ϕ_2	ρ/ (kg m^{-3})	$10^6 V^E$/ (m^3 kg^{-1})
0.0	773.7	0	0.67	774.8	12.5
0.02	772.7	2.1	0.70	775.7	11.6
0.03	772.3	3.0	0.75	777.0	10.5
0.04	771.9	3.9	0.80	778.5	9.1
	immiscible region		0.90	782.2	5.1
0.66	774.7	12.4	1.00	786.64	0

solubility in mixed solvents composed of nonpolar, polar, and associating solvents is more affected by the solvent composition than is the solution density. Therefore, it may not be accurate to assume that excess solubility can be directly related to excess volume for such mixed solvent solutions.

The vapor pressures for the mixed solvent solutions are shown in Figure 3 and listed in Table III. Vapor pressure data of Nitta et al. (*6*) and of Li and Lu (*10*) for cyclohexane + benzene solutions at 298.15 K are essentially duplicated in this work, confirming that the method for vapor pressure measurements is satisfactory. In spite of a detailed search of the literature, no directly comparable vapor pressure data were found for either the benzene + methanol or the cyclohexane + methanol solutions at 298.15 K. However, single values of vapor pressure for benzene + methanol solutions are available for a low benzene content at 313.15 K of Oracz and Kolasinska (*11*) and at the azeotropic composition at 307.65 K of Tochigi et al. (*12*). These latter values were extrapolated to the temperature of 298.15 K and are shown in Figure 3 for comparison.

Positive deviations from Raoult's law as indicated by all three mixed solvents are characteristic of solutions which form minimum boiling azeotropes, whereas those with very large positive deviations tend to form separate, immiscible liquid phases as discussed by Van Ness (*13*) and Smith and Van Ness (*14*). These deviations from Raoult's law are ascribed to strong forces of attraction between like molecules and weak forces between unlike molecules so that partial pressures of such solutions are greater than those of ideal solutions, almost as if the liquids separated into two liquid phases which exerted separate vapor pressures. Benzene and cyclohexane form a minimum boiling azeotrope at a cyclohexane composition of 48.1 mass %. Methanol and benzene also form a minimum boiling azeotrope at a benzene composition of 60.9 mass %. Finally, although at 298.15 K two phases are formed with mixtures of methanol and cyclohexane, a minimum boiling azeotrope is also formed at a higher temperature, at a cyclohexane composition of 60.9 mass %. The azeotropic compositions are from Weast (*15*). At 298.15 K, the equilibrium compositions of the two liquid phases for methanol and cyclohexane are 0.045 volume fraction methanol in the cyclohexane-rich liquid and 0.66 volume fraction methanol in the methanol-rich liquid. These values were determined from a combination of the density, solubility, and vapor pressure measurements. The solution densities and excess volumes are listed in Table IV.

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Registry Numbers Supplied by Author. Benzene, 71-43-2; cyclohexane, 110-82-7; methanol, 67-56-1; *n*-butane, 106-97-8.

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