Vapor−Liquid Equilibrium in Binary Systems Formed by Allyl Alcohol with Benzene and with Cyclohexane

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The vapor−liquid equilibrium (VLE = pressure, temperature, and composition of coexisting vapor and liquid phases) was measured at 313.15, 323.15, and 333.15 K by the ebulliometric method for the binary systems formed by allyl alcohol with benzene and with cyclohexane. Positive azeotropes were found in both systems. The experimental VLE results were correlated with equations representing liquid-phase activity coefficients and by the AEOS (association + equation of state) equation of state.

Introduction

The purpose of the presented work was to determine the vapor−liquid equilibrium in binary systems formed by allyl alcohol with cyclic hydrocarbons (benzene and cyclohexane). There are no literature data for these systems. The only available data for mixtures formed by allyl alcohol with hydrocarbons are for systems with toluene,[1,2] 1,4-xylene, and ethylbenzene.[3]

Experimental Section

Chemicals. Allyl alcohol (CA Registry no. 107-18-6) was purchased from Fluka Chemie GmbH, as a physicochemical standard of purity > 99.5%. A purity > 99.75% (by mass) was found by GLC. The content of water determined by GLC analysis with a TDC detector, on a HP-FFAP (poly(ethylene glycol)−TPA modified) 30 m × 0.53 mm × 0.1 μm film thickness column, was <0.01 mass %. The substance was used without further purification. Benzene (CA Registry no. 71-43-2) was purchased from Sigma-Aldrich Chemie GmbH as a HPLC standard of purity > 99.9%. A purity > 99.69% (by mass) was found by GLC. The substance was further purified by fractional distillation through a 25 theoretical plates laboratory column. Finally, purity > 99.9% was obtained. The content of water determined by GLC was >0.04 mass %. Cyclohexane (CA Registry no. 110-82-7) was purchased from Polskie Odczynniki Chemiczne, Gliwice. The substance was purified by fractional distillation through a 25 theoretical plates laboratory column. The purity > 99.9% by mass was obtained. The content of water determined by GLC was > 0.01 mass %.

Vapor Pressure Measurements. In the handling of vapor−liquid equilibrium, the most crucial data are the saturation vapor pressure data of pure components. For allyl alcohol, measurements were made by Bauer and Burschkies[4] and by Evert.[5] Both data sets are for temperatures lower than 313 K, except that the Evert data are accompanied by boiling temperatures. All other literature data are derived from these two sets. Statistical analysis proves that the Bauer and Burschkies data are more accurate. For benzene and cyclohexane, numerous literature data are available. The most reliable of these are by Williamham et al.[6] For benzene, a newer set of data was published by the same laboratory[7] without comments. As a source of auxiliary data, the compilation by Dauber and Danner[8] was used.

In this work measurements of pure compound vapor pressure were carried out in a modified Świętosławski ebulliometer.[9] The ebulliometer was connected to the pressure stabilizing system consisting of a 0.6 m^3 buffer vessel combined with a vacuum pump and a pressurized argon container. The pressure was adjusted manually by using the system of vessels and a rubber balloon filled with argon until the boiling temperature of the compound in the ebulliometer was reached within ±0.005 K. The temperature was measured by using a SYSTEMTEKNIK AB S1228 thermometer with a platinum resistance probe and the resolution of 0.001 K. The calibration was made by comparison with readings of the Tinsley & Co Muller Bridge type 4772 with a platinum resistance temperature sensor type 5187 SA calibrated by NPL to Class I. In the course of measurements, the ice point temperature was checked. No systematic deviations of the temperature during measurements were observed. The pressure was measured with a Texas Instruments 144-01 precision pressure gauge with a No. 8, type 2 quartz Bourdon tube frequently recalibrated against a high precision mercury manometer (resolution ±0.01 mm) connected to the same system. The pressure resolution was ±0.3 Pa, and a pressure vacuum better than 10^{-2} Pa was used for the 144-01 null point. The estimated accuracy of the pressure measurement was ±10 Pa, and that of the temperature measurement was ±10 mK on the ITS-90 scale. The results obtained are given in Table 1. The results for allyl alcohol can be treated as an extension of Bauer and Burschkies[4] data (Figure 1).

Vapor Pressure Correlation. The vapor pressure data are correlated by means of the Antoine equation

$$\log(p/\text{kPa}) = A - \frac{B}{T/\text{K} - C} \qquad (1)$$

where p is the pressure, T is the temperature, and A, B, and C are adjustable parameters.

To obtain a good representation of VLE data in a wider range of pressure and temperature, the equation of state method is more useful. The development of the equation of state methods[10,11] shows that these methods are able to

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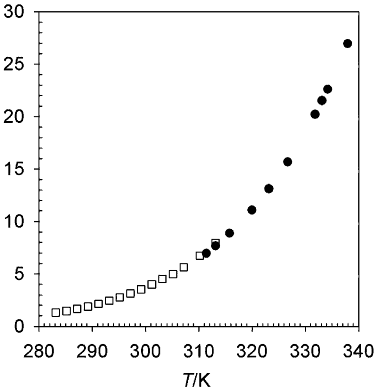

Figure 1. Vapor pressure of allyl alcohol: ●, own measurements; □, Bauer and Burschkies.[4]

Table 1. Pure Component Vapor Pressures

allyl alcohol		benzene		cyclohexane	
T/K	P/kPa	T/K	P/kPa	T/K	P/kPa
311.42	6.954	308.19	19.752	302.20	15.519
313.15	7.672	312.17	23.345	306.55	18.709
315.81	8.869	313.15	24.323	310.76	22.298
319.97	11.079	315.56	26.817	313.15	24.561
323.15	13.112	318.79	30.495	314.28	25.709
326.70	15.672	321.67	34.124	317.52	29.208
331.84	20.194	323.15	36.114	320.77	33.073
333.15	21.510	324.33	37.765	323.15	36.208
334.20	22.603	326.85	41.498	323.62	36.831
337.96	26.943	329.23	45.299	326.47	40.921
		331.52	49.195	330.29	46.944
		333.15	52.151	333.15	51.924
		333.64	53.057	333.47	52.484
		335.65	56.941	336.63	58.495
		338.01	61.766	339.05	63.460
		340.17	66.471	340.23	65.994

reproduce VLE data almost within the accuracy of the measurement.

In previous work,[12] it has been found that the AEOS[13] (association + equation of state) equation of state is most suitable for representation of phase equilibria in the systems formed by associating or even chemically reacting compounds.[14] The superiority of the AEOS equation over other EOSs in the representation of pressure and molar volume (V) of mixtures containing alcohols and inert components has been shown by Pfohol et al.[15]

In the AEOS model, the thermodynamic properties of an associated mixture are viewed as a result of chemical equilibria between associated species and physical interactions between all, associated or inert, species existing in a mixture. The chemical equilibria are assumed to follow a simplified, yet plausible association model, which, in general, depends on the nature of the associating compound. An equilibrium mixture of associated species is treated as a mixture of nonreacting polymeric aggregates.

The use of the AEOS equation leads to the split of the compressibility factor into two parts

$$z = z^{(\mathrm{ph})} + z^{(\mathrm{ch})} - 1 \tag{2}$$

where $z^{(\mathrm{ph})}$ and $z^{(\mathrm{ch})}$ are the physical and chemical contributions to the compressibility factor, respectively. The $z^{(\mathrm{ph})}$ contribution is equivalent to the equation of state for nonreacting monomeric species. In this work, it was calculated from the cubic equation of state of Yu et al.[16]

$$z^{(\mathrm{ph})} = \frac{V}{V - b} - \frac{a(T)V}{RT[V(V + c) + b(3V + c)]} \tag{3}$$

where $a(T)$, b, and c are generalized as functions of the critical temperature T_c, critical pressure P_c, and acentric factor ω of a pure component.

The $z^{(\mathrm{ch})}$ contribution is a function of the reciprocal mean association number, and its shape depends on the association model applied. In this work, the linear Mecke−Kempter-type association model has been used. It well represents the behavior of alcohols, phenols, ketones, amines, pyridine bases, and other similar compounds.[14,15] In the Mecke−Kempter model, all subsequent association constants (K) are equal. This leads to the following term for the chemical contribution:

$$z^{(\mathrm{ch})} = \frac{2}{1 + \sqrt{1 + 4RTK/V}} \tag{4}$$

The association constant is a function of standard enthalpy ($\Delta H°$) and entropy ($\Delta S°$) of association, both linearly dependent on temperature, through heat capacity ($\Delta C_p°$) according to eq 5 (the appropriate values of C_p are given in Table 2).

$$\ln K = \frac{-\Delta H°(T_0) + \Delta C_p° T_0}{RT} + \frac{1}{R}[\Delta S°(T_0) - \Delta C_p° - \Delta C_p° \ln T_0] + \frac{\Delta C_p°}{R} \ln T \tag{5}$$

The complete equation of state for an associating compound (eq 2) has the following characteristic parameters besides the association constant: the critical temperature (T'_c), the critical pressure (P'_c), and the acentric factor (ω') of a hypothetical monomeric compound with nonspecific interactions identical to those in the associating substance but incapable of forming associates. All these parameters were determined by fitting the equation adjustable parameters with pure compound experimental vapor pressure and liquid density as functions of temperature. No binary data were used in the fit. The obtained values of parameters are given in Table 2.

The determination of pure component parameters is a crucial step in the application of the AEOS equation. It is not sufficient to obtain a good fit to pure component data. It must also be ensured that the relative magnitude of the $z^{(\mathrm{ph})}$ and $z^{(\mathrm{ch})}$ terms be correct; that is, the effects of association and nonspecific interactions on the compressibility factor must be correctly divided. In principle, this can be accomplished by using physically meaningful values of the association parameters $\Delta H_{ii}°$ and $\Delta S_{ii}°$ (Table 2).

The correlation results for pure component vapor pressures by means of Antoine and AEOS equations are given in Figures 2−4. The comparison of correlation results (Table 2) shows that there is almost no difference between the two equations. The values for the AEOS equation are slightly higher. The enthalpies of vaporization (ΔH) calculated from vapor pressure differ from values obtained by direct calorimetric measurements about 0.1%, which is very satisfactory. The root-mean-square deviations [RMSD(p)] of property p are calculated from the equation

$$\mathrm{RMSD}(p) = \sqrt{\frac{\sum_{i=1}^{n}(p_i^{\mathrm{calcd}} - p_i^{\mathrm{exptl}})^2}{n}} \tag{6}$$

where p_i^{calcd} is the property calculated at point number i, p_i^{exptl} is the property measured at point number i, and n is

Table 2. Correlation of Pure Component Vapor Pressures

	allyl alcohol 310−340 K	benzene 305−345 K	cyclohexane 300−345 K
Parameters of Antoine Equation (*T*/K, *p*/kPa)			
A	6.897 934	6.041 781	6.022 001
B	1493.989	1215.142	1228.051
C	64.708	52.163	48.007
Error: RMSD(*p*)/Pa	8.9	4.8	8.26
Parameters of AEOS Equation			
T'_c/K	456.221	515.17	544.64
P'_c/bar	50.619 94	28.07	37.17
ω'	0.3605	0.3464	0.2538
$\Delta H°$/kJ·mol^{-1}	−20.283 66		
$\Delta S°$/J·mol^{-1}	−81.627		
$\Delta C_p°$/J·mol^{-1}	−28.932		
Error: RMSD(*p*)/Pa	13.5	12.2	9.7
Enthalpy of Vaporization (ΔH/kJ·mol^{-1}) at T = 298.15 K			
calc from own vapor pressure	46.45	33.89	34.03
calorimetric		33.85[17]	33.03[17]
calc from lit. vapor pressure	44.47[4]	33.88[6]	33.96[6]
Daubert and Danner prediction[8]	45.09	39.83	32.84

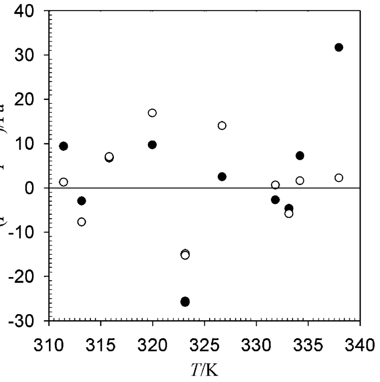

Figure 2. Deviation of allyl alcohol saturation pressure data from correlation with the AEOS (●) and Antoine (○) equations.

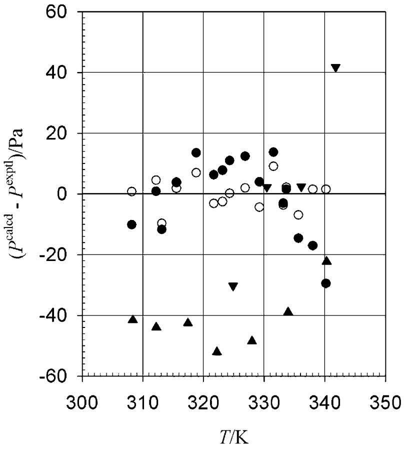

Figure 3. Deviation of benzene saturation pressure data from correlation with the AEOS (●) and Antoine (○) equations. Deviations of Forzati et al.[7] (▲) and Zmaczynski[21] (▼) data from the AEOS correlation.

the number of experimental points. The values obtained for pressure (*P*) are given in Table 2.

Vapor−Liquid Equilibrium Measurements. The vapor−liquid equilibrium measurements were carried out for both systems at isothermal conditions by means of the ebulliometric method described elsewhere.[9]

The pure compound was introduced to the ebulliometer and heated until the boiling point was reached at the adjusted pressure. The readouts of pressure and temperature were taken at the equilibrium stage. The specified temperature was measured with an accuracy of ±0.01 K. Pressure was measured with an accuracy of ±10 Pa. The samples of boiling liquid and vapor condensate were collected for analysis. Next, the precise amount of the second component was added and the steady state was reached at the desired temperature. The measure of steady state was the stability of temperature within ±0.003 K for about 5 min. In general, the time between introducing samples was about 30 min. The whole procedure was repeated until the concentration of the second component exceeded 50%.

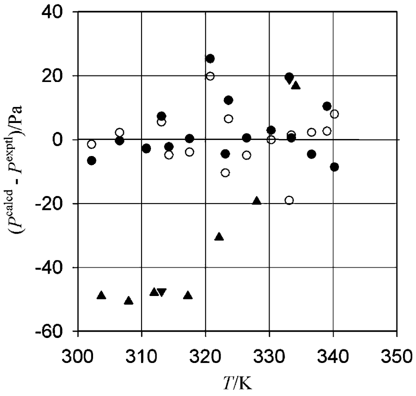

Figure 4. Deviation of cyclohexane saturation pressure data from correlation with the AEOS (●) and Antoine (○) equations. Deviations of Wilingham et al.[6] (▲) and Wu et al.[22] (▼) data from the AEOS correlation.

The sample composition was determined by using an HP 5890 series II gas chromatograph. The HP-FFAP column (poly(ethylene glycol)−TPA modified; 30 m × 0.53 mm × 0.01 μm film thickness), HP 3396 injector, and flame ionization detector (FID) were used for both systems investigated. For each system, a calibration procedure was used. The accuracy of the determination of the liquid phase was ±0.001; that of the vapor phase was ±0.002.

The vapor−liquid measurements were carried out under isothermal conditions at the temperatures 313.15, 323.15, and 333.15 K for both examined systems. The results obtained for the equilibrium pressure (*P*/kPa), temperature (*T*/K), and mole fraction of the first component in the liquid (x_1) and vapor (y_1) phases are listed in Tables 3 and 4 and given in Figures 5 and 6.

Vapor−Liquid Equilibrium Consistency. The consistency check was made for each isotherm with the method proposed by Eubank et al.[18] (Test 1). The auxiliary data

Table 3. Experimental Results of Vapor−Liquid Equilibrium[a]

$T = 313.15$ K			$T = 323.15$ K			$T = 333.15$ K		
x_1	y_1	P/kPa	x_1	y_1	P/kPa	x_1	y_1	P/kPa
			Allyl Alcohol (1) + Benzene (2)					
0.0000	0.0000	24.323	0.0000	0.0000	36.115	0.0000	0.0000	52.151
0.0720	0.1021	26.057	0.0786	0.1142	38.954	0.0710	0.1245	56.552
0.1200	0.1200	26.071	0.1410	0.1410	39.051	0.1620	0.1620	57.000
0.1705	0.1297	26.106	0.1685	0.1466	39.147	0.1695	0.1639	57.097
0.2710	0.1599	25.767	0.2731	0.1724	38.751	0.2695	0.1845	56.746
0.4514	0.1715	24.819	0.4536	0.1953	37.373	0.4532	0.2242	54.802
0.7367	0.2411	20.751	0.5562	0.2146	35.982	0.5511	0.2367	52.868
0.9274	0.4709	13.334	0.7297	0.2605	31.565	0.7292	0.3016	46.722
0.9654	0.6312	10.887	0.9221	0.4505	21.719	0.9227	0.4897	33.510
1.0000	1.0000	7.673	0.9296	0.4852	20.943	0.9325	0.5175	32.181
			0.9645	0.6294	17.896	0.9605	0.6929	28.227
			1.0000	1.0000	13.088	1.0000	1.0000	21.510
			Allyl Alcohol (1) + Cyclohexane (2)					
0.0000	0.0000	24.562	0.0000	0.0000	36.209	0.0000	0.0000	51.925
0.1236	0.2093	28.940	0.1345	0.2337	43.449	0.1211	0.2586	63.271
0.2203	0.2251	29.082	0.2205	0.2464	43.741	0.2200	0.2768	63.958
0.2260	0.2260	29.091	0.2470	0.2470	43.761	0.2790	0.2790	63.981
0.3224	0.2329	29.034	0.3195	0.2599	43.700	0.3202	0.2875	63.973
0.4251	0.2438	28.916	0.4184	0.2681	43.522	0.4143	0.2969	63.790
0.4822	0.2467	28.742	0.4806	0.2728	43.288	0.4821	0.3016	63.439
0.5486	0.2518	28.524	0.5274	0.2757	43.001	0.5303	0.3027	63.105
0.7860	0.2577	26.138	0.7835	0.2844	39.376	0.7795	0.3161	57.959
0.8995	0.3275	20.552	0.8606	0.3034	34.848	0.8586	0.3789	51.282
0.9274	0.4057	17.783	0.8930	0.3592	31.428	0.9299	0.4691	40.717
0.9502	0.4794	15.900	0.9316	0.4351	27.276	0.9440	0.5108	38.530
0.9619	0.5395	13.718	0.9461	0.4933	25.203	0.9602	0.5885	34.380
1.0000	1.0000	7.672	0.9622	0.5610	22.141	1.0000	1.0000	21.510
			1.0000	1.0000	13.112			

[a] x_1 = mole fraction of the first component in the liquid phase; y_1 = mole fraction of the first component in the vapor phase; T = equilibrium temperature; P = equilibrium pressure.

Table 4. Correlation of VLE

	$T = 313.15$ K		$T = 323.15$ K		$T = 333.15$ K		$T = 313.15-333.15$ K	
equation	RMSD(y)	RMSD(P)/Pa	RMSD(y)	RMSD(P)/Pa	RMSD(y)	RMSD(P)/Pa	RMSD(y)	RMSD(P)/Pa
				Allyl Alcohol (1) + Benzene (2)				
NRTL_2	0.0417	396.6	0.0629	664.5	0.0629	664.5		
NRTL_3	0.0389	74.8	0.0610	124.2	0.0592	152.9		
UNIQUAC	0.0449	271.4	0.0659	452.9	0.0633	665.0		
WILSON	0.0360	949.9	0.0636	299.2	0.0615	476.8		
SSF_2	0.0458	323.9	0.0745	690.7	0.1745	12053.6		
SSF_4	0.0390	73.5	0.0693	552.9	0.0561	643.8		
RK_3	0.0395	138.2	0.0610	126.7	0.0593	185.2		
RK_4	0.0391	84.7	0.0610	125.4	0.0591	165.4		
θ = adjusted	0.0284	41.2	0.0381	167.14	0.0368	318.5	0.0351	214.9
$\theta = -0.0033$	0.0249	62.5	0.0407	132.0	0.0391	263.0	0.0361	172.9
				Allyl Alcohol (1) + Cyclohexane (2)				
NRTL_2	0.0128	299.2	0.0195	387.3	0.0202	480.8		
NRTL_3	0.0146	180.5	0.0198	182.0	0.0201	212.8		
UNIQUAC	0.0128	347.7	0.0196	451.7	0.0206	566.5		
WILSON	0.0260	1517.5	0.0217	223.3	0.0220	284.5		
SSF_2	0.0138	458.9	0.0274	771.8	0.0236	879.2		
SSF_4	0.0136	427.0	0.0215	554.0	0.0226	714.8		
RK_3	0.0147	209.3	0.0194	206.1	0.0196	237.8		
RK_4	0.0150	148.7	0.0199	186.6	0.0203	216.5		
θ = adjust	0.0118	67.5	0.0090	220.3	0.0101	402.8	0.0103	266.9
$\theta = a + bT$	0.0117	71.3	0.0094	200.0	0.0100	418.9	0.0104	269.7

were taken from Daubert and Danner.[8] The obtained differences between calculated and experimental liquid-phase compositions do not exceed 0.015, which is still a reasonable value, taking into account that the $P^*(y)$ is in this process a high order polynomial.

Vapor−Liquid Equilibrium Correlation. The results of measurements (each isotherm) have been correlated with the Redlich−Kister and SSF (sum of symmetric functions) equations with two to four adjustable parameters, the NRTL equation with two and three adjustable parameters, and the UNIQUAC and Wilson equations. The exact form of these equations was given previously.[9] The odd parameters of the SSF equation correspond to A_i, while even ones correspond to a_i. The vapor-phase nonideality was taken into account using the Daubert and Danner[8] pure component virial coefficients and liquid density data. The cross virial coefficients and excess volume data were neglected in these calculations. The correlation results are summarized in Table 4. The best results for single isobars were obtained with equations having a larger number of adjustable parameters (Redlich−Kister, SSF). Similar results were obtained with the AEOS equation of state. The

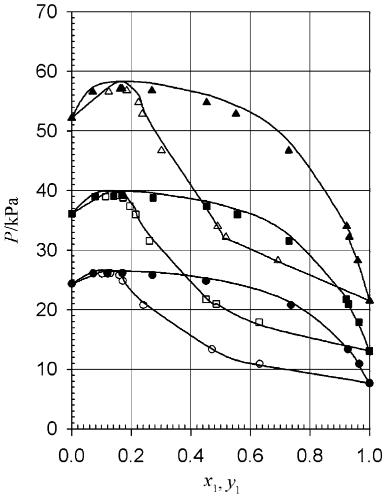

Figure 5. Vapor−liquid equilibrium in the system formed by allyl alcohol with benzene. Experimental data: ●, $T = 313.15$ K; ■, $T = 323.15$ K; ▲, $T = 333.15$ K; bubble points (solid symbols); dew points (hollow symbols). Solid line = correlation and prediction with AEOS equation.

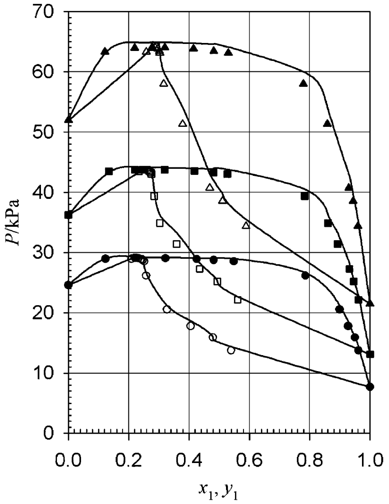

Figure 6. Vapor−liquid equilibrium in the system formed by allyl alcohol with cyclohexane. Experimental data: ●, $T = 313.15$ K; ■, $T = 323.15$ K; ▲, $T = 333.15$ K; bubble points (solid symbols); dew points (hollow symbols). Solid line = correlation and prediction with AEOS equation.

parameters of the AEOS equation obtained from pure component data (Table 2) were used unchanged for mixture calculations. The binary parameters *a, b,* and *c* of the Yu et al.[11] equation ($z^{(\mathrm{ph})}$ term) were calculated using the classical mixing rules,

$$a = \sum_{i=1}^{2}\sum_{i=1}^{2} x_i x_j (1 - \theta_{ij})\sqrt{a_i a_j} \qquad (7)$$

$$b = \sum_{i=1}^{2} x_i b_i \qquad (8)$$

$$c = \sum_{i=1}^{2} x_i c_i \qquad (9)$$

The AEOS equation has only one binary adjustable parameter θ_{12}. The correlation has been carried out in two ways. First, the results for each isotherm were fitted separately. Then, for the system with benzene, the mean value of $\theta_{12} = -0.0033$ was calculated, and for the system with cyclohexane, the temperature dependence was established

$$\theta = -0.08628 + 0.000428T \qquad (10)$$

Table 5. Azeotropic Parameters

$x_1 = y_1$	T_{az}/K	P_{az}/kPa	lit.
	Allyl Alcohol (1) + Benzene (2)		
0.120	313.15	26.07	this work
0.141	323.15	39.05	this work
0.162	333.15	57.00	this work
0.2203	349.9	101.1	Lecat[14]
0.2196	349.95	101.1	Lecat[14]
	Allyl Alcohol (1) + Cyclohexane (2)		
0.225	313.15	29.09	this work
0.247	323.15	43.76	this work
0.279	333.15	63.98	this work
0.2659	347.25	101.1	Lecat[14]

Using y_1 and P values, VLE was calculated for experimental x_1 and P. The results are compared in Table 4 and Figures 5 and 6. The differences between RMSD (eq 6) values obtained for y_1 and P by direct correlation and by prediction with a constant value of θ_{12} (system with benzene) or computed from eq 10 (system with cyclohexane) are negligible. This shows that the AEOS equation of state is able to represent the VLE with almost the same accuracy in the 30 K interval and for individual isotherms.

Azeotropes. The positive azeotropes, first reported by Lecat,[19] were observed. The azeotropic parameters determined by the ebulliometric method[20] are given in Table 5.

Acknowledgment

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