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# Vapor−Liquid Equilibrium of Two Aqueous Systems that Exhibit Liquid−Liquid Phase Separation

**Gonzalo N. Escobedo-Alvarado and Stanley I. Sandler***

Center for Molecular and Engineering Thermodynamics, Department of Chemical Engineering,
University of Delaware, Newark, Delaware 19716

Accurate vapor−liquid equilibrium (VLE) data have been measured for the systems 2-butanol + water at 318.19 K, 338.37 K, and 353.45 K and for 2-butoxyethanol + water at 363.18 K and 371.19 K. The VLE measurements were done with a Stage−Muller dynamic still. The binary data reported satisfy thermodynamic consistency tests. In addition, the liquid−liquid equilibria for the system 2-butoxyethanol + water have also been measured at these temperatures.

## Introduction

Accurate vapor−liquid equilibrium (VLE) data are important in the chemical industry and for the development and testing of thermodynamic models. The systems 2-butanol + water and 2-butoxyethanol + water are particularly interesting, since they exhibit the so-called type VI critical behavior characterized by a closed-loop liquid−liquid solubility gap. Although good liquid−liquid equilibrium data are available in the literature (Moriyoshi et al., 1975; Schneider, 1963) for these systems, there are very few reliable VLE data. All reported measurements in the DECHEMA data series (Gmehling et al., 1977) for the system 2-butanol + water failed to pass the point-to-point consistency test. Using a static apparatus, Fischer and Gmehling (1994) measured the VLE for this mixture at 323 K and Gaube et al. (1987) at 298 K. These authors only provide measured liquid-phase composition data.

Several data sets are available in the literature for the system 2-butoxyethanol + water. Scatchard and Wilson (1963) used the static method to measure VLE from 278.15 to 358.15 K, and the vapor-phase composition was obtained indirectly. Schneider and Wilhem (1959) measured the same system at 383.15, 403.15, and 418.15 K using a dynamic method. These measurements go up to 399.98 kPa and are the highest pressure data set available. Koga (1991) reported saturation pressure data at 298.15 K for this mixture using a static method; so again, no vapor-phase composition data were obtained. Chiavone et al. (1993) used a dynamic still to measure the VLE at 358.15 and 368.15 K, and these data passed the thermodynamic consistency data.

In this study we report the results of our isothermal VLE measurements for binary mixtures of 2-butanol + water at 318.19 K, 338.37 K, and 353.45 K and for 2-butoxyethanol + water at 363.18 K and 371.19 K. These are the first reported measurements using a dynamic still for the system 2-butanol + water.

## Experimental Procedure

***Apparatus.*** The VLE measurements were made using the dynamic method in an all-glass Stage−Muller still in which both the liquid and condensed vapor phases are recirculated and sampled. The still was manufactured by Fischer Labor-und-Verfahrenstechnik of Germany and has been described previously (Eng and Sandler, 1984).

**Table 1. Vapor Pressures of Pure Substances**

| 2-butanol | | 2-butoxyethanol | |
|---|---|---|---|
| *T* (K) | *P* (kPa) | *T* (K) | *P* (kPa) |
| 328.97 | 14.584 | 363.18 | 5.522 |
| 334.96 | 19.856 | 365.07 | 5.919 |
| 334.97 | 19.863 | 371.19 | 7.953 |
| 339.58 | 24.925 | 378.55 | 10.935 |
| 339.60 | 24.932 | 381.53 | 12.370 |
| 339.61 | 24.939 | | |
| 343.60 | 30.237 | | |
| 343.61 | 30.244 | | |
| 346.94 | 35.226 | | |
| 346.95 | 35.232 | | |
| 346.96 | 35.239 | | |
| 349.72 | 39.883 | | |
| 353.45 | 46.950 | | |

Temperature measurements were made with a high-precision platinum resistance thermometer (Rosemount model 162N) accurate to 0.02 K with a resolution of 0.001 K. The platinum resistance thermometer was calibrated by Rosemount in the temperature range −200 to 380 °C at 1.00 °C intervals, and the calibration is directly traceable to the National Institute of Standards and Technology.

Pressure measurements were done with a precision, absolute mercury manometer (Wallace & Tiernan Division, Pennwalt Corp., model FA-187) to an accuracy of 1.1 kPa. This manometer was calibrated using a standard bar, whose calibration is also directly traceable to the National Institute of Standards and Technology. The resolution of the instrument is 0.345 kPa.

***Composition Calibration and Measurement.*** Approximately 20 mixtures of known composition were prepared gravimetrically for each binary system, with a precision of 0.02% in weight. These mixtures were analyzed using a Hewlett-Packard model 5890 gas chromatograph with a model 3390A integrator. A 6 ft × 1/8 in. Chromosorb 102 packed column and a thermal conductivity detector were used. The oven temperature conditions and helium flow rate were set to 170 °C and 40 mL/min for 2-butanol + water and to 240 °C and 50 mL/min for 2-butoxyethanol + water, respectively.

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* To whom correspondence should be addressed.

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**Table 2. Experimental Vapor−Liquid Equilibria Data for the System 2-Butanol (1) + Water (2)**

| $T = 318.19$ K | | | $T = 338.37$ K | | | $T = 353.45$ K | | |
|---|---|---|---|---|---|---|---|---|
| $P$ (kPa) | $x_1$ | $y_1$ | $P$ (kPa) | $x_1$ | $y_1$ | $P$ (kPa) | $x_1$ | $y_1$ |
| 9.660 | 0.0000 | 0.0000 | 25.252 | 0.0000 | 0.0000 | 48.026 | 0.0000 | 0.0000 |
| 10.408 | 0.0026 | 0.0866 | 26.278 | 0.0015 | 0.0395 | 53.634 | 0.0019 | 0.1061 |
| 11.192 | 0.0056 | 0.1506 | 26.426 | 0.0018 | 0.0471 | 55.906 | 0.0028 | 0.1431 |
| 13.081 | 0.0119 | 0.2625 | 27.283 | 0.0029 | 0.0693 | 60.244 | 0.0040 | 0.1889 |
| 13.891 | 0.0146 | 0.3090 | 28.944 | 0.0053 | 0.1313 | 64.948 | 0.0060 | 0.2417 |
| 14.533 | 0.0209 | 0.3420 | 29.668 | 0.0058 | 0.1516 | 71.054 | 0.0138 | 0.3304 |
| 14.701 | 0.0293 | 0.3429 | 31.725 | 0.0117 | 0.2070 | 75.998 | 0.0269 | 0.3718 |
| 14.782 | 0.3439 | 0.3533 | 33.615 | 0.0129 | 0.2598 | 75.047 | 0.0292 | 0.3631 |
| 14.715 | 0.4627 | 0.3713 | 33.548 | 0.0169 | 0.2586 | 76.538 | 0.0387 | 0.3730 |
| 14.708 | 0.5036 | 0.3763 | 35.606 | 0.0204 | 0.2977 | 76.936 | 0.4031 | 0.3917 |
| 14.681 | 0.5245 | 0.3806 | 37.510 | 0.0280 | 0.3303 | 76.714 | 0.4732 | 0.4220 |
| 14.620 | 0.5419 | 0.3844 | 38.522 | 0.0300 | 0.3552 | 76.126 | 0.5360 | 0.4197 |
| 14.414 | 0.6000 | 0.4099 | 40.095 | 0.0406 | 0.3730 | 74.027 | 0.6361 | 0.4635 |
| 14.350 | 0.6164 | 0.4100 | 40.048 | 0.2930 | 0.3742 | 71.834 | 0.7009 | 0.5042 |
| 14.155 | 0.6462 | 0.4217 | 40.055 | 0.3704 | 0.3788 | 69.207 | 0.7555 | 0.5450 |
| 13.784 | 0.6897 | 0.4498 | 40.014 | 0.4503 | 0.3911 | 66.320 | 0.8023 | 0.6149 |
| 13.271 | 0.7450 | 0.4866 | 39.825 | 0.5242 | 0.4085 | 64.259 | 0.8391 | 0.6542 |
| 12.906 | 0.7825 | 0.5207 | 39.440 | 0.5693 | 0.4273 | 63.215 | 0.8597 | 0.6740 |
| 12.440 | 0.8163 | 0.5485 | 38.097 | 0.6685 | 0.4809 | 60.130 | 0.9167 | 0.7296 |
| 12.177 | 0.8392 | 0.5847 | 36.830 | 0.7126 | 0.5016 | 58.505 | 0.9355 | 0.7581 |
| 11.678 | 0.8711 | 0.6170 | 35.424 | 0.7629 | 0.5592 | 56.099 | 0.9545 | 0.8016 |
| 10.975 | 0.9135 | 0.6721 | 33.899 | 0.8116 | 0.5951 | 51.975 | 0.9769 | 0.8830 |
| 10.476 | 0.9400 | 0.7269 | 32.650 | 0.8448 | 0.6221 | 46.950 | 1.0000 | 1.0000 |
| 10.118 | 0.9534 | 0.7571 | 30.233 | 0.9120 | 0.6953 | | | |
| 9.518 | 0.9670 | 0.8378 | 23.450 | 1.0000 | 1.0000 | | | |
| 8.795 | 0.9841 | 0.9139 | | | | | | |
| 8.190 | 1.0000 | 1.0000 | | | | | | |

**Table 3. Experimental Vapor−Liquid Equilibria Data for the System 2-Butoxyethanol (1) + Water (2)**

| $T = 363.18$ K | | | $T = 371.19$ K | | |
|---|---|---|---|---|---|
| $P$ (kPa) | $x_1$ | $y_1$ | $P$ (kPa) | $x_1$ | $y_1$ |
| 70.231 | 0.0000 | 0.0000 | 94.527 | 0.0000 | 0.0000 |
| 70.848 | 0.0024 | 0.0107 | 95.546 | 0.0024 | 0.0128 |
| 71.752 | 0.0058 | 0.0197 | 96.727 | 0.0054 | 0.0247 |
| 72.468 | 0.0113 | 0.0303 | 97.092 | 0.0072 | 0.0281 |
| 72.583 | 0.1874 | 0.0339 | 97.740 | 0.0112 | 0.0330 |
| 72.374 | 0.2202 | 0.0344 | 98.003 | 0.0161 | 0.0335 |
| 71.901 | 0.2585 | 0.0372 | 97.997 | 0.1623 | 0.0331 |
| 70.875 | 0.3358 | 0.0446 | 97.794 | 0.1653 | 0.0334 |
| 69.782 | 0.3708 | 0.0455 | 97.052 | 0.2446 | 0.0336 |
| 66.852 | 0.4586 | 0.0508 | 96.228 | 0.2749 | 0.0347 |
| 64.678 | 0.5042 | 0.0528 | 95.404 | 0.3048 | 0.0374 |
| 62.167 | 0.5429 | 0.0565 | 92.178 | 0.4009 | 0.0410 |
| 58.198 | 0.5960 | 0.0609 | 82.949 | 0.5454 | 0.0573 |
| 54.594 | 0.6349 | 0.0645 | 77.389 | 0.6015 | 0.0645 |
| 49.532 | 0.6846 | 0.0712 | 70.308 | 0.6571 | 0.0748 |
| 44.699 | 0.7217 | 0.0809 | 62.370 | 0.7103 | 0.0899 |
| 39.974 | 0.7556 | 0.0903 | 55.485 | 0.7518 | 0.1072 |
| 30.787 | 0.8274 | 0.1312 | 48.100 | 0.7939 | 0.1326 |
| 25.961 | 0.8642 | 0.1636 | 39.852 | 0.8348 | 0.1688 |
| 21.789 | 0.8916 | 0.2058 | 33.406 | 0.8670 | 0.2093 |
| 16.187 | 0.9323 | 0.3139 | 26.190 | 0.9030 | 0.2754 |
| 13.905 | 0.9501 | 0.3847 | 20.594 | 0.9328 | 0.3599 |
| 9.464 | 0.9757 | 0.5564 | 17.469 | 0.9523 | 0.4336 |
| 5.522 | 1.0000 | 1.0000 | 13.574 | 0.9716 | 0.5698 |
| *a* | 0.1669 | 0.0123 | 11.947 | 0.9777 | 0.6396 |
| | | | 7.953 | 1.0000 | 1.0000 |
| | | | *a* | 0.1107 | 0.0138 |

[a] Liquid−liquid equilibrium measured at atmospheric pressure.

The following expansion was used for the gas chromatograph (GC) calibration:

$$m_1 = A_1 + A_1(1 - A_1)\sum_{j=0}^{n} p_j(2A_1 - 1)^j \qquad (1)$$

where $m_1$ is the mass fraction of component 1, $A_1$ is its area fraction from GC analysis, and $p_j$ are the adjustable parameters. In the case of the system 2-butanol + water we developed two calibration curves: one for the water-rich region and another for the 2-butanol-rich region. In both cases we used $n = 2$ in the equation above. For the system 2-butoxyethanol + water, which covered the whole concentration range, a value of $n = 4$ proved to give a better fit of the experimental data. The compositions determined in this manner are accurate to better than 0.0007 in mole fraction.

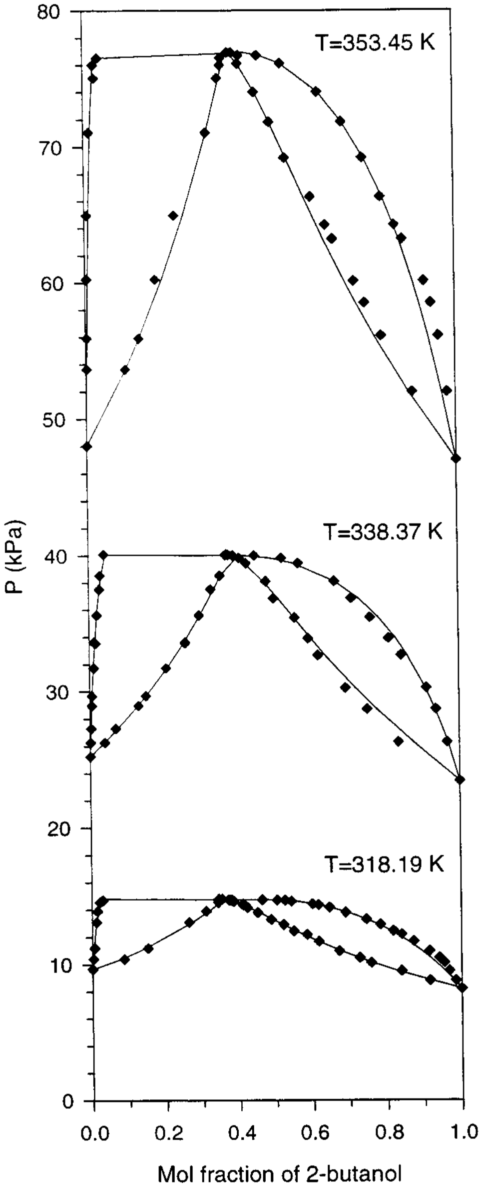

**Figure 1.** VLE of the system 2-butanol + water. The points are the experimental data, and the lines are the results obtained using the PRSV EOS with the WS mixing rule and the UNIQUAC model.

**Table 4. Thermodynamic Consistency Tests**

| | area test | | | point-to-point test | | |
|---|---|---|---|---|---|---|
| system | $T$ (K) | A1 | A2 | % diff | AAD $\Delta y$ | AAD $\Delta P$ (kPa) |
| 2-butanol/water | 318.19 | 0.512 | 0.504 | 0.761 | 0.0080 | 0.07 |
| | 338.37 | 0.494 | 0.49 | 0.396 | 0.0100 | 0.17 |
| | 353.45 | 0.381 | 0.432 | 6.27 | 0.0060 | 0.32 |
| 2-butoxyethanol/ water | 363.18 | 0.397 | 0.379 | 2.345 | 0.0060 | 0.21 |
| | 371.19 | 0.36 | 0.367 | 0.91 | 0.0030 | 0.26 |

***Materials.*** The chemicals used in this study were supplied by Aldrich Chemicals. The guaranteed purity of 2-butanol (anhydrous) was 99.5% and that of 2-butoxyethanol (spectrophotometric grade) was 99.0%+. Doubly

**Table 5. Binary Interaction Parameters Using Method A ($\gamma-\phi$ Method)**

| system | $T$ (K) | UNIQUAC $\Delta U_{12}$ (kJ mol$^{-1}$) | $\Delta U_{21}$ (kJ mol$^{-1}$) | AAD $\Delta P$ (kPa) | AAD $\Delta y$ | NRTL $\alpha_{12}$ | $\Delta g_{12}$ (kJ mol$^{-1}$) | $\Delta g_{21}$ (kJ mol$^{-1}$) | AAD $\Delta P$ (kPa) | AAD $\Delta y$ |
|---|---|---|---|---|---|---|---|---|---|---|
| 2-butanol (1)/ water (2) | 318.19 | 0.7717 | 1.4627 | 0.300 | 0.0220 | 0.2 | −1.6504 | 11.6447 | 0. 380 | 0.0276 |
|  | 338.37 | 1.1323 | 1.0564 | 0.330 | 0.0170 | 0.2 | −1.6900 | 11.4788 | 0.400 | 0.022 |
|  | 353.45 | 0.7368 | 1.6384 | 2.300 | 0.0320 | 0.2 | −1.9347 | 12.7717 | 2.690 | 0.0370 |
| 2-butoxyethanol (1)/ water (2) | 363.18 | 2.1515 | 0.0936 | 0.510 | 0.0090 | 0.15 | −4.8743 | 14.9245 | 0.660 | 0.0141 |
|  | 371.19 | 2.1163 | 0.1225 | 0.980 | 0.0070 | 0.15 | −5.0983 | 15.5581 | 1.000 | 0.0088 |

**Table 6. Binary Interaction Parameters Using Method B ($\phi-\phi$ Method)**

Wong−Sandler Mixing Rule + the UNIQUAC Model

| system | $T$ (K) | $k_{12}$ | $\Delta U_{12}$ (kJ mol$^{-1}$) | $\Delta U_{21}$ (kJ mol$^{-1}$) | AAD $\Delta P$ (kPa) | AAD $\Delta y$ |
|---|---|---|---|---|---|---|
| 2-butanol (1)/water (2) | 318.19 | −0.116 | 2.6319 | 3.5476 | 0.180 | 0.014 |
|  | 338.37 | −0.110 | 3.1779 | 3.1969 | 0.310 | 0.011 |
|  | 353.45 | −0.168 | 3.2434 | 4.1145 | 1.530 | 0.022 |
| 2-butoxyethanol (1)/water (2) | 363.18 | 0.415 | 3.7416 | 0.4860 | 0.560 | 0.007 |
|  | 371.19 | 0.380 | 3.1155 | 0.9092 | 1.050 | 0.007 |

Wong−Sandler Mixing Rule + the NRTL Model

| system | $T$ (K) | $k_{12}$ | $\alpha_{12}$ | $\Delta g_{12}$ (kJ mol$^{-1}$) | $\Delta g_{21}$ (kJ mol$^{-1}$) | AAD $\Delta P$ (kPa) | AAD $\Delta y$ |
|---|---|---|---|---|---|---|---|
| 2-butanol (1)/water (2) | 318.19 | −0.150 | 0.2 | 2.9358 | 17.3063 | 0.140 | 0.013 |
|  | 338.37 | −0.148 | 0.2 | 3.2635 | 17.1316 | 0.300 | 0.009 |
|  | 353.45 | −0.153 | 0.2 | 3.4918 | 18.9895 | 1.530 | 0.020 |
| 2-butoxyethanol (1)/water (2) | 363.18 | 0.020 | 0.15 | −0.0909 | 27.0768 | 0.000 | 0.009 |
|  | 371.19 | 0.001 | 0.15 | −0.1786 | 27.2231 | 0.000 | 0.006 |

distilled water with a conductivity of 18.2 MΩ cm was used. We checked the purity of each organic component first by GC analysis and then by comparing the vapor pressure measured in the Stage−Muller still with data in the literature. Since the area fraction of each chemical was greater than 0.995 and the vapor pressures were within 0.1 kPa of the literature values, the chemicals were used without further purification. The vapor pressures of the pure substances are reported in Table 1.

## Results and Discussion

The results of the VLE measurements are presented in Tables 2 and 3. Both systems are very nonideal, showing azeotropic behavior with liquid−liquid phase separation. In the case of the system 2-butoxyethanol + water it was not possible to go to higher temperatures because of the pressure limitation of the glass still used. Liquid−liquid equilibrium was also measured for this system using previously described equipment (Peschke and Sandler, 1995), and these data are included in Table 3.

***Thermodynamic Consistency Tests.*** The experimental data were tested for thermodynamic consistency using two conventional methods: the integral area consistency test and the point-to-point consistency test proposed by Fredenslund et al. (1977). The integral area compares the areas above and below the $x$-axis in a plot of $\ln(\gamma_1/\gamma_2)$ versus mole fraction. These two areas should be equal for a thermodynamically consistent data set. The integral area test was used by comparing the area differences as a percentage of the total area. These percent differences are shown in Table 4. These areas were obtained by integration of a polynomial fitted to the experimental data. The data satisfy the criterion for a consistent data set [2% error suggested by Prausnitz (1969) and 10% as proposed by Herington (1947)].

The point-to-point consistency test compares the experimental data with the values obtained from the Gibbs−Duhem equation. In this study we followed the procedure proposed by Fredenslund (1977), fitting the excess Gibbs free energy with Legendre polynomials. In general, as shown in Table 4, the consistency of each data set is within the generally accepted criterion of AAD ($\Delta y$) < 0.01 for consistent data.

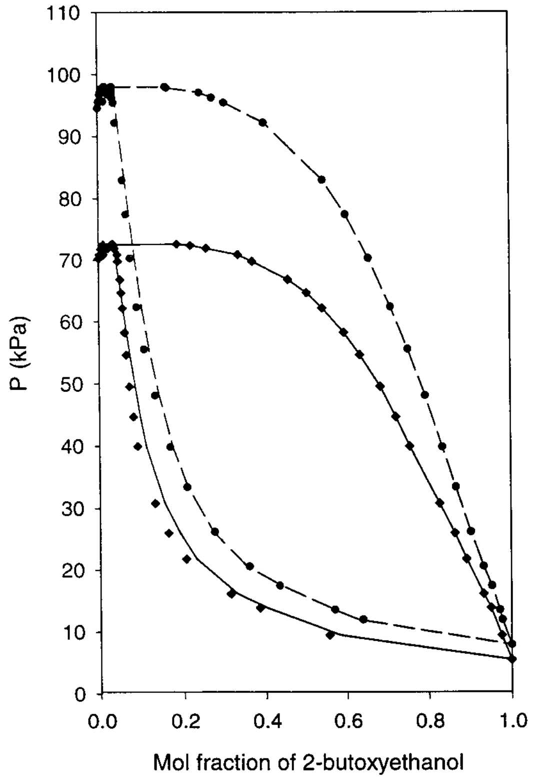

**Figure 2.** VLE of the system 2-butoxyethanol + water. The diamonds are the experimental data at $T$ = 363.18 K, and the circles are the experimental data at $T$ = 371.19 K. The solid and the broken lines are the results obtained using the PRSV EOS with the WS mixing rule and the NRTL model at 363.18 K and 371.19 K, respectively.

***Data Reduction.*** The simplex method has been used to reduce the VLE data. The objective function was

$$F = \frac{1}{N}\sum_{i=1}^{N}(P_i^{\mathrm{exp}} - P_i^{\mathrm{calc}})^2 \tag{2}$$

where $N$ is the number of data points.

Two methods were used.

In method A (the $\gamma-\phi$ method), the vapor phase is described by the Lewis−Randall rule and an activity coefficient model is used for the liquid. The pure component fugacities of the liquid and the vapor states were computed using the Peng−Robinson equation of state as modified by Stryjek and Vera (1986). The phase equilibrium equation in this case is

$$f_i^{V}(T,P)y_i = x_i\gamma_i f_i^{L}(T,P) \tag{3}$$

where $f_i^V$ and $f_i^L$ are the fugacities of pure component $i$ in the vapor and liquid states, respectively.

In method B **(**the $\phi-\phi$ method), the Wong−Sandler (WS) (1992) mixing rule with the Peng−Robinson equation of state as modified by Stryjek and Vera (PRSV) and the UNIQUAC (Abrams and Prausnitz, 1975) and NRTL (Renon and Prausnitz, 1965) models were used for both the vapor and liquid phases. The optimized binary parameters are shown in Tables 5 and 6. The results of the correlations using the WS mixing rule are shown in Figures 1 and 2. The results in the tables show that for these systems, the $\phi-\phi$ method with the WS mixing rule describes these systems better than the $\gamma-\phi$ method.

## Conclusions

We have shown with our results that the Stage−Muller still can be used to measure VLE of the systems 2-butanol + water and 2-butoxyethanol + water. Both these systems show liquid−liquid phase separation and have a large difference between the vapor pressures of the pure components. We have also shown that the PRSV equation of state in combination with the Wong−Sandler mixing rule is a useful model for describing the phase behavior of these systems.

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