# Multiphase Equilibrium Behavior of the Mixture Carbon Dioxide + Ethane + Methanol

## Seung-Pyo Hong, Christi L. Patton, and Kraemer D. Luks*

Department of Chemical Engineering, The University of Tulsa, Tulsa, Oklahoma 74104-3189

The three-phase liquid–liquid–vapor (llg) phase equilibrium behavior of the mixture carbon dioxide + ethane + methanol was experimentally studied. The boundaries of the three-phase llg region in pressure–temperature space are delineated. Compositions and molar volumes of the two liquid phases and the vapor phase along the 288.15 and 298.15 K isotherms are reported.

## Introduction

The solvent gas ethane displays pronounced liquid–liquid–vapor (llg) partial miscibility with the solute methanol (*1–6*). The binary mixture has an llg locus that extends from an upper critical end point (UCEP, or $l-l = g$) determined to be at 303.26 K and 51.38 bar (*5*) (in close agreement with refs 4 and 6) down to a quadruple point (sllg) where crystals of methanol are present, estimated to be in the vicinity of 175.4 K (*3*). Ethane is llg partially miscible with all 1-alkanols up to 1-pentadecanol (*5*); for longer 1-alkanols, the formation of a solid 1-alkanol phase supersedes the occurrence of llg partial miscibility.

The partial miscibility behavior of the homologous series of ethane + 1-alkanol mixtures is complicated by the interplay between 1-alkanol association and chain length. The 1-alkanol solutes can associate through hydrogen bonding, the effect of which apparently diminishes with the chain length of the solute. For example, the substantial hydrogen bonding in methanol (*7*) leads to its sizable llg locus. However, the association that still occurs with the higher 1-alkanols causes them to behave like even longer *n*-paraffins in the presence of ethane. [It is known that ethane is llg partially miscible with the *n*-paraffins from *n*-octadecane to *n*-pentacosane (*8–10*)]. The result is that ethane has a minimum llg locus extent in the homologous series of mixtures for 1-butanol (*4, 5*); the llg loci then increase in extent with 1-alkanol carbon number until sllg points become the lower termini of the loci.

Like ethane, carbon dioxide has no permanent dipole moment. It has a large quadrupole moment, about 6-fold that of ethane. In the system carbon dioxide + methanol, the evidence is that complexes between the two species form (*7*). This solvation effect prevents partial miscibility from occurring for 1-alkanols of carbon number less than 3; llg partial miscibility occurs for the binary mixtures carbon dioxide + 1-propanol to 1-tetradecanol (*11*). llg partial miscibility for higher 1-alkanols is superseded by sllg point formation as in the case of the gas solvent ethane.

There is considerable interest in the use of methanol, ethanol, and other polar molecules as cosolvents in supercritical extraction processes, wherein the cosolvent methanol would enhance the ability of a solvent gas, carbon dioxide, e.g., to solubilize a liquid- or solid-phase species. A relatively modest amount of methanol, on the order of 5 mol % in supercritical carbon dioxide, has been shown to be an effective cosolvent for certain applications. The temperature required for this supercritical extraction process is usually not far above ambient, as the process is most effective near and above the critical point of pure carbon dioxide.

Extraction at subcritical temperatures with the gas solvent carbon dioxide, using an llg configuration, is an alternative process approach. The presence of a cosolvent such as methanol in this system could enhance the extractive capability of the second (carbon dioxide-rich) liquid phase in much the same manner as it would in the supercritical configuration. The second liquid phase ($l_2$) is typically quite dense, at a pressure not too far removed from that of the pure carbon dioxide vapor pressure curve at the same temperature. However, given the above cited miscibility of the species pair carbon dioxide and methanol, such an llg process would not be feasible unless the solute species being contacted were llg partially miscible with carbon dioxide to begin with. Even then, it is possible that the addition of methanol to the solvent-solute system would reduce the llg system to an lg system due to carbon dioxide–methanol solvation. Establishing a means for avoiding this reduction is the motivation for this experimental study. In this study, we map out in some detail in thermodynamic state space the llg region (surface) of the ternary mixture carbon dioxide + ethane + methanol. It is recognized that ethane is already llg partially miscible with methanol. This study will quantitatively address how much gas solvent carbon dioxide, a broadly effective extracting fluid, can be added to the ethane + methanol mixture before one leaves ("loses") the llg surface. The boundaries of the ternary llg surface in thermodynamic phase space will be identified. In addition, it will be determined how the methanol distributes itself between the two liquid phases of the llg surface, with the understanding that the presence of methanol in the $l_2$ phase could enhance solute solubility in that phase.

In a strict sense, the surface being studied here is the domain of a possible llg extraction configuration *without* the species to be extracted being present. Ethane in combination with carbon dioxide and a cosolvent may enable the formation of a subcritical dense fluid phase at a modest pressure. That phase may exhibit a solubility typical of a carbon dioxide–methanol solvent–cosolvent system. This possibility will be discussed further at the end of this paper.

Some limited data, upper and lower critical end points at temperatures above 297 K, on the three-phase system carbon dioxide + ethane + methanol have been reported by Efremova and Shvarts (*12*). Also, some two-phase critical points are reported on the same mixture.

Other ternary llg system studies in the literature that have some thermodynamic similarity to the present carbon dioxide + ethane + methanol llg system study are those of nitrogen + ethane + methanol (*13*) and carbon dioxide + ethane + *n*-tetradecylbenzene (*14*). The first study is a prototype of an absorption process wherein the methanol is the absorbing species. The second study is a prototype of a carbon dioxide

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+ "rich gas" + oil liquid reservoir system; given the low volatility of the n-tetradecylbenzene, which is llg partially miscible with both gas solvents, the location of the llg surface as a pressure–temperature projection mimicked that of the binary lg surface of the carbon dioxide + ethane mixture (*15*).

## Experimental Section

A detailed description of the experimental apparatus is given in an earlier paper by Fall and Luks (*16*). The procedure for performing llg studies was described in another paper by Fall et al. (*17*). These descriptions were updated by Jangkamolkulchai and Luks (*18*). The experimental apparatus employs a stoichiometric approach wherein a known amount of methanol is added to a volumetrically calibrated visual (glass) equilibrium cell. The total volume of the cell is typically 7–9 cm$^3$. The cell vapor space is thoroughly flushed with ethane gas at moderate pressures (less than 15 bar), after which measured amounts of ethane gas are added to the cell from a high-pressure bomb using a manual pump, until a second liquid phase forms. At the temperatures studied, no measurable amount of methanol is removed from the cell by this flushing procedure. Then, starting with a desired proportion of the three fluid phases in this binary system, measured amounts of carbon dioxide are added using a second high-pressure bomb plus manual pump arrangement. The cell contents are brought to equilibrium by a magnetically actuated steel ball stirrer mechanism as each solvent gas is added to the cell.

The manual pumps are precise to ±0.001 cm$^3$; typically, 20 cm$^3$ of gas are added to the cell to create a three-phase system. We estimate the accuracy of gas additions by manual pump to be ±0.02 cm$^3$. The precision of weighing the methanol liquid prior to flushing the cell is ±0.0001 g. Typically, about 1 g of methanol is weighed into the cell for an experimental run.

With the use of mass balances, the compositions and molar volumes of the three phases can be determined from the three "conjugate" measurements at a given temperature and pressure. For a ternary three-phase system, an invariant point requires that both the temperature and the pressure be fixed. Since pressure along an llg isotherm is dictated to some degree by the solvent gas addition process, some smoothing of the laboratory data was required prior to performing the stoichiometric computations. Conjugate measurements in this study involved taking phase volume and overall composition raw data for a set of three experimental runs at the same pressure and temperature; in each run a different phase is caused to be volumetrically dominant relative to the other two phases. Two isotherms, 288.15 and 298.15 K, were studied within the llg region.

The boundaries of the llg region consisting of l = 1 − g and 1 − 1 = g critical end points were determined by visual observation. One boundary of the llg surface is the binary ethane + methanol llg locus. As mentioned in the Introduction, this system was studied earlier by several investigators.

Temperature was measured by a platinum-resistance thermometer to an estimated accuracy of ±0.02 K. Pressure was generally measured to ±0.07 bar with pressure transducers which were frequently calibrated using a dead weight gauge. Pressures could be measured to a precision of ±0.01 bar, if desired, by calibrating the pressure transducers before and after each run with the dead weight gauge. Phase volumes in the calibrated cell were determined by a cathetometer to an accuracy of ±0.005 cm$^3$.

The methanol used was purchased from Aldrich Chemical Co., with a stated purity of 99+ %, with a H$_2$O content of less

**Table 1. Temperature and Pressure Raw Data for the Critical End Point Boundaries for the llg Region of the System Carbon Dioxide + Ethane + Methanol**

| UCEP (l − 1 = g) | | LCEP (l = 1 − g) | |
|---|---|---|---|
| T/K | P/bar | T/K | P/bar |
| 309.26[a] | 51.38 | 280.55 | 36.88 |
| 307.69 | 52.69 | 281.69 | 38.62 |
| 306.44 | 53.55 | 283.55 | 41.40 |
| 305.25 | 54.32 | 284.74 | 43.44 |
| 303.97 | 55.27 | 285.64 | 44.97 |
| 302.75 | 56.11 | 286.92 | 47.40 |
| 301.02 | 57.34 | 288.29 | 49.81 |
| 299.79 | 58.30 | 289.26 | 51.71 |
| 298.15 | 60.32 | 290.19 | 53.13 |
| 297.40 | 61.71 | 292.15 | 56.54 |
| 297.77 | 63.51 | 293.67 | 58.77 |
| 299.17 | 66.78 | 295.38 | 61.41 |
| 299.83 | 67.89 | 296.27 | 62.78 |
| 300.31 | 68.67 | 297.33 | 64.33 |
| 300.97 | 69.60 | 298.21 | 65.62 |
| 301.98 | 71.00 | 299.45 | 67.38 |
| 300.97 | 69.81 | | |

[a] Binary ethane + methanol llg system.

than 0.005%. Chromatographic analysis indicated a purity consistent with the supplier's stated purity. No identifiable impurity was detected, and thus no further purification of the methanol was performed. During this study, the methanol was accessed by syringe through a barrier designed to protect it from hydroscopic contamination.

The carbon dioxide was purchased from Air Products, Inc., as "Coleman Grade", 99.99 mol % pure. It was transferred to an evacuated storage bomb immersed in an ice bath. The vapor phase was vented to remove light impurities, if present. The purity was then verified by liquefying the carbon dioxide at 298.15 K. The vapor pressure at this temperature was within 0.03 bar of that reported by Vargaftik (*19*). The difference between the bubble and dew point pressures was 0.1 bar. The critical temperature and pressure were within 0.06 K and 0.03 bar of the literature values (*20*).

The ethane was obtained from the Matheson Co. as a Research Purity Grade gas with a rating of 99.99 mol % pure. The ethane was loaded and purified like the carbon dioxide. The purity was checked by liquefying it at 298.15 K. The average of the bubble point and dew point pressures was within 0.02 bar of the vapor pressure reported by Goodwin et al. (*21*). The difference between the bubble and dew point pressures was 0.06 bar. The critical temperature and pressure were within 0.07 K and 0.04 bar, respectively, of the literature values (*21*).

The data formulations of Angus et al. (*20*) for carbon dioxide and Goodwin et al. (*21*) for ethane were used to perform the stoichiometric computations.

## Results and Discussion

Table 1 presents pressure and temperature data for the lower critical end point (LCEP, or l = 1 − g) and upper critical end point (UCEP, or l − 1 = g) loci that bound the three-phase llg region of the ternary system carbon dioxide + ethane + methanol. Figure 1 shows these data in pressure–temperature space, along with the ethane + methanol binary llg locus. The critical end point data should be considered accurate to ±0.05 K and ±0.15 bar. These three loci form the upper boundaries of the llg surface for this ternary system. The llg surface narrows in extent as temperature decreases. No attempt was made to study l = 1 − g critical end points below 280 K. The LCEP and UCEP loci come together to form a tricritical point (l = 1 = g), the precise location of which is difficult to estimate. The critical end point data of Efremova and Shvarts are shown in Figure 1, including a tricritical point, denoted "t". Given

**Table 2. Phase Compositions and Molar Volumes for the llg Region of the System Carbon Dioxide + Ethane + Methanol as a function of Pressure at 288.15 K**

| | $l_1$ phase | | | $l_2$ phase | | | g phase | |
|---|---|---|---|---|---|---|---|---|
| $P$/bar | $x(C_2H_6)$ | $x(CO_2)$ | $v$/(cm$^3$/mol) | $x(C_2H_6)$ | $x(CO_2)$ | $v$/(cm$^3$/mol) | $x(C_2H_6)$ | $v$/(cm$^3$/mol) |
| 33.21[a] | 0.3936 | 0.0000 | 51.9 | 0.9056 | 0.0000 | 76.3 | 1.0000[c] | |
| 35.00 | 0.3938 | 0.0259 | 52.4 | 0.8628 | 0.0373 | 76.1 | 0.9586 | 378.7 |
| 36.00 | 0.3930 | 0.0405 | 52.5 | 0.8353 | 0.0638 | 76.0 | 0.9375 | 369.7 |
| 37.00 | 0.3948 | 0.0520 | 52.7 | 0.8175 | 0.0780 | 75.6 | 0.9121 | 361.8 |
| 38.00 | 0.3929 | 0.0692 | 52.9 | 0.7892 | 0.1042 | 74.9 | 0.8893 | 353.2 |
| 39.00 | 0.3941 | 0.0799 | 53.1 | 0.7747 | 0.1162 | 74.8 | 0.8666 | 345.6 |
| 40.00 | 0.3919 | 0.0984 | 53.3 | 0.7469 | 0.1427 | 74.3 | 0.8406 | 337.8 |
| 41.00 | 0.3913 | 0.1093 | 53.6 | 0.7324 | 0.1534 | 74.1 | 0.8124 | 325.7 |
| 42.00 | 0.3883 | 0.1298 | 53.7 | 0.7051 | 0.1777 | 73.6 | 0.7890 | 316.7 |
| 43.00 | 0.3860 | 0.1439 | 53.9 | 0.6852 | 0.1940 | 73.2 | 0.7612 | 306.7 |
| 44.00 | 0.3819 | 0.1657 | 54.2 | 0.6602 | 0.2162 | 72.7 | 0.7341 | 296.4 |
| 45.00 | 0.3779 | 0.1842 | 54.4 | 0.6313 | 0.2453 | 71.9 | 0.7104 | 286.6 |
| 46.00 | 0.3749 | 0.2043 | 54.9 | 0.6047 | 0.2656 | 70.9 | 0.6763 | 278.8 |
| 47.00 | 0.3705 | 0.2284 | 55.1 | 0.5700 | 0.2958 | 70.0 | 0.6453 | 272.2 |
| 48.25 | 0.3676 | 0.2621 | 55.8 | 0.5302 | 0.3211 | 69.0 | 0.5933 | 257.9 |
| 49.64[b] | (0.4190) | (0.3125) | (61.4) | (0.4190) | (0.3215) | (61.4) | 0.538[c] | 243.[c] |

[a] Binary ethane + methanol. [b] LCEP (l = l − g): values in parentheses obtained by extrapolation of mean molar densities. [c] Estimated.

**Table 3. Phase Compositions and Molar Volumes for the llg Region of the System Carbon Dioxide + Ethane + Methanol as a Function of Pressure at 298.15 K**

| | $l_1$ phase | | | $l_2$ phase | | | g phase | |
|---|---|---|---|---|---|---|---|---|
| $P$/bar | $x(C_2H_6)$ | $x(CO_2)$ | $v$/(cm$^3$/mol) | $x(C_2H_6)$ | $x(CO_2)$ | $v$/(cm$^3$/mol) | $x(C_2H_6)$ | $v$/(cm$^3$/mol) |
| 41.06[a] | 0.3705 | 0.0000 | 51.9 | 0.9244 | 0.0000 | 84.3 | 1.0000[c] | |
| 44.00 | 0.3661 | 0.0312 | 51.9 | 0.8713 | 0.0522 | 85.7 | 0.9283 | 263.9 |
| 45.00 | 0.3648 | 0.0411 | 52.0 | 0.8526 | 0.0707 | 85.7 | 0.9074 | 255.0 |
| 46.00 | 0.3615 | 0.0523 | 52.1 | 0.8353 | 0.0884 | 85.8 | 0.8884 | 246.7 |
| 47.00 | 0.3582 | 0.0625 | 52.1 | 0.8194 | 0.1052 | 86.2 | 0.8663 | 237.6 |
| 48.00 | 0.3555 | 0.0731 | 52.2 | 0.8019 | 0.1233 | 86.5 | 0.8450 | 228.8 |
| 49.00 | 0.3494 | 0.0867 | 52.1 | 0.7866 | 0.1396 | 86.9 | 0.8249 | 220.5 |
| 50.00 | 0.3477 | 0.0958 | 52.2 | 0.7702 | 0.1573 | 87.2 | 0.8018 | 212.0 |
| 51.00 | 0.3403 | 0.1098 | 52.1 | 0.7543 | 0.1762 | 87.6 | 0.7816 | 203.7 |
| 52.00 | 0.3373 | 0.1223 | 52.0 | 0.7373 | 0.1957 | 88.0 | 0.7600 | 196.0 |
| 53.00 | 0.3308 | 0.1346 | 52.0 | 0.7193 | 0.2168 | 88.6 | 0.7401 | 187.8 |
| 54.00 | 0.3282 | 0.1471 | 52.0 | 0.6988 | 0.2395 | 89.3 | 0.7232 | 180.9 |
| 55.00 | 0.3214 | 0.1597 | 52.1 | 0.6783 | 0.2630 | 90.0 | 0.7033 | 172.8 |
| 56.00 | 0.3193 | 0.1718 | 52.0 | 0.6575 | 0.2860 | 91.0 | 0.6823 | 165.1 |
| 57.00 | 0.3109 | 0.1835 | 52.1 | 0.6384 | 0.3092 | 92.0 | 0.6602 | 156.7 |
| 60.32[b] | (0.296) | (0.219) | (52.) | | | | | |

[a] Binary ethane + methanol. [b] UCEP (l − l ≈ g): values in parentheses obtained by graphical extrapolation. [c] Estimated.
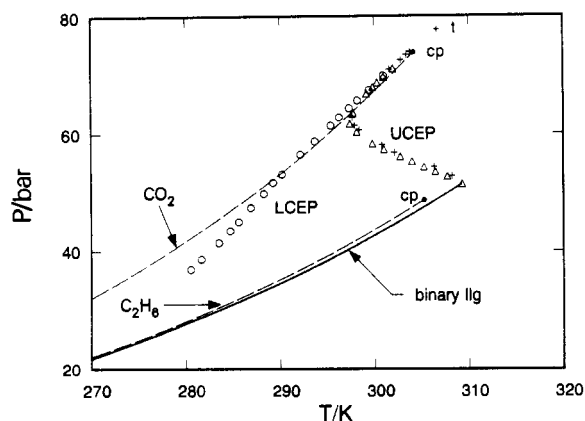


**Figure 1.** Liquid–liquid–vapor region of the system carbon dioxide + ethane + methanol in pressure–temperature space. Dashed curves are the vapor pressure curves of the species carbon dioxide ($CO_2$) and ethane ($C_2H_6$). The curve labeled binary llg is the locus of the binary ethane + methanol llg system. The data labeled + are from ref 12.

that a ternary tricritical point is invariant, we consider this point to be an estimate, albeit reasonable, at best.

The llg isothermal data for the ternary system at 288.15 and 298.15 K are presented in Tables 2 and 3. Figures 2–5 are plots of the ethane compositions and the liquid-phase
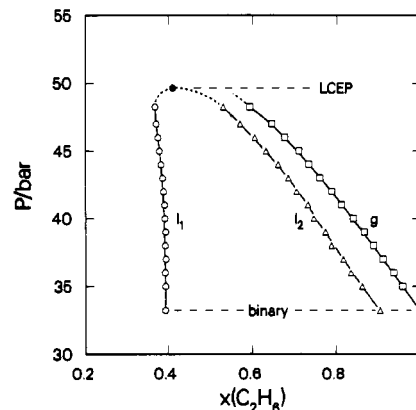


**Figure 2.** Ethane mole fraction $x(C_2H_6)$ as a function of pressure $P$ for the three phases of the llg region of carbon dioxide + ethane + methanol at 288.15 K.

molar volumes as a function of pressure. The ternary mole fractions are estimated to be accurate to ±0.005, and the liquid-phase molar volumes are estimated to be accurate to ±1.0 cm$^3$ g mol$^{-1}$. These are conservative estimates, based primarily on the average absolute deviation (AAD) of the raw data from smoothed curves as well as our experience at being able to generate reproducible data using the stoichiometric technique described herein. Our earlier studies on ethane + 1-alkanol mixtures indicated that the amount of methanol in
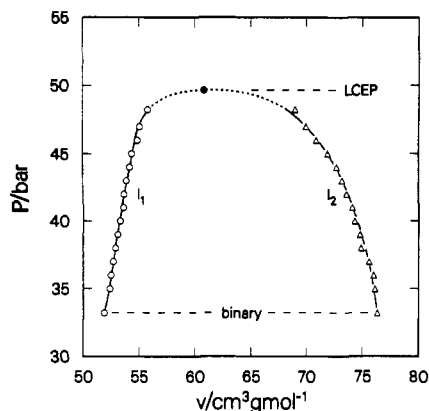
**Figure 3.** Molar volume $v$ as a function of pressure $P$ for the $l_1$ and $l_2$ phases of the llg region of carbon dioxide + ethane + methanol at 288.15 K.
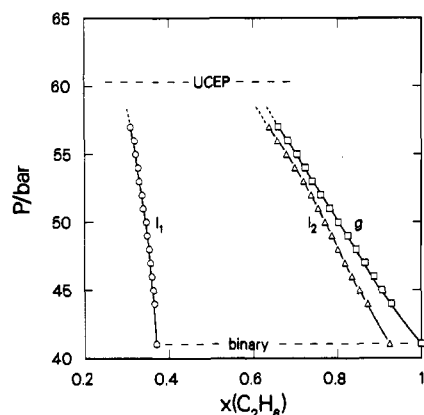


**Figure 4.** Ethane mole fraction $x(C_2H_6)$ as a function of pressure $P$ for the three phases of the llg region of carbon dioxide + ethane + methanol at 298.15 K.



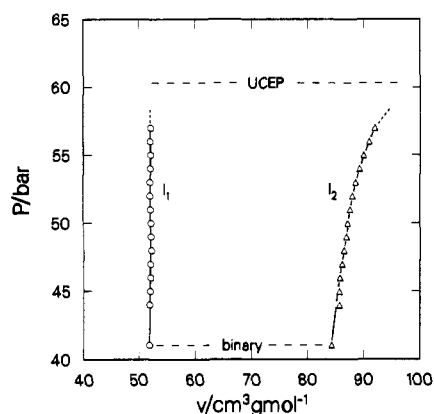**Figure 5.** Molar volume $v$ as a function of pressure $P$ for the $l_1$ and $l_2$ phases of the llg region of carbon dioxide + ethane + methanol + 298.15 K.

the vapor phase was essentially negligible, so we assumed that the vapor phase was solute-free in all stoichiometric calculations. This assumption would be weakest near a l – l = g point. Graphical extrapolation was used to estimate the properties of the g phase at the LCEP at 288.15 K and of the $l_1$ phase at the UCEP at 298.15 K.

Certain estimation techniques were used for identifying the properties at the end points of the isotherms in Tables 2 and 3. The properties of the $l_1$ and $l_2$ phases at the LCEP ($l_1 = l_2$) were calculated using a ternary system variation based on the Cailletet and Mathias method for pure substances (22). Arithmetic averages of the three species molar densities for the two phases were extrapolated as the LCEP was

approached. These averages were well-behaved and provide estimates of the composition and molar volume of the critical phase $l_1 = l_2$. This approach was shown to be as accurate as experimentally determining the properties of the critical phase $l_1 = l_2$ in an earlier study for binary llg systems (5).

The llg surface is eventually terminated by addition of carbon dioxide to the ethane + methanol system. For a solute that is llg partially miscible with both ethane and carbon dioxide, such as n-tetradecylbenzene (14), the llg surface will exhibit a pronounced "fold" or extremum, in this case a maximum in terms of pressure. This phenomenon is attributable to the facts that the mixture carbon dioxide + ethane has a correspondingly pronounced azeotrope (15) and n-tetradecylbenzene is not especially volatile or soluble in the $l_2$ phase. The location of the lg surface in pressure–temperature space for carbon dioxide + ethane is similar to the location of the llg surface for carbon dioxide + ethane + n-tetradecylbenzene. However, for methanol (instead of n-tetradecylbenzene), the llg surface terminates, as shown in Figure 1, before it reaches the expected extremum in pressure.

The llg data in Tables 2 and 3 indicate that about 42 mol % carbon dioxide on a methanol-free basis is required to achieve critical end point termination of the llg region. The addition of carbon dioxide to the $l_1$ phase appears to act primarily as a diluent. It is accompanied by modest decreases in the mole fractions in both ethane and methanol.

With respect to the capacity of the $l_2$ phase for methanol, which would be important for enhancing the ability of that phase to solubilize solutes that may be present in the $l_1$, or original mixture, phase, the concentrations are fairly constant with pressure (7.5 mol % at 298.15 K and 9–12 mol % at 288.15). The constancy is probably attributable to the pressure variations along these isotherms being less than 20 bar. It is noteworthy that the methanol levels in the $l_2$ phase are substantial enough to make that phase a good candidate for (subcritical) extraction. The ultimate utility of this configuration for llg extraction would of course be dictated by the nature of the solute(s) being extracted. Future work by our group will address such applications.

## Glossary

| | |
|---|---|
| AAD | average absolute deviation |
| g | vapor phase |
| $l,l_1,l_2$ | liquid phases |
| LCEP | lower critical end point (l = l – g) |
| P | pressure |
| s | solid phase |
| T | temperature |
| UCEP | upper critical end point (l – l = g) |
| $v$ | molar volume |
| $x$ | mole fraction |

## Literature Cited

(1) Kuenen, J. P.; Robson, W. G. *Philos. Mag. Ser. 5* **1899**, *148*, 180.
(2) Kuenen, J. P.; *Philos. Mag. Ser. 6* **1903**, *6*, 637.
(3) Ma, Y. H.; Kohn, J. P. *J. Chem. Eng. Data* **1964**, *9*, 3.
(4) Brunner, E. *J. Chem. Thermodyn.* **1985**, *17*, 871.
(5) Lam, D. H.; Jangkamolkulchai, A.; Luks, K. D. *Fluid Phase Equilib.* **1990**, *59*, 263.
(6) Lam, D. H.; Luks, K. D. *J. Chem. Eng. Data* **1991**, *36*, 307.
(7) Fulton, J. L.; Yee, G. G.; Smith, R. D. *J. Am. Chem. Soc.* **1991**, *113*, 8327.
(8) Specovius, J.; Leiva, M. A.; Scott, L.; Knobler, C. M. *J. Phys. Chem.* **1981**, *85*, 2313.
(9) Peters, C. J.; Lichtenthaler, R. N.; de Swaan Arons, J. *Fluid Phase Equilib.* **1986**, *29*, 495.
(10) Estrera, S. S.; Luks, K. D. *J. Chem. Eng. Data* **1987**, *32*, 201.
(11) Lam, D. H.; Jangkamolkulchai, A.; Luks, K. D. *Fluid Phase Equilib.* **1990**, *60*, 131.

(12) Efremova, G. D.; Shvarts, A. V. *Russ. J. Phys. Chem.* **1972**, *46* (2), 237.
(13) Zeck, S.; Knapp, H. *Fluid Phase Equilib.* **1986**, *25*, 303.
(14) Miller, M. M.; Luks, K. D. *Fluid Phase Equilib.* **1989**, *51*, 315.
(15) Fredenslund, A.; Mollerup, J. *J. Chem. Soc., Faraday Trans. 1* **1973**, *70*, 1653.
(16) Fall, D. J.; Luks, K. D. *J. Chem. Eng. Data* **1984**, *29*, 413.
(17) Fall, D. J.; Fall, J. L.; Luks, K. D. *J. Chem. Eng. Data* **1985**, *30*, 82.
(18) Jangkamolkulchai, A.; Luks, K. D. *J. Chem. Eng. Data* **1989**, *34*, 92.
(19) Vargaftik, N. B. *Tables on the Thermophysical Properties of Liquids and Gases*, 2nd ed.; John Wiley and Sons: New York, 1975; pp 167–168.
(20) Angus, S.; Armstrong, B.; de Reuck, K. M. *Thermodynamic Tables of the Fluid State Carbon Dioxide*; Pergamon Press: Oxford, 1976.
(21) Goodwin, R. D.; Roder, H. M.; Straty, G. C. NBS Note No. 684, 1976.
(22) Maron, S. H.; Prutton, C. F. *Principles of Physical Chemistry*; Macmillan: London, 1958; pp 84–85.