Reliable Prediction of Phase Stability Using an Interval Newton Method

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A key step in phase equilibrium calculations is determining if, in fact, multiple phases are present. Reliably solving the phase stability and, ultimately the phase equilibrium problem, is a significant challenge for high pressure vapor/liquid, liquid/liquid and vapor/liquid/liquid equilibrium. We present the first general-purpose computational method, applicable to any arbitrary equation of state or activity coefficient model, that can mathematically guarantee a correct solution to the phase stability problem. In this paper, we demonstrate the use of this new method, which uses techniques from interval mathematics, for the van der Waals equation of state to determine liquid/liquid and liquid/vapor phase stability for a variety of representative systems. Specifically, we describe and test interval methods for phase stability computations for binary mixtures that exhibit Type I and Type II behavior, as well as for a relatively simple ternary mixture. This shows that interval techniques can find with absolute certainty all stationary points, and thus solve the phase stability problem with complete reliability.

1. INTRODUCTION

The determination of phase stability, i.e., whether or not a given phase will split into multiple phases, is a key step in phase equilibrium calculations. This is especially true for high pressure phase equilibria where even simple binary solutions can exhibit complex phase behavior. The phase stability problem is frequently formulated in terms of the tangent plane condition [1]. Minima in the tangent plane distance are sought, usually by solving a system of nonlinear equations for the stationary points [2]. If any of these yield a negative tangent plane distance, indicating that the tangent plane intersects (or lies above) the Gibbs energy of mixing surface, the phase is unstable. The difficulty lies in that, in general, given any arbitrary equation of state (EOS) or activity coefficient (G^E) model, most computational methods cannot find with complete certainty all the stationary points, and thus no guarantee of stability can be provided.

What is needed are robust techniques that can find *all* solutions to a system of nonlinear equations, and do so *with certainty*, or techniques that can find the *global* optimum of a function, and do so again *with certainty*. We first reported the implementation of such a technique, using interval mathematics, for the solution of the phase stability problem for the Margules equation and the NRTL equation [3]. Here we extend this technique to systems modeled with equations of state, using the van der Waals (VDW) equation as an example.

Interval mathematics involves computation with intervals as opposed to real numbers. Interval Newton methods, when combined with a generalized bisection approach, provide the power to find with confidence all solutions of a system of nonlinear equations [5,6], and to find with total reliability the global minimum of a nonlinear objective function [7], provided only that upper and lower bounds are available for all variables. Efficient techniques for implementing interval Newton/generalized bisection are a relatively recent development, and thus such methods have not yet been widely applied. Schnepper and Stadtherr [8] suggested the use of this method for solving chemical process modeling problems, and recently described an implementation [9].

For the phase stability problem various approaches have been proposed recently. For example, Sun and Seider [10] apply a homotopy-continuation method, which will often find all stationary points. Although it is easier to initialize than Michelsen's method [2], it may still be initialization dependent and provides no theoretical guarantee that all solutions have been found. The "area" method of Eubank et al. [11], which is based on exhaustive search over a grid, can also be very reliable. They suggest [11] that a course grid be used first to find the approximate location of solutions. Then, regions appearing not to contain a solution are arbitrarily eliminated from consideration and the search continues with a finer grid in the remaining regions. However, there is no mathematical guarantee provided that the regions eliminated do not contain solutions. McDonald and Floudas [12] show that for certain activity coefficient models, the problem can be reformulated to make it amenable to solution by global optimization techniques, which do mathematically guarantee that the correct answer is found. However, in general there appears to remain a need for an efficient *general-purpose* method that can perform phase stability calculations with *mathematical certainty* for any arbitrary EOS or activity coefficient model.

In this paper, we demonstrate the use of interval methods for phase stability computations of binary and ternary VDW mixtures that exhibit Type I and Type II behavior [4]. These methods can be applied in connection with any EOS or activity coefficient model, and when properly implemented are completely reliable.

2. PHASE STABILITY ANALYSIS

The determination of phase stability is often done using tangent plane analysis [1,2]. A phase at specified T, P, and feed mole fractions $\mathbf{z}$ is unstable if the Gibbs energy of mixing versus composition surface $m = \Delta g^{\mathrm{M}}/RT$ ever falls below a plane tangent to the surface at $\mathbf{z}$. That is, if the tangent plane distance

$$D = m - m_0 - \sum_{i=1}^{c} \left(\frac{\partial m}{\partial x_i}\right)_0 (x_i - z_i) \tag{1}$$

is negative for any composition $\mathbf{x}$, the phase is unstable. The subscript zero indicates evaluation at $\mathbf{x} = \mathbf{z}$, and c is the number of components. A common approach for determining if D is ever negative is to minimize D subject to the mole fractions summing to one. It is readily shown that the stationary points in this optimization problem can be found by solving the system of nonlinear equations:

$$\left(\frac{\partial m}{\partial x_i}\right) - \left(\frac{\partial m}{\partial x_i}\right)_0 - \left(\frac{\partial m}{\partial x_c}\right) + \left(\frac{\partial m}{\partial x_c}\right)_0 = 0, \quad i = 1, 2, ..., c\text{-}1 \tag{2}$$

$$1 - \sum_{i=1}^{c} x_i = 0 \tag{3}$$

If an equation of state model for m is used, so that m is expressed in terms of $\mathbf{x}$ and v, the molar volume of the mixture, then (2) and (3) must be solved simultaneously with the equation of state, which for the problems considered here is the van der Waals (VDW) equation:

$$P = \frac{RT}{v - b} - \frac{a}{v^2} \tag{4}$$

The standard mixing rules, $b = \sum x_i b_i$ and $a = \sum\sum x_i x_j a_{ij}$, are used.

The $(c+1) \times (c+1)$ system given by equations (2) - (4) above has a trivial root at $\mathbf{x} = \mathbf{z}$ and $v = v_0$ and frequently has multiple nontrivial roots as well. Thus conventional equation solving techniques such as Newton's method may fail by converging to the trivial root or give an incorrect answer to the phase stability problem by converging to a stationary point that is not the global minimum of D. This is aptly demonstrated by the experiments of Green *et al.* [13], who show that the pattern of convergence from different initial guesses demonstrates a complex fractal-like response for even the simple ternary mixture used as an example below. We demonstrate here the use of an interval Newton/generalized bisection (IN/GB) method for solving the system (2) - (4). The method requires no initial guess, and will find *with certainty* all the stationary points of D.

3. INTERVAL COMPUTATIONS

The interval Newton/generalized bisection (IN/GB) method involves computations on intervals rather than real numbers. A real *interval*, X, is defined as a set of real numbers lying between (and including) upper and lower bounds; i.e., $X = [a,b] = \{x \in \Re \mid a \leq x \leq b\}$, where $a,b \in \Re$ and $a \leq b$. A real interval vector $\mathbf{X} = (X_i) = (X_1,X_2,...,X_n)^{\mathrm{T}}$ has n real interval components X_i, and since it can be interpreted geometrically as an n-dimensional rectangle, is frequently referred to as a *box*. Note that in this section lower case quantities are real numbers and upper case quantities are intervals. Several good introductions to computation with intervals are available, including recent monographs by Neumaier [5] and Hansen [7].

Of particular interest here are interval Newton methods. Consider the solution of the system of real nonlinear equations $\mathbf{f}(\mathbf{x}) = \mathbf{0}$, where it is desired to find all solutions in an specified initial interval $\mathbf{X}^{(0)}$. The basic iteration step in interval Newton methods is, given an interval $\mathbf{X}^{(k)}$, to solve the linear interval equation system

$$\mathrm{F}'(\mathbf{X}^{(k)})(\mathbf{N}^{(k)} - \mathbf{x}^{(k)}) = -\mathbf{f}(\mathbf{x}^{(k)}) \tag{5}$$

for a new interval $\mathbf{N}^{(k)}$, where k is an iteration counter, $\mathrm{F}'(\mathbf{X}^{(k)})$ is an interval extension of the real Jacobian $\mathrm{f}'(\mathbf{x})$ of $\mathbf{f}(\mathbf{x})$ over the current interval $\mathbf{X}^{(k)}$, and $\mathbf{x}^{(k)}$ is a point in the interior of $\mathbf{X}^{(k)}$, usually taken to be the midpoint. The interval extension $\mathrm{F}'(\mathbf{X}^{(k)})$ of the Jacobian is determined by substituting the interval $\mathbf{X}^{(k)}$ for $\mathbf{x}$ in the expression $\mathrm{f}'(\mathbf{x})$ for the real Jacobian, and performing interval operations in place of real operations. It can be shown [14] that any root $\mathbf{x}^*$ of the set of equations that is within the current interval, i.e. $\mathbf{x}^* \in \mathbf{X}^{(k)}$, is also contained in the newly computed interval $\mathbf{N}^{(k)}$. This suggests that the next iteration for $\mathbf{X}$ should be the intersection of $\mathbf{X}^{(k)}$ with $\mathbf{N}^{(k)}$, i.e. $\mathbf{X}^{(k+1)} = \mathbf{X}^{(k)} \cap \mathbf{N}^{(k)}$. There are various interval Newton methods, which differ in how they determine $\mathbf{N}^{(k)}$ from equation (5) and thus in the tightness with which $\mathbf{N}^{(k)}$ encloses the solution set of (5).

While the iteration scheme discussed above can be used to tightly enclose a solution, what is of most significance here is the power of (5) to provide a test of whether a solution exists within a given interval and whether it is a unique solution. For several techniques for finding $\mathbf{N}^{(k)}$ from (5), it can be proven (e.g., [5]) that if $\mathbf{N}^{(k)}$ is totally contained within $\mathbf{X}^{(k)}$, i.e. $\mathbf{N}^{(k)} \subset \mathbf{X}^{(k)}$, then there is a *unique* zero of the set of nonlinear equations $\mathbf{f}(\mathbf{x}) = \mathbf{0}$ in $\mathbf{X}^{(k)}$, and furthermore that Newton's method with real arithmetic *will converge* to that solution starting from *any* point in $\mathbf{X}^{(k)}$. Thus, if $\mathbf{N}^{(k)}$ is determined using one of these techniques, the computation can be used as a root inclusion test for any interval $\mathbf{X}^{(k)}$: 1) If $\mathbf{X}^{(k)}$ and $\mathbf{N}^{(k)}$ do not intersect, i.e., $\mathbf{X}^{(k)} \cap \mathbf{N}^{(k)} = \emptyset$, then there is no root in $\mathbf{X}^{(k)}$. 2) If $\mathbf{N}^{(k)}$ is totally contained in $\mathbf{X}^{(k)}$, then there is exactly one root in $\mathbf{X}^{(k)}$ and Newton's method with real arithmetic will find it. Otherwise, 3) no conclusion can be drawn. In the last case, one could then repeat the root inclusion test on the next interval Newton iterate $\mathbf{X}^{(k+1)}$, assuming it is sufficiently smaller than $\mathbf{X}^{(k)}$, or one could bisect $\mathbf{X}^{(k+1)}$ and repeat the root inclusion test on the resulting intervals. This is the basic idea of IN/GB methods. If $\mathbf{f}(\mathbf{x}) = \mathbf{0}$ has a finite number of real solutions in the specified initial box, a properly implemented IN/GB method can find *with mathematical certainty* any and all solutions to a specified tolerance, or can determine *with mathematical certainty* that there are no solutions in the given box [6,15]. The technique used here for computing $\mathbf{N}^{(k)}$ from (5) is the preconditioned Gauss-Seidel-like technique developed by Hansen and Sengupta [16]. A detailed step-by-step description of the IN/GB algorithm used here is given by Schnepper and Stadtherr [9].

To solve the set of equations (2) - (4) described above for the VDW phase stability problem involves c+1 variables, the c component mole fractions and the molar volume. For the mole fractions, the initial box $\mathbf{X}^{(0)} = [\mathbf{0},\mathbf{1}]$ is suitable. In practice the initial lower bound is set to an arbitrarily small positive number ε (10^{-10} was used) to avoid taking the logarithm of zero in subsequent calculations. This can be done without the loss of reliability providing a sufficiently small value of ε is used. The lower limit on the molar volume was taken to be the smallest pure component VDW size parameter b, and the upper bound was taken to be the ideal gas molar volume for the T and P under investigation. Although it is possible to have compressibility factors greater than one at very high P_{r}, this was deemed satisfactory for the range of T_{r} and P_{r} explored below. Our implementation of the IN/GB method for the phase stability problem is based on appropriately modified routines from the packages INTBIS [6] and INTLIB [17].

4. RESULTS

Phase stability calculations, as described above, were performed for three different mixtures using the VDW equation and the standard mixing rules. For the binary systems the pure component parameters were calculated from critical properties, $b = RT_c/8P_c$ and $a = 27R^2T_c^2/64P_c$. The pure component parameters for the ternary mixture and unlike-pair interaction parameters for both binary and ternary mixtures are described below.

The first test case is for a binary mixture that exhibits Type I behavior according to the classification of van Konynenburg and Scott [4]. The second test case is a binary mixture that exhibits Type II behavior. The third test case is the ternary mixture studied by Green et al. [13]. These systems were chosen to be representative tests of the implementation of the IN/GB method for solution of the phase stability problem using equations of state. Included are systems that exhibit vapor/liquid (VLE) , liquid/liquid (LLE), and vapor/liquid/liquid (VLLE) equilibria and that span regions in which three real volume roots to the VDW equation are obtained.

4.1 Binary Mixture Type I

The first test case is a binary mixture that exhibits Type I behavior [4]. Type I systems only show VLE and have a continuous critical line extending from the critical point of pure component 1 to pure component 2. The *PT* projection of this system is shown in Figure 1. In the computations component 1 was arbitrarily chosen to be carbon dioxide (T_c=304.2 K, P_c=73.8 bar). The properties of component 2 were chosen to replicate the example used by van Konynenburg and Scott for a Type I mixture (this corresponds to their values of the dimensionless parameters ξ=0, ζ=0.5 and Λ=-0.05). The resulting size and energy parameters are b_1=b_2=42.8374 cm^3/mol, a_{11}=3.6565 x 10^6 (cm^6 bar)/mol^2, a_{22}=1.097 x 10^7 (cm^6 bar)/mol^2 and a_{12}=7.6792 x 10^6 (cm^6 bar)/mol^2. The system has a vapor/liquid two-phase envelope over a wide range of *T* and *P* but above P_r=3.23 only one phase exists at any T and *x*. The *T*-x_2 diagram for this system at P_r=1.00 is shown in Figure 2 [4]. In Figure 2 the *T* and *P* are reduced with T_c and P_c of component 1 (CO$_2$).

We performed phase stability calculations for three different feed compositions at P_r=1.00 and one feed composition at P_r=3.24, just above the highest pressure at which two phases can exist. The results are shown in Table 1. The newly implemented IN/GB method correctly predicted a feed at T_r=1.5, P_r=1.0, z_1=0.6 to be unstable; i.e., that it will split into two phases, as indicated by the negative value of the tangent plane distance D. The other three feed points were correctly shown to be stable. Also included in Table 1 are the roots found that correspond to stationary points in the tangent plane distance function, the number of root inclusion tests

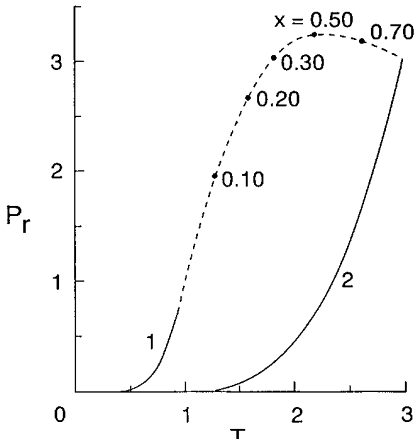

Figure 1 *PT* projection of Type I binary system

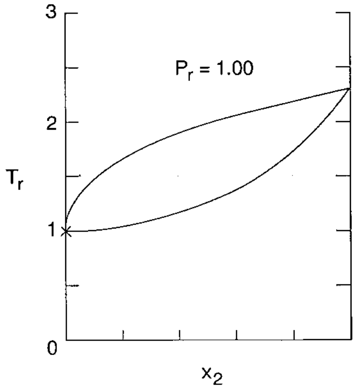

Figure 2 *T*-*x* diagram of Type I binary at P_r=1.00

performed in the computation, the level reached in the binary tree (number of bisections that were required on the longest branch in the bisection scheme) and the total CPU time on an HP 9000/735 workstation, which by today's standards can be considered a moderately fast machine. The last three items give an indication of the performance of the algorithm. We would expect standard approaches (e.g., [2]) to the phase stability problem to be faster, but these methods do not reliably solve the problem in all cases. Thus, as one might expect, to obtain guaranteed reliability some premium must be paid in terms of computation time. Nonetheless, the CPU times reported here compare very well with other reliable methods. For instance, on one liquid/liquid phase stability problem using the NRTL equation, an implementation [3] of the IN/GB method required 0.06 s on an HP 9000/735, while the C implementation of McDonald and Floudas' model-specific technique [12] required 0.11 s on an HP 9000/730. (Note that, based on the well-known LINPACK-100 benchmark, the Model 735 used here is about 70% faster than the Model 730). In addition to being very efficient for a mathematically guaranteed method, the technique used here has the advantage of being model-independent; it can be easily used to solve the phase stability problem for any excess Gibbs free energy or equation of state model.

4.2 Binary Mixture Type II

The second test case is a binary Type II system, which has VLLE at relatively low T and P and, like Type I, has a continuous critical line connecting the two pure component critical points. The PT projection of this system is shown in Figure 3. Component 1 was chosen to be CO_2 and the properties of component 2 were chosen to replicate the example used [4] for a Type II mixture (this corresponds to their values of the dimensionless parameters ξ=0, ζ=0.473 and Λ=0.105). The resulting size and energy parameters are $b_1=b_2=$ 42.8374 cm^3/mol, $a_{11}=3.6565 \times 10^6$ (cm^6 bar)/mol^2, $a_{22}=1.022 \times 10^7$ (cm^6 bar)/mol^2 and $a_{12}=6.20981 \times 10^6$ (cm^6 bar)/mol^2. The T-x_2 diagram for this system at P_r=0.4 is shown in Figure 4, where the T and P are reduced with T_c and P_c of CO_2. At P_r=0.4 the system exhibits both VLE and LLE with three-phase VLLE existing at a T somewhat below T_r=1, as shown in the figure. At higher P_r=1.5, the VLE and LLE phase envelopes separate since this is above the upper critical endpoint and VLLE can no longer exist.

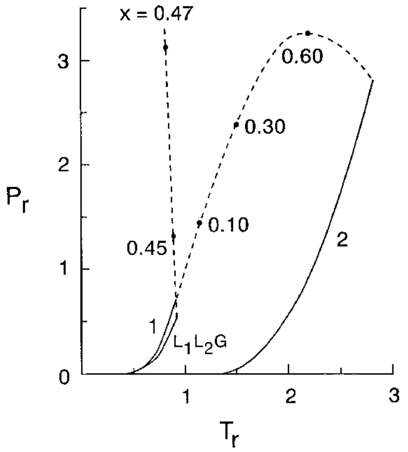

Figure 3 PT projection of Type II binary system

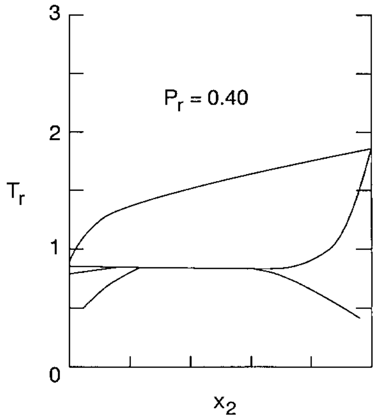

Figure 4 T-x diagram of Type II binary at P_r=0.4

We performed phase stability calculations for five different feed compositions at P_r=0.4 and seven feed compositions at P_r=1.5 and the results are shown in Table 1. In all cases our computations correctly predict whether or not the feed will split into multiple phases. At P_r=0.4 (Figure 4) and T_r=1.5 we have chosen one composition inside and one outside the VLE envelope. At the same P_r and T_r=0.7, which is not far below the VLLE line, we have chosen compositions contained within and those outside the LLE region. At P_r=1.5 we have examined three temperatures, which sample the VLE envelope, the LLE envelope and the single phase region. Also listed in Table 1 is the performance data, indicating reasonable solutions times to these problems.

Table 1
Binary Type I and Type II mixtures: roots found and performance

Feed (P_r,T_r,z_1,z_2)	Roots (x_1,x_2,v)	D	root inclusion tests	level reached in binary tree	CPU time: HP 9000/ 735 (sec)
Type I System:					
1.0, 1.5, 0.6, 0.4	0.6,0.4,279.5	0.0	1355	33	0.36
	0.5987,0.4013,277.2	5.513x10^{-8}			
	0.0963,0.9037,52.92	-0.8780			
1.0, 1.5, 0.2, 0.8	0.2.0.8.54.14	0.0	727	17	0.19
	0.8101,0.1899,401.3	0.3836			
	0.6577,0.3423,152.1	0.4720			
1.0, 1.5, 0.95, 0.05	0.95,0.05,445.3	0.0	1632	24	0.36
3.24, 2.0, 0.4, 0.6	0.4,0.6,67.76	0.0	707	18	0.18
Type II System:					
0.4, 1.5, 0.8, 0.2	0.8,0.2,119.9	0.0	702	20	0.19
	0.5019,0.4981,115.0	0.7884			
	0.0990,0.9010,54.79	0.4809			
0.4, 1.5, 0.2, 0.8	0.2,0.8,106.3	0.0	223	19	0.06
	0.1135,0.8865,237.5	1.643x10^{-5}			
	0.0057,0.9943,53.37	-0.7907			
0.4, 0.7, 0.97, 0.03	0.97,0.03,58.12	0.0	723	19	0.16
0.4, 0.7, 0.5, 0.5	0.5,0.5,49.32	0.0	1997	26	0.44
	0.8589,0.1411,54.57	-0.0280			
	0.2233,0.7767,47.50	-0.0121			
0.4, 0.7, 0.1, 0.9	0.1,0.9,46.95	0.0	934	23	0.21
1.5, 1.5, 0.5, 0.5	0.5,0.5,66.76	0.0	3250	28	0.76
	0.8856,0.1144,263.1	-0.1399			
	0.5521,0.4479,70.94	1.270x10^{-4}			
1.5, 1.5,0.2, 0.8	0.2,0.8,56.05	0.0	1190	19	0.31
	0.8181,0.1819,244.2	0.1251			
	0.6719,0.3281,114.5	0.1665			
1.5, 1.5, 0.9, 0.1	0.9,0.1,266.6	0.0	1296	18	0.30
1.5, 1.0, 0.6, 0.4	0.6,0.4,55.08	0.0	2254	17	0.49
1.5, 0.6, 0.97, 0.03	0.97,0.03,53.28	0.0	654	17	0.14
1.5, 0.6, 0.5, 0.5	0.5,0.5,47.95	0.0	1267	23	0.29
	0.8965,0.1035,51.93	-0.0589			
	0.1414,0.8586,46.32	-0.0404			
1.5, 0.6, 0.1, 0.9	0.1,0.9,46.19	0.0	1393	23	0.32
	0.8303,0.1697,50.96	0.1243			
	0.6744,0.3256,49.25	0.1286			

4.3 Ternary Mixture Type I

The third test case was chosen to replicate the example used by Green et al. [13] to demonstrate the wide regions of initial guesses for which the standard Newton's method will not converge, or converges to trivial or non-physical solutions. It is a ternary mixture with b_{11}=b_{22}=b_{33}=61.5375 cm^3/mol, a_{11}=5.11 x 10^6 (cm^6 atm)/mol^2, a_{22}=1.19 x 10^7 (cm^6 atm)/mol^2, a_{33}=8.52 x 10^6 (cm^6 atm)/mol^2, a_{12}=2a_{11}, a_{13}=(a_{11} a_{33})$^{1/2}$, and a_{23}=(a_{22} a_{33})$^{1/2}$. A binary mixture of components 1 and 2 would exhibit Type I behavior. Species 3 has a molecular description between that of components 1 and 2. At 400K and 80 atm the ternary mixture has a small two phase region, as shown in Figure 5a. At 400K and 60 atm the two phase region has enlarged, as shown in Figure 5b. At both 60 and 80 atm we examined the same feeds studied by Green et al. (1993). At 80 atm three of the four feeds tested were stable. At 60 atm the five feeds examined were all stable. The results of these computations are shown in Table 2, along with performance data.

At 60 atm there is a region with three real roots of the VDW equation, as is the case for some of the test points examined in the binary Type I and Type II systems discussed above. The possibility that there may be three real volume roots does not present any difficulty when this method is used, since the EOS is solved simultaneously with the stationary point conditions and the method will find all roots to the combined system.

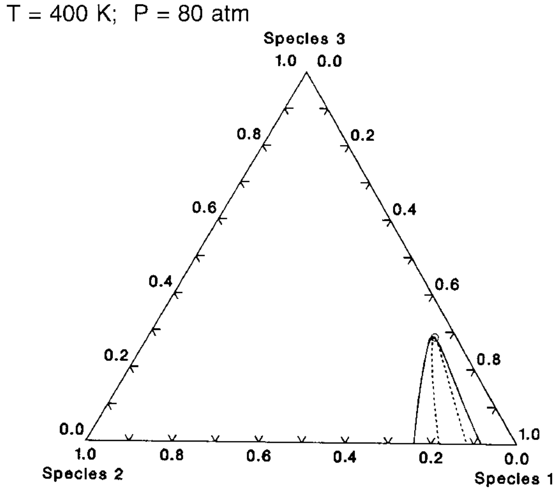

Figure 5a Ternary mixture at 400K and 80 atm.

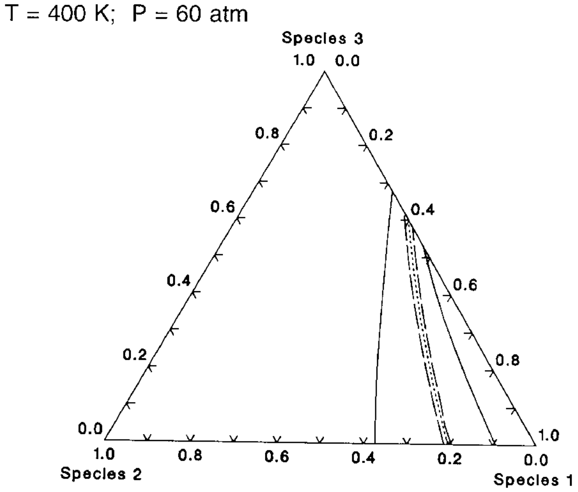

Figure 5b Ternary mixture at 400K and 60 atm.

5. CONCLUSIONS

In this paper we have presented the results of the first implementation of an interval Newton/generalized bisection technique for the determination of the phase stability of binary and ternary van der Waals mixtures. The advantage of this technique is that it is a general-purpose computational method, applicable to any arbitrary equation of state or activity coefficient model, and that it can find with mathematical certainty all the stationary points, thus guaranteeing a correct solution to the phase stability problem. We have demonstrated the use of this new method for the determination of phase stability of binary van der Waals mixtures that exhibit both Type I and Type II phase behavior, as well as a ternary van der Waals mixture.

6. ACKNOWLEDGMENTS

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Table 2
Ternary mixture: roots found and performance

Feed (P(atm),T(K), z_1,z_2,z_3)	Roots (x_1,x_2,x_3,v)	D	root inclusion tests	level reached in binary tree	CPU time: HP 9000/ 735 (sec)
80, 400, 0.83, 0.085, 0.085	0.83,0.085,0.085,214.8	0.0	3581	29	1.76
	0.8049,0.1065,0.0886,177.1	0.00046			
	0.6967,0.2090,0.0943,107.7	-0.00988			
80, 400, 0.77,0.115,0.115	0.77,0.115,0.115,150.0	0.0	4065	31	2.09
	0.8267,0.0690,0.1043.227.7	-0.00358			
	0.7186,0.1617.0.1197,118.2	-0.00133			
80, 400, 0.72,0.14,0.14	0.72,0.14,0.14,124.6	0.0	4680	34	2.45
	0.8132,0.0648,0.112,226.3	-0.00364			
	0.7494,0.1144,0.1362,143.3	0.00026			
80, 400, 0.69, 0.155,0.155	0.69,0.155,0.155,116.9	0.0	4469	30	2.28
	0.7940,0.0700,0.1360,214.3	0.00183			
	0.7579,0.0970,0.1451,163.4	0.00284			
60, 400, 0.80,0.10,0.10	0.80,0.10,0.10,368.6	0.0	2574	23	1.28
	0.7187,0.1735,0.1079,186.5	0.02559			
	0.5379,0.3625,0.0996,92.54	-0.04865			
60, 400, 0.69, 0.155, 0.155	0.69,0.155,0.155,295.9	0.0	2512	25	1.32
	0.6770,0.1669,0.1561,265.4	0.00011			
	0.4367,0.4288,0.1345,88.34	-0.1683			
60, 400, 0.67,0.165,0.165	0.67,0.165,0.165,126.9	0.0	4134	30	2.01
	0.8119,0.0531,0.1350,392.7	-0.07515			
	0.6477,0.1862,0.1661,115.9	-0.00017			
60, 400, 0.66, 0.170,0.170	0.66,0.170,0.170,122.7	0.0	4988	36	2.5
	0.8095,0.0523,0.1382,392.6	-0.0758			
	0.6543,0.1754,0.1703,119.9	$-0.29\text{x}10^{-5}$			
60, 400, 0.65,0.175,0.175	0.65,0.175,0.175,119.4	0.0	4887	35	2.46
	0.8061,0.0520,0.1419,392.0	-0.07495			
	0.6583,0.1672,0.1745,123.5	$0.87\text{x}10^{-5}$			

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