Improved Reduced Flash Calculations Using Two-Parameter BIP Equation

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Abstract

Phase equilibrium calculations constitute a significant percentage of computational time in compositional simulation, especially as the number of components and phases increase. Reduced methods address this problem by carrying out phase equilibrium calculations using a reduced number of parameters. These methods have shown to speed up batch flash calculations, decrease simulation times, and also improve convergence behaviour.

In this paper, we present new reduced parameters using the two-parameter binary interaction parameter formula originally proposed by Li and Johns (2006). The new reduced parameters are applied to solve two-phase flash calculations for several five different fluid descriptions. The results show significant reduction in the number of iterations required to achieve convergence compared to the Li and Johns original approach. We further compare the results to other reduced methods, including the dominant eigenvalue method and a recently improved version of that method.

Introduction

Gas injection achieves enhanced oil recovery primarily by developing miscibility in situ. Compositional simulation of gas floods is an important tool for recovery estimation, but a significant drawback is that compositional simulation is costly when the number of components and phases is large. Thus, one approach is to lump components into several pseudocomponents and to limit the number of phases that can form. Reducing the number of components, however, decreases the accuracy of the phase behaviour calculation in compositional simulation, and also the detailed type of compositional information required for better surface facility designs.

Reduced methods are one approach to speed up flash and stability calculations so that more components and phases can be used. Michelsen (1986) reduced the number of parameters to be solved to three by setting all binary interaction parameters ($k_{ij}$ - BIPs) to zero. Jensen and Fredenslund (1987) extended Michelsen’s research by the addition of two reduced parameters for every column of the BIP matrix with at least one nonzero element. Thus, for one column of nonzero BIPS his approach has five reduced parameters. Most real fluids, however, have multiple columns of nonzero BIPs so that these methods are not practical.

Hendriks (1988) and Hendriks and Van Bergen (1992) proposed a dominant eigenvalue method (DEM) that involves a spectral decomposition of the $1-k_{ij}$ matrix. The number of reduced parameters depends
on the number of eigenvalues retained; they suggested neglecting eigenvalues less than 0.001. In the DEM approach the phase behaviour is therefore altered depending on the magnitude of the eigenvalues neglected. Nichita and Minescu (2004) extended the DEM approach for two-phase flash calculations to multiphase equilibrium.

Pan and Firoozabadi (2002) also considered the DEM approach and showed that flash calculation with reduced parameters are more robust than conventional flash calculations. They showed that the tangent plane distance function (TPD) is smoother in reduced space than in compositional space.

Li and Johns (2006) introduced another reduction method where there are six reduced parameters independent of the number of components. Their approach, denoted as LJ in this paper, is based on a specific formulation of the BIP matrix, which in general must be tuned to PVT data. Okuno, Johns and Sepehrnoori (2010a) improved convergence of Li and Johns’ method by adding a successive substitution (SS) step after each Newton-Raphson iteration. This modification increased computational time per iteration, but decreased the number of iterations to convergence, and overall computational time.

Okuno, Johns and Sepehrnoori (2010b) showed that the reduced methods can be applied to three-phase stability and flash calculations. They implemented their approach in compositional simulation and showed significant computational time reductions.

Pan and Tchelepi (2010) applied the DEM reduced method to compositional simulation and concluded reduced methods are significantly faster than conventional methods. However, they neglected eigenvalues smaller than 0.07, which could significantly alter the phase behaviour.

More recently, Nichita and Graciaa (2010) improved the dominant eigenvalue method (IDEM) by developing a new set of reduced parameters that increased linearity in the residual functions. They showed a significant reduction in the number of NR iterations compared to the standard DEM method.

Haugen and Beckner (2011) compared the DEM reduced method and the conventional method for both stability and two-phase flash calculations using similar programming optimization levels. They concluded that reduced methods (DEM) are worthwhile only when the number of components is ten or higher.

In this paper, we apply the approach by Nichita and Graciaa (2010) to the BIP formula of Li and Johns (2006). The first section summarizes the equations and approach for the various reduced methods. The second section presents our new reduced parameters and two-phase flash calculation algorithm. We then compare all of these various reduced methods for five fluid characterizations in the literature.

Background

Flash calculations solve for the equilibrium phase compositions that result from specifying the temperature, pressure, and overall composition. All flash calculation methods, whether reduced or conventional, require an initial set of K-values estimated either from a prior flash in simulation, stability analysis, or from correlations (Pan and Firoozabadi 2003, Wilson 1969). For flash calculations, \( N_C \) nonlinear equations are solved by successive substitution (SS) and/or Newton-Raphson iteration (NR) to obtain equality of chemical potential.
(Sandler 1994). Stability analysis is often performed prior to solving the fugacity equations to determine if a phase is stable or not (Michelsen 1982a). For NR iterations, a \( N_C \times N_C \) Jacobian matrix must be solved at each iteration, which takes significant computational effort especially as the number of components and number of phases become large.

The main idea behind the reduced methods is that if fugacity coefficients can be expressed solely as a function of a few scalars, then the size of the Jacobian matrix can be reduced. The BIP matrix is manipulated so that the fugacity coefficients are only a function of a few parameters, reducing the size of the Jacobian matrix. Such a transformation makes the Jacobian calculation nearly independent of the number of components, which can result in a substantial reduction in computational time.

For a generalized cubic equation-of-state, the fugacity coefficient equations can be written as a function of composition by,

\[
\ln \phi_i = \frac{(Z - 1)}{B} B_i - \ln(Z - B) - \frac{A}{(\delta_1 - \delta_2)B} \left( \frac{2 \sum_{j=1}^{N_C} A_{ij} x_j}{A} - \frac{B_1}{B} \right) \ln \left( \frac{Z + \delta_1 B}{Z + \delta_2 B} \right)
\]

\( i = 1, \ldots N_C \)  

(1)

where \( \delta_1 \) and \( \delta_2 \) are EOS-type specific coefficients.

The main difference in the reduced methods is how they treat parameter \( A \) and \( \sum_{k=1}^{N_C} A_{ik} x_k \) in Eq. (1). These two parameters are defined by,

\[
\sum_{j=1}^{N_C} A_j x_j = \sqrt{A} \sum_{j=1}^{N_C} A_j x_j (1 - k_{ij})
\]

and,

\[
A = \sum_{i=1}^{N_C} \sum_{j=1}^{N_C} A_{ij} x_i = \sum_{i=1}^{N_C} \sum_{j=1}^{N_C} \sqrt{A} A_{ij} x_i (1 - k_{ij}) = \sum_{i=1}^{N_C} \sqrt{A} x_i \left( \sum_{j=1}^{N_C} \sqrt{A} x_j (1 - k_{ij}) \right)
\]

Michelsen (1986) set the BIPs in Eqs. (2) and (3) to zero \( (k_{ij} = 0) \) and recognized that these equations can be expressed as a function of the reduced parameter:

\[
\alpha = \sum_{i=1}^{N_C} \sqrt{A} x_i
\]

Michelsen retained \( B \) as a second reduced parameter, and used the mole fraction of liquid as a third reduced parameter. The fugacity coefficient in Eq. (1) can then be expressed as a function of the first two reduced parameters, since the compressibility factor \( Z \) is also a function of \( A \) and \( B \).

Newton-Raphson (NR) iterations are typically done after several successive substitution (SS) steps to ensure a good initial guess of the reduced parameters, which depend on equilibrium phase compositions. In Michelsen’s approach, the following three residual functions were used to estimate the three reduced parameters from NR:

\[
e_{i} = \sum_{i=1}^{N_C} (y_i - x_i)
\]
At convergence, the residual functions in Eqs. (5) should be less than a small specified tolerance. Alternatively, one could calculate the fugacities from the reduced parameters and use a component fugacity convergence criterion. Michelsen’s reduced method, however, is not used in practice since most fluid characterizations have nonzero BIPs.

Hendriks and Van Bergen (1992) developed new reduced parameters to account for nonzero BIPs. They applied spectral decomposition to the BIP matrix and obtained,

\[
(1 - k_j) = \sum_{k=1}^{N_c} \lambda_k q_i^k q_j^k
\]

(6)

where \( q_i^k \) is the eigenvector corresponding to the \( k^{th} \) eigenvalue. For \( n \) non-zero (or very small) eigenvalues, new reduced parameters are defined as,

\[
Q_a = \sum_{i=1}^{N_c} q_i^\alpha \sqrt{A_i} x_i \quad \alpha = 1, \ldots, n
\]

(7)

Using Eqs. (7), the EOS parameters can then be written as,

\[
A = \sum_{a=1}^{n} \lambda_a Q_a^2
\]

\[
\sum_{j=1}^{N_c} A_j x_j = \sum_{a=1}^{n} \lambda_a Q_a \sqrt{A_i} q_i^a
\]

(8)

Equations (8) are only approximate since small eigenvalues are set to zero, which changes the BIP values. This method is referred to as the dominant eigenvalue method (DEM), where the number of reduced parameters depends on the number of eigenvalues (\( n \)) retained. In total, there are \( n+2 \) reduced parameters when \( B \) and the mole fraction of liquid are also included. The Newton-Raphson iterations then solve \( n+2 \) error functions:

\[
e_a = \sum_{i=1}^{N_c} x_i \sqrt{A_i} q_i^a - Q_a \quad a = 1, \ldots, n
\]

\[
e_{n+1} = \sum_{i=1}^{N_c} x_i B_i - B
\]

\[
e_{n+2} = \sum_{i=1}^{N_c} (y_i - x_i)
\]

(9)

Li and Johns (2006) took a different approach to develop new reduced parameters when BIPs are nonzero. They expressed the BIP matrix using two parameters through the function

\[
k_{ij} = (h_i - h_j)^2 g_i g_j
\]

(10)

where \( h_i \) and \( g_i \) are component tuning parameters determined from a best fit to PVT data (or in some cases by doing a best fit of the BIPs from prior tuned characterizations). Using Eq. (10), they derived the following expressions:

\[
A = \Theta_1^2 + 2\Theta_2^2 - 2\Theta_1\Theta_4
\]

\[
\sum_{j=1}^{N_c} A_j x_j = \sum_{a=1}^{4} \Theta_a q_{ia}
\]

(11)

where the four reduced parameters are: 

\[
\Theta_a = \left\{ \sum_{i=1}^{N_c} x_i \sqrt{A_i}, \sum_{i=1}^{N_c} x_i \sqrt{A_i h_i g_i}, \sum_{i=1}^{N_c} x_i \sqrt{A_i h_i^2 g_i}, \sum_{i=1}^{N_c} x_i \sqrt{A_i g_i} \right\}
\]

and the vector of known parameters: 

\[
\left\{ \sum_{i=1}^{N_c} x_i \sqrt{A_i}, \sum_{i=1}^{N_c} x_i \sqrt{A_i h_i g_i}, \sum_{i=1}^{N_c} x_i \sqrt{A_i h_i^2 g_i}, \sum_{i=1}^{N_c} x_i \sqrt{A_i g_i} \right\}
\]
\[ q_{\alpha} = \left\{ A_i, 2A_i A_{\alpha}^{-1}, -\sqrt{A_i g_i}, -\sqrt{A_i h_i^2 g_i} \right\}. \]

(13)

Together with the EOS parameter \( B \) and the mole fraction of liquid, Li and Johns (LJ) used a total of six reduced parameters. Unlike the DEM approach, the number of reduced parameters in their approach is always six, eliminating the need to determine how many eigenvalues to retain.

For all reduced methods, the reduced parameters based on one of the phases are updated after each NR iteration. The reduced parameters for the second phase are calculated by a simple material balance equation. The fugacity coefficients are then calculated from the reduced parameters at each iteration to update the K-values. Because the mole fraction of liquid is known (it is a reduced parameter), the liquid equilibrium phase compositions are then directly computed from the equation

\[ x_i = z_i / \left[ (1 - L) K_i + L \right]. \]

Since the equilibrium phase compositions are calculated from the reduced parameters, the phase compositions only exactly satisfy

\[ \sum_{i=1}^{N_c} x_i = \sum_{i=1}^{N_c} y_i = 1 \] at convergence.

Further, the values of the residual functions based on equality of fugacity coefficients could increase in the first NR iteration even though the residual parameters are converging.

Okuno, Johns and Sepehrnoori (2010a) corrected the mass balance error in the LJ method by adding a SS step after each NR iteration of the reduced parameters. That is, after the equilibrium phase compositions are first calculated from the reduced parameters following NR, the reduced parameters are recalculated based on these equilibrium phase compositions. These reduced parameters are then used to solve for new equilibrium phase compositions after calculation of fugacity coefficients and K-values. Although the SS step required additional computational time, Okuno et al. (2010a) showed that by calculating the fugacity coefficients at the same iteration as the reduced parameters they achieved faster convergence. In this paper, we denote their approach as LJ+SS.

Nichita and Graciaa (2010) introduced new reduced parameters for the DEM method. In their novel approach, they expressed the fugacity coefficient for a given phase as a linear function of the new reduced parameters:

\[ \ln \phi_i = \sum_{\alpha=1}^{n+2} h_{\alpha} q_{\alpha} \quad i = 1, \ldots, N_c \]

(14)

where \( h_{\alpha} \) are the reduced parameters and \( q_{\alpha} \) are vectors of length \( N_C \) with known elements. A significant advantage of this approach over the standard DEM method is that the fugacity coefficients are easy to calculate and the expression is linear with the reduced parameters. Nichita and Graciaa showed that fewer iterations are required for convergence with their improved approach (IDEM) over the DEM method. Further this approach naturally uses the equality of fugacity coefficients as the convergence criterion and Rachford-Rice are solved for the phase mole fractions after each iteration because the phase mole fractions are no longer reduced parameters.

**The Improved LJ Method**

In this section, we extend the Nichita and Graciaa approach to the Li and Johns (2006) BIP formula and develop new reduced parameters that improve convergence. The improved LJ method is denoted ILJ.
The fugacity coefficients can be written as a linear combination of six reduced parameters. That is,

$$\ln \phi_i = \sum_{\alpha=1}^{6} h_{\alpha} q_{i\alpha}$$  \hspace{1cm} (15)$$

where for each phase:

$$h_\alpha = \frac{-2\Theta_\alpha}{\Delta B} \ln \left( \frac{Z + \delta B}{Z + \delta Z B} \right) \alpha = 1,\ldots,4$$  \hspace{1cm} (16)$$

$$h_5 = \frac{(Z-1)}{B} + \frac{A}{\Delta B^2} \ln \left( \frac{Z + \delta B}{Z + \delta Z B} \right)$$  \hspace{1cm} (17)$$

$$h_6 = -\ln(Z - B)$$  \hspace{1cm} (18)$$

$$q_{i\alpha} = \left\{ \sqrt{A_i}, 2 \sqrt{A_i h_{i\alpha}}, -\sqrt{A_i g_{i\alpha}}, -\sqrt{A_i h_{i\alpha}^2 g_{i\alpha}}, B_i, 1 \right\}$$  \hspace{1cm} (19)$$

Eq. (19) is very similar to Eq. (13), but the reduced parameters given by Eqs. (16) – (18) are completely different. The LJ reduced parameters defined in Eqs. (12) are embedded in the new reduced parameters through Eqs. (16). However, we do not use the LJ reduced parameters as independent parameters for the Jacobian. Further, there is no direct relationship to relate $h_\alpha$ from one phase to another phase. Thus, we solve for the difference in the reduced parameters defined by,

$$h_{\delta\alpha} = h_{\alpha} - h_{\alpha}$$  \hspace{1cm} (20)$$

The residual functions to be minimized are then:

$$e_\alpha = h_{\alpha} - h_{\alpha} - h_{\delta\alpha} \alpha = 1,\ldots,6$$  \hspace{1cm} (21)$$

and the Jacobian matrix is given by,

$$J_{\alpha\beta} = \frac{\partial e_\alpha}{\partial h_{\delta\beta}} = \frac{\partial h_{\alpha}}{\partial h_{\delta\beta}} - \frac{\partial h_{\alpha}}{\partial h_{\delta\beta}} - \delta_{\alpha\beta}$$  \hspace{1cm} (22)$$

where $\delta_{\alpha\beta} = 1$ when $\alpha = \beta$ or zero otherwise.

The procedure for the improved LJ method (ILJ) begins by making SS iterations to get a good initial guess for the Newton-Raphson iterations. The approach is similar to the conventional SS method since the fugacity coefficients can be computed directly from the new reduced parameters. The procedure for the two-phase SS iterations are:

1. Perform Rachford-Rice iterations to determine an initial guess of the equilibrium phase compositions and mole fraction of liquid (Rachford and Rice 1952). The K-values can be determined from Wilson’s correlation, a prior stability analysis, or a prior solution.

2. Calculate LJ’s reduced parameters for the liquid phase using Eqs. (12), and

$$B = \sum_{i=1}^{N} B_i x_i .$$

3. Calculate LJ’s reduced parameters for the vapor phase using the linear relation,

$$\Theta_{va} = \frac{\Theta_{va} - L\Theta_{va}}{1 - L}$$  \hspace{1cm} (23)$$

$$B_v = \frac{B_i - LB_i}{1 - L}$$

where $z$ indicates that the overall composition $z_i$ is used in the calculation.


5. Calculate compressibility factors $Z_L$ and $Z_V$ by solving the cubic EOS equation. We used Peng-Robinson EOS in this paper (Peng-Robinson 1976).

6. Calculate $h_{\alpha\alpha}$ and $h_{\alpha\alpha}$ from Eqs. (16) – (18). Also, calculate $h_{\delta\alpha}$ from Eq. (20).

7. Update the logarithm of the component fugacity coefficients for each phase by Eqs. (15).
8. Calculate K-values using,

\[ \ln K_i = \ln \frac{\phi_{li}}{\phi_{li}^{\alpha}} = \sum_{\alpha=1}^{\delta} h_{\alpha i} q_{\alpha i}. \] (24)

9. Make Rachford-Rice calculations to update the equilibrium phase compositions \( x_i \) and \( y_i \).

10. Check for convergence based on the equality of component fugacities where we used \( \max_{i} \left| \ln \left( \frac{\phi_{li} x_i}{\phi_{li}^{\alpha} y_i} \right) \right| < 10^{-3} \). If the tolerance is achieved, start the NR iterations described in the paragraph below. Otherwise, go to step 2) and do another SS step.

The procedure for the two-phase Newton-Raphson (NR) iterations are:
1. Repeat steps 2) – 8) of the SS iteration procedure.
2. Calculate the Jacobian matrix as shown in the Appendix.
3. Solve for \( h_{\alpha i} \) using Newton-Raphson.
5. Solve the Rachford-Rice equations to update the equilibrium phase compositions and liquid mole fraction.
6. Check for NR convergence when \( \max_{i} \left| \ln \left( \frac{\phi_{li} x_i}{\phi_{li}^{\alpha} y_i} \right) \right| < 10^{-10} \). Other criteria and tolerances can be used as needed. If not converged, go to step 1).

The above procedure could be extended to three or more phases if needed. The robust and rapid Rachford-Rice procedure by Okuno, Johns, and Sepehrnoori (2010b) can be used for multiphase calculations.

Results

In this section, we compare five different reduced methods using five different fluids, namely oil A, B and C from Li and Johns (2006); and mixtures 1 and 2 from Pan and Firoozabadi (2002). Table 1 shows the temperature and pressure conditions for 45 different flash calculations. The table also shows the variation in the \( \text{CO}_2 \) mole percentage of mixture 2.

Convergence Behavior

Figure 1 shows the residuals for each NR iteration for example oil A near the critical region (484°F, 1044.41 psia). The residuals for the standard DEM and LJ reduced methods increase for the first NR iteration owing to the material balance error discussed in the previous section. That is, the equilibrium phase compositions are not consistent with the fugacity coefficients and overall material balances. This material balance error eventually decays to zero as iterations converge.

The other three reduced methods (LJ+SS, ILJ, and IDEM) do not have this problem and converge in fewer iterations as shown in Fig. 1. This is also true for all 45 flash calculations in Table 1.

Figure 2 shows the number of iterations required to achieve convergence for the 45 flash calculations as a function of the length of the tie line, i.e. \( \sqrt{\sum_{i=1}^{N_c} (y_i - x_i)^2} \). The tie-line length approaches zero at the critical point and therefore gives a good indication of how far the tie-line is from the critical locus. As shown, the number of iterations increases substantially as the flash approaches a critical point. In general, the number of iterations is fairly independent of the number of eigenvalues used in the DEM method, except for one value that does not converge. The phase behavior changes only slightly in the DEM approach when all eigenvalues are used (denoted DEM(all) in Fig. 2) compared to when eigenvalues greater than 0.001 are used (denoted
DEM>0.001). The phase behavior change is more significant when only eigenvalues greater than 0.07 are retained, as was suggested by Pan and Tchelepi. Table 2 shows the number of eigenvalues were used for each fluid based on these criteria. The LJ method always has six reduced parameters and does not have the problem of selecting the number of eigenvalues to retain.

The number of iterations for the improved versions (IDEM and ILJ) are shown in Fig. 3. Both ILJ, LJ+SS, and IDEM converge on average in one less iteration compared to DEM, and LJ for flash calculations far from from the critical locus. Near the critical region, the ILJ, LJ+SS, and IDEM converge in two fewer iterations. Slight differences in convergence between ILJ and IDEM result from slightly different fluid characterizations owing to the eigenvalue approximations.

**Computational Time**

Another factor that affects overall computational time is the computational time per iteration. Figure 4 compares the computational time required per iteration for the reduced algorithms for all 45 flash calculations in Table 1. The compositions in the figure are arranged left to right in order of increasing number of components (oil A is a seven-component model, while oil C is fifteen components). The flash calculation times are computed with an Intel Xeon CPU with 3.06 GHz and 12 GB RAM.

Figure 4 shows that the improved reduced method by Nichita and Graciaa (denoted IDEM(all)) shows a significant increase in computational time as the number of components increases. More components mean that more eigenvalues are used. The other methods (LJ, ILJ, and IDEM with fewer eigenvalues) are all about the same computational time. Because ILJ requires less iterations than LJ, the total computational time for ILJ is faster, especially near a critical point. ILJ is typically also faster than IDEM>0.001, depending on the number of eigenvalues used.

**Phase Behaviour Accuracy**

As shown in Figure 5, standard and improved reduced methods give about the same accuracy on the PT diagram for oil C, except for the case when BIPs are zero (Michelsen’s method). LJ and ILJ are exactly the same since they use the same BIPs ($h_i$ and $g_i$ parameters are the same).

The mole fraction of liquid shows a much larger error as the number of eigenvalues used are reduced compared to the case when all eigenvalues are retained (see Fig. 5). Figure 5 also shows that a best fit of the $h_i$ and $g_i$ parameters to the original BIP matrix (with all eigenvalues retained) performed well compared to DEM>0.07. There is no generally accepted method for determining how many eigenvalues to keep with the DEM or IDEM approach. With the LJ and ILJ method, the $h_i$ and $g_i$ parameters should be tuned to the PVT data for best accuracy. Figure 6 shows that DEM and LJ are significantly more accurate than Michelsen’s method.

Figure 7 shows the total average computational time for the flash calculations shown in Table 1 using the LJ and ILJ methods. The speed up with ILJ increases from 15% to 35% as the number of components in the fluid characterization increase. Figure 8 shows the average number of iterations required for each method. The ILJ method requires fewer iterations, which explains partly why there is some speed up with ILJ over LJ. Figure 9
shows a similar advantage of IDEM over DEM.

**Conclusions**

We extended the approach by Nichita and Graciaa (2010) to the method of Li and Johns (2006) and developed new reduced parameters based on their two-parameter BIP formulation. The main conclusions are:

- The improved LJ method (ILJ) requires fewer iterations, and less computational time than the standard LJ method.
- The improved LJ method eliminates the SS step required in the LJ method developed by Okuno et al. (2010a).
- The improved dominant eigenvalue method (IDEM) of Nichita and Graciaa (2010) gave similar improvements compared to the standard DEM method.
- Neglecting eigenvalues less than 0.07 in the DEM and IDEM approach can result in significantly less phase behavior accuracy than if all eigenvalues greater than 0.001 are retained.

### Table 1. Temperatures and pressures for different flash calculations

<table>
<thead>
<tr>
<th>Mixture 1</th>
<th>Mixture 2</th>
<th>Oil A</th>
<th>Oil B</th>
<th>Oil C</th>
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<td>psia</td>
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<td>22</td>
<td>400</td>
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### Table 2. Number of eigenvalues used for various fluids based on the eigenvalue magnitude

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<th>Mixture 1</th>
<th>Mixture 2</th>
<th>Oil A</th>
<th>Oil B</th>
<th>Oil C</th>
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<td>11</td>
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Figure 1. Convergence of flash calculations for oil A near the critical point for five different reduced methods. DEM(4) and IDEM(4) denote that four eigenvalues are retained. LJ: (Li and Johns, 2006), LJ+SSI (Okuno, Johns, and Sepehrnoori, 2010a).

Figure 2. Number of iterations needed for convergence for the two standard reduced methods. DEM (all) denotes that all eigenvalues were retained, while DEM>0.001 indicates that only eigenvalues greater than 0.001 were retained. DEM>0.07 retained eigenvalues > 0.07.

Figure 3. The improved methods reduce the number of iterations required to achieve convergence.

Figure 4. Comparison of computational time per iteration for all five fluids from Table 1 using the improved reduced methods.
Figure 5. Average error in liquid phase mole fractions with the reduced methods compared to the original fluid characterization with all eigenvalues retained. The improved methods yield identical accuracy to the standard methods.

Figure 6. Calculation of bubble point curve for oil C using various reduced methods. The accuracy is reduced when too many eigenvalues are discarded.

Figure 7. Average computational time for LJ and ILJ for the flash calculations shown in Table 1.

Figure 8. Average number of iterations required for convergence with the LJ and ILJ methods.
Nomenclature

J Jacobian matrix
N\textsubscript{C} Number of components
N\textsubscript{p} Number of phases
Q DEM reduced parameters
Z Compressibility factor
e Error function
h\textsubscript{i}, g\textsubscript{i} LJ vectors
h Independent parameters for improved algorithms
k\textsubscript{ij} BIP matrix
n\textsubscript{i} Number of eigenvalues considered
q Eigenvector
x Liquid molar composition
y Vapor composition

Greek letters

δ\textsubscript{1}, δ\textsubscript{2} EOS parameters
φ Fugacity coefficient
Θ LJ reduced parameters
λ Eigenvalue

Subscripts

i Composition
j Composition or phase
L Liquid
V Vapor

Abbreviations

BFGS Broyden–Fletcher–Goldfarb–Shanno
BIP Binary interaction parameters
DEM Dominant eigenvalue method
EOS Equation of State
LJ Algorithm based on Li et al.
IDEM Improved dominant eigenvalue method
ILJ Improved algorithm based on Li et al.
NR Newton Raphson
PR Peng Robinson
SS Successive substitution
TLL Tie-line length
TPD Tangent plane distance

References


Equilibria Calculations. Fluid Phase Equilibria 74: 17-34


Figure 9. Average computational time for DEM and IDEM for the flash calculations shown in Table 1.


Pan, H. and Tchelepi, H.A. 2010. Reduced-Variables Method for General-Purpose Compositional Reservoir Simulation. Paper presented at the CPS/SPE International Oil & Gas Conference and Exhibition in China, Beijing, 8-10 June


Wilson, G. 1969. A Modified Redlich-Kwong Equation of State , Application to General Physical Data Calculations. Paper presented at AIChE 65th National Meeting, Cleveland, Ohio, 4-7 May

### Appendix. Jacobian for Newton-Raphson Iterations

The elements of the Jacobian matrix are given by

\[
\frac{\partial h_{\alpha j}}{\partial h_{\beta \gamma}} = \sum_{y=1}^{5} \frac{\partial h_{\alpha j}}{\partial Q_{\gamma y}} \frac{\partial Q_{\gamma y}}{\partial h_{\beta \gamma}} \quad \alpha, \beta = 1, ..., 6; \quad \gamma = 1, ..., 5; \quad \text{and} \quad j = L, V \quad (A.1)
\]

The derivatives on the right side of eq. A.1. are determined as,

\[
\frac{\partial h_{\alpha j}}{\partial Q_{\gamma y}} = \left( \frac{\partial h_{\alpha j}}{\partial Q_{\gamma y}} \right)_{Z_{j}} + \left( \frac{\partial h_{\alpha j}}{\partial Z_{j}} \right)_{Q_{y}} \frac{\partial Z_{j}}{\partial Q_{\sigma y}} \quad (A.2)
\]

For liquid,

\[
\frac{\partial Q_{\gamma y}}{\partial h_{\alpha \beta}} = \sum_{i=1}^{N_{L}} \theta_{\gamma i} \frac{\partial x_{i}}{\partial h_{\alpha \beta}^i}.
\]
and for vapor, 
\[ \frac{\partial Q_{\gamma j}}{\partial h_{\alpha \beta}} = \sum_{i=1}^{N_{\gamma}} \theta_{\gamma i} \frac{\partial y_i}{\partial h_{\alpha \beta}}, \]  
(A.3)

where \( \theta_{\gamma i} = \{ \sqrt{A_{\gamma}}, \sqrt{A_{\gamma} h_{\gamma}}, \sqrt{A_{\gamma} h_{\gamma}^2}, \sqrt{A_{\gamma} g_{\gamma}}, B_{\gamma} \} \).

The analytical expression for Eq. (A.2) is:

\[ \left( \frac{\partial h_{\alpha j}}{\partial Q_{\gamma j}} \right)_{Z_j} = \begin{cases} 
\frac{h_{\alpha j} \delta_{\alpha \gamma}}{Q_{\gamma j}} & \text{for} \ \alpha = 1, \ldots, 4; \ \gamma = 1, \ldots, 4 \\
\frac{1}{B_j} \ln \left( \frac{Z_j + \delta_j B_j}{Z_j + \delta_j B_j} \right) (\Delta B_j^2) & \text{for} \ \alpha = 5; \ \gamma = 1, \ldots, 4 \\
\frac{1}{B_j} \left( h_{\alpha j} + 2Q_{\alpha j} \frac{Z_j}{\pi_j} \right) & \text{for} \ \alpha = 1, \ldots, 4; \ \gamma = 5 \\
\frac{1}{B_j} \left( -2h_{\gamma j} B_j - 1 + Z_j \left( 1 + \frac{A_j}{\pi_j} \right) \right) & \text{for} \ \alpha = 5; \ \gamma = 5 \\
\frac{1}{Z_j - B_j} & \text{for} \ \alpha = 6; \ \gamma = 5 \\
0 & \text{otherwise} \\
\end{cases} \]  
(A.4)

\[ \left( \frac{\partial h_{\alpha j}}{\partial Z_j} \right)_{Q_j} = \begin{cases} 
\frac{2Q_{\alpha j}}{\pi_j} & \text{for} \ \alpha = 1, \ldots, 4 \\
\frac{1}{B_j} \left( 1 - \frac{A_j}{\pi_j} \right) & \text{for} \ \alpha = 5 \\
\frac{1}{Z_j - B_j} & \text{for} \ \alpha = 6 \\
\end{cases} \]  
(A.5)

where \( \pi_j = (Z_j + \delta_j B_j)(Z_j + \delta_j B_j) \)

\[ \frac{\partial Z_j}{\partial Q_{\gamma j}} = \begin{cases} 
\frac{\partial Z_j}{\partial A_j} \frac{\partial A_j}{\partial Q_{\gamma j}} & \text{for} \ \gamma = 1, \ldots, 4 \\
\frac{1}{B_j} \left( \frac{\partial F_j}{\partial Z_j} \right)_{Q_j} & \text{for} \ \gamma = 5 \\
\end{cases} \]  
(A.6)
where

\[
\frac{\partial Z_j}{\partial A_j} = -\frac{Z_j - B_j}{\frac{\partial F_j}{\partial Z_j}} , \quad \frac{\partial A_j}{\partial Q_{rj}} = \begin{bmatrix} 2Q_{1j}, 4Q_{2j}, -2Q_{3j}, -2Q_{4j} \end{bmatrix}
\] (A.7-8)

and

\[
\left( \frac{\partial F_j}{\partial Z_j} \right)_{Q_j} = 3Z_j^2 + 2[(\delta_1 + \delta_2 - 1)B_j - 1]Z_j + \left[ A_j + \delta_i \delta_2 B_j^2 - (\delta_1 + \delta_2)B_j(B_j + 1) \right]
\] (A.9)

The expressions for Eqs. (A3) are

\[
\frac{\partial x_i}{\partial h_\beta} = -d_i \left[ \frac{\partial K_i}{\partial h_\beta} V + \frac{\partial V}{\partial h_\beta} (K_i - 1) \right]
\]

and

\[
\frac{\partial y_i}{\partial h_\beta} = d_i \left[ \frac{\partial K_i}{\partial h_\beta} (1 - V) - \frac{\partial V}{\partial h_\beta} (K_i (K_i - 1)) \right]
\] (A.10)

where,

\[
\frac{\partial V}{\partial h_\beta} = \sum_{i=1}^{N_i} d_i \frac{\partial K_i}{\partial h_\beta} \left( K_i - 1 \right)^2
\] (A.11)

\[
\frac{\partial K_i}{\partial h_\beta} = K_i q_i \beta
\] (A.12)

\[
d_i = \frac{z_i}{(1 + V(K_i - 1))^2}
\] (A.13)