An Improved Multicomponent Diffusion Model for Compositional Simulation of Fractured Unconventional Reservoirs

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Summary

Most simulators currently use the advection/diffusion model (ADM), where the total flux comprises Darcian advection and Fickian diffusion. However, significant errors can arise, especially for modeling diffusion processes in fractured unconventional reservoirs, if diffusion is modeled by the conventional Fick's law using molar concentration. Hence, we propose an improved multicomponent diffusion model for fractured reservoirs to better quantify the multiphase multicomponent transport across the fracture/matrix interface.

We first give a modified formulation of the Maxwell-Stefan (MS) equation to model the multicomponent diffusion driven by the chemical potential gradients. A physics-based modification is proposed for the ADM in fractured reservoirs, where fracture, matrix, and their interface are represented by three different yet interconnected flow domains to honor the flux continuity at the fracture/matrix interface. The added interface using a more representative fluid saturation and composition of the interface can hence better capture the transient mass fluxes between fracture and matrix. The proposed approach is also implemented in an in-house compositional simulator. The multicomponent diffusion model is validated with both intraphase and interphase diffusion experiments. Then, the improved model for fracture/matrix interaction is compared with a fine-grid model. The proposed multiple interacting continua (MINC) model with three continua (MINC3) can better match the fine-grid model's result than the double-porosity (DP) model, which only obtains a fair match at an early time.

Then, we simulate a gas huff 'n' puff (HnP) well in the Permian Basin to investigate the effect of diffusion within the fractured tight oil reservoir. The simulation reveals that diffusion has a minor effect on the performance of depletion when oil is the dominant phase. For gas HnP, the simulation neglecting diffusion will underestimate the oil recovery factor (RF) but overestimate the gas rate. The DP approach tends to overestimate the RF of heavy components but leads to a similar cumulative oil RF compared with MINC3. With the diffusion included in the simulation, gas HnP performance becomes more sensitive to the soaking time than the model without diffusion. Although increasing the soaking time will lead to a higher RF after considering diffusion, the incremental oil is not sufficiently large to justify a prolonged soaking time.

Introduction

In conventional reservoirs, the advection often dominates the transport process over diffusion because of the porous rock's high permeability, where diffusion is mostly negligible. However, for the low-permeability unconventional reservoir, such as tight oil reservoirs, the role of diffusion might no longer be negligible within the matrix (Fu et al. 2019) or at the matrix/fracture interface (Hoteit 2013; Alharthy et al. 2018), where the ratio of advective mass flux to diffusive mass flux (defined as the dimensionless Péclet number Pe) approaches unity. The Pe might still be large enough in fracture networks for us to reasonably neglect diffusive flux when modeling transport in fractured tight reservoirs (Hoteit and Firoozabadi 2009), but for gas injection into the stimulated reservoir volume (SRV) of unconventional oil reservoirs, an accurate inclusion of multicomponent molecular diffusion within the tight matrix or at the fracture/ matrix interface is critical.

The most commonly used diffusion model is the classic Fick's law, which states that the diffusive flux of the *i*th component is linearly related to the gradient of its own concentration (c_i or $c_i x_i$ in a binary system), as shown in **Fig. 1a**.

where c_t is the total molar concentration of all species and D_{ij} is the binary diffusion coefficient. It can be measured or estimated using correlations such as Sigmund (1976).

For a multicomponent system, the generalized Fick's law (Cussler 2009) can be written as

where \mathcal{D}_{ij} is the multicomponent diffusion coefficient and often is not equal to the binary diffusion coefficient D_{ij} .

The challenge is the measurement for the multicomponent diffusion coefficient, which is very limited even for the ternary system, let alone systems with more than three components. Eq. 2 is often simplified as

$$\vec{r}_i = -c_t D_{i,\beta} \nabla x_i, \qquad (3)$$

where $D_{i,\beta}$ is the diffusion coefficient of component *i* within the bulk phase β and can be estimated using empirical correlations such as Wilke and Chang (1955). However, this approach might not be suitable for modeling gas injection in unconventional oil reservoirs because $D_{i,\beta}$ is often treated as an ad hoc parameter and can neglect key physics such as composition dependency. For example, Fu et al. (2019) had to adjust the effective-diffusion coefficients after every cycle of gas HnP to match the experimental results.

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Original SPE manuscript received for review 19 January 2021. Revised manuscript received for review 9 March 2021. Paper (SPE 204010) peer approved 16 March 2021.



Fig. 1—(a) Binary diffusion vs. (b) multicomponent diffusion (after Taylor and Krishna 1993).

Another widely adopted approach is to relate $D_{i,\beta}$ with the binary diffusion coefficients to include some extent of composition dependency, such as

$$D_{i,\beta} = \frac{1 - x_i}{\sum_{j \neq i}^{nc} x_j / \mathcal{D}_{ij}}, \qquad (4)$$

which was originally proposed by Fairbanks and Wilke (1950), assuming that all components other than i have the identical velocity when calculating the Fickian-type diffusion coefficient for component i, treating the mixture as a pseudobinary system including a solute component i and all other components lumped together as the solvent.

It is often used in many previous studies (Qiao 2015; Xiong 2015; Eker 2018; Yu et al. 2019), as well as commercial simulators [e.g., GEM (CMG 2018) and ECLIPSE 300 (Schlumberger, Houston, Texas, USA)], because of its simplicity. However, it can easily violate the equimolar constraints unless all the diffusion coefficients are taken to be equal, as discussed by Hoteit (2013). More importantly, such simplification is essentially equivalent to the binary diffusion and cannot account for the cross-diffusion effects.

For a multicomponent system with more than two components, the diffusive flux of a species is not just proportional to its own chemical potential gradient, also known as the cross-diffusion effects. The counterintuitive phenomena observed in experiments (Duncan and Toor 1962; Arnold and Toor 1967) include diffusion barrier (zero diffusive flux with a nonzero gradient), reverse diffusion (molecules diffuse against the gradient), and osmotic diffusion (nonzero diffusive flux without a gradient), as shown in Fig. 1b.

Though the generalized Fick's law (Eq. 2) can be used to quantify the discussed cross-diffusion effects, the multicomponent diffusion coefficients cannot be evaluated accurately because of the lack of experimental measurements. A better alternative is the MS formulation based on the diffusion coefficients that can be evaluated more easily from molecular dynamics simulation (Liu 2013) or empirical correlations (Leahy-Dios and Firoozabadi 2007).

Multicomponent Diffusion Model

We first present the mathematical formulation for multicomponent diffusion in the bulk phase. It is then incorporated into the general ADM in porous media, which allows a more accurate characterization of the diffusive flux within the tight matrix and at the matrix/fracture interface.

Multicomponent Diffusion in the Bulk Phase. The most comprehensive form of the MS diffusion formulation in the bulk phase can be written as

where \mathcal{D}_{ij} is the MS diffusion coefficient between components *i* and *j*. It can be proved that the three kinds of diffusion coefficients discussed are equal only for a binary system. It shall be noted that the chemical potential gradient here is the driving force of multicomponent diffusion, which is more rigorous and has more fundamental roots in irreversible thermodynamics (Cussler 2009).

With the equimolar constraint $\left(\sum_{i=1}^{m} \vec{J}_{i} = 0\right)$, we could rewrite the MS equation (Firoozabadi 2015) in the matrix form with (nc-1) independent fluxes,

where **B** is an $(nc - 1) \times (nc - 1)$ matrix. Each element in **B** can be expressed as

$$B_{ij} = \begin{cases} \sum_{j \neq i}^{m} \frac{x_j}{\mathscr{D}_{ij}} + \frac{x_i}{\mathscr{D}_{inc}}, & i = j, \\ \frac{x_i}{\mathscr{D}_{inc}} - \frac{x_i}{\mathscr{D}_{ij}}, & i \neq j. \end{cases}$$
(7)

J is also an $(nc-1) \times 3$ matrix composed of (nc-1) independent rows of diffusive flux vector. **X** is a diagonal matrix with **X** = diag{ $x_1, ..., x_{nc-1}$ }. μ is an $(nc-1) \times 1$ matrix, representing the chemical potential of (nc-1) independent components. $\nabla \mu$ is an $(nc-1) \times 3$ matrix,

$$\nabla \boldsymbol{\mu} = \begin{bmatrix} \frac{\partial \mu_1}{\partial x} & \frac{\partial \mu_1}{\partial y} & \frac{\partial \mu_1}{\partial z} \\ \vdots & \vdots & \vdots \\ \frac{\partial \mu_{nc-1}}{\partial x} & \frac{\partial \mu_{nc-1}}{\partial y} & \frac{\partial \mu_{nc-1}}{\partial z} \end{bmatrix}.$$

Eq. 6 can also be written to explicitly express the diffusive flux as

$$\mathbf{J} = -\frac{c_t}{RT} \mathbf{B}^{-1} \mathbf{X} \nabla \boldsymbol{\mu}, \qquad (8)$$

which calculates the diffusive flux using the chemical potentials from the phase-equilibrium calculation.

To relate the diffusive flux with the concentration gradient like those in Fick's law, the right-hand side of Eq. 8 could be rewritten by expressing the chemical potential with respect to fugacity and molar fractions (Hoteit 2013). Finally, we can rewrite Eq. 8 using the molar fraction,

where Γ , an $(nc-1) \times (nc-1)$ matrix, is known as the thermodynamic correction factor, representing the nonideality of the fluid mixture. Each element of Γ can be calculated as

$$\Gamma_{ij} = x_i \frac{\partial \ln f_i}{\partial x_j}.$$
 (10)

Hence, the multicomponent diffusion-coefficient matrix can be expressed as the product of two matrices,

where **D** is a matrix composed of D_{ij} . D_{ii} is the diagonal diffusion coefficient, often an order of magnitude larger than the off-diagonal term D_{ij} . In general, the diffusion-coefficient matrix **D** is not symmetric (i.e., $D_{ij} \neq D_{ji}$). The multicomponent diffusion coefficient D_{ij} in the bulk phase can be calculated in the matrix form by Eq. 11.

Because the diffusive flux can be calculated using Eq. 8 without explicitly evaluating the diffusion-coefficient matrix **D**, we choose only to calculate Γ or **D** when benchmarking the measured diffusion coefficients. To calculate the diffusive flux in compositional simulation, we only need to compute \mathcal{D}_{ij} and **B** using empirical models as detailed in the Appendix A.

Diffusion Model in Rock Matrix. The porous media in an unconventional tight reservoir can often be conceptualized as two subdomains, low-permeability rock matrix and high-permeability fracture network, including large-dimension hydraulic fractures (HFs) and small-dimension natural fractures (NFs). Advection often dominates the fluid transport over diffusion in fracture networks because of its high conductivity ($Pe \gg 10$), and diffusion is hence negligible. However, for the tight matrix with low permeability ($\ll 1$ md), the role of diffusion might no longer be negligible within the matrix as Pe approaches unity. During gas injection, the fracture network might be filled with highly mobile gases, while the less-mobile oil phase saturates the matrix, leading to a large concentration gradient at the matrix/fracture interface, making the diffusive flux a significant component in the total flux. Hence, an accurate prediction of multicomponent diffusion within the tight matrix and at the matrix/fracture interface is critical to capture the complex physics when simulating unconventional reservoirs.

For the diffusive flux in the porous media, the influence of porosity ϕ , tortuosity $\tilde{\tau}$, and the phase saturation S_{β} needs to be included in the effective diffusion-coefficient matrix (\mathbf{D}_{eff}) for porous media as

 $\mathbf{D}_{\rm eff} = \frac{\phi S_{\beta}}{\tilde{\tau}} \mathbf{D}.$ (12)

Because we only need to explicitly calculate the diffusive fluxes rather than the diffusion coefficients in this work, we can introduce the properties of porous media into Eq. 8 as

$$\mathbf{J}_{\beta} = -\frac{\rho_{\beta}}{RT} \frac{\phi S_{\beta}}{\tilde{\tau}} \mathbf{B}^{-1} \mathbf{X} \nabla \boldsymbol{\mu}, \qquad (13)$$

where the subscript β denotes the phase label. Here we can account for the diffusion both in oil ($\beta = o$) and gas phase ($\beta = g$). The molar density of phase ρ_{β} replaces the total molar concentration c_i because we do not designate a component as the solvent, and any component might disappear during the transport process. Chemical potential does not need the phase label, because μ_i as chemical potential of component *i* shall be the same across all different phases to achieve the thermodynamic equilibrium.

The diffusion model based on the molar concentration (e.g., Eq. 2) might encounter problems handling the intraphase diffusive fluxes (diffusion within the same phase) between two neighboring gridblocks, as shown in **Fig. 2.** Gridblock *n* has zero S_o and gridblock *m* has zero S_g , which is a very common scenario during gas injection.

The molar composition of the gas phase in gridblock m is clearly undefined. However, even if the gas phase does not exist in gridblock m, we can find the chemical potential of component i in the gas phase, $\mu_{g,i}$, the same as $\mu_{o,i}$, which can be calculated using the results from the phase-equilibrium calculation (Moortgat and Firoozabadi 2013). Similarly, the preceding formulation can handle multicomponent diffusion within the oil phase. Hence, the formulation by Eq. 13 can accurately capture the intraphase diffusive flux between these two neighboring gridblocks in Fig. 2. For the interphase diffusion (diffusive flux across the phase boundary), the flux can be handled implicitly by the phase-equilibrium calculation after every Newton's iteration once the governing mass-balance equation is solved.

Finally, we need to discretize Eq. 13 to calculate the total flux-term calculation when assembling the mass-balance equations. For the two neighboring gridblocks shown in Fig. 2, the diffusive flux from gridblock m to its interface with gridblock n can be written as

$$(\vec{J}_i)_m = \sum_{\beta} (\vec{J}_{\beta,i})_m = \sum_{\beta} \sum_{j=1}^{nc-1} (L_{\beta,ij})_m \frac{(\mu_j)_m - (\mu_j)_{mn+1/2}}{d_m},$$

where the subscript *m* denotes the properties of gridblock *m*, and the subscript mn + 1/2 denotes the properties at the interface of connected gridblocks *n* and *m*. The effective phenomenological coefficients for diffusion in porous media $L_{\beta,ij}$ can be expressed as

$$L_{\beta,ij} = \frac{\rho_{\beta}}{RT} \frac{\phi S_{\beta}}{\tilde{\tau}} B_{\beta,ij}^{-1} x_{\beta,j}, \qquad (14)$$

which modified the formulation in Moortgat and Firoozabadi (2013) by introducing the tortuosity. It is also worth mentioning that \mathbf{L}_{β} is not symmetric, like the phenomenological coefficients under constraints. We also take out the distance of gridblock *m* to the interface d_m because we are going to use the gridblock with different sizes. All the parameters (including temperature, fluid, and rock properties, but with the exception of chemical potential) are lumped into this $L_{\beta,ij}$ for the convenience of discretization. $L_{\beta,ij}$ does not have the same unit as a diffusion coefficient, but instead the same as the original Onsager phenomenological coefficient. Hence, we choose to name $L_{\beta,ij}$ as the effective Onsager coefficients for diffusion in porous media, and its corresponding matrix **L** as the matrix of effective Onsager coefficients.



Fig. 2—Connection between two neighboring gridblocks.

Similarly, the diffusive flux from gridblock n to its interface with gridblock m can also be given, and the sum of these two fluxes shall be zero because of the continuity of diffusive fluxes. Hence, we have

which represents a system of (nc - 1) equations. We can rewrite Eq. 15 in the matrix form as

where $\mathbf{L}_m = \sum_{\beta} (\mathbf{L}_{\beta})_m$.

Each element of \mathbf{L}_{β} has been given in Eq. 14. $(\mu_j)_{mn+1/2}$ can hence be explicitly expressed as

where $\boldsymbol{\mu}_{mn+1/2}$ is an $(nc-1) \times 1$ matrix.

Hence, we can write the diffusive flux between gridblocks m and n as

which is more general than those presented in Moortgat and Firoozabadi (2013), because now it can account for different gridblock sizes.

Because the matrix operations presented in Eq. 18 are computationally expensive, Moortgat and Firoozabadi (2013) recommended using $0.5(\mathbf{L}_m + \mathbf{L}_n)/d$, where *d* is the uniform gridblock size with $d = 2d_m = 2d_n$. Or, to be more consistent with this work, we can write

However, this simplification is not valid when gridblock size changes abruptly (e.g., local grid refinements near wells or fractures). Eq. 19 can also fail to be equal to the rigorous formulation of Eq. 18 when the difference between \mathbf{L}_m and \mathbf{L}_n is large (e.g., when $\mathbf{L}_m \gg \mathbf{L}_n$). Hence, we propose using another formulation that is equivalent to Eq. 18,

$$\mathbf{J} = \left(d_m \mathbf{L}_m^{-1} + d_n \mathbf{L}_n^{-1}\right)^{-1} (\boldsymbol{\mu}_m - \boldsymbol{\mu}_n).$$
(20)

The general mathematical proof of their equivalence is detailed in the Appendix B. Specifically for this problem, we can prove the equivalence of Eqs. 18 and 20 by writing the diffusive flux from gridblock m to the interface as

$$\mathbf{J} = \mathbf{L}_m \frac{\boldsymbol{\mu}_m - \boldsymbol{\mu}_{mn+1/2}}{d_m} \Rightarrow d_m \mathbf{L}_m^{-1} \mathbf{J} = \boldsymbol{\mu}_m - \boldsymbol{\mu}_{mn+1/2}.$$

Similarly, we can write the diffusive flux from gridblock n to the interface as

$$-\mathbf{J} = \mathbf{L}_n \frac{\boldsymbol{\mu}_n - \boldsymbol{\mu}_{mn+1/2}}{d_n} \Rightarrow d_n \mathbf{L}_n^{-1} \mathbf{J} = \boldsymbol{\mu}_{mn+1/2} - \boldsymbol{\mu}_n.$$

Combining these two expressions leads to

$$d_m \mathbf{L}_m^{-1} \mathbf{J} + d_n \mathbf{L}_n^{-1} \mathbf{J} = \boldsymbol{\mu}_m - \boldsymbol{\mu}_n \Rightarrow \mathbf{J} = \left(d_m \mathbf{L}_m^{-1} + d_n \mathbf{L}_n^{-1} \right)^{-1} (\boldsymbol{\mu}_m - \boldsymbol{\mu}_n).$$

At the first glance, Eq. 20 looks more computationally expensive than Eq. 18, let alone Eq. 19. But in Eq. 14, \mathbf{B}_{β}^{-1} needs to be evaluated first for both phases, and hence using Eq. 20 can skip the costly numerical matrix inversion in some cases. For example, for the scenario in Fig. 2, the formulation of Eq. 20 will only require performing numerical inversion once, as we have

$$\mathbf{L}_{m} = \frac{\rho_{o}}{RT} \frac{\phi S_{o}}{\tilde{\tau}} \mathbf{B}_{o}^{-1} \mathbf{X}_{o} \Rightarrow \mathbf{L}_{m}^{-1} = \frac{\tilde{\tau}RT}{\rho_{o}\phi S_{o}} \mathbf{X}_{o}^{-1} \mathbf{B}_{o},$$
$$\mathbf{L}_{n} = \frac{\rho_{g}}{RT} \frac{\phi S_{g}}{\tilde{\tau}} \mathbf{B}_{g}^{-1} \mathbf{X}_{g} \Rightarrow \mathbf{L}_{n}^{-1} = \frac{\tilde{\tau}RT}{\rho_{g}\phi S_{g}} \mathbf{X}_{g}^{-1} \mathbf{B}_{g}.$$

Recalling X_g is a diagonal matrix, and its inversion can be performed easily by replacing each element in the diagonal with its reciprocal, the computational complexity of which is only O(n). Hence, to complete Eq. 20, we just need to do

$$\left(d_m \mathbf{L}_m^{-1} + d_n \mathbf{L}_n^{-1}\right)^{-1} = \left(d_m \frac{\tilde{\tau} R T}{\rho_o \phi S_o} \mathbf{X}_o^{-1} \mathbf{B}_o + d_n \frac{\tilde{\tau} R T}{\rho_g \phi S_g} \mathbf{X}_g^{-1} \mathbf{B}_g\right)^{-1}$$

where only one matrix inversion is actually needed, and the computational complexity is $O(n^3)$. But even for Eq. 19, which is simplified but less accurate, we need to perform numerical matrix inversion twice as

$$\frac{1}{2} \left(\frac{\mathbf{L}_m}{d_m} + \frac{\mathbf{L}_n}{d_n} \right) = \frac{1}{2} \left(\frac{\rho_o}{d_m RT} \frac{\phi S_o}{\tilde{\tau}} \mathbf{B}_o^{-1} \mathbf{X}_o + \frac{\rho_g}{d_n RT} \frac{\phi S_g}{\tilde{\tau}} \mathbf{B}_g^{-1} \mathbf{X}_g \right).$$

Although equivalent to Eq. 20, Eq. 18 will require the performance of three numerical matrix inversions first and then two expensive matrix multiplications, resulting in computational complexity of $5O(n^3)$. We here summarize the comparison of the preceding three formulations in different scenarios, as shown in **Table 1**. It can be seen that the formulation proposed in this work (Eq. 20) is more efficient computationally than the original formulation (Eq. 18), while having the same computational accuracy. When the oil and gas coexist in both gridblocks, we can only account for the intraphase diffusion in the gas phase and neglect the intraphase diffusion in the oil phase, which can greatly reduce the computational cost from $7O(n^3)$ to $O(n^3)$. This is because the diffusion coefficient in the liquid phase is often many orders of magnitude smaller than that in the gas phase (Cussler 2009).

Hydrocarbon F	hase Number	Computati	Computational Complexity		
Gridblock m	Gridblock n	Eq. 18	Eq. 19	Eq. 20	
1	1	$5\mathcal{O}(n^3)$	$2\mathcal{O}(n^3)$	$\mathcal{O}(n^3)$	
2	1	$6\mathcal{O}(n^3)$	$3\mathcal{O}(n^3)$	$4\mathcal{O}(n^3)$	
2	2	$7\mathcal{O}(n^3)$	$4\mathcal{O}(n^3)$	$7\mathcal{O}(n^3)$ or $\mathcal{O}(n^3)$ if needed	

Table 1—Comparison of computational complexity among different formulations for diffusive flux.

Our formulation detailed previously can account for both intraphase and interphase diffusion, the nonideality of the hydrocarbon system, and the effect of porous media. It can better capture the physics during multicomponent diffusion in porous media compared with the pseudobinary diffusion model based on the classic Fick's law. After diffusive flux is explicitly calculated, it is linearly added to the advective flux (governed by Darcy's law) to form the total mass flux (Webb and Pruess 2003), which is the essence of the ADM. It is worth mentioning that the diffusive fluxes in this work are calculated from the MS equation or equivalently the generalized Fick's law, which is far more accurate than the conventional ADM based on the classic Fick's law.

An Improved Diffusion Model for Fracture/Matrix Interaction. Significant errors can arise, especially for modeling injection processes in fractured reservoirs using the conventional ADM, where advection is simply evaluated using the fracture mobility because of the single-point upstream weighting (Wu et al. 2004) and diffusion is solely calculated by the concentration gradient (Hoteit 2013). We hereby present a physics-based modification for multiphase multicomponent fracture/matrix interaction in fractured reservoirs.

To model the mass fluxes between fracture and matrix, different conceptual models have been developed, including discrete approaches such as local grid refinement, the discrete-fracture-network model, the embedded discrete-fracture model, and continuum approaches, such as DP, dual porosity/dual permeability (DPDK), or MINC.

In this work, we assume that an HF is explicitly modeled using any of the discrete approaches (local grid refinement, the discrete-fracture-network model, or the embedded discrete-fracture model), and the other part of the fractured reservoir is handled by a continuous approach (DP, DPDK, or MINC). This assumption is reasonable because other than a finite number of discrete HFs, the main flow channel for an unconventional reservoir is the continuously distributed NFs within the SRV, without which the reservoir rock is almost unproductive (i.e., the region outside SRV). The HF gridblock is de facto treated as an NF gridblock with a very high permeability and small grid size. As a result, the method presented here does not distinguish between HF and NF gridblocks but only requires that a matrix gridblock can only be connected with either an HF or NF gridblock (hereafter both referred to as NF gridblock), while connections with matrix gridblocks are not limited. A thin cell is added between fracture and matrix gridblocks to represent the fracture/matrix interface, equivalent to using three continua in the MINC, as shown in **Fig. 3**.



Fig. 3—A thin cell representing the interface between fracture and matrix.

For transport in porous media, thermodynamic variables (pressure, temperature, chemical potential) are slowly varying in space and with time. Within each timestep, the thermodynamic equilibrium is assumed for each gridblock, which is the most crucial assumption for compositional simulation. A gridblock of large volume is only acceptable where variations of thermodynamic variables are small (Pruess and Narasimhan 1985). The direct neighboring region between matrix and fracture is often subject to a large gradient of thermodynamic variables, which calls for the need to use this interface continuum to better characterize fracture/matrix interaction. A good example is a comparison work by Wu and Pruess (1988), where the authors showed that MINC could match the water-imbibition rate of fine-grid modeling of water injection, while the DP approach mismatches the 0- to 10-day results for a core-scale case and 0- to 100-day results for a field-scale case.

The added interface gridblock has the same rock properties as the matrix, but its fluid saturation and composition can equilibrate quickly with the fracture because both the advective flux and diffusive flux are directly affected by the connected fracture gridblock. The interface gridblock is treated in the same manner as a common gridblock, and their governing equations are also being solved simultaneously with the neighboring fracture and matrix gridblocks. The flux between fracture and interface gridblock is quantified in a fully transient manner instead of using a pseudosteady-state transfer function with only two subgrids (DP or DPDK).

In conventional reservoir simulation, the simple upstream (also known as upwind) weighting scheme is routinely used (Aziz and Settari 1979). As a result, the fracture relative permeability is commonly selected in estimating the mobility for flow toward the matrix (e.g., water or gas injection). However, fracture/matrix flow is physically controlled by the matrix-flow properties, which can lead to unphysical solutions or significant numerical errors using simple upstream weighting. A good analogy is the way of handling mobility for an injector well, where de facto "downstream" mobility is used [i.e., the relative permeabilities are calculated using the saturation of the rock gridblock rather than that of the well gridblock (Kazemi et al. 1978; CMG 2018)]. The added interface will act as a buffer zone to prevent overestimation of the advective flux caused by single-point upstream weighting using direct fracture/matrix connection, as discussed by Wu et al. (2004).

Specifically for gas injection, the injected gas often has a composition that is very different from the reservoir fluid, resulting in a huge concentration gradient (or more rigorously speaking, chemical potential gradient) and further substantial diffusive fluxes (Coats 1989). A direct connection between fracture and matrix will inevitably introduce errors to the early-time fluxes. This is because the average fluid composition of a large matrix gridblock apparently cannot represent the fluid composition at the fracture/matrix interface that physically controls the fracture/matrix interaction. For tight reservoirs, the pseudosteady-state transport between fracture and matrix might never be reached (Sherafati 2018), especially because of the frequent well change during HnP. The added continuum using a more representative fluid saturation and composition of the interface can hence better capture the transient mass exchange between fracture and matrix instead of simply assuming pseudosteady state.

The original MINC approach places the centroid of the fracture gridblock at the fracture/matrix interface. That is, $d_n = 0$ in Fig. 2, assuming gridblock *n* representing the NF and *m* representing fracture/matrix interface. The reason is that the harmonic average permeability controlling the fracture/matrix advection can reduce to the matrix permeability as

$$\frac{d_{nm}}{k_{mn+1/2}} = \frac{d_n}{k_n} + \frac{d_m}{k_m} = \frac{d_m}{k_m} \Rightarrow k_{mn+1/2} = k_m,$$

where d_{nm} is the nodal distance between two gridblocks and $d_{nm} = d_n + d_m$. Using matrix permeability for the fracture/matrix interface is physically correct for advection-dominant fracture/matrix interaction (e.g., water injection), which is also included in transfer function for other continuous fracture-modeling approaches, such as DP or DPDK (Lim and Aziz 1995).

However, for diffusion calculation, $d_n = 0$ will further reduce the diffusive flux by Eq. 18 or Eq. 20 into

which means the fluid-diffusion coefficients of the fracture gridblock will not influence the diffusive fluxes across the fracture/matrix interface. This is physically wrong. For example, during gas injection, the fracture is saturated with gas, and the matrix is saturated with oil. However, the diffusion coefficients now only depend on the oil-phase diffusion coefficients in the matrix, which are several orders of magnitude lower than those of the gas phase, making the diffusive fluxes across the fracture/matrix interface unimportant.

$$k_f = \frac{\phi_f w_f^2}{12}.$$
 (22)

For the fracture/matrix-interface gridblock, we assume its pore volume is an adjustable parameter to match the results of the finegrid model. Then d_m can be calculated using the proximity functions given in Pruess (1983). It is expected that its thickness $(2d_m)$ is between the width of the fracture and the dimension of the actual matrix block. The inner matrix gridblock takes the value as the total pore volume minus the pore volume of the interface gridblock. Its nodal distance to the interface gridblock is calculated using the original MINC method by Pruess (1983).

Compositional Model

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The black-oil model is the most widely used model because of its simplicity. However, compositional models are becoming more popular in recently published studies, especially for unconventional tight reservoirs (Qiao 2015; Wan 2015; Xiong 2015; Sherafati 2018; Uzun et al. 2018). This is mainly because black-oil models could not accurately handle the transport process when it is highly composition dependent (e.g., when diffusion is nonnegligible). We use a fully implicit compositional model, which can account for the nanopore confinement and geomechanical effects as presented in Tian et al. (2019). Its associated in-house simulator MSFLOW-COM is first developed by Xiong (2015) and updated by Tian et al. (2019) to better model the gas-injection process. It has been validated with the results of a commercial simulator, experimental data, and analytical solutions (Xiong 2015; Tian et al. 2019).

Transport Equations. The transport equations are nc + 1 mass-balance equations corresponding with nc + 1 components. It is assumed that a component existing in the oil and gas phases does not dissolve in water, which is a reasonable assumption except for some rare cases related to carbon dioxide (CO₂) injection. Similarly, the water component is assumed to only distribute in the water phase, with its mole fraction in the oil and gas phases being negligible. The mass-balance equation for the water component, which is assumed to be present only in the aqueous phase, can hence be written as

$$-\nabla \cdot (\rho_w \vec{v}_w) + q_w = \frac{\partial (\phi \rho_w S_w)}{\partial t}.$$
 (23)

Then, transport equations for the nc components existing in both oil and gas phases can be written as

where component *i* can be a pure hydrocarbon, nonhydrocarbon [e.g., CO_2 or nitrogen (N₂)], or pseudocomponent as long as the thermodynamic properties required by the equation of state (EOS) are specified. For this reason, it is generalized as a "hydrocarbon" component in the context of this work.

These partial-differential equations are integrated within the arbitrary representative elementary volume by following the integral finite-difference method, which is essentially equivalent to the finite-volume method (Pruess 1991).

where V_n represents the volume of a gridblock. By applying the divergence theorem, volume integrals are converted to surface integrals.

where S_n denotes the surface area of a gridblock, and \hat{n} is the outward normal unit vector that describes the spatial orientation of dS. Fluids and rock properties are evaluated by averaging them over explicitly defined gridblocks, while mass fluxes across the interface between neighboring gridblocks are approximated by the finite-difference method. The discretization of time is implemented with the backward first-order finite-difference approximation. The upstream mobility weighting is implemented. Finally, the discretized equations are written into a residual form, ready to be solved by Newton's method.

$$R_{i,n}^{t+1} = \frac{\left[V_n \phi(\rho_o S_o x_i + \rho_g S_g y_i)\right]_n^{t+1} - \left[V_n \phi(\rho_o S_o x_i + \rho_g S_g y_i)\right]_n^t}{\Delta t} - (V_n q_i)_n^{t+1} \\ -\sum_{m \in \eta_n} \left[(\rho_o x_i \lambda_o)_{nm+1/2}^{t+1} \gamma_{nm}^{t+1} \left(\Phi_{om}^{t+1} - \Phi_{om}^{t+1}\right) + \left(\rho_g y_i \lambda_g\right)_{nm+1/2}^{t+1} \gamma_{nm}^{t+1} \left(\Phi_{gm}^{t+1} - \Phi_{gm}^{t+1}\right)\right] \\ -\sum_{m \in \eta_n} \sum_{j=1}^{nc-1} A_{nm}^{t+1} (L_{ij})_{nm+1/2}^{t+1} \frac{(\mu_j)_m^{t+1} - (\mu_j)_n^{t+1}}{d_m^{t+1} + d_m^{t+1}}, \qquad (27)$$

where λ_{β} is the mobility of phase β , defined as $\lambda_{\beta} = k_{r\beta}/\mu_{\beta}$, and $(L_{ij})_{nm+1/2}^{t+1}$ is the element in the effective phenomenological coefficient matrix $\mathbf{L}_{nm+1/2}$, given earlier as

$$\mathbf{L}_{nm+1/2} = (d_m + d_n) \left(d_m \mathbf{L}_m^{-1} + d_n \mathbf{L}_n^{-1} \right)^{-1}.$$
 (28)

Similarly, the residual form of the mass-balance equation for the water component can be written as

where Φ_{β} is the flow potential of a phase and can be related to Darcy's law as

. . 1

$$\vec{v}_{\beta} = -\frac{kk_{r\beta}}{\mu_{\beta}}\nabla\Phi_{\beta} = -\frac{kk_{r\beta}}{\mu_{\beta}}\nabla(P_{\beta} + \rho_{\beta}gZ).$$
(30)

The transmissibility γ_{nm} between two connected gridblocks is defined as

where A_{nm} is the common interface area between connected gridblocks *n* and *m*; d_n is the distance from the center of gridblock *n* to the common interface between gridblocks *n* and *m*, as shown in Fig. 2; and $k_{nm+1/2}$ is the harmonically averaged absolute permeability along the connection between gridblocks *n* and *m*. Also to be noted, there is a superscript t + 1 even for A_{nm} or d_n because the area and distance both depend on rock deformation and hence are time dependent.

To handle the disappearance of certain components in the transport equations, which is a common phenomenon during injection, the transport equations are reformulated from those originally presented by Xiong (2015). The first transport equation is still the massbalance equation of the water component, which corresponds with the primary variable S_w . If the water component disappears in some instances (e.g., gas injection into fractures resulting in zero S_w), the diagonal element of the Jacobian matrix will become zero. Then, this diagonal term can be manually set to unity to avoid singularity, and S_w will be held constant as zero. The second transport equation is the sum of all nc + 1 mass-balance equations,

$$R_{2,n}^{\text{new}} = R_{w,n} + \sum_{i=1}^{n} R_{i,n}, \qquad (32)$$

which corresponds with the primary variable P_o . Based on the equimolar constraint, the total diffusive flux will be zero for $R_{2,n}^{ewc}$. As long as there is one component remaining, the diagonal element of the Jacobian matrix will still be nonzero because the total advective flow can always be driven by the pressure gradient.

The third transport equation is now the mass-balance equation of the first hydrocarbon component, which corresponds with the primary variable z_1 . If z_1 stays zero in certain cases (e.g., injecting a large volume of gas with other components), then the diagonal block matrix can become singular. Then, this diagonal element can be manually set to unity to avoid singularity, and z_1 will be held constant as zero in this case. The new system of transport equations now becomes $(R_{w,n}, R_{2,n}^{new}, R_{1,n}, ..., R_{nc-1,n})$, with the same ordering of primary variables as $(S_w, P_o, z_1, ..., z_{nc-1})$.

Phase-Equilibrium Calculation. In the compositional simulation, it is commonly assumed that the mass transfer between phases occurs much faster than fluid flow in the reservoir rock. Hence, an EOS-based phase-equilibrium calculation is a must to accurately predict the number and composition of phases at a given pressure, temperature, and overall composition. The phase-equilibrium calculation in this work is dependent on a hybrid algorithm combining successive substitution and Newton's method, as shown in **Fig. 4**. This algorithm was originally proposed to study the nanopore confinement with a dynamic pore size (Tian et al. 2019). In this work, we turn off the nanopore-confinement option in the simulator. The Peng and Robinson (1976) EOS is used in this work because of its popularity in reservoir-fluid characterization, although our phase-equilibrium algorithm can generally work with any cubic EOS.



Fig. 4—EOS-based phase-equilibrium-calculation algorithm in this work.

The phase-equilibrium calculation has two main modules: the stability test and phase-split calculation. Both are computed using the hybrid algorithm combining successive substitution and Newton's method. For the stability test considering nanopore confinement, Eq. 33 needs to be solved (Sherafati and Jessen 2017),

$$\ln\varphi_i(\mathbf{w}) + \ln W_i + \ln P_w - \ln\varphi_i(\mathbf{z}) - \ln z_i - \ln P_z = 0, \qquad (33)$$

where P_w is the pressure of the trial phase with composition w, and P_z is the phase pressure of the feed with composition z. W_i is the primary unknown of Eq. 33 and is defined as

where w_i is the molar composition of the trial phase.

The original system is stable if, and only if, $\sum_{i=1}^{\infty} W_i \leq 1$. The phase-split calculation can be skipped if the system is deemed as stable. Still, μ_i as the chemical potential of component *i* needs to be calculated to complete the diffusive flux calculation.

$$\mu_i = \mu_i^0 + RT \ln(z_i \varphi_i P_z). \tag{35}$$

If the stability test has determined that the initial feed is not stable at given pressure P and temperature T, phase-split computation is then invoked, which essentially solves the equality of fugacities,

under the material-balance constraints $z_i = \beta_L x_i + \beta_V y_i$ and $1 = \beta_L + \beta_V$. β_L is the mole fraction of the liquid phase and β_V is the mole fraction of the vapor phase; x_i is the molar fraction of component *i* in the liquid phase; and y_i is the molar fraction of component *i* in the vapor phase. The Rachford-Rice equation is used to calculate β_L . After it is solved, the molar compositions of liquid and vapor can then be computed.

After the phase-equilibrium calculation is completed, the compositional variables can be used to update the key properties of phase β such as saturation S_{β} , molar density ρ_{β} , viscosity, and many other key parameters. The chemical potential of component *i* is the same for all the phases. Hence, we can evaluate it based on any phase,

$$\mu_i = \mu_i^0 + RT \ln(y_i \varphi_i^V P_V). \tag{37}$$

The molar density of a hydrocarbon phase β can be calculated as $\rho_{\beta} = P/(Z_{\beta}RT)$, where the compressibility factor is calculated from an EOS and adjusted by a constant volume shift (Jhaveri and Youngren 1988). The pore space occupied by hydrocarbon can be evaluated as

 $1 - S_w = \frac{\beta_L}{\rho_L} + \frac{\beta_V}{\rho_V}.$ (38)

The phase saturation can then be calculated accordingly; for example, the oil saturation can be calculated as

$$S_o = \frac{\rho_V \beta_L (1 - S_w)}{\rho_L \beta_V + \rho_V \beta_L}, \qquad (39)$$

which is vital when calculating the governing transport equations.

Compositional Simulator. With a more general diffusion formulation proposed in this work, major modifications are implemented in this updated version of MSFLOW-COM to better model the gas-injection process in tight oil reservoirs. The flow chart of the updated version of MSFLOW-COM is shown in **Fig. 5**.

The data-input module reads the input from a formatted data file. The simulation-control module controls the main time loop, and it will start a run with a user-specified initial timestep size. The timestep size can be adjusted automatically to accommodate the changes of well constraint. The modules in the shaded area are all within a timestep and nested in the simulation-control module.

The data-initialization module can initialize all the secondary variables with the given primary variables and prepare for the first Newton's iteration within a timestep. Besides, it can update the well status at the beginning of each timestep if necessary.

The nonlinear solver linearizes the system of nonlinear equations using Newton's method (i.e., calculating the Jacobian matrix and the residual of governing equations, as detailed previously). After the system of linear equations is established, an iterative sparse matrix solver derived from the generalized minimal residual method is called to solve the linear system and pass the changes of primary variables δx as the solution. If δx for the current iteration level *l* is flagged as acceptable, the secondary variables can be computed with the updated primary variables x^{l+1} , similar to variable initialization.

In this work, a timestep will be cut if the program detects a change in one of the primary variables being greater than a threshold value to avoid the unphysical value of secondary variables. For example, δSw might be too large, making $S_w > 1$. If the current timestep is not converged, the program will decide whether the simulation needs another Newton's iteration (when *l* is less than the maximum allowable iterations l_{max}) or a timestep cut. If the current timestep is converged, the simulator will estimate the step size for the next timestep, reset the iteration level *l* to one, and start a new timestep. The data-output module will also be invoked to write the converged results of the current timestep in the required format according to the user's need, such as outputting the saturation data of all grids at the user-specified time. It can also write the calculated results of a well vs. time in a user-specified frequency. It can output all the primary variables after certain timesteps for the purpose of restarting calculation.

Finally, if the current time has reached the user-specified stopping time t_{stop} , the simulation will end, and the output data file will be saved and closed.



Fig. 5—Flow chart of the in-house compositional simulator MSFLOW-COM.

Model Validation

This section first compares the results of the proposed multicomponent diffusion formulation with the results of both intraphase and interphase diffusion experiments. Then, the proposed MINC3 (with three continua being fracture, interface, and matrix) is compared with the results of the fine-grid model, which is often more accurate.

Modeling Multicomponent Diffusion in the Bulk Phase. Two simulation cases were established and run to validate the implementation of the multicomponent diffusion model against the experimental data. Both laboratory measurements are obtained using the Loschmidt diffusion apparatus (Taylor and Krishna 1993). Two vertical tubes (or cells) are filled with fluids of different compositions but at the same temperature and pressure. Within the same tube, the concentration is uniform. At the time t = 0, the barrier isolating the two tubes is removed, and the diffusion is allowed to occur. After some time, the two tubes containing different fluids are separated again, and the composition of each cell can be measured for one data point. For the next data point, the experiments need to be started all over again, from the initial composition at time t = 0.

The first case simulates a ternary intraphase diffusion experiment by Arnold and Toor (1967). The ternary system composed of methane, argon, and hydrogen was maintained at 34°C (307.15 K) and 1 atm (101 325 Pa). The initial composition of the upper tube is 50.9% argon and 49.1% hydrogen. The initial composition of the lower tube is 51.5% methane and 48.5% argon. The diffusion-tube length for the upper tube, L_{up} , is 39.59 cm, with $(\pi/L_{up})^2 = 5.58 \text{ ft}^{-2}$. The diffusion-tube length for the lower tube, L_{low} , is 40.57 cm, with $(\pi/L_{low})^2 = 5.57 \text{ ft}^{-2}$. In this 1D experiment, the radius of the tube does not affect the results. A radius of 6.35 mm (¼ in.) is specified for the radial grid in this model. A uniform grid size of 8 cm is used in the axial direction, but the sizes of the uppermost and lowermost gridblocks are slightly modified to account for the length difference between the upper and lower tubes, respectively. Both the porosity and tortuosity are set as unity to simulate the bulk-fluid diffusion. After tuning the ratio between diagonal and off-diagonal terms to 1.0 : 2.3 using the method by Hoteit (2013), the agreement of laboratory data (discrete points) and simulation results (lines) becomes satisfactory. For methane, the mole fractions of the two tubes tend to become equal over time, as shown in **Fig. 6**.

But for argon, the mole fraction in the top tube increases with time first and then starts to decline as shown in **Fig. 7**, which manifests the reverse diffusion. At the beginning of the experiment, the diffusive flux of argon almost completely depends on the interactions with the other two components (off-diagonal diffusion) because the initial diagonal diffusion term for argon driven by its own concentration gradient is very small. However, as argon diffuses from the bottom to the top, the diagonal diffusion term increases and starts to dominate the process. Eventually, it is large enough to overcome the off-diagonal terms, ending the reverse diffusion, and argon diffuses from top to bottom.



Fig. 6—Comparison of methane mole fraction between simulation and laboratory data.



Fig. 7—Comparison of argon mole fraction between simulation and laboratory data.

The second case simulates a nine-component interphase diffusion experiment by McKay (1971), who investigated the bulk-phase diffusion between gas-cap gas and synthetic solvent (oil). The experiments were run with the gas at the top tube and the oil at the bottom tube to avoid the gravity-driven advection. The system was at the typical reservoir condition with a constant temperature of 158°F (343.15 K). The initial pressure was 1,763 psia (12.16 MPa), and the pressure decreased to 1,759 psia (12.13 MPa) at the end of the experiment. The thermodynamic properties of each component are summarized in **Table 2**, where C₁ represents alkane with one carbon atom (i.e., methane), *i*-C₄ represents isobutane, *n*-C₄ represents normal butane, and so forth. The critical pressure P_c , critical temperature T_c , critical volume V_c , and acentric factor ω are from NIST (2020). The constant volume-shift parameters are from Hoteit (2013).

The binary interaction coefficient δ_{ij}^{BIC} between components *i* and *j* is calculated by the critical volume V_c , and an exponent of 1.2 suggested by Oellrich et al. (1981) is used,

$$\delta_{ij}^{\text{BIC}} = 1 - \left(\frac{2\sqrt{V_{c,i}^{1/3}V_{c,j}^{1/3}}}{V_{c,i}^{1/3} + V_{c,j}^{1/3}}\right)^{1.2}.$$
(40)

The initial fluid compositions of two tubes are listed in Table 3.

In the original measurement, only the molar densities of methane, ethane, propane, and normal butane were recorded. Hence, the simulated molar densities of these components without any tuning are compared with the laboratory measurement by McKay (1971), as shown in **Figs. 8 through 11.** Some minor mismatches exist, such as methane in oil and ethane in gas, which are possibly caused by the errors in density prediction due to inaccurate volume-shift parameters or errors in the estimation of diffusion coefficients. A better match can be achieved by slightly increasing the diffusion coefficients (e.g., by a factor of 1.4), as discussed by Hoteit (2013). But overall, the simulation results derived from the multicomponent diffusion formulations presented in this work can satisfactorily match the laboratory measurement, even without any major tuning.

Component	P_c (MPa)	T_c (K)	V _c (L/mol)	ω	Volume Shift	Σ_v (cm ³)
N ₂	3.396	126.19	0.0894	0.0372	-0.176	18.50
CO ₂	7.377	304.13	0.0941	0.22394	-0.052	26.90
C ₁	4.599	190.56	0.0986	0.01142	-0.054	25.14
C ₂	4.872	305.33	0.1456	0.0993	-0.143	45.66
C ₃	4.248	369.83	0.2000	0.1524	-0.005	66.18
i-C4	3.629	407.81	0.2577	0.184	-0.006	86.70
n-C ₄	3.796	425.13	0.2549	0.201	-0.001	86.70
<i>i</i> -C ₅	3.396	460.35	0.3057	0.2296	-0.007	107.22
<i>n</i> -C ₅	3.370	469.70	0.3110	0.251	-0.006	107.22

Table 2—Fluid properties of the interphase diffusion experiment (McKay 1971; Hoteit 2013).

	N ₂	CO ₂	C ₁	C ₂	C ₃	i-C ₄	n-C ₄	<i>i</i> -C ₅	<i>n</i> -C ₅
Gas	3.68%	0.67%	75.79%	10.41%	6.72%	0.63%	1.57%	0.27%	0.26%
Oil	1.43%	0.63%	33.18%	17.09%	30.12%	4.38%	12.45%	0.40%	0.32%

Table 3—Initial fluid compositions of the top gas and bottom oil (McKay 1971).



Fig. 8—Comparison of methane molar density between simulation and laboratory data.



Fig. 9—Comparison of ethane molar density between simulation and laboratory data.



Fig. 10—Comparison of propane molar density between simulation and laboratory data.



Fig. 11—Comparison of n-butane molar density between simulation and laboratory data.

Modeling Multicomponent Diffusion between Matrix and Fracture. Now that the proposed diffusion formulation in this work has been validated with experimental data, we would like to further validate the multicomponent diffusion model for fracture/matrix interaction. Because of the lack of diffusion experiments for fracture/matrix interaction, the proposed MINC3 (with fracture, interface, and matrix) is validated with the case where a matrix block is explicitly refined (125 grids), as shown in **Fig. 12.** Although such refinement is impractical for large-scale reservoir-simulation problems, it can serve very well for the sole purpose of model validation (Wu and Pruess 1988).

The result is also compared with the conventional DP approach, which uses two continua (fracture and matrix) to model the fracture/matrix interaction. The matrix block is assumed to be a cube with an edge of 10 m. The fracture with a half-width of 0.3048 m (0.01 ft) is assumed to be connected with the matrix cube with all six faces. The matrix has a porosity of 0.12 and a permeability of 9×10^{-4} md. The fracture has a porosity of 1.0 and a conductivity of 4.5 md·m, which are typical values for tight oil reservoirs.

The fluid composition and properties are modeled after the published fluid characterization (Zhang et al. 2020a) established for a typical oil sample from the Wolfcamp Shale with eight pseudocomponents: methane (CH₄), CO₂, N₂-C₂, C₃, C_{4–6}, C_{7–15}, C_{16–24}, and C₂₅₊, as shown in **Table 4.** For a pseudocomponent, by assuming it is an alkane and has a general chemical formula as C_nH_{2n+2} , the carbon number *n* can be calculated from the molecular weight. Then, the diffusion volume Σ_{ν} can be calculated using Fuller et al. (1969). The binary interaction coefficients are also given by Zhang et al. (2020a), as listed in **Table 5**.

It is assumed that the produced gas from the separator is injected through the fracture. Initially, the fracture is filled with the injected gas, the matrix is saturated with oil, and their compositions are listed in **Table 6**. The fracture gridblock is assumed to have a volume of 100 m^3 because the fracture gridblock in the field-scale model can often exchange mass more quickly with other fracture gridblocks or wells to maintain a composition close to the injected gas during gas injection. The initial reservoir pressure of the model is 27.58 MPa (4,000 psi). The model is assumed to be isothermal at 349.82 K (170°F).

The average molar density of a component in the matrix cube vs. time can be calculated and used to demonstrate the difference among the fine grid, DP, and MINC3.



Fig. 12—(Top) Comparison among the fine grid, (bottom-left) DP, and (bottom-right) MINC3 conceptual model.

	P_c (MPa)	T_c (K)	V_c (L/mol)	ω	MW (g/mol)	Volume Shift	Σ_{v} (cm ³)
CO ₂	7.38	304.20	0.094	0.2250	44.0	0.000	26.90
CH_4	4.60	190.60	0.099	0.0080	16.0	0.000	25.14
N_2 - C_2	4.87	301.16	0.146	0.0966	30.0	0.000	45.66
C ₃	4.25	369.80	0.203	0.1520	44.1	0.000	66.18
C ₄₋₆	3.53	457.42	0.292	0.2260	69.2	0.000	107.22
C ₇₋₁₅	2.58	581.65	0.510	0.4020	138.1	0.042	209.82
C ₁₆₋₂₄	2.16	828.11	0.983	0.7647	259.5	-0.154	373.98
C_{25+}	1.08	987.81	1.483	0.8755	377.6	0.263	558.66

Table 4—Thermodynamic properties of the Wolfcamp oil (after Zhang et al. 2020a).

	CO ₂	CH_4	N ₂ -C ₂	C ₃	C ₄₋₆	C ₇₋₁₅	C ₁₆₋₂₄	C ₂₅₊
CO ₂	0.00	0.08	0.08	0.10	0.06	0.06	0.05	0.05
CH_4	0.08	0.00	1.71×10^{-3}	5.74×10^{-3}	1.38×10^{-2}	3.13×10^{-2}	8.14×10^{-2}	5.98×10^{-2}
N ₂ -C ₂	0.08	1.71×10^{-3}	0.00	1.19×10^{-3}	5.29×10^{-3}	1.71×10^{-2}	5.68×10^{-2}	3.91×10^{-2}
C ₃	0.10	5.74×10^{-3}	1.19×10^{-3}	0.00	1.47×10^{-3}	9.39×10^{-3}	4.25×10^{-2}	2.71×10^{-2}
C ₄₋₆	0.06	1.38×10^{-2}	5.29×10^{-3}	1.47×10^{-3}	0.00	3.47×10^{-3}	2.88×10^{-2}	1.63×10^{-2}
C ₇₋₁₅	0.06	3.13×10^{-2}	1.71×10^{-2}	9.39×10^{-3}	3.47×10^{-3}	0.00	1.26×10^{-2}	4.82×10^{-3}
C ₁₆₋₂₄	0.05	8.14×10^{-2}	5.68×10^{-2}	4.25×10^{-2}	2.88×10^{-2}	1.26×10^{-2}	0.00	1.89×10^{-3}
C_{25+}	0.05	5.98×10^{-2}	3.91×10^{-2}	2.71×10^{-2}	1.63×10^{-2}	4.82×10^{-3}	1.89×10^{-3}	0.00

Table 5—Binary interaction coefficients of the fluid system (Zhang et al. 2020a)

	CO_2	CH_4	N_2 - C_2	C ₃	C ₄₋₆	C ₇₋₁₅	C ₁₆₋₂₄	C_{25+}
Gas	0.80%	70.67%	16.32%	8.82%	3.33%	0.06%	0.00%	0.00%
Oil	0.48%	35.17%	9.72%	8.01%	11.73%	24.00%	5.92%	4.96%

Table 6—Fluid compositions of the injected gas and matrix oil.

As shown in Table 6, methane (or C_1) is the main component for injected gas. Its changing average molar density with respect to time is plotted in **Fig. 13**. The MINC3 can match the result with the fine-grid model satisfactorily after tuning the volume ratio of the interface gridblock to the total matrix block (or equivalently, the thickness of the interface). For the case that achieves the match, the interface has a thickness of 1.65 m, taking approximately 70% of the total matrix volume. The conventional DP starts to deviate at approximately 70 days and generally overestimates the diffusive flux of C_1 into the matrix, where the average molar density of C_1 in the matrix block is much higher than that predicted by the fine-grid model, as shown in Fig. 13. After 600 days, MINC3 also starts to deviate from the fine-grid model but is still much better than DP, as shown in **Fig. 14**. This is somewhat acceptable for practical reservoir simulation because gas injection rarely lasts 1 year, let alone 600 days.



Fig. 13—Comparison of C_1 molar density among the three models (t < 600 days).



Fig. 14—Comparison of C₁ molar density among the three models.

As demonstrated in Table 6, C_{4-6} and C_{7-15} are the main components of reservoir oil, and their average molar density in the matrix cube vs. time are also plotted and compared among the fine grid, DP, and MINC3, as shown in **Figs. 15 and 16**. Except for some deviation at the early time, MINC3 can reasonably match the result of the fine-grid model, while DP can only match the fine-grid model at a very early time (t < 70 days). It can also be seen that the classic DP approach will overestimate the diffusive flux of C_{4-6} leaving the matrix, where the average molar density of C_{4-6} in the matrix block is significantly lower than that predicted by the fine-grid model, as

shown in Fig. 15. For a single matrix cube, DP predicts a recovery factor of 58.2% for C_{4-6} , higher than that predicted by the fine-grid case as 50.7%. Similarly, overestimation of diffusive flux can also be found for C_{7-15} , as shown in Fig. 16. For the heaviest component, C_{25+} , MINC3 achieves a better match with the fine-grid model than DP, as shown in **Fig. 17**. This is because the interface's fluid composition is treated the same as that of the matrix in the conventional DP model, which is physically wrong. The fracture/matrix interface, where the largest composition gradient (or, more fundamentally, chemical potential gradient) exists, shall not have the same fluid composition as the matrix in the model.



Fig. 15—Comparison of C_{4-6} molar density among the three models.



Fig. 16—Comparison of C_{7-15} molar density among the three models.

Simulation of Gas HnP with Multicomponent Diffusion

A conceptual model is established using the typical completion design in the Wolfcamp Shale to investigate the multicomponent diffusion in fractured unconventional reservoirs. The conceptual model for the SRV uses a Cartesian grid system consisting of cubic gridblocks with a uniform size of 10 m. There are three gridblocks in the *x*-direction, leading to a stage spacing of 30 m (98.43 ft), which matches the typical completion design in this region as detailed in Zhang et al. (2020a). There are seven gridblocks in the *y*-direction of the SRV, which corresponds with a fracture half-length of 70 m (229.66 ft). There are seven gridblocks in the *z*-direction of the SRV, which corresponds with a fracture height of 70 m (229.66 ft). By assuming each stage is identical, a closed flow boundary is imposed because of symmetry. It is worth mentioning that the "stage" in our model is a half-stage by assuming a mirror plane perpendicular to the *y*-direction. Hence, a factor of 200 should be used to scale any production or injection rates for a horizontal well drilled in the *x*-direction with 100 full stages.



Fig. 17—Comparison of C_{25+} molar density among the three models.

The SRV within a stage is assumed to behave like a fractured reservoir with a dense fracture network as a result of massive reservoir stimulation. Hence, we choose not to explicitly place a discrete HF in the model. Instead, the MINC approach with three continua (including fracture, matrix, and their interface) is used, as proposed previously. Each primary gridblock is subdivided into three secondary gridblocks. The thickness (or, equivalently, the volume) of the fracture/matrix interface is obtained by matching the fine-grid model. The rock properties of the matrix and fracture are summarized in **Table 7**, most of which are modified from Zhang et al. (2020a). A matrix tortuosity of 4.1 is used for diffusion calculation, which is from Gupta et al. (2020).

Rock Type	Properties	Value	Unit
	Permeability	2.961×10 ⁻¹⁶ (0.30)	m ² (md)
	Porosity	0.1	%
Fracture	Spacing	10	m
	Width	6.096×10 ⁻³ (0.02)	m (ft)
	Tortuosity	1.0	unitless
	Permeability	2.961×10 ⁻¹⁹ (0.30)	m² (µd)
Matrix	Porosity	12.0	%
	Tortuosity	4.1	unitless

Table 7—Rock properties of the conceptual model for Wolfcamp Shale.

The fluid properties are the same as the validation case presented previously. The initial water saturation is 0.36. As mentioned in Zhang et al. (2020a), water is first injected to mimic the typical hydraulic-fracturing process in this region. The SRV model is then depleted for 4 years with a minimum bottomhole pressure of 8.27 MPa (1,200 psi). Because depletion is still the primary production method for tight oil reservoirs, a case with depletion only is first simulated for 10 years to serve as a base case in contrast to the gas HnP cases.

Three different scenarios are simulated and compared: the DP model with multicomponent diffusion (DP w/Diff), the MINC3 model with multicomponent diffusion (MINC w/Diff), and the MINC3 model without multicomponent diffusion (MINC w/o Diff). The oil rates of the three scenarios are indistinguishable, as shown in **Fig. 18. Fig. 19** can further confirm this observation by comparing the cumulative production of C_{7-15} , which accounts for 24.00% of the reservoir oil and 52.41% of the stock-tank oil in molar fraction.

For the gas rates, depletion scenarios considering the multicomponent diffusion have a slightly higher value than the scenario neglecting the multicomponent diffusion, as shown in **Fig. 20**. This is because the diffusion fluxes are more prominent in the gas phase than in the oil phase. Also, it can be seen that for both oil and gas rates, the DP model yields the same result as the MINC3 model during the primary depletion.

Then, the well is injected with gas under a constant surface rate of 2×10^3 m³/d (or 4×10^5 m³/d for the entire horizontal well) and maximum bottomhole pressure of 48.3 MPa (7,000 psi) for 50 days. The base scenario uses a soaking time of 10 days. The well is put back into production with a minimum bottomhole pressure of 8.27 MPa (1,200 psi) for 300 days. The duration of each HnP stage is the same as detailed by Zhang et al. (2020a).

The differences between MINC w/Diff and DP w/Diff are compared for gas HnP. It can be seen that gas HnP can substantially improve the oil rate in addition to the depletion case (Dep w/Diff), as shown in **Fig. 21**, which is consistent with the field observations. Also, **Fig. 22** shows that the DP approach will lead to a higher cumulative production of C_{7-15} , which is the main component of the produced oil. This can be explained by recalling the single-matrix-block case presented previously in this work, where the DP approach tends to overestimate the diffusive fluxes of heavy components from the matrix to fracture compared with the fine-grid and MINC approaches.











Fig. 20—Comparison of gas rates for three depletion scenarios.







Fig. 22—Comparison of cumulative C7-15 production between MINC and DP.

The difference between MINC and DP is less noticeable with respect to the cumulative oil production measured by the volume, as shown in **Fig. 23.** Also, **Fig. 24** shows that the gas rate predicted by the DP approach is slightly higher than that predicted by the MINC approach, especially at the early time of each production cycle. This can be explained by recalling the single-matrix-block case presented previously, where the DP approach tends to overestimate the flux of light components compared with the fine-grid and MINC approaches. With nearly the same volume of oil produced, the DP approach predicted a much higher component RF for C₇₋₁₅, meaning that the oil produced in the DP model is richer in C₇₋₁₅ and hence heavier. This implies that light components in gas will find it easier to mix with the matrix oil using the DP approach rather than the three-subgrid MINC approach. Numerically, the MINC approach can better match the fine-grid model than the DP approach. Physically, this is because the added continuum uses a more representative fluid saturation and composition of the matrix/fracture interface and hence can better capture the transient mass fluxes between the fracture and matrix.

Gas HnP with and without considering the multicomponent diffusion is then simulated using the MINC3 approach to show the effect of molecular diffusion on oil rates. As shown in **Fig. 25**, considering multicomponent diffusion can significantly enhance the oilproduction rate. Using the total oil in place as 2.2×10^4 m³ (stock-tank condition), the oil RF for depletion (Dep) is estimated to be 11.33% after 10 years. Gas HnP with diffusion (MINC w/Diff) yields an oil RF of 16.74% using the MINC approach. The gas HnP without diffusion (MINC w/o Diff) will underestimate the cumulative oil production and yield an RF of 15.13%, as shown in **Fig. 26**. The improvement factor for oil with diffusion is 1.48, which is defined as the ratio of cumulative production after an improved oil recovery/enhanced oil recovery approach to that after the primary depletion. Typically, this improvement factor is between 1.3 and 1.8 for gas HnP according to various field observations (Wang et al. 2017). Without accounting for the multicomponent diffusion, the improvement factor is estimated as 1.34. When neglecting diffusion, the gas/oil mixing will be driven by advection only, and hence fewer light components can enter the matrix. More injected gas will stay in fracture networks and can easily flow back to the surface (depicted by the early gas-rate peaks in **Fig. 27**) once the well is open for production, leading to an overestimation of the gas rate.











Fig. 25—Comparison of oil rates for gas HnP with and without diffusion.



Fig. 26—Comparison of cumulative oil production for gas HnP with and without diffusion.



Fig. 27—Comparison of gas rates for gas HnP with and without diffusion.

Soaking is a very important phase within a gas HnP cycle. During the soaking period, the main recovery mechanism is the mass exchange between the injected gas and the in-situ oil in the matrix, which enables the subsequent oil swelling, oil-viscosity reduction, gas/oil interfacial-tension reduction, and vaporizing or condensing (Wang et al. 2017). A recent sensitivity study using a commercial simulator (Zhang et al. 2020b) concluded that without considering the multicomponent diffusion, the effect of soaking time on the oil production is negligible when the injection and production times are held constant.

Here, a sensitivity study on soaking time is performed for gas HnP using the in-house simulator accounting for multicomponent diffusion. The soaking time is set to be 0, 10, 20, and 30 days (not plotted), with the injection and production times being held as constant. As shown in **Fig. 28**, a longer soaking time will lead to a slightly higher oil rate. It is more visible in **Fig. 29** that a longer soaking time will lead to a lower peak gas rate, implying a better oil/gas mixing during the soaking stage because of diffusion. The simulated RFs after six HnP cycles are 16.604, 16.743, 16.774, and 16.835% for a soaking time of 0, 10, 20, and 30 days, respectively.

However, the sensitivity study for cases without the multicomponent diffusion demonstrates a much smaller oil-rate difference regarding different soaking times, as shown in **Fig. 30**. The peak gas rate is almost not affected by increased soaking duration, as shown in **Fig. 31**, which implies that the oil/gas mass exchange solely by advection is small during soaking. The simulated RFs after six HnP cycles are 15.094, 15.131, 15.088, and 15.108% for a soaking time of 0, 10, 20, and 30 days, respectively.

Overall, gas HnP performance is more sensitive to the soaking duration when the multicomponent diffusion is included in the model. Although increasing soaking time will lead to a higher RF after considering diffusion, the incremental of RF is not large enough to support a prolonged soaking time, which is similar to the finding by Cronin et al. (2020). This is because increased soaking time will reduce the production time in the long run, which can extend the payback period for a field project. However, an operator might consider increasing the soaking time to reduce the excessive gas flowback right after the gas injection because soaking does not do any harm from the technical perspective.











Fig. 30-Effect of soaking time on oil rates for gas HnP without diffusion.



Fig. 31—Effect of soaking time on gas rates for gas HnP without diffusion.

Conclusions

We present a modified formulation to model the multicomponent diffusion driven by the chemical potential gradients. An improved ADM is proposed for the fractured reservoirs, where fracture, matrix, and their interface are represented by three different yet interconnected flow domains to honor the continuity of multiphase flux at the fracture/matrix interface. The added interface using a more accurate fluid saturation and composition of the interface can hence better capture the transient mass fluxes between fracture and matrix. The proposed approach is also implemented in an in-house compositional simulator, MSFLOW-COM.

Our model is validated with both intraphase and interphase diffusion experiments. Then, the improved multicomponent diffusion model for fracture/matrix interaction is also compared with a fine-grid model. The proposed MINC3 model (with continua being fracture, interface, and matrix) can better match the result of the fine-grid model than the DP model, which only obtains a fair match at the early time.

Then, a gas HnP well in the Permian Basin is simulated to investigate the effect of multicomponent diffusion within the fractured tight oil reservoir. Our simulation shows that diffusion has a minor effect on the oil RF of primary depletion because oil is the dominant phase in the reservoir. However, for the gas rates, depletion considering diffusion has a slightly higher value than simulation neglecting diffusion.

Simulating gas injection without considering diffusion will underestimate the oil RF but overestimate the gas rate compared with the simulation considering diffusion. This is because when neglecting diffusion, the gas/oil mixing will be driven by advection only, and the injected gas will mainly stay in fracture networks and will easily flow back to the surface after the well is open for production.

When diffusion is included in the model, gas HnP performance becomes more sensitive to the soaking time than the model without diffusion. Although increasing soaking time will lead to a higher RF after considering diffusion, the incremental of RF is not sufficiently large to justify a prolonged soaking.

Simulation using the DP approach will overestimate the RFs of heavy components but yield a similar cumulative oil RF compared with the MINC approach. This is because the DP approach overestimates the diffusive flux between matrix and fracture compared with the fine-grid or MINC approaches.

Nomenclature

- A_{nm} = interface area between connected gridblocks *n* and *m*, m²
 - $c_i = \text{molar concentration of component } i, \text{ mol m}^{-2}$
- d_n = distance from the center of gridblock *n* to the interface, m
- $D_{i,\beta}$ = effective diffusion coefficient of component *i* in phase β , m²s⁻¹
- D_{ij} = binary diffusion coefficient, m²s⁻
- \mathcal{D}_{ij} = multicomponent diffusion coefficient, m²s⁻¹
- $\mathcal{D}_{ij} = Maxwell-Stefan diffusion coefficient, m^2 s^{-1}$
- $f_i =$ fugacity of component *i*, Pa
- g =gravitational acceleration, m s⁻²
- $\vec{J}_i =$ molar diffusive flux of component *i*, mol m² s⁻¹
- $k = permeability, m^2$

 $k_{nm+1/2}$ = harmonically averaged permeability between gridblocks *n* and *m*, m²

- $k_{r\beta}$ = relative permeability of phase β , unitless
- \vec{K}_i = phase equilibrium ratio of component *i*, unitless
- L_{ij} = effective phenomenological coefficients in porous media, mol² J⁻¹ m⁻¹ s⁻¹
- $P_c = critical pressure, Pa$
- $P_{\beta} =$ fluid pressure of phase β , Pa
- $q_i = \text{sink/source term of component } i, \text{ mol m}^{-3} \text{ s}^{-1}$
- R =universal gas constant, J K⁻¹ mol⁻¹
- S_n = surface area of gridblock n, m²
- $S_{\beta} =$ fluid saturation of phase β , unitless

- t = time, seconds
- T =temperature, K
- $T_c =$ critical temperature, K
- $\vec{v}_{\beta} =$ velocity vector of phase β , m s⁻¹
- $V_c = \text{critical volume, m}^3$
- V_n = bulk volume of gridblock n, m³
- $w_f =$ fracture width, m
- w_i = mole fraction of component *i* in the trial phase, unitless
- x_i = mole fraction of component *i* in the oil phase, unitless
- \mathbf{x}^{l} = vectore of the primary unknowns at the *l*th iteration
- y_i = mole fraction of component *i* in the gas phase, unitless
- z_i = overall mole fraction of component *i*, unitless
- Z = height above mean sea level, m
- $Z_{\beta} =$ compressibility factor of phase β , unitless
- β_L = mole fraction of liquid phase, unitless
- β_V = mole fraction of vapor phase, unitless
- η_n = set of gridblocks that are connected with gridblock *n*
- δ_{ij}^{BIC} = binary interaction coefficient between components *i* and *j*, unitless
- γ_{nm} = transmissibility between connected gridblocks *n* and *m*, m²
- Γ = matrix of thermodynamic correction factors, unitless
- $\lambda_{\beta} = \text{mobility of phase } \beta, \text{Pa·s}$
- μ_i = chemical potential of component *i*, J mol⁻¹
- $\mu_{\beta} = \text{viscosity of phase } \beta, \text{Pa·s}$
- $p_i^{\beta} =$ fugacity coefficient of component *i* in phase β , unitless

 $\phi = \text{porosity}, \text{unitless}$

- $\Phi_{\beta n}$ = flow potential of phase β at gridblock *n*, Pa
- $\rho_{\beta} = \text{molar density of phase } \beta, \text{ mol m}^{-3}$
- $\tilde{\tilde{\tau}} = \text{rock tortuosity, unitless}$
- ω_i = acentric factor of component *i*, unitless

Acknowledgments

The National Natural Science Foundation of China is thanked for providing financial support (Grant No. 51974356) to this study. Support by the Research Institute of Petroleum Exploration and Development is also acknowledged.

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Appendix A—Calculation of the MS Diffusivities

In this work, the methodology proposed by Leahy-Dios and Firoozabadi (2007) is slightly modified to compute the MS diffusion coefficient (or MS diffusivity) \mathcal{D}_{ij} , and subsequently **B**. First, the generalized Vignes equation is used to estimate \mathcal{D}_{ij} using the diffusion coefficients at infinite dilution.

where $D_{ij}^{x_i \to 1}$ is the binary diffusion coefficient when component *i* is infinitely diluted in component *j*, and $D_{ij}^{x_i \to 1}$ is a ternary diffusion coefficient representing the interaction between components *i* and *j* while both are infinitely diluted in a third component *k*.

For estimating $\mathcal{D}_{ii}^{x_k \to 1}$, Kooijman and Taylor (1991) proposed

$$\mathcal{D}_{ij}^{\mathbf{x}_k \to 1} = \left(D_{ik}^{\mathbf{x}_k \to 1} D_{jk}^{\mathbf{x}_k \to 1} \right)^{1/2}, \quad \dots \quad (A-2)$$

which was implemented in this work. The empirical equation by Leahy-Dios and Firoozabadi (2007) correlates $D_{ij}^{x_j \to 1}$ at given *P* and *T* with the diffusivity at low pressure, D_{ij}^0 , using the reduced pressure and temperature,

$$\frac{\rho_j D_{ij}^{x_j \to 1}}{\rho^0 D_{ij}^0} = A_0 \left(\frac{T_{r,j} P_{r,i}}{T_{r,i} P_{r,j}} \right)^{A_1} \left(\frac{\tilde{\mu}_j}{\tilde{\mu}^0} \right)^{A_2 + A_3}, \quad \dots \quad (A-3)$$

where the reduced pressure is defined as $P_{r,i} = P/P_{c,i}$ and the reduced temperature is defined as $T_{r,i} = T/T_{c,i}$. The superscript 0 denotes the properties at low pressure (approximately 1 atm). The molar density at low pressure ρ^0 is evaluated using the ideal-gas law. The binary diffusion coefficient at low pressure D_{ij}^0 is evaluated using the method by Fuller et al. (1969),

$$D_{ij}^{0} = \frac{101.325 \left(1/MW_{j} + 1/MW_{i}\right)^{1/2} T^{1.75}}{P\left[\left(\Sigma \nu\right)_{i}^{1/3} + \left(\Sigma \nu\right)_{j}^{1/3}\right]^{2}}, \qquad (A-4)$$

where *T* is the temperature (in K), *P* is the pressure (in Pa), and MW_i is the molecular weight (in g/mol). The diffusion volume of component *i* is $(\Sigma v)_i$ (in cm³), which is the sum of the atomic diffusion volume based on the chemical formula of *i* (Fuller et al. 1969).

The low-pressure viscosity $\tilde{\mu}^0$ is evaluated using the correlation by Stiel and Thodos (1961). ρ_j is the molar density of pure component *j* at given *P* and *T* and is evaluated using the Peng and Robinson (1976) EOS with constant volume shifts. $\tilde{\mu}_j$ is the viscosity of pure component *j* at given *P* and *T* and is evaluated using the corresponding-state method by Pedersen and Fredenslund (1987). From our experience, we believe that the Jossi-Stiel-Thodos correlation (Jossi et al. 1962) can give a better estimation of viscosity for light and intermediate hydrocarbon components (C₁₋₉), while the Pedersen and Fredenslund (1987) method can give a fair estimation for heavy components (C₁₀₊) without being tuned by the experimental data. Hence, we choose to combine the above two correlations to calculate the viscosity of pure component at given *P* and *T* in this work. The constants in Eq. A-3 are given by Leahy-Dios and Firoozabadi (2007) using the nonlinear least-squares analysis. After \mathcal{D}_{ij} is calculated from Eq. A-1, we can proceed to calculate each element in the matrix **B** using Eq. 7. Then, its inverse matrix \mathbf{B}^{-1} can be computed (solving $\mathbf{BB}^{-1} = \mathbf{I}$ using lower–upper decomposition in this work) and further used to calculate the diffusive flux.

Appendix B—Proof for the Equivalence of Two Diffusion Formulations

Our objective is to prove the equivalence of two diffusion formulations in matrix form,

$$\mathbf{J} = \left(d_m \mathbf{L}_m^{-1} + d_n \mathbf{L}_n^{-1} \right)^{-1} (\boldsymbol{\mu}_m - \boldsymbol{\mu}_n), \mathbf{J} = \mathbf{L}_m (\mathbf{L}_m d_n + \mathbf{L}_n d_m)^{-1} \mathbf{L}_n (\boldsymbol{\mu}_m - \boldsymbol{\mu}_n)$$

which is equivalent to proving a more general mathematical form as

where $\mathbf{A} = \mathbf{L}_m/d_m$ and $\mathbf{B} = \mathbf{L}_n/d_n$. First, we can have

$$\begin{split} \mathbf{A}(\mathbf{A}+\mathbf{B})^{-1}\mathbf{B} &= (\mathbf{A}+\mathbf{B}-\mathbf{B})(\mathbf{A}+\mathbf{B})^{-1}\mathbf{B},\\ &= (\mathbf{A}+\mathbf{B})(\mathbf{A}+\mathbf{B})^{-1}\mathbf{B} - \mathbf{B}(\mathbf{A}+\mathbf{B})^{-1}\mathbf{B}\\ &= \mathbf{B} - \mathbf{B}(\mathbf{A}+\mathbf{B})^{-1}\mathbf{B}. \end{split}$$

Similarly, we can have

$$\begin{split} \mathbf{B}(\mathbf{A} + \mathbf{B})^{-1}\mathbf{A} &= \mathbf{B}(\mathbf{A} + \mathbf{B})^{-1}(\mathbf{A} + \mathbf{B} - \mathbf{B}), \\ &= \mathbf{B}(\mathbf{A} + \mathbf{B})^{-1}(\mathbf{A} + \mathbf{B}) - \mathbf{B}(\mathbf{A} + \mathbf{B})^{-1}\mathbf{B}, \\ &= \mathbf{B} - \mathbf{B}(\mathbf{A} + \mathbf{B})^{-1}\mathbf{B}. \end{split}$$

Combining the preceding two will give us

$$\mathbf{A}(\mathbf{A} + \mathbf{B})^{-1}\mathbf{B} = \mathbf{B}(\mathbf{A} + \mathbf{B})^{-1}\mathbf{A}.$$
 (B-2)

Thus, we can derive

$$\begin{aligned} \mathbf{A}(\mathbf{A} + \mathbf{B})^{-1}\mathbf{B}\big(\mathbf{A}^{-1} + \mathbf{B}^{-1}\big) &= \mathbf{A}(\mathbf{A} + \mathbf{B})^{-1}\mathbf{B}\mathbf{A}^{-1} + \mathbf{A}(\mathbf{A} + \mathbf{B})^{-1}\mathbf{B}\mathbf{B}^{-1}, \\ &= \mathbf{B}(\mathbf{A} + \mathbf{B})^{-1}\mathbf{A}\mathbf{A}^{-1} + \mathbf{A}(\mathbf{A} + \mathbf{B})^{-1}, \\ &= \mathbf{B}(\mathbf{A} + \mathbf{B})^{-1} + \mathbf{A}(\mathbf{A} + \mathbf{B})^{-1}, \\ &= \mathbf{I} \end{aligned}$$

Finally, we can use the definition of the inverse matrix and complete the proof as

 $\mathbf{A}(\mathbf{A} + \mathbf{B})^{-1}\mathbf{B}(\mathbf{A}^{-1} + \mathbf{B}^{-1}) = \mathbf{I} \Rightarrow \mathbf{A}(\mathbf{A} + \mathbf{B})^{-1}\mathbf{B} = (\mathbf{A}^{-1} + \mathbf{B}^{-1})^{-1}.$