Modelling and simulation of wormhole formation during acidization of fractured carbonate rocks

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Abstract

Acidizing is a commonly used stimulation treatment for carbonate reservoirs, performed by injecting acid into the near-wellbore rocks to remedy drilling damage, or to create, by mineral dissolution, deeply-penetrating, narrow, high-permeability channels, termed wormholes. Wormhole formation in un-fractured matrix has been widely studied by numerical modelling, but little work has been done for fractured cases, which may cause wormholes to propagate quite differently. In this study, a continuum model with explicit fractures is developed, where the governing equations are discretized by the finite-volume method. This model produces the correct dissolution patterns on a 2-D un-fractured domain discretized by Delaunay triangulation. Then it is used to examine wormhole formation in cases with single and multiple fractures, including consideration of characteristics such as fracture orientation, pattern and aperture. Fractures oriented along the flow direction can dominate the wormhole propagation, while fractures perpendicular to the flow not only change the number of branches, but also the orientation of the wormhole. Because of the positive feedback associated with fractures, the flow+dissolution process typically uses only some of the available fracture pathways. Perhaps the most significant result is that the optimum injection rate is almost unchanged from that of the same un-fractured medium.

Keywords: Reactive flow; Acidization; Wormhole; Fractured carbonate rocks; Discrete fracture; Finite volume method.

1. Introduction

Many subsurface operations – such as drilling, cementing, well completion, and production – can result in damage to the rock formation near the wellbore, decreasing the permeability and resulting in a reduction in oil or gas production. Acidizing is one type of treatment that is employed to increase the permeability around the wellbore. During this process, acid is injected into the rock under a pressure that is lower than the pressure that would cause the rock to fracture. The acid reacts with and dissolves some of the rock components, which can – in favourable cases – overcome some of the induced damage, or just enhance the local permeability. In carbonate rocks, acidizing can not only reduce the damage, but it also can create conductive channels that extend some distance into the rock mass, known as wormholes, whose hydraulic conductivity is several orders of magnitude larger than that of the porous medium.

A large number of experimental observations and numerical simulations (Fredd and Fogler, 1998;
Golfier et al., 2002; Kalia and Balakotaiah, 2007; Kalia and Balakotaiah, 2009; Maheshwari and Balakotaiah, 2013b; Maheshwari et al., 2014; Mostofizadeh and Economides, 1994; Panga et al., 2002) have demonstrated that the dissolution structure and wormhole morphology depend on the combined effects of injection rate, reaction kinetics, heterogeneity of rock and reservoir temperature. For example, at very low injection rate, all acid reacts with the rock before penetrating deeply into the medium. As a result, the borehole wall is essentially dissolved completely, and this leads to the face dissolution pattern. On the contrary, when acid is injected into the rock very quickly, it invades the majority of the pores of the rock, increasing the porosity uniformly, leading to the uniform dissolution pattern. As the injection rate increases, between these lower and upper extremes, conical, wormhole and ramified dissolution patterns are formed. As a practical matter, laboratory core floods show that the volume of acid required to break through the core, i.e. the breakthrough volume, is a minimum when the wormhole dissolution pattern is formed. Consequently, for acidizing in a carbonate reservoir, the creation of deep and thin wormholes, using a minimal volume of injected acid, is economically favourable. This pattern is also the one that is believed to be most desired for flow enhancement (Economides and Nolte, 2000). Therefore, there is a practical motivation for seeking to understand the sets of conditions and characteristics which lead to the development of wormholes.

To describe the acidization process and wormhole formation, several models have been proposed over the last few decades, based on different approaches and focused on different scales. These models can be broadly classified into four types (Maheshwari et al., 2013): (1) dimensionless model (Daccord et al., 1993a; Daccord et al., 1993b; Daccord et al., 1989); (2) capillary tube model (Buijse, 2000; Gdanski, 1999; Hung et al., 1989; Schechter and Gidley, 1969); (3) network model (Budek and Szymczak, 2012; Fredd and Fogler, 1998; Hoefner and Fogler, 1988; Kang et al., 2010; Tansey, 2014) and (4) continuum model (Cohen et al., 2008b; Cohen et al., 2007; Ghommem et al., 2015; Golfier et al., 2002; Izgec et al., 2010; Kalia and Balakotaiah, 2007; Kalia and Balakotaiah, 2009; Liu et al., 1997; Maheshwari et al., 2014; Maheshwari et al., 2013; Panga et al., 2005; Ratnakar et al., 2013; Safari et al., 2016; Tabasy and Rashidi, 2015). Reviews of these models, along with summaries of their assumptions and limitations, can be found in (Fredd and Miller, 2000; Ghommem et al., 2015; Golfier et al., 2002; Maheshwari et al., 2013; Panga et al., 2005).

The continuum model has been widely used over the last few years because it is better at forecasting the dissolution patterns observed in experiments, and it accurately estimates the breakthrough volumes. The continuum model for carbonate acidization that was first developed by Liu et al. (1997), is, unfortunately, not valid in the mass transfer controlled regime. Subsequently, Golfier et al. (2002) presented a model considering the effect of mass transfer but it is not valid in the kinetic regime. Based on previous work, Panga et al. (2005) presented a continuum model that can capture both the kinetic and mass transfer controlled regime simultaneously, by defining two concentration variables and a mass transfer coefficient. Afterwards, the continuum model has been widely used to investigate the effect of flow geometry (linear and radial) (COHEN et al., 2008a; Cohen et al., 2008b; Kalia and Balakotaiah, 2007), the type of injected acid (Maheshwari and Balakotaiah, 2013a; Maheshwari et al., 2015; Maheshwari et al., 2014; Ratnakar et al., 2012; Ratnakar et al., 2013), medium heterogeneity (Izgec et al., 2010; Kalia and Balakotaiah, 2009), completion methods (Kalia and Balakotaiah, 2010), and reservoir temperature (Kalia and Glasbergen, 2009; Kalia and Glasbergen, 2010) on wormhole propagation and optimum injection
It is generally accepted that carbonate reservoirs are heterogeneous, with complex spatial arrangements of rock types distributed over multiple length-scales (Pickup et al., 2012). It is also appreciated that carbonate reservoirs can be affected by fractures that may have complex and multi-scale spatial distributions (Questiaux et al., 2010; Spence et al., 2014). Predicting the flow effects of the fractures is a major topic in itself, but one of the methods is to treat the fractures as equivalent porous media, where the elements of the mesh that represent fractures are usually assigned to have lower porosity and higher permeability compared with the properties of the matrix rocks. That approach will be adopted here, using line-like elements to capture the geometrical localization of the fractures and their properties. Relative to the focus of this paper, the existing studies show that the trajectory of a developing wormhole, through the rock matrix, follows pathways defined by local regions of higher permeability rock. By extending that idea, it is reasonable to infer that the presence of fractures will also influence the development of wormhole patterns. Therefore, it is relevant to develop understanding as to how fractures influence the dissolution patterns during acid injection in carbonate rocks, and to use that understanding to better design acidizing treatments in field operations. As noted previously, there are few studies that focus on the effect of pre-existing fractures during matrix acidizing of carbonate rocks. Most of the work on the reactive flow in fractured media has focused on the study of the wormhole formation and propagation in a single fracture. Some numerical models, which are based on mass conservation for fluid flow and reactant transport, and equations for chemical kinetics within the fracture space, are developed to investigate the influence of the initial aperture field, the flow rate, and the chemical kinetics on dissolution patterns (Deng et al., 2016; Detwiler and Rajaram, 2007; Detwiler et al., 2001; Dong et al., 2002; Hanna and Rajaram, 1998; Hill et al., 2001; O’Brien et al., 2003; Szymczak and Ladd, 2009; Upadhyay et al., 2015). These models provide a useful starting point for numerical analysis of reactive flow in fractured porous media. However, these models cannot be used to study the interaction between fractures, and cannot explain the dissolution process in a fractured rock mass that has more than one fracture. Kalia and Balakotaiah (2009) studied the effect of heterogeneities, including fractures on wormhole formation. Yuan et al. (2016) developed a model by combining the Stokes-Brinkman and reactive transport equations to the mineral dissolution in fractured carbonate formations. However, they treated fractures as anomalous pores that have high porosity, and their method does not describe the fracture characteristics of lower porosity with higher permeability.

The main aim of this paper is to develop a reactive-transport simulation model that permits an evaluation of the effect of fractures on reactive flow in carbonate rocks. This desire requires a choice of methods which can be classified into two categories: (a) discrete fracture plus matrix models (DFM) (Frampton and Cvetkovic, 2007; Garipov et al., 2016; Geiger et al., 2004; Ghogomu and Therrien, 2000; Graf and Therrien, 2008; Huang et al., 2014; MacQuarrie and Mayer, 2005; Sandve et al., 2012; Steefel and Lichtner, 1998a; Steefel and Lichtner, 1998b; Therrien and Sudicky, 1996; Yan et al., 2014; Yan et al., 2016); and (b) continuum-only models (Cvetkovic and Gotovac, 2014; Doughty, 1999; Fahs et al., 2014; Hao et al., 2013; Lichtner, 2000; Samper et al., 2008; Zheng and Samper, 2015), which also can be called effective-medium models. This work is based on the DFM concept.

In discrete fracture plus matrix models, there is a background composed of matrix material, with
an array of superposed fractures that partition it into either: a partly-disrupted continuum, or into an assembly of completely isolated blocks, or a combination of these. The locations of fractures are explicitly defined, and the dimensionality of the fractures is reduced from $n$ to $(n-1)$. The model accounts explicitly for the effect of each individual fracture on fluid flow and solute transport. However, the discrete fracture plus matrix model poses a challenge for the discretization of the domain into a suitable mesh. To accurately capture the complexity of a fractured porous media, it is necessary to use an unstructured discretization scheme. It should be noted that a low-quality mesh can result in numerical instability, so the design of the mesh is not trivial.

In continuum models, the flow effects of both the fractures and matrix are combined within representative elementary volumes (REV), and there is locally mass exchange between the fracture and matrix system that results in homogenization into an equivalent behaviour. The continuum models are valid only for modeling systems containing a dense network of highly interconnected fractures, where the homogenization assumptions are sufficiently valid. It can reduce the computational complexity compared with the discrete fracture model. However, the major shortcoming with the continuum model is that it requires formulation of a fluid exchange function which involves a geometry-dependent parameter whose meaning can only be defined within certain flow regimes (Geiger et al., 2013; Hardebol et al., 2015). These models are not well suited for the modeling of a small number of larger fractures, especially when the purpose (as here) is to examine the local effects of the fractures relative to a specific process such as dissolution. A detailed discussion of model formulations for reactive transport in fractured media can be found in (MacQuarrie and Mayer, 2005). In the present work, the discrete fracture model was adopted to model the solute transport and reaction in fractured porous media. The detail of the solution method is given and the effect of fractures on dissolution patterns and breakthrough volume is analyzed.

This paper is organised as follows. In Section 2, the mathematical model, which describes the fluid flow, solute transport, and chemical reaction in both matrix and fracture system, is presented. In Section 3 we give a detailed numerical solution method that uses Delaunay triangulation to discretize the physical domain, and the finite volume method to discretize the governing equations. In Section 4 we compare the simulation results obtained from a degenerate model, with a previous computational study, to verify our work. In Section 5 we present 2-D simulation results on the dissolution process and its sensitivity to various factors such as fracture orientation, fracture density, fracture aperture, and injection rate. Finally, the paper is summarized by conclusions in Section 6.

2. Mathematical model

We study the case where acid is injected into a 2D rectangular domain, from the left side only, at a constant velocity (Fig. 1). The pressure at the right side is fixed and there is no fluid flowing across the top and bottom boundaries. The domain is composed of an otherwise-continuous matrix material that is segmented by an array of fractures. The fracture array is adjusted in a series of models to discover the role of fracture properties in terms of the resulting patterns of wormhole formation.

The governing equations for the reaction-transport problem in fractured porous media are the coupled equations for fluid flow, solute transport, and rock dissolution in both matrix system and fracture system. These governing equations describe the phenomenon of reactive flow in fractured porous media at the Darcy scale. However, some sample-scale information, such as the local
permeability, dispersion tensor, and mass-transfer coefficient, is needed to complete the Darcy scale model. These quantities could be derived from detailed pore-scale models of the several components of the system (Fredd and Fogler, 1998; Mostaghimi et al., 2013; Tansey, 2014; Van der Land et al., 2013), but that is not done here. Instead, empirical formulas – known as structure-property relations – are used in this paper. The following assumptions are made for this study: the fluid is incompressible, the fracture length does not change with time, but the fracture aperture can increase as dissolution occurs, and the system is under isothermal conditions.

![No flow boundary](image)

**Fig. 1 Geometry and boundary conditions of the physical model**

### 2.1 Darcy scale model

#### 2.1.1 Fluid flow

The description of fluid flow in a fractured porous medium requires governing equations for both the porous matrix and for the fracture system. Following Panga’s work (Panga et al., 2005), the continuity equation in the matrix is given by

$$\frac{\partial \phi}{\partial t} + \nabla \cdot \mathbf{v} = 0$$

(1)

where \( \mathbf{v} \) is the Darcy or superficial velocity vector, and \( \phi \) is the porosity of the matrix. The flow field is given by Darcy’s law

$$\mathbf{v} = \frac{k}{\mu} \nabla P$$

(2)

where \( k \) is the permeability tensor, which is calculated from a pore-scale model or determined by lab measurement, \( \mu \) is the viscosity of the fluid phase, and \( P \) is the pore-fluid pressure.

A mass balance of the fluid in a fracture gives

$$\frac{\partial}{\partial t} \left( \rho b \right) d\xi = -\frac{\partial (\rho b u_i)}{\partial \xi} \cdot d\xi + \left( \rho \mathbf{v} \cdot \mathbf{n} \right)_i \cdot d\xi - \left( \rho \mathbf{v} \cdot \mathbf{n} \right)_i \cdot d\xi$$

(3)

where \( b \) is the fracture aperture, \( u \) is the fluid velocity in the fracture, \( \mathbf{n} \) is the unit normal vector of the interface between the matrix and fracture, and \( d\xi \) is the differential element along the fracture direction. As the fluid is incompressible, the density \( \rho \) is a constant. Dividing by \( \rho d\xi \), we obtain
\[
\frac{\partial b}{\partial t} = -\frac{\partial (bu)}{\partial \xi} + q^+ - q^-
\]  \hspace{1cm} (4)

where \( q^+ \) and \( q^- \) represent normal components of fluid flux across the interface between the fracture and matrix. The value of the flux is given by:

\[
q^+ - q^- = \left[-\nabla \cdot (k \cdot \nabla P)\right]_{\Gamma_{mf}}
\]  \hspace{1cm} (5)

where \( \Gamma_{mf} \) refers to the matrix-fracture interface. The average flow velocity in a fracture is related to the pressure, using the well-known Poiseuille equation for laminar flow between parallel plates (Hanna and Rajaram, 1998), as follows:

\[ u = -\frac{b^2}{12\mu} \frac{\partial P}{\partial \xi} \]  \hspace{1cm} (6)

This expression implies that the local permeability of the fracture is \( k^\text{fr} = b^2/12 \), where, the superscript \( \text{fr} \) stands for the fracture. Note that this approach treats the fracture as an equivalent porous material, so a model built this way (as DFM) is most appropriately viewed as a full continuum with extreme heterogeneities within it.

2.1.2 Solute transport

The solute transport equation in the matrix is

\[
\frac{\partial \left( \phi C_f \right)}{\partial t} + \nabla \cdot (v C_f) = \nabla \cdot \left( \phi D_e \cdot \nabla C_f \right) - a_v \cdot R_c \cdot (C_f - C_s)
\]  \hspace{1cm} (7)

where \( C_f \) is the cup-mixing concentration of the solute in the fluid phase, \( C_s \) is the concentration of the solute at the fluid-solid interface, \( D_e \) is the effective diffusion tensor, \( a_v \) is the interfacial area available for reaction per unit volume of the medium, and \( R_c \) is the mass transfer coefficient.

A fluid-phase balance for the solute in a fracture gives

\[
\frac{\partial (bC_f)}{\partial t} = -\frac{\partial (buC_f)}{\partial \xi} + \frac{\partial}{\partial \xi} \left( b \cdot D_e \frac{\partial C_f}{\partial \xi} \right) + \Gamma^+ - \Gamma^-
\]  \hspace{1cm} (8)

Where the last two terms on the right hand side are: advective-diffusive gain and loss of solute mass across the fracture-matrix interfaces, respectively. This source terms are found to be

\[
\Gamma^+ - \Gamma^- = \left[-\nabla \cdot (D_e \cdot \nabla C_f) + \nabla \cdot (C_f \cdot v)\right]_{\Gamma_{mf}}
\]  \hspace{1cm} (9)

2.1.3 Solute reaction

The equation relating the mass transfer of solute in matrix transport to the fluid-solid interface to the reaction kinetics that are represented by \( Ra(C_s) \) is given as

\[ R_c \cdot (C_f - C_s) = Ra(C_s) \]  \hspace{1cm} (10)

For the case of HCl-CaCO_3 reaction which is a first order irreversible reaction, \( Ra(C_s)=R_cC_s \), where \( R_c \) is the dissolution rate constant, with units of velocity.

The change of porosity in the matrix due to dissolution is described by
\[
\frac{\partial \phi}{\partial t} = \frac{Ra(C_i)a_{\alpha}}{\rho_s}
\]  
(11)

Where \( a \) is the dissolving power of the acid, defined as grams of solid dissolved per mole of acid reacted, and \( \rho_s \) is the density of the solid.

Similarly, the flux balance at the fluid-solid interface in a fracture can be written as

\[
2R_f^\phi \cdot (C_f - C_i) = Ra^\phi (C_i)
\]  
(12)

where \( R_f^\phi \) is the velocity of solute transfer in fracture.

Dissolution of the fracture walls leads to alterations of the fracture aperture, here expressed as an average value

\[
\frac{\partial b}{\partial t} = \frac{Ra^\phi (C_i)\alpha}{\rho_s}
\]  
(13)

In this paper, we do not consider the reaction at the fracture wall, and focus only on the effect of the presence of fracture on the wormhole formation. This choice allows us to focus on the patterns of wormhole development and their relationship to the fracture distribution, without any complications from changes to the fracture system itself. That simplification will be relaxed in future work.

### 2.2 Pore-scale model

The Darcy-scale model describes the transport and reaction of solute at the scale of the finite elements. But the parameters of the Darcy-scale model depend on phenomena that take place at the pore scale, where changes of the rock texture, and the expression of those changes in certain characteristics (matrix porosity, permeability, average pore radius and the interfacial surface area as dissolution proceeds), are the result of processes that depend on each rock type. To complete the Darcy-scale model, we adopt here a simple pore-scale model for the matrix, following Kalia and Balakotaiah (2009):

\[
\frac{k}{k_0} = \frac{\phi}{\phi_0} \left( \frac{\phi(1-\phi)}{\phi_0(1-\phi)} \right)^{\beta}
\]  
(14)

\[
\frac{r_p}{r_0} = \sqrt{\frac{k\phi}{k_0\phi}}
\]  
(15)

\[
\frac{a_s}{a_0} = \frac{\phi r_0}{\phi_0 r_p}
\]  
(16)

where \( k_0 \) is the initial permeability of the matrix, \( \beta \) is a constant that depends on the structure of the medium, \( r_p \) is the pore radius, \( r_0 \) and \( a_0 \) are the initial values of average pore radius and interfacial area, respectively.

The mass transfer coefficient, and the effective dispersion coefficients, in the matrix are expressed (in the simple model) by the relations (Kalia and Balakotaiah, 2009):
\[ Sh = \frac{2R e_p}{D_m} = Sh_a + \frac{0.7}{m^{1/2}} \text{Re}_{p}^{1/2} \text{Sc}^{1/3} \] (17)

\[ D_{\infty} = \alpha_m D_m + \frac{2\lambda_x |u| r_p}{\phi} \] (18)

\[ D_{T} = \alpha_m D_m + \frac{2\lambda_T |u| r_p}{\phi} \] (19)

where \( Sh \) is the Sherwood number represents dimensionless mass transfer coefficient; \( D_m \) is the magnitude of fluid velocity; \( R e_p \) is the pore Reynolds number defined as \( R e_p = 2\mu r_p / \nu \), \( \nu \) is the kinematic viscosity ; \( Sc \) is the Schmidt number defined as \( Sc = \nu / D_m \); \( D_{\infty} \) and \( D_T \) are the longitudinal and transverse dispersion coefficient, respectively. \( \alpha_m, \lambda_x \) and \( \lambda_T \) are constants that depend upon the pore structure and their typical values are 0.5, 0.5, 0.1 for a packed-bed of spheres, respectively.

As mentioned before, the permeability of a fracture is related to the fracture aperture by

\[ k^f = \frac{h^2}{12} \] (20)

For solute transport in a fracture, we define the hydrodynamic dispersion coefficient in the fracture according to Steefel and Lichtner (1998b) as

\[ D^f = \alpha^f u + D_m \] (21)

where \( \alpha^f \) refers to the dispersity along the fracture.

2.3 Boundary and initial conditions

Here, we use the Dirichlet boundary condition at the inlet, and no-flux conditions at the transverse (top and bottom) boundaries. The initial and boundary conditions are given by

\[ C_{fl} = C_0, \text{ at } x = 0 \] (22)

\[ \nu_0 = -\frac{k_s}{\mu} \frac{\partial P}{\partial x}, \quad \frac{\partial P}{\partial y} = 0, \text{ at } x = 0 \] (23)

\[ \frac{\partial C_{fl}}{\partial x} = 0, P = P_e, \text{ at } x = L \] (24)

\[ \frac{\partial P}{\partial y} = 0, \frac{\partial C_{fl}}{\partial y} = 0, \text{at } y = 0 \text{ and } y = H \] (25)

\[ C_{fl} = 0, \text{ at } t = 0 \] (26)

where \( v_0 \) is the injection velocity, \( C_0 \) is the concentration of the injected acid, and \( P_e \) is the pressure at the exit boundary of the domain.

2.4 Dimensionless Variables
The following dimensionless variables and parameters are defined,

\[ x_D = \frac{x}{L}, \quad y_D = \frac{y}{L}, \quad \beta_D = \frac{\beta}{L}, \quad U = \frac{u}{v_0}, \quad V = \frac{v}{v_0}, \quad t_D = \frac{t}{L/v_0}, \quad r_D = \frac{r_p}{r_0}, \quad a_D = \frac{a}{a_0}, \quad \kappa = \frac{k}{k_0}, \quad \kappa_f = \frac{k_f}{k_0}. \]

\[ D = \frac{D}{D_m}, \quad D_f = \frac{D_f}{D_m}, \quad C_D = \frac{C_D}{C_0}, \quad C_f = \frac{C_f}{C_0}, \quad P_D = \frac{P - P_e}{(\mu v_0 L)/k_0}, \quad h_f^2 = \frac{2R_f r_0}{D_m}, \quad Da = \frac{R_a L}{v_0}, \]

\[ Pe_L = \frac{v_0 L}{D_m}, \quad N_{ac} = \frac{a C_0}{\rho}, \quad \Phi^2 = \frac{R_a a_L^2}{D_m}, \quad \eta = \frac{2R_f}{L}, \quad \alpha = \frac{H}{L}, \quad B = \frac{b}{L} \]

where the subscript D represents dimensionless variable, L is the characteristic length scale in the flow direction, H is the height of the domain, and U and V are dimensionless velocities in the matrix and fracture, respectively. \( \eta \) is the pore-to-domain length ratio, and \( \alpha \) is the aspect ratio. \( \kappa \) and \( \kappa_f \) are the dimensionless permeability of the matrix and fracture, respectively. \( B \) is the dimensionless fracture aperture. The parameters obtained from the non-dimensionalization are the pore scale Thiele modulus \( h_f^2 \), defined as the ratio of diffusion time to reaction time, Damköhler number \( Da \), defined as the ratio of convection time to reaction time; the axial Peclet number \( Pe_L \), defined as the ratio of advective transport rate to diffusive transport rate; the acid capacity number \( N_{ac} \), defined as the volume of solid dissolved per unit volume of acid consumed and the macroscopic Thiele modulus \( \Phi^2 \) is core-scale equivalent of the pore-scale Thiele modulus. More details on the definition of these dimensionless groups can be found in Kalia (2008). The PDEs after conversion into the non-dimensionalized form are as follows:

\[ U = -\kappa \cdot \nabla P_D \]  

(27)

\[ \frac{\partial \phi}{\partial t_D} + \nabla U = 0 \]  

(28)

\[ \frac{\partial (\phi C_D)}{\partial t_D} + \nabla \cdot (U C_D) = \nabla \cdot (D \cdot \nabla C_D) - Q_S \]  

(29)

\[ \frac{\partial \phi}{\partial t_D} = \frac{Da \cdot N_{ac} \cdot a_D C_D}{1 + \frac{h_f^2 r_D}{Sh}} \]  

(30)

\[ V = -\kappa_f \cdot \frac{\partial P_D}{\partial \xi_D} \]  

(31)

\[ \frac{\partial (BV)}{\partial \xi_D} - Q^+ + Q^- = 0 \]  

(32)

\[ \frac{\partial (BC_D)}{\partial t_D} + \frac{\partial (BVC_D)}{\partial \xi_D} = \frac{\partial}{\partial \xi_D} \left( BD_f \cdot \frac{\partial C_D}{\partial \xi_D} \right) + \Gamma_D^+ - \Gamma_D^- \]  

(33)

And the resulting boundary and initial conditions are
\[
\begin{align*}
-k_r \frac{\partial P}{\partial x} \bigg|_{x=a} &= 1, \\
P \bigg|_{x=0} &= 0, \\
-k_r \frac{\partial P}{\partial y} \bigg|_{y=a} &= 0, \\
C_D \bigg|_{x=0} &= 1, \\
\frac{\partial C_D}{\partial x} \bigg|_{x=1} &= 0, \\
\frac{\partial C_D}{\partial y} \bigg|_{y=0} &= 0.
\end{align*}
\] (34)

\[
\begin{align*}
C_D \bigg|_{x=0} &= 0, \\
\phi(x_D, y_D) \bigg|_{x=0} &= \phi_0 + \hat{f}, \\
B(x_D, y_D) \bigg|_{x=0} &= B_0 + \tilde{f}
\end{align*}
\] (35)

where

\[
Q_S = \frac{D_a \cdot a_D C_D}{1 + \frac{k_r^2 r_0^2}{Sh}}
\] (36)

(\![D_a, D_r] = \left(\frac{\alpha_a \phi Da}{\phi} + \lambda_x |U||r_0|\eta, \frac{\alpha_a \phi Da}{\phi} + \lambda_r |U||r_0|\eta\right)
\] (37)

In these formulas, \(B_0\) is the average initial fracture aperture (which is held constant in the results presented here). \(\hat{f}\) and \(\tilde{f}\) are random fluctuations in the initial porosity field and fracture aperture field, respectively. These random fluctuations can be either uniformly distributed or normally distributed in the specified interval, and are introduced to represent some degree of heterogeneity of the two media.

### 3. Numerical Solution

Several numerical methods have been employed in the literature for the simulation of reactive flow in porous media, such as the finite element method (FEM), the finite difference method (FDM), and the finite volume method (FVM). Due to the fact that FVM is locally conservative and has a clear physical interpretation (Eymard et al., 2000; Eymard et al., 2006; Gallouët et al., 2000; Karimi-Fard et al., 2003; Moukalled et al., 2016; Versteeg and Malalasekera, 2007), the FVM is used in this paper to solve the reactive flow equations which describe the fluid flow, solute transport and rock dissolution in fractured porous media. The numerical solution of the PDEs consists of the discretization of the domain, a process known as meshing, and the discretization of the equations as described in the following.

In the discrete fracture plus matrix model, the total domain is decomposed into the matrix and the fracture subdomains, both of which are Darcy continua. Fig. 2(a) shows a two-dimensional physical domain of a fractured porous rock mass. The complexity of a fractured rock mass needs
to be represented by the use of an unstructured mesh. Fig. 2(b) depicts a discretization of this
domain using triangles for the matrix, and segments for the fractures, where the thick red line
represents the fracture trace. The fracture thickness is not represented in the grid domain, but it is
considered in the computational domain as shown in Fig. 2(c). The errors associated with the
“expansion” of the fractures when constructing the computational domain are small (Ma et al.,
2006). Unknowns such as pressure, concentration, porosity, and fracture aperture are stored at the
centroids of grid elements, which are called the cell-centered variable arrangement method.

![Fig. 2. Sketches for discretization of a 2-D fractured porous medium. (a) Physical domain; (b) Grid domain; (c) Computational domain](image)

As shown in Fig. 2(b), the thick line that represents a fracture is divided into several linear
segments that are the edges of the triangles that comprise the matrix mesh. There are three types of
connections in the grid domain, named matrix-matrix, matrix-fracture and fracture-fracture
connections (Fig. 3). This geometrical information is used to discretize the governing equations.

![Fig. 3. Different connections in grid domain. (a) matrix-matrix, (b) matrix-fracture, (c) fracture-fracture.](image)

The solution procedure of the overall system can be briefly described as follows: The pressure
field and the Darcy velocity distribution at time $t^{n+1}$ are first calculated from Eqs. (27) (28) (31)
(32) by using the concentration field at time $t^n$. Next, the Darcy velocity at time $t^{n+1}$ is used to
update the pore scale parameters and is substituted into Eq. (29) and Eq. (33). The concentration
field and porosity field at time \( t^n+1 \) are then determined by coupling Eqs. (29)–(33) and Eq. (30). For each step of the calculation, the dynamic time step scheme is used to guarantee the stability of the simulation, i.e. if the difference in variables calculated from using time step \( \Delta t \) and \( \Delta t/2 \) is less than a specified differential tolerance \( \epsilon \), the time step \( \Delta t \) is selected as the computational step. Otherwise, the time step is reduced until the time step criterion is satisfied. The detailed derivation of the finite volume approximation is shown in Appendix 1. A system of linear equations of the form \( Ax = b \) is obtained by substituting the discretized forms of the transient, convection and diffusion terms into the general equation. The structure of the coefficient matrix \( A \) is depicted in Fig. 4. It consists of matrix-matrix transmissibility, matrix-fracture transmissibility, and fracture-fracture transmissibility, which all are assembled together in one matrix. The system of algebraic equations is solved using the iterative method. Note that large variations in dependent variables between iterations may result in large source terms and large changes in the coefficients, which may cause divergence of the iterative solution procedure. The under-relaxation method is used to slow down the change of the dependent variable values and improve the convergence. It should be noted that the numerical method described above is only feasible on 2D fractured porous media. Another challenge arises when extending this method to a 3D domain, associated with the mesh discretization of the 3D fractured porous media. The theory and methods of the discretization of the 3D fractured media are beyond the scope of this work.

![Fig. 4 The structure of the coefficient matrix of the system of algebraic equations](image)

### 4. Model testing

For the reason that no numerical or experimental result for wormhole formation in fractured masses is available in the literature, the model described in this work is tested by comparing it with the reactive flow problem (described in previous works) without consideration of the effect of fractures. This demonstrates that the discretization method used in the unstructured grid formulation is suitable for the reactive flow problem, and that the governing equations are solved correctly. Furthermore, we must ensure that the method to characterize the fractured medium based, on DFM, can be used for this kind of convection-diffusion problem. Fortunately, the accuracy of using DFM has been demonstrated by Therrien and Sudicky (1996), Steefel and Lichtner (1998a), Ghogomu and Therrien (2000), and Karimi-Fard et al. (2003), to cite a few, who solve the transport problem in water resource engineering and petroleum engineering.
We compare the results, using our method, with the results obtained by Kalia (2008). Firstly, to keep the mesh size consistent with Kalia’s work, which uses a square cell of size 0.003×0.003, we set the length of the triangular mesh side not smaller than 0.005. After discretization of the grid domain using Delaunay triangulation, we get 19,430 elements for simulation. Furthermore, a random initial porosity field is generated by adding a random fluctuation which is assumed to be uniformly distributed in the interval [0.05, 0.35] which is a variation of +/- 0.15 about the initial average porosity value of 0.2. The values of other parameters and dimensionless groups used in this example are the same as those given in Kalia (2008). As Kalia (2008) mentioned, we define the pore volume of acid required to breakthrough, $PV_{BT}$, as the amount of acid consumed per pore volume when acid reaches the downstream end of the domain. Numerically, we take the breakthrough time as the time when the overall pressure drop across the medium falls to $1/100^{th}$ of its initial value.

The calculations of $PV_{BT}$ obtained from our work and Kalia’s are compared in Fig. 5, as the reciprocal of Damköhler number $1/Da$ varies from $10^{-5}$ to 1. It can be seen that our calculations of $PV_{BT}$, and thus the optimum injection rate at the minimum, are in good agreement with Kalia’s work. The slight deviation observed for low injection rates is mostly due to the difference in the size and number of elements in the mesh, and the randomness in the initial porosity field.

![Fig. 5. Breakthrough curves for comparison with results obtained by Kalia (2008)](image)

### 5. Results with fractures

This section presents the results from simulating the model described by Eqs. (27)–(35), considering characteristics of the fracture arrays and injection rates, aiming to better understand the effect of fractures on wormhole formation. The parameters and dimensionless numbers used in the simulations are shown in Table 1. All the values remain fixed throughout the study, unless otherwise stated.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>N. Kalia 2008</td>
<td></td>
</tr>
<tr>
<td>Current work</td>
<td></td>
</tr>
</tbody>
</table>

Table 1. List of parameters and dimensionless numbers used in simulation.
5.1 Effect of fracture orientation

Here, we examine the effect of fracture orientation on wormhole formation, using the rectangular domain and the set of variables summarized in Table 1. The approach is to add a single fracture of specified orientation into a model with no fracture, and evaluate whether the fracture has an impact on the dissolution pattern. For each of these configurations, we show (Fig. 6) the pattern of enhanced porosity soon after the start of injection (left panel in each row), and then the pattern at the breakthrough time (right panel). For the case without any fracture (top row), the locations, where dissolution has occurred, advance to the right, away from the line of injection. The positive feedback – where flow is faster in those cells with higher permeability, results in a faster dissolution, and thus an increase in permeability that leads to increased flow and further dissolution – progresses by becoming localized into a prominent wormhole that exhibits some branching (Fig. 6 (a)). This is the classic pattern expected for injection rates that are near the minimum that is shown in Fig. 6.

The subsequent rows of Fig. 6 show early-time and breakthrough dissolution patterns in cases with a single fracture. The early-time patterns are not significantly different to those seen in the case without any fracture, although there is a hint that the initiation of the early-time dissolution process may be influenced by the downwind flux increase that is related to the “entry” point of the fracture in cases (b) and (c). The three fracture orientation arrangements, including axial, diagonal and perpendicular, all result in localization of the dissolution along the fracture, with additional wormholes leading off of the fracture in the downwind direction. Therefore, it is possible to say that the pattern and the extent of reactive dissolution mainly depends on the location and orientation of the fracture. However, if the fracture orientation is perpendicular to the flow direction, as is shown in Fig. 6(d), the fracture effects are less obvious in terms of dominating the orientation of the wormholes, but there are nevertheless differences relative to the case without a
fracture. Similar effects relative to wormhole propagation, resulting from varying the fracture orientation, have been found by Kalia and Balakotaiah (2009) where the fracture is treated as a kind of higher-porosity matrix and the fracture width is represented by one mesh cell. These outcomes suggest that enhanced flow pathways of any kind may play a substantial role in governing how wormholes develop.

![Wormhole patterns in the presence of fractures](image)

Fig. 6. Wormhole patterns in the presence of fractures (the pictures on the left are the porosity fields when the simulation time equals to 0.2 and the pictures at right are the porosity fields when acid was breakthrough) (a) no fracture, (b) axial fracture, (c) diagonal fracture, (d) transverse fracture. $t_D$ is the dimensionless time.

5.2 Multiple fractures

Multiple, parallel fractures, with a single orientation impact on wormhole formation and propagation in ways that might be surprising. Fig. 7(a) shows the dissolution pattern with three parallel fractures along the flow direction. It can be seen that only one fracture has contributed to the wormhole process by the time acid breakthrough occurs. This happens because the injection acid mainly flows along the path with the least resistance. In this model, the fractures have much higher effective permeability than does the matrix. However, once again, the subtleties of flow are apparent: because the matrix is slightly heterogeneous, acid flows preferentially in one or more zones with locally higher values of permeability, and increases the permeabilities within those zones by etching the pore throats. Although more than one flow channel might be formed initially,
once one channel connects to a fracture, the easy-flow path captures the majority of the flow. Thereafter, the propagation of the other channels stops or at least reduces, due to their lower acid fluxes. This is a classic example of non-linearity of outcome due to positive feedbacks. A similar result is observed in Fig. 7(b), where three parallel fractures, with different lengths, are present in a diagonal direction. In this case, the middle fracture has become the dominant flow pathway, possibly because it is longer and thus captures more of the flow, but it is also possible that this result is primarily due to a favoured upwind flow path that just happens to lead to the capture zone of that particular fracture.

However, this dominance of a single fracture disappears when the fractures are not arranged in a way that causes one of them to capture the majority of the flux (Fig. 7 (c)). The dominant wormhole in this example makes use of all the fractures. This is because in this situation, the smaller flow resistance in the fractures acts to provide a local “short cut” within the overall flow field. It can also be seen from Fig. 7(c) that the acid in this fracture first leaks out into the matrix from the point that has the highest porosity and permeability. The subsequent local flow direction is not along the direction of the fracture, but is, instead, somewhat aligned with the global flow pattern. To see this phenomenon clearly, we show the porosity fields and corresponding flux field of this model at different time steps in Fig. 8 and the pressure contour at the same time steps in Fig. 9. We can see that the multiple fingers propagating downstream have equivalent driving energies at the time they form, and that later, small changes in energy still allow them to propagate due to their already-started high perm parts. During this period, a few branches are formed on the wormhole and the amount of the branch depends on the heterogeneity of the matrix around the fracture.

Fig. 7. Influence of fracture quantity on wormhole patterns (the pictures at the left are the porosity fields when the simulation time equals to 0.2 and the pictures at right are the porosity fields when acid was breakthrough) (a) three horizontal parallel fractures; (b) three diagonal parallel fractures; (c) four discrete fractures. \( t_D \) is the dimensionless time.
Fig. 8 Porosity fields (left) and corresponding flux fields (right) as a function of time for fractured porous medium that has four discreet fractures. (a) $t_D=0.05$; (b) $t_D=0.2$; (c) $t_D=0.35$; (d) $t_D=0.55$; (e) $t_D=0.65$; (f) $t_D=0.81$ (breakthrough).
Fig. 9 Pressure contour as a function of time for fractured porous medium that has four discreet fractures. (a) $t_D=0.05$; (b) $t_D=0.2$; (c) $t_D=0.35$; (d) $t_D=0.55$; (e) $t_D=0.65$; (f) $t_D=0.81$ (breakthrough).

5.3 Effect of fracture aperture

The effect of fracture apertures is studied by comparing the dissolution patterns at the breakthrough time, and at the volume of acid required to achieve breakthrough. As an example to be compared, the dissolution pattern created when acid is injected into the un-fractured porous medium is depicted in Fig. 10(a). Fig. 10(b, c, d) shows the dissolution patterns that result from the same numerical parameters except for assigning the fracture apertures as $10^{-6}$m, $10^{-5}$m, $10^{-4}$m, respectively. It can be seen from comparing Fig. 10(a, b) that the fracture array with smallest of these apertures has almost no effect on wormhole propagation. The injected acid tends to flow into the bigger pores rather than flow into the fracture, because the permeability in the matrix is the same order of magnitude as in the fractures. With the increase of fracture aperture, the permeability of the fracture increases, and the flow in the fracture network gradually dominates. Therefore, the wormhole propagates in a manner that makes partial use of the fractures. As a result, the dissolution pattern is dramatically different (Fig. 10(c)) even though the injection rate and porosity field are kept the same. However, as the fracture aperture further increases, the dissolution pattern does not seem to change, and only a little difference in wormhole diameter and extent can be observed in Fig. 10(d). This is because the flow is now limited by the characteristics of the matrix areas between the fracture ends. Due to the same matrix property field in all models, injected acid goes though the same matrix pores and etches the same matrix pore throats that
provide the connections between the fractures, and this results in the same dissolution pattern in both (c) and (d). Fig. 11 displays the acid volume required to achieve breakthrough for the un-fractured medium and in models with the different fracture apertures. It shows clearly that the acid volume required to breakthrough decreases as the fracture aperture increases, as expected. We can also see that the acid volume consumed in the fractured medium with small fracture aperture ($10^{-6}$ m) is the same as that consumed in the un-fractured case. Again, it demonstrates that a fracture array with smaller apertures has almost no effect on wormhole propagation. Actually, in our simulation cases, the breakthrough volume in case (b) is slightly higher than that in case (a). This might because the existence of fractures increases the branching of the wormhole and this needs more acid to etch the rock.

![Fig. 10. Effect of fracture aperture on wormhole patterns. (a) no fracture; (b) fracture aperture $b=10^{-6}$ m; (c) fracture aperture $b=10^{-5}$ m; (d) fracture aperture $b=10^{-4}$ m. $t_D$ is the dimensionless time.](image)

![Fig. 11. Acid volume required to achieve breakthrough for un-fracture medium and fracture aperture values of $10^{-6}$ m, $10^{-5}$ m, and $10^{-4}$ m.](image)
5.4 Effect of injection rate

In the absence of fracture, five types of dissolution patterns are formed with the increase of injection rate, namely ((a) to (e) in Fig. 12): face dissolution, conical wormhole, dominant wormhole, ramified wormhole, and uniform dissolution. The numerical simulation results depicted in Fig. 12 highlight the role of injection rate in a model domain without any fractures and lacking strong material heterogeneities. As explained by Kalia and Balakotaiah (2009), the differences between these dissolution patterns are the result of competition between axial advection, transverse dispersion and reaction mechanism processes.

Fig. 12. Dissolution patterns (Porosity fields) and corresponding concentration fields (right) from 2D simulations in un-fractured porous medium at different injection rates. (a) face dissolution at $q=0.002 \text{cm}^3/\text{s}$; (b) conical dissolution at $0.02 \text{cm}^3/\text{s}$; (c) dominate wormhole at $q=0.2 \text{cm}^3/\text{s}$; (d) ramified wormhole at $q=4 \text{cm}^3/\text{s}$; (e) uniform dissolution at $q=200 \text{cm}^3/\text{s}$. $t_D$ is the dimensionless time.
As has been demonstrated in the preceding examples, a fracture can dominate the flow path, and thus the magnitude of axial advection such that fractures can transform the dissolution patterns, in comparison to a similar un-fractured medium. Five cases are now considered, each containing the same spatial arrangement of short and dis-connected fractures, and the same matrix porosity distribution. These cases are simulated at the same series of injection rates as used in the un-fractured medium simulations shown in Fig. 13. The outcomes illustrate the differences in dissolution patterns compared with those created in the un-fractured model domain (Fig. 13), illustrated here by the resulting porosity field and the corresponding concentration field at each of the injection rates. It can be seen that the dissolution patterns generated in fractured porous media are similar with those generated in un-fractured media. Face dissolution, conical wormhole, dominant wormhole, ramified wormhole, and uniform dissolution patterns are created in our simulations when the injection rate increases from lower values to higher values. However, it can also be observed that all dissolution patterns have fewer branches than the patterns formed in the porous medium that does not have fractures. This is because the permeability of the fractures is substantially greater than that of the matrix material, creating a dominant flow path that becomes increasingly enhanced in the conditions that lead to the dominant wormhole response (middle panels).

We compare the breakthrough volume of the different dissolution patterns obtained in both fractured and un-fractured simulations (Fig. 14). The first observation is that the acid volume for breakthrough in a fractured medium is less than that in the equivalent un-fractured medium for all of the dissolution patterns (which depend on the flux rates). This global decrease of the breakthrough volume may be explained by the higher conductivity of the fractures. It is conceivable that this decrease could be even greater for a particularly-favourable fracture that has large aperture and is aligned with the flow direction. Moreover, it is interesting to note that, in our example, the optimum injection rates in fractured and un-fractured porous medium are same. This means that the optimum conditions determined from un-fractured reservoir can also be used in fractured reservoir when the acid-mineral system is same.
Fig. 13. Dissolution patterns (Porosity fields) and corresponding concentration fields (right) from 2D simulations in fractured porous medium at different injection rates. (a) face dissolution at $q=0.002\text{cm}^3/\text{s}$; (b) conical dissolution at $0.02\text{cm}^3/\text{s}$; (c) dominate wormhole at $q=0.2\text{cm}^3/\text{s}$; (d) ramified wormhole at $q=4\text{cm}^3/\text{s}$; (e) uniform dissolution at $q=200\text{cm}^3/\text{s}$. $t_D$ is the dimensionless time.
6. Conclusions and discussion

We have described a continuum model that can calculate the reactive flow of acid in a fractured rock mass, here assumed to be a carbonate rock type, based on the discrete-fracture+matrix (DFM) model. The model calculates the dissolution of rock mass, increasing the porosity in the reacted locations, with feedbacks to the flow system. The derivation of the numerical method is detailed for a 2-D version of the model. The model implementation is used to simulate some simple cases that highlight the difference in dissolution patterns with and without the presence of fractures. In particular, the effect of the characteristic parameters of fracture arrays, such as fracture orientations, fracture-array geometry, and fracture aperture, are investigated in relation to wormhole formation. The effect of fractures on the resulting dissolution patterns at different injection rates is also studied. The main results can be summarized as follows:

(1) The wormhole propagation depends on the orientation of any fractures. When the fracture or fractures form a flow pathway along the flow direction, or create paths that have an acute angle with flow direction, the wormhole propagation is dominated by the presence of the fracture(s). When the fracture orientation is perpendicular to the flow direction, the influence of fractures on main wormhole propagation is small, but the fracture may alter the direction and amount of branches.

(2) The geometry of multiple fractures influences the propagation of a wormhole. Simulation results indicate that one fracture will dominate the wormhole for a group of parallel fractures no matter whether they are oriented along the flow direction or not.

(3) The dissolution patterns vary with the increase of fracture aperture, up to a point. Fractures with small apertures will not dominate the wormhole propagation and have no influence on breakthrough volume, and hence they can be neglected in the computational domain.

(4) Finally, perhaps the most useful conclusion is that the optimum injection rate is almost unchanged from un-fractured medium to fractured medium if the acid-mineral reaction system is same. The existence of fracture only affects the dissolution structures that are associated with different injection rates.
Although the mathematical model proposed is completely general and can be extended to other kinetics, it should be noted that all the simulation results presented in this work are for first order irreversible reactions. Moreover, the numerical example simulated for linear flow can also be extended to radial flow situations. These extensions will be studied in future work. It should also be noted that our focus has been on the dissolution patterns in a fractured medium, and the effect of complex fracture systems on the optimum injection rate has not been studied in detail. This will be addressed in future work. In addition, a limitation should be mentioned: that the numerical method used in this work cannot accurately simulate the reactive flow problem in a model with intersecting fractures. This is because an intermediate mesh should be introduced at the intersection point to redistribute flux along every fracture. As the size of the intermediate mesh is much smaller than other elements, the time step will be limited by the smallest mesh and the numerical error occurs. Again, the numerical method will be improved in the future work.

Acknowledgements

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Appendix A

The mass conservation equation can be considered as a special case of the convection-diffusion-reaction equation, without diffusion and source terms. So, only the discretization of the convection-diffusion-reaction equation is presented in this paper. After discretizing the physical domain into computational domain, the grid property of the fracture is similar to the matrix, except that the grid shape of the fracture is rectangular while the grid shape of the matrix is triangular. Therefore, we only focus the description of the discretization of the convection-diffusion-reaction equation in the matrix-matrix grid (Fig. 3(a)). It should be noted that the discretization in matrix-fracture grid (Fig. 3(b)) and fracture-fracture grid (Fig. 3(c)) are same. By integrating Eq. (29) over the element P shown in Fig. 3, we obtain

\[ \int_{P} \frac{\partial (\phi C_{D})}{\partial t_D} dV + \int_{P} \nabla \cdot (UC_{D}) dV = \int_{P} \nabla \cdot (DV C_{D}) dV - \int_{P} Q_{s} dV \]  

Replacing the volume integrals of the convection and diffusion terms by surface integrals through the use of the divergence theorem, and then, replacing the surface integral by a summation of fluxes over the element faces, Eq. (A-1) becomes

\[ \int_{P} \frac{\partial (\phi C_{D})}{\partial t_D} dV + \sum_{f=\text{faces}} \left( \int_{f} (UC_{D}) \cdot dS \right) = \sum_{f=\text{faces}} \left( \int_{f} (DV C_{D}) \cdot dS \right) - \int_{P} Q_{s} dV \]  

Using a single Gaussian point for the face integral, the diffusion term is approximated as

\[ \sum_{f=\text{faces}} \left( \int_{f} (DV C_{D}) \cdot dS \right) \approx \sum_{f=\text{faces}} (D \cdot \nabla C_{D})_{f} \cdot \mathbf{S}_{f} \]  

Therefore, the gradient in the direction normal to the interface should be calculated and the expression should involve the variable values at centroids P and N of the cells. If the direction of
$S_f$ is along the line joining the two points $P$ and $N$, the gradient of the variable in the direction of $S_f$ can be written as

$$\nabla C_D \cdot S_f = \frac{C_D(N) - C_D(P)}{d_{PN}} |S_f|$$

(A-4)

However, in unstructured meshes the lines joining the centroids of the elements straddling the interface are usually not parallel to the normal vector, as shown in Fig. 3(a). In this case, the gradient expressed above should be corrected by adding a contribution arising from non-orthogonality. There are different ways to correct the gradient (Haselbacher, 1999; Kim and Choi, 2000; Moukalled et al., 2016; Sandve et al., 2012; Versteeg and Malalasekera, 2007), but the most common form is to introduce a term known as cross-diffusion. According to this method, the surface vector can be written as the sum of two vectors $E_f$ and $T_f$, i.e.

$$S_f = E_f + T_f$$

(A-5)

Where, the subscript $f$ represent the face connect two elements; $E_f$ is in the direction of line $PN$, then Eq. (A-4) can be modified into the form

$$\nabla C_D \cdot S_f = \nabla C_D \cdot E_f + \nabla C_D \cdot T_f$$

(A-6)

$$= |E_f| \frac{C_D(N) - C_D(P)}{d_{PN}} + \nabla C_D \cdot T_f$$

The second term on the right hand side is called cross-diffusion and is treated as source term. To evaluate the cross-diffusion term, the definition of vector $T_f$ is required. Several options can be used to defining the direction of $T_f$ such as minimum correction, orthogonal correction and over-relaxed correction (Moukalled et al., 2016). The over-relaxed approach is adopted in this paper, because the diagonal of the coefficient matrix is greater than others and increases as grid non-orthogonality increases. To achieve this, the direction of $T_f$ is set to be parallel to line $ab$ as shown in Fig. A-1. Mathematically $E_f$ and $T_f$ are computed as

$$E_f = \left( \frac{|S_f|}{\cos \theta} \right) e = \left( \frac{|S_f|}{S_f \cos \theta} \right) e = \frac{S_f \cdot S_f}{e \cdot S_f} e$$

(A-7)

$$T_f = S_f - E_f = S_f - \frac{S_f \cdot S_f}{e \cdot S_f} e$$

(A-8)

It should be noted that the gradient on the right hand side of Eq. (A-6) is still unknown. Fortunately, it can be calculated using the value at the previous iteration. The detail of computing the gradient will be presented later.
Fig. A-1. Decomposing $S_f$ via the over-relaxed approach

Similarly, by using a single Gaussian point for the face integral, the convection term can be approximated as

$$\sum_{f \in N_p} \left( \int (UC_f) \cdot dS \right) = \sum_{f \in N_p} (UC_f) \cdot S_f = \sum_{f \in N_p} \bar{m}_f \cdot (C_D)_f$$  \hspace{1cm} (A-9)

Where,

$$\bar{m}_f = U \cdot S_f$$  \hspace{1cm} (A-10)

As the convective transport is more dependent on upstream conditions, it may lead to unphysical results if we were to use a symmetrical linear profile similar to the one used in the discretization of the diffusion term. On the other hand, using the upwind scheme can produce physical and bounded solutions, however, it is achieved at the cost of low accuracy. To balance the accuracy and stability, a large number of schemes used to discretize the convection term have been created (Eymard et al., 2000; Moukalled et al., 2016). The Normalized Weighing Factor (NWF) method based on the Normalized Variable Formulation (NVF) framework (Darwish and Moukalled, 1996) is adopted to simulate the reactive flow in fractured porous media in this paper. The NVF is a face formulation which relies on the upwind, downwind, and far upwind node values. Using this formulation, the normalized variable $\tilde{C}_D$ can be expressed in term of $C_D$ as

$$\tilde{C}_D = \frac{C_D - C_D(U)}{C_D(D) - C_D(U)}$$  \hspace{1cm} (A-11)

Where $D$, $C$, and $U$ in the brackets denote the Downwind, Upwind, and far Upwind nodes at any particular face. In unstructured grid, far Upwind node $U$ is a ‘dummy’ node defined by assuming $U$ to lie on the line joining the nodes $C$ and $D$ such that $C$ is the midpoint of the segment joining the points $U$ and $D$ as depicted in Fig. A-2. Thus, the value of $C_D(U)$ can computed as

$$C_D(U) = C_D(D) - 2V C_D(C) \cdot d_{CD}$$  \hspace{1cm} (A-12)
Fig. A-2. Downwind node, Upwind node, and dummy Far Upwind node in unstructured grid

Since the calculation of \( C_D(U) \) needs the gradient of node \( C \), the term involving \( C_D(U) \) is treated in a deferred correction fashion. The NWF method is developed by linearizing the normalized interpolation profile such that

\[
(C_D)_f = lC_D(C) + k
\]  

(A-13)

Where, \( l \) and \( k \) are constants which values depend on the scheme used. Combining Eq. (A-11) and Eq. (A-13) yields

\[
(C_D)_f = lC_D(C) + kC_D(D) + (1-l-k)C_D(U)
\]  

(A-14)

The MINMOD scheme (Moukalled et al., 2016) is used in this work, in term of the NWF method, the values of \( l \) and \( k \) are given by

\[
[l, k] = \begin{cases} 
  \left[ \frac{3}{2}, 0 \right] & 0 < C_D(C) < \frac{1}{2} \\
  \left[ \frac{1}{2}, \frac{1}{2} \right] & \frac{1}{2} \leq C_D(C) < 1 \\
  [1, 0] & \text{elsewhere}
\end{cases}
\]  

(A-15)

As Darwish and Moukalled (1996) mentioned, the NWF method has a higher robustness, since the value of \( l \) is not smaller than that of \( k \), resulting in the diagonal dominance of the coefficient matrix.

To discretize the source term and the transient term, combining Eq. (30) and Eq. (36) yields

\[
Q_s = \frac{\partial \phi}{\partial t_D} \frac{1}{N_w}
\]  

(A-16)

As shown in Eq. (A-16), now the source term is in terms of the transient. Letting

\[
\Pi = \phi \left( C_D + \frac{1}{N_w} \right)
\]  

(A-17)

The source term and transient term in Eq. (29) can be expressed as

\[
\int_{V_r} \frac{\partial (\phi C_D)}{\partial t_D} dV + \int_{V_r} Q_s dV = \int_{V_r} \frac{\partial}{\partial t_D} \left[ \phi \left( C_D + \frac{1}{N_w} \right) \right] dV = \int_{V_r} \frac{\partial \Pi}{\partial t_D} dV = \frac{\partial \Pi}{\partial t_D} \cdot V_p
\]  

(A-18)

Eq. (A-1) becomes
Integrate Eq. (A-19) over the time interval \([t_D - \Delta t_D/2, t_D + \Delta t_D/2]\) and then divide all terms by the temporal element volume \(\Delta t_D\), the transient term can be written as

\[
\frac{\partial \Pi}{\partial t_D} V_p + \int_{V_p} \nabla \cdot (\mathbf{UC}_D) \, dV = \int_{V_p} \nabla \cdot (\mathbf{D} \nabla C_D) \, dV \tag{A-19}
\]

To express the face values at \(t_D + \Delta t_D/2\) and \(t_D - \Delta t_D/2\) in terms of the element values at \(t_D\), an interpolation profile is used on a pseudo time element in a similar fashion to what was used in the convection term (Moukalled et al., 2016). Using the second order upwind interpolation profile, the transient term is approximated as

\[
\frac{\Pi^{i_0+\Delta t_D/2}_k - \Pi^{i_0-\Delta t_D/2}_k}{\Delta t_D} \cdot V_p = \frac{3\Pi^{i_0}_k - 4\Pi^{i_0-\Delta t_D}_k + \Pi^{i_0-2\Delta t_D}_k}{2\Delta t_D} \cdot V_p \tag{A-20}
\]

As noted above, the discretization of both the diffusion term and the convection term need the calculation of the gradient at cell centroids and faces. There are several methods available in the literature to calculate a gradient at the cell centroid. One of the most commonly used techniques is to use the Least-Square method (Moukalled et al., 2016; Ollivier-Gooch and Van Altena, 2002; Versteeg and Malalasekera, 2007). Based on this method, the following set of two equations with two unknowns (for a 2-D domain) is obtained

\[
\begin{bmatrix}
\sum_{i=1}^{N_x} \omega_k \Delta x_i \Delta y_i \\
\sum_{j=1}^{N_y} \omega_k \Delta x_i \Delta y_j
\end{bmatrix}
\begin{bmatrix}
\frac{\partial C_D}{\partial x} \\
\frac{\partial C_D}{\partial y}
\end{bmatrix}_p
= \begin{bmatrix}
\sum_{i=1}^{N_x} \omega_k \Delta x_i \Delta y_i \Delta C_{i,k} \\
\sum_{j=1}^{N_y} \omega_k \Delta x_i \Delta y_j \Delta C_{i,k}
\end{bmatrix} \tag{A-22}
\]

where \(\Delta x = \mathbf{PF} \cdot \mathbf{i} \), \(\Delta y = \mathbf{PF} \cdot \mathbf{j}\), and \(\Delta C_D = C_D (P) - C_D (N)\). \(\omega_k\) is a weighting factor, whose choice is important in determining the properties of the gradient. As proposed in Mavriplis (2003), \(\omega_k\) is taken as the inverse distance between P and N, and is given by

\[
\omega_k = \frac{1}{d_{PN_k}} \tag{A-23}
\]

References


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