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Coupled fluid flow, solute transport and dissolution processes in discrete fracture networks: An advanced Discontinuous Galerkin model

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► To cite this version:

Sara Tabrizinejadas, Anis Younes, Hussein Hoteit, Jerome Carrayrou, Marwan Fahs. Coupled fluid flow, solute transport and dissolution processes in discrete fracture networks: An advanced Discontinuous Galerkin model. *Advances in Water Resources*, 2023, 180, pp.104540. 10.1016/j.advwatres.2023.104540 . hal-04296444

HAL Id: hal-04296444

<https://hal.science/hal-04296444v1>

Submitted on 22 Nov 2023

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4 2 **Coupled fluid flow, solute transport and dissolution processes in discrete**
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6 3 **fracture networks: an advanced Discontinuous Galerkin model**
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51 20 Submitted to *Advances in Water Resources*
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24 **Abstract**

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3 25 Modeling dissolution processes in discrete fracture networks (DFNs) is a challenging task.
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5 26 Challenges are related to the highly nonlinear coupling between flow, mass transport, and
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7 27 reactive processes associated with fracture aperture evolution by dissolution. Further,
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9 28 advection-dominated transport due to fast fluid flow in fractures renders the problem more
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11 29 complex from a computational point of view, as traditional numerical methods may introduce
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13 30 unphysical oscillations or excessive numerical diffusion. The Discontinuous Galerkin (DG)
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15 31 method is known to be suitable for the simulation of advection-dominated transport. In this
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17 32 work, an advanced DG model is developed to model transport with dissolution in DFNs. We
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19 33 propose an upwind formulation to deal with the upstream concentration at the intersection of
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21 34 several fractures. The upstream concentration at an intersection node is calculated based on
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23 35 the average nodal concentrations of all the fractures having an inflow at that node, weighted
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25 36 by the volumetric fluxes of these fractures. The dispersion term is discretized with the Mixed
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27 37 Finite Element (MFE) method, which ensures the continuity of the dispersive flux at the
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29 38 intersection of fractures with different apertures. The obtained nonlinear coupled flow-
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31 39 transport-dissolution equations are discretized in time with a high-order scheme via the
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33 40 method of lines (MOL). Numerical examples and comparisons with standard finite element
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35 41 (FE) and finite volume (FV) solutions are performed to investigate the correctness and
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37 42 efficiency of the developed model. Results show that the new DG-DFN model avoids
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39 43 unphysical oscillations encountered with the standard FE method and strongly reduces the
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41 44 numerical diffusion observed with the upwind FV scheme. The DG-DFN model is then used
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43 45 to investigate the effect of the dissolution rate on the flow, transport, and aperture evolution
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45 46 processes for a single fracture and for a DFN. A quasi-linear evolution of the fracture aperture
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47 47 is observed for low dissolution rates. For high dissolution rates, a funnel-shaped enlargement
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49 48 is observed with a significant widening for the fractures near the inlet and minor effects for
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51 49 those away from the injection location.

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51 51 **Keywords:** Dissolution processes; Fractured rocks; Discrete Fracture Network;
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53 52 Discontinuous Galerkin; advection-dominated transport.
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1. Introduction

The flow of water in fractured rocks is usually associated with dissolution processes and transport of the dissolved species (Liu *et al.*, 2020). Naturally existing fractures as well as chemically-induced fractures can significantly affect the domain transmissivity that, in turn, affect flow, transport and dissolution processes. When the porosity and permeability of the rocks are low, the fractures act as preferential fluid pathways (Mukhametdinova *et al.*, 2020). Therefore, fluid flow in the rock matrix can be neglected and therefore, the domain can be represented as a discrete fracture network (DFN) (Medici *et al.*, 2016, Lopes *et al.*, 2022; Aliouache *et al.*, 2019). Dissolution processes in DFNs are important in several environmental and engineering applications such as in heat extraction in geothermal reservoirs (Kim *et al.*, 2015), recovery in oil reservoirs (Medekenova and Jones, 2014; Khoei *et al.* 2020; Sahu *et al.* 2023), radioactive and nuclear waste disposal (Graf and Therrien, 2007; Natarajan and Suresh Kumar, 2010) and carbon sequestration (Hosseini *et al.* 2020; Tran and Jha, 2021; Li *et al.* 2021).

Despite the wide range of applications, dissolution processes in DFNs are still poorly investigated (Lopes *et al.*, 2022; Aliouache *et al.*, 2019). Most of the existing studies have focused on dissolution processes in unfractured domains and how these processes can create preferential flow pathways (Hanna and Rajaram, 1998; Dijk *et al.*, 2002; Detwiler *et al.*, 2003; Detwiler and Rajaram, 2007; Deng and Spycher, 2019). Experimental investigations have shown that, depending on the experimental conditions, different dissolution behaviors can be observed. This complicates the interpretation of the effects of controlling parameters on the flow, transport, dissolution and fracture evolution processes (Detwiler and Rajaram, 2007). Modeling has been also used to understand fractures initiation and propagation in unfractured domains due to dissolution processes (Detwiler and Rajaram, 2007; Ameli *et al.*, 2014; Deng and Spycher, 2019; Sahu *et al.* 2023). However, modeling-based studies for dissolution

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2 78 processes in well-developed discrete fracture networks and the effect of the reaction rate on
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6 79 the dissolution of connected fractures are poorly investigated (Deng and Spycher, 2019).
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10 80 Modeling dissolution processes in DFNs reveals specific challenges that are not present in
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12 81 unfractured media (Viswanathan et al., 2022). The main challenge is the dynamic evolution of
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14 82 the fractured system due to the geochemical dissolution (Tenthorey and Fitzgerald, 2006).
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16 83 The enlargement of fractures as a result of the dissolution process continuously affects the
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18 84 fluid flow, which in turn, affects the dissolution process and the transport of dissolved species.
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20 85 Therefore, the four processes (*i.e.* flow, transport, dissolution and fracture enlargement) are
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22 86 strongly coupled and nonlinear. In addition, in DFNs, the transport of the dissolved species is
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24 87 often advection-dominated due to fast fluid flow in fractures. In such a case, traditional
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26 88 numerical methods, such as standard finite element (FE) or finite volume (FV) methods can
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28 89 generate excessive numerical diffusion that may distort the dissolution process. Traditional
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30 90 numerical methods can also generate non-physical oscillations which can be the origin of
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32 91 convergence issues that hamper the applicability of the numerical models.
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36 92 The Discontinuous Galerkin (DG) finite element method is well-adapted to capture sharp
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38 93 moving fronts occurring in advection-dominant transports (Younes and Ackerer, 2008). The
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40 94 discretization of the hyperbolic term of the transport equation with DG has been largely
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42 95 reported in the literature. For instance, Younes *et al.*, (2014) have developed an efficient DG
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44 96 model for buoyancy-driven flow problems in unfractured porous media. In fractured domains,
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46 97 Zidane and Firoozabadi (2014) and Moortgat *et al.*, (2016) have developed a DG numerical
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48 98 model for multicomponent compressible flow in 2D and 3D domains. However, the
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50 99 developed schemes employed the DG method only for the transport in the matrix continuum.
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55 100 In these studies, transport in fractures, where advection is dominant, is modeled with an
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57 101 upwind first order FV scheme which is known to introduce excessive numerical diffusion
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59 102 (Konz *et al.*, 2009). Recently, Younes et al. (2023) have used the DG method for modeling
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103 coupled flow and mass transport processes in fractured porous media with no reactive
104 processes and with constant fracture aperture. To the best of our knowledge, the DG method
105 has never been used for modeling coupled flow, mass transport and reactive dissolution
106 processes in a discrete fracture network.

107 The objective of this paper is to develop an advanced numerical model based on the DG
108 method for the simulation of dissolution processes in DFNs, and to take advantage of this
109 model for investigating the effects of geochemical dissolution rate on fracture evolution, flow
110 and transport processes. The upwind P1-DG scheme developed by Younes *et al.* (2009) is
111 adopted to simulate the advection processes in the network of 1D fractures. With this scheme,
112 the degrees of freedom are the mean concentration on elements and the concentration
113 gradient. The Riemann solver is then used to evaluate the concentration at the nodes. The
114 main challenge in applying this technique for a DFN is the evaluation of the concentration at
115 nodes of intersection of several fractures. A new formulation is developed in this work where
116 the upstream concentration at a fracture intersection node is calculated using the average of
117 the concentrations of all fractures providing flow to that node, weighted by the flow rate in the
118 corresponding fractures.

119 Darcy's law combined with the cubic law is used to simulate flow within the fractures. The
120 flow equations are discretized with the hybrid formulation of the Mixed Finite Element
121 (MFE) method (Younes *et al.*, 2009). The mass lumping technique developed by Koohbor *et*
122 *al.* (2020) is employed to avoid over- and under-shoots observed in transient flow simulations
123 with small time steps. The MFE method is also used for the discretization of the dispersion
124 term to ensure the continuity of the dispersive flux at nodes where several fractures with
125 different apertures intersect. The highly nonlinear coupled equations of fluid flow, advection-
126 dispersion transport and aperture evolution due to dissolution are solved simultaneously in
127 order to avoid operator-splitting errors. The time discretization is performed using a high

128 order integration method via the method of lines (MOL). MOL has been demonstrated to be
 129 very efficient in solving high nonlinear systems of equations, such as unsaturated flow in
 130 porous media (Fahs et al., 2009; Farthing et al., 2003; Miller et al., 2006). With MOL, the
 131 spatial derivatives are discretized while the time derivatives are kept in their continuous form.
 132 This allows for converting the partial differential equations to a system of ordinary differential
 133 equations. Then, we use high order implicit time discretization with the DASPK solver
 134 (Brown et al., 1994) which is highly efficient and accurate for solving nonlinear systems with
 135 a large number of unknowns (Younes *et al.*, 2011).

136 The paper is organized as follows: First, the partial differential equations describing flow,
 137 transport and dissolution processes are presented. Second, the new DG-DFN numerical model
 138 is described. Then, the validation and accuracy of the new model is investigated by
 139 comparison against a standard FE model, obtained using COMSOL Multiphysics, and an in-
 140 house upwind FV model. Finally, the DG model is employed to investigate the effect of the
 141 dissolution rate on flow, transport and aperture evolution for a single fracture and for a DFN.

142 2. The Mathematical model

143 The fractures are simulated as highly permeable porous media. The permeability of the
 144 fractures is estimated using the well-known Poiseuille equation (Hanna and Rajaram, 1998).
 145 The flow process in a fracture with a variable aperture $b[L]$ is governed by the fluid mass
 146 conservation equation:

$$\frac{\partial(\theta b)}{\partial t} + \nabla \cdot (b\mathbf{q}) = 0 \quad (1)$$

147 and the Darcy law:

$$\mathbf{q} = -\frac{\rho_0 g \kappa}{\mu} \nabla H = -\frac{\rho_0 g b^2}{12\mu} \nabla H \quad (2)$$

148 where, $H[L]$ is the hydraulic head, $t[T]$ is the time and $\mathbf{q}[LT^{-1}]$ is the Darcy velocity, $b[L]$
 149 is the local fracture aperture, $\theta[-]$ is the porosity, $g[LT^{-2}]$ is the gravity acceleration,
 150 $\mu[M.L^{-1}.T^{-1}]$ is the dynamic viscosity of the fluid and $\kappa[L^2]$ is the permeability calculated
 151 using the cubic law $\kappa = \frac{b^2}{12}$ (Witherspoon *et al.*, 1980; Graf and Therrien, 2007; He *et al.*,
 152 2021).

153 Under isothermal conditions and no precipitation processes, the mass conservation of a
 154 contaminant species through a fracture, taking into account the dissolution process, is ruled by
 155 the advection-dispersion-reaction equation (Detwiler and Rajaram, 2007):

$$\frac{\partial(\theta b C)}{\partial t} + \nabla \cdot (b \mathbf{q} C) - \nabla \cdot (b \mathbf{D} \nabla C) = R(C) \quad (3)$$

156 where, $C[M.L^{-3}]$ is the concentration of the dissolved species, $R(C)[M.L^{-2}.T^{-1}]$ is the
 157 reaction term and $\mathbf{D}[L^2.T^{-1}]$ is the velocity-dependent dispersion tensor for a fracture filled
 158 by a porous medium.

159 The transport Eq. (3) writes

$$\theta b \frac{\partial C}{\partial t} + C \left(\frac{\partial(\theta b)}{\partial t} + \nabla \cdot (b \mathbf{q}) \right) + b \mathbf{q} \cdot \nabla C - \nabla \cdot (b \mathbf{D} \nabla C) = R(C) \quad (4)$$

160 which, using Eq. (1), simplifies to

$$\theta b \frac{\partial C}{\partial t} + b \mathbf{q} \cdot \nabla C - \nabla \cdot (b \mathbf{D} \nabla C) = R(C) \quad (5)$$

161 In the case of one-dimensional fracture, D is given by:

$$D = \alpha_L \times |\mathbf{q}| + D_m \quad (6)$$

162 where, $\alpha_L [L]$ is the longitudinal dispersivity in the fracture and $D_m [L^2.T^{-1}]$ is the diffusion
163 coefficient of the dissolved species.

164 Due to dissolution, the fracture aperture evolves during time as follows:

$$\rho_r \times \delta \times \frac{\partial b}{\partial t} - R(C) = 0 \quad (7)$$

165 where $\rho_r [M.L^{-3}]$ is the density of the rock and $\delta [-]$ is a stoichiometric coefficient that
166 represents the mass of mineral entering solution for a unit mass of dissolved rock.

167 In this work, under the assumption of no precipitation processes, the reaction term $R(C)$
168 representing the dissolution processes is modeled using a first-order approximation with a
169 constant reaction rate (Hanna and Rajaram, 1998; Detwiler and Rajaram, 2007; Steefel and
170 Lasaga, 1994; Steefel and Lichtner, 1998):

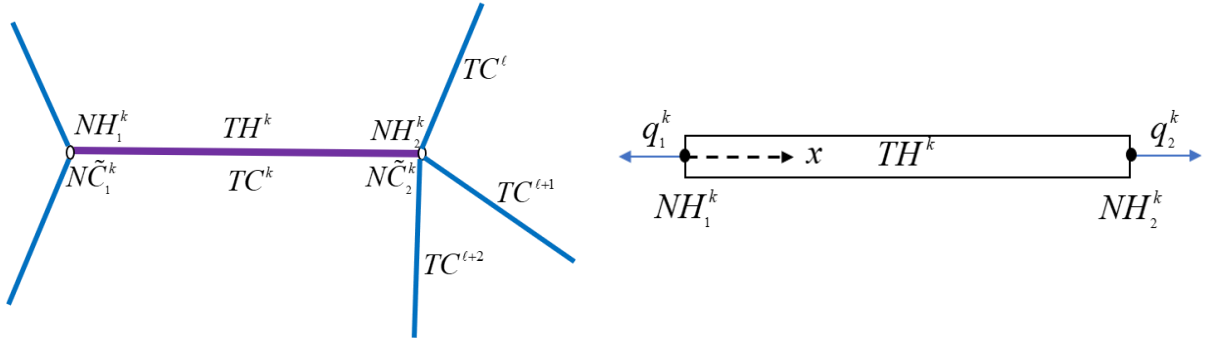
$$R(C) = K_C (C_s - C) \quad (8)$$

171 where, $K_C [L.T^{-1}]$ is the constant reaction rate and $C_s [M.L^{-3}]$ is the concentration of the
172 contaminant species at saturation.

3. The numerical model

174 To solve the set of equations (1-8) for flow, transport, and aperture evolution, we selected
175 numerical methods that provide accurate and consistent solutions for each kind of equation.
176 Thereby, the spatial discretization is based on the combination of the MFE method for flow
177 and dispersion, the upwind DG method for advection, and the FV method for reaction. We
178 summarize the main steps of the spatial discretization that allows for obtaining a system of
179 coupled ordinary differential equations. This system is then solved with the DASPK time
180 integration solver. The notations used for the spatial discretization of the flow and transport

181 are shown in Figure 1. In this figure NC_i (resp. NH_i) represents the concentration (resp.
 182 hydraulic head) at the intersection node i . TC^k (resp. TH^k) represents the concentration
 183 (resp. hydraulic head) at the fracture k . q_i^k is the outlet water flux at the node i of the
 184 fracture k .



185 **Figure 1.** Flow and transport notations in a fracture k .

186 **3.1 Spatial discretization of the flow**

187 The flow in the fracture network is discretized with the MFE method. Thus, for a fracture k
 188 of length l^k and width b^k , the velocity is approximated with linear test functions as follows:

$$189 \quad q^k = \sum_{j=1}^2 q_j^k w_j^k \quad (9)$$

190 where, q^k is the velocity inside the fracture k , $w_1^k = \frac{x - l^k}{b^k l^k}$ and $w_2^k = \frac{x}{b^k l^k}$ are the linear
 191 interpolation functions of the MFE method and q_j^k is the flux leaving the node j of the
 192 fracture k .

193 The continuity Eq. (1) is integrated over the fracture k yielding the following mass
 conservation equation:

$$194 \quad \theta^k l^k \frac{\partial b^k}{\partial t} + q_1^k + q_2^k = 0 \quad (10)$$

194 The variational formulation of Darcy's law Eq. (2) using w_i^k as a test function gives:

$$\sum_{j=1}^2 q_j^k \int_0^{\ell^k} w_i^k w_j^k = -\frac{\rho_0 g (b^k)^2}{12\mu} \int_0^{\ell^k} \nabla H w_i^k \quad (11)$$

195 Integration by part leads to:

$$\sum_{j=1}^2 q_j^k \int_0^{\ell^k} w_i^k w_j^k = -\frac{\rho_0 g (b^k)^2}{12\mu} \left(\int_0^{\ell^k} H (\nabla w_i^k) - [H w_i^k]_{x=0}^{x=\ell^k} \right) \quad (12)$$

196 Using the trapezoidal rule for the evaluation of the left integral yields:

$$q_i^k = \frac{\rho_0 g (b^k)^2}{12\mu} \times \frac{2b^k}{\ell^k} (TH^k - NH_i^k) \quad (13)$$

197 Eq. (13) is then substituted into Eq. (10) to obtain:

$$-\theta^k \ell^k \frac{\partial b^k}{\partial t} + \frac{\rho_0 g (b^k)^3}{6\ell^k \mu} (NH_1^k + NH_2^k - 2TH^k) = 0 \quad (14)$$

198 To close system (14), mass conservation is written at each node of fracture intersection. At the

199 node i surrounded by n fractures, we have:

$$\sum_n q_i^n = Q_i^* \quad (15)$$

200 where Q_i^* is the pumped/injected quantity at the intersection node i .

201 Substituting Eq. (13) into Eq. (15) yields:

$$\sum_n \frac{\rho_0 g (b^n)^3}{6\ell^n \mu} (TH^n - NH_i^n) = Q_i^* \quad (16)$$

202 Hence, the head NH_i at the intersection node i writes

$$NH_i = \frac{\sum_n \left(\frac{\rho_0 g (b^n)^3}{6 \ell^n \mu} TH^n \right) - Q_i^*}{\sum_n \frac{\rho_0 g (b^n)^3}{6 \ell^n \mu}} \quad (17)$$

For a given fracture network, the flow system is formed by writing Eq. (14) for all the fractures k in which, NH_1^k and NH_2^k are substituted using Eq. (17). This system is solved for the head traces TH^k at all mesh edges. Hence, the flow system has the number of fracture cells as degrees of freedom (DOF).

3.2 Spatial discretization of the transport

The DG method, which is well adapted for transport with sharp interface fronts, is employed for the discretization of the advection equation. With this method, we assume a linear discontinuous approximation of the concentration:

$$C^k(x, t)|_k = \phi_1^k(x) \times TC^k + \phi_2^k(x) \times C_x^k \quad (18)$$

where C^k is the concentration at the fracture k , which has two degrees of freedom: TC^k and C_x^k . TC^k is the average value of concentration in the fracture k and C_x^k is the concentration variation along the fracture direction. The corresponding interpolation functions are:

$$\phi_1^k(x) = 1, \quad \phi_2^k(x) = x - \bar{x}_k \quad (19)$$

where \bar{x}_k is the center of the fracture k .

The weak formulation of the transport Eq. (5) writes:

$$\theta^k b^k \int_0^{\ell^k} \frac{\partial C}{\partial t} \phi_i + b^k \int_0^{\ell^k} \nabla \cdot (\mathbf{q} C \phi_i) - b^k \int_0^{\ell^k} C \phi_i \nabla \cdot \mathbf{q} - b^k \int_0^{\ell^k} C \mathbf{q} \nabla \phi_i - \int_0^{\ell^k} \nabla \cdot (b \mathbf{D} \nabla C) \phi_i = \int_0^{\ell^k} R(C) \phi_i \quad (20)$$

In the following, we develop the calculation of the second term, for which a specific treatment is developed to solve the Riemann problem, and the fifth dispersive term, which is calculated

218 with the MFE to ensure the continuity of the dispersive flux at the intersection of several
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219 fractures.
 220 The second term is transformed into a boundary integral where the Riemann problem is
 221 solved at the interface as follows:

$$b^k \int_0^{\ell^k} \nabla \cdot (\mathbf{q} C \phi_i) = b^k \left(q_1^k C_1^{k,*} \phi_i \Big|_{x=0} + q_2^k C_2^{k,*} \phi_i \Big|_{x=\ell^k} \right) \quad (21)$$

222 The upstream concentration $C_j^{k,*}$ is given by

$$\left(C_j^{k,*} \right)_{j=1,2} = \begin{cases} NC_{j,in}^k & \text{if } q_j^k \geq 0 \\ NC_{j,out}^k & \text{if } q_j^k < 0 \end{cases} \quad (22)$$

223 where, $NC_{j,in}^k$ and $NC_{j,out}^k$ are the interior and outer concentrations at the node j of the
 224 fracture k , respectively.

225 The interior concentration $NC_{j,in}^k$ at the node j is obtained from Eq. (18):

$$NC_{j,in}^k = TC^k + \tau_j^k C_x^k \quad (23)$$

226 with $\tau_1^k = -\frac{\ell^k}{2}$ corresponding to the first node and $\tau_2^k = \frac{\ell^k}{2}$ to the second node of k .

227 The concentration $NC_{j,out}^k$ is calculated using the interior concentration ($NC_{j,in}^n$) of all the
 228 fractures n sharing the node j and having a positive flux q_j^n as follows:

$$NC_{j,out}^k = \frac{\sum_n q_j^n NC_{j,in}^n}{\sum_n q_j^n} \Bigg|_{q_j^n \geq 0} \quad (24)$$

229 Thus, the second integral in Eq. (20) can be written in the following form:

$$b^k \int_0^{\ell^k} \nabla \cdot (\mathbf{q} C \phi_i) = b^k q_1^k \begin{bmatrix} \varepsilon_1^k & (1-\varepsilon_1^k) \\ \varepsilon_1^k \tau_1^k & (1-\varepsilon_1^k) \tau_1^k \end{bmatrix} \begin{bmatrix} NC_{1,in}^k \\ NC_{1,out}^k \end{bmatrix} + b^k q_2^k \begin{bmatrix} \varepsilon_2^k & (1-\varepsilon_2^k) \\ \varepsilon_2^k \tau_2^k & (1-\varepsilon_2^k) \tau_2^k \end{bmatrix} \begin{bmatrix} NC_{2,in}^k \\ NC_{2,out}^k \end{bmatrix} \quad (25)$$

230 where ε_j^k is such that $\varepsilon_j^k = 1$ if $q_j^k \geq 0$, else $\varepsilon_j^k = 0$.

231 Using the MFE approximation for the dispersive flux $\tilde{q} = -D\nabla C$, the fifth dispersive integral
 232 in Eq. (20) writes:

$$-\int_0^{\ell^k} \nabla \cdot (bD\nabla C) \phi_i = \int_0^{\ell^k} \nabla \cdot (b\tilde{q}) \phi_i = \frac{\tilde{q}_1^k + \tilde{q}_2^k}{\ell^k} \int_0^{\ell^k} \phi_i = \begin{bmatrix} (\tilde{q}_1^k + \tilde{q}_2^k) \\ 0 \end{bmatrix} \quad (26)$$

233 where, \tilde{q}_j^k is the dispersive flux at the node j of the fracture k , calculated in a similar
 234 manner to the Darcy flux Eq. (13):

$$\tilde{q}_j^k = b^k D_k \frac{TC^k - N\tilde{C}_j}{\ell^k/2} \quad (27)$$

235 where $N\tilde{C}_j$ is the concentration at the node j .

236 $N\tilde{C}_j$ is calculated by imposing the continuity of the dispersive flux of all the fractures
 237 (including k) sharing the node j . For a node j shared by n fractures, $N\tilde{C}_j$ is such that:

$$\sum_n 2b^n D_n \frac{TC^n - N\tilde{C}_j}{\ell^n} = 0 \quad (28)$$

238 which yields:

$$N\tilde{C}_j = \frac{\sum_n \frac{b^n}{\ell^n} D_n TC^n}{\sum_n \frac{b^n}{\ell^n} D_n} \quad (29)$$

239 Thus, Eq. (27) becomes:

$$\tilde{q}_j = 2 \frac{b^k}{\ell^k} D_k \left(TC^k - \frac{\sum_n \frac{b^n}{\ell^n} D_n TC^n}{\sum_n \frac{b^n}{\ell^n} D_n} \right) \quad (30)$$

240 If the fracture k is filled by a porous medium, the dispersion coefficient is approximated by:

$$D_k = \alpha_L^k |\bar{q}_k| + D_m \quad (31)$$

241 where, α_L^k is the longitudinal dispersivity through the fracture k and $\bar{q}_k = \frac{(q_2^k - q_1^k)}{2b_k}$ is the

242 mean Darcy velocity in k .

243 Finally, the calculation of all integrals of the transport Eq. (20) gives

$$\begin{aligned} [M_1] \begin{bmatrix} \frac{\partial TC^k}{\partial t} \\ \frac{\partial C_x^k}{\partial t} \end{bmatrix} + b^k q_1^k \begin{bmatrix} \varepsilon_1^k & (1 - \varepsilon_1^k) \\ \varepsilon_1^k \tau_1^k & (1 - \varepsilon_1^k) \tau_1^k \end{bmatrix} \begin{bmatrix} NC_{1,in}^k \\ NC_{1,out}^k \end{bmatrix} + \\ b^k q_2^k \begin{bmatrix} \varepsilon_2^k & (1 - \varepsilon_2^k) \\ \varepsilon_2^k \tau_2^k & (1 - \varepsilon_2^k) \tau_2^k \end{bmatrix} \begin{bmatrix} NC_{2,in}^k \\ NC_{2,out}^k \end{bmatrix} + [M_2] \begin{bmatrix} TC^k \\ C_x^k \end{bmatrix} + \begin{bmatrix} (\tilde{q}_1^k + \tilde{q}_2^k) \\ 0 \end{bmatrix} = \begin{bmatrix} \ell^k R(TC^k) \\ 0 \end{bmatrix} \end{aligned} \quad (32)$$

244 where the terms of the (2×2) local matrices are $M_{1,ij} = \theta^k b^k \int_0^{\ell^k} \phi_i^k \phi_j^k$ and

245 $M_{2,ij} = -(q_1^k + q_2^k) \int_0^{\ell^k} \phi_i^k \phi_j^k - b^k \int_0^{\ell^k} \phi_j^k q \nabla \phi_i^k$, the expressions of $NC_{1,in}^k$ and $NC_{1,out}^k$ are given by

246 Eq. (23) and Eq. (24) and the dispersive fluxes \tilde{q}_1^k and \tilde{q}_2^k are given by Eq. (30).

247 If dissolution is considered, the aperture of the fracture can evolve with time, as ruled by Eq.

248 (7). The integration of this equation on the fracture k gives

$$\rho_r \delta \frac{\partial b^k}{\partial t} \ell^k - \ell^k R(TC^k) = 0 \quad (33)$$

249 Plugging Eq. (8) into Eq. (33) yields

$$\rho_r \delta \frac{\partial b^k}{\partial t} - K_c (C_s - TC^k) = 0 \quad (34)$$

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4 250 The final coupled nonlinear flow-transport-dissolution system to solve is formed by: (i) the
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6 251 flow Eq. (14) in which we substitute Eq. (17) yielding the hydraulic head at the fracture cells
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9 252 of the DFN, (ii) the transport Eqs. (23), (24), (30) and (32), which yield the mean and the
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11 253 variation of the concentration in each fracture and the (iii) the dissolution Eq. (34), which
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14 254 gives the aperture for each fracture.

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17 255 The obtained nonlinear system is discretized in time, via the method of lines. The main idea
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19 256 of this method is to keep the time derivative in its continuous form and to discretize the space
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22 257 derivatives. This results in a system of ODEs. The ODE system is solved using the DASPK
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24 258 solver which is based on a higher time integration scheme (Backward differentiation formula
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26 259 or Adams method). The resulting nonlinear system is solved using the Newton-Raphson
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29 260 method. The linear system arising at each time step is solved with the preconditioned Krylov
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31 261 iterative method. The solver adapts both the time step size and the order of the time
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34 262 integration scheme in order to reach the prescribed accuracy. Time step and order of
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36 263 integration are updated based on error control. In this work the accuracy is prescribed with a
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39 264 relative tolerance of 10^{-6} . Details of the MOL and the use of DASPK for nonlinear coupled
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41 265 flow transport systems are given in Younes *et al.* (2011; 2022).
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45 266 **4. Accuracy of the DG-DFN model**

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48 267 This section aims at verifying the correctness of the developed DG-DFN model and
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51 268 evaluating its performance for the simulation of advection dominant transport with dissolution
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53 269 in a fracture network. To this aim, the results of the DG-DFN model are compared to the
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56 270 results of COMSOL Multiphysics, which is based on the standard FE method, and to an in-
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58 271 house upwind FV model. Three examples are investigated. The two first examples deal
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272 respectively with dispersion dominant and advection dominant linear transport in a DFN. The
273 third example deals with coupled flow, transport, and aperture fracture evolution due to
274 dissolution in a single fracture with high diffusion.

4.1 Transport with high dispersion in a DFN (example 1)

276 Analytical solutions are often of great interest for the verification of numerical codes. For
277 coupled flow, transport and dissolution processes, some analytical and reference solutions
278 were developed under simplified conditions (Hayek *et al.*, 2012; Suk 2016). However, these
279 solutions cannot deal with fractured domains. Verification in fractured domains cannot be
280 performed against analytical solution. Therefore, to verify our newly developed code, we use
281 a standard FE numerical model developed in COMSOL Multiphysics software and an in-
282 house model based on the upwind FV method. The generation of the stochastic network is
283 performed with the “Discrete Fracture Network Add-In” which is available in the recent
284 versions of COMSOL. This Add-In creates a randomized distribution of position, size,
285 orientation and aperture for the fracture network. We used power law stochastic rule to create
286 the network as in Barton and Zoback (1992) and Hooker *et al.*, (2009). The network of
287 fracture for the Example 1 is shown in Figure 2a. A constant head gradient is imposed on the
288 network from left to right. The horizontal domain is initially free of contaminant. The left
289 inlet boundary of the domain has a fixed concentration. An outflow boundary condition is
290 considered for the right side of the domain. Impermeable boundary conditions are considered
291 on the top and bottom boundaries. The parameters for Example 1 are given in Table 1. This
292 first example involves high values of dispersion and diffusion to avoid unphysical
293 oscillations.

294 The results of the numerical models are compared on two observation points (O_1 and O_2) and
295 for a monitoring fracture (F_1) which is highlighted in Figure 2b.

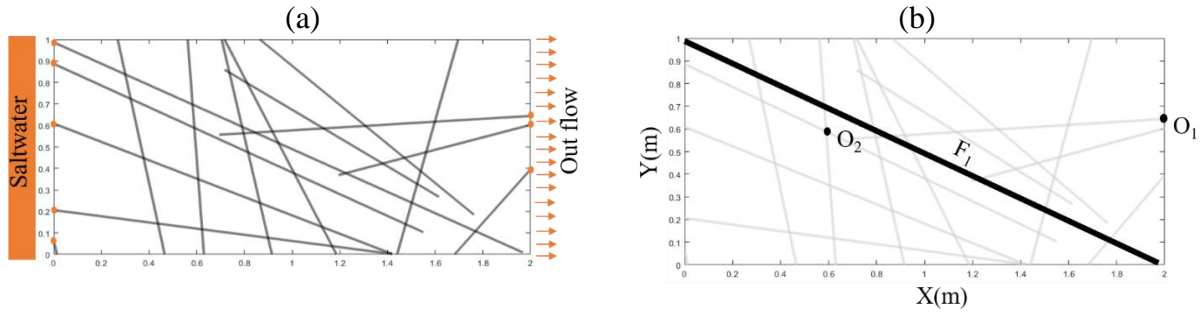


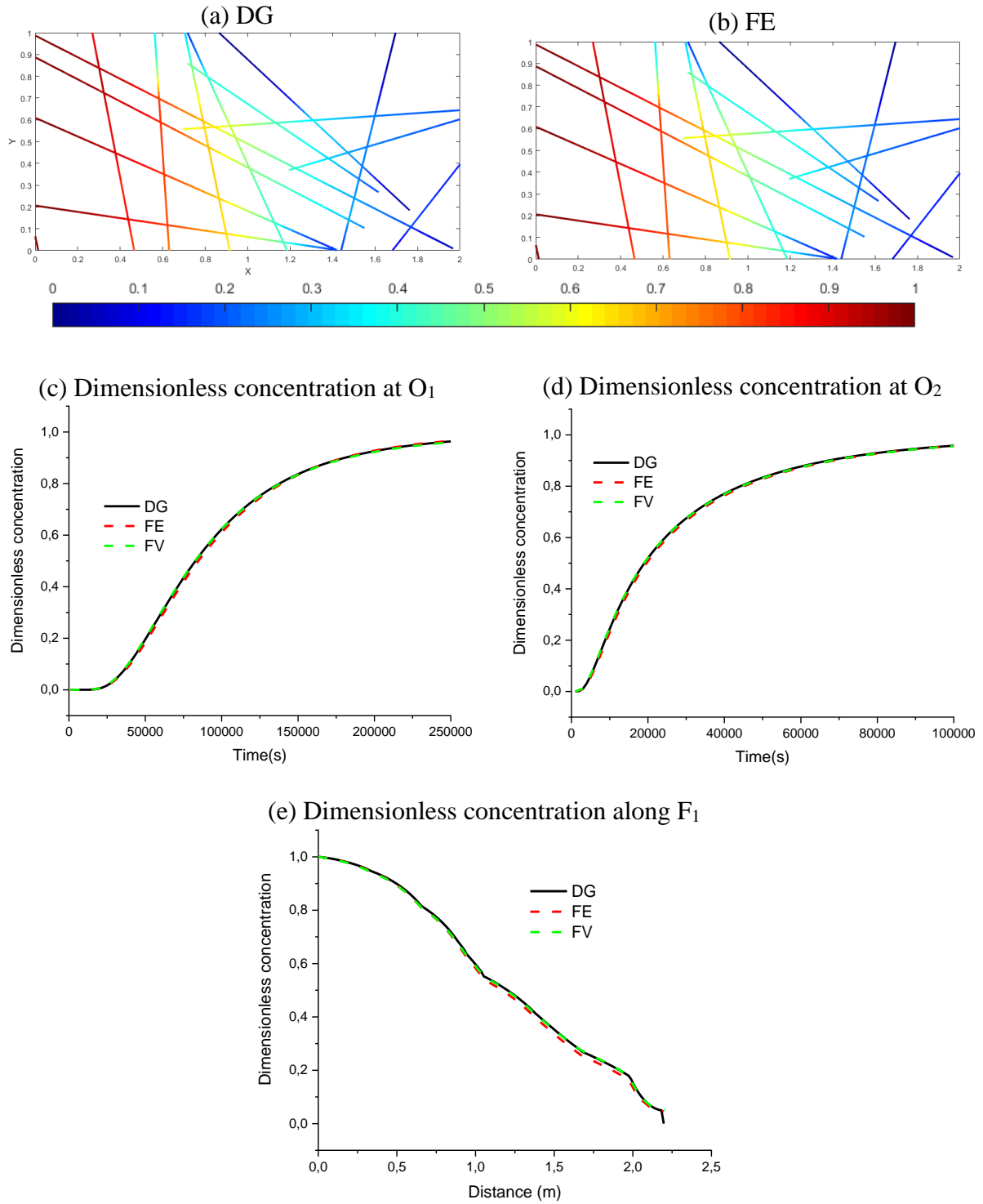
Figure 2. Problem description: (a) fracture network and boundary conditions, (b) Highlighted observation points (O_1 , O_2), and a monitoring fracture (F_1)

Table 1: Parameters used in the three test examples.

Freshwater density	$\rho_0 = 1000 \text{ kg}\cdot\text{m}^{-3}$		
Porosity	1.0		
Viscosity	$\mu = 0.001 \text{ Pa}\cdot\text{s}$		
Rock density	$\rho_r = 27000 \text{ mol}/\text{m}^3$		
Stoichiometric coefficient	$\delta = 0.4 \text{ kg}/\text{m}^3$		
	Example 1	Example 2	Example 3
Diffusion	$D_m = 10^{-6} \text{ m}^2/\text{s}$	$D_m = 10^{-9} \text{ m}^2/\text{s}$	$D_m = 10^{-7} \text{ m}^2/\text{s}$
Dispersivity	$\alpha = 0.2 \text{ m}$	$\alpha = 0$	$\alpha = 0$
Initial Thickness	$b = 1\text{cm}$	$b = 1\text{cm}$	$b_0 = 1\text{mm}$
Permeability	$\kappa = 10^{-11} \text{ m}^2$	$\kappa = 10^{-9} \text{ m}^2$	$\kappa = b^2/12$
Head at the upstream	$H_1 = 1\text{m}$	$H_1 = 1\text{m}$	$H_1 = 0.01\text{m}$
Head at the downstream	$H_0 = 0$	$H_0 = 0$	$H_0 = 0$
Reaction rate	(-)	(-)	$K_C = 10^{-5} \text{ m}/\text{s}$
Initial concentration	$C_0 = 0$	$C_0 = 0$	$C_0 = 0.1768 \text{ mol}/\text{m}^3$
Inlet concentration	$C_{inj} = 35 \text{ g}/\text{l}$	$C_{inj} = 35 \text{ g}/\text{l}$	$C_{inj} = 0$

The distribution of dimensionless concentration in the fracture network after 14 hours with DG and FE models are shown in Figures 3a and 3b, respectively. The dimensionless concentration is calculated as the ratio of the concentration to the concentration of injected fluid. Similar concentration distributions are obtained with DG and FE solutions. The evolution of the concentration with time with DG, FE and FV methods at the two observation points O_1 and O_2 are depicted in Figures 3c and 3d, respectively. The concentration with the

307 three models after 14 hours along F_1 are also plotted in Figure 3e. Excellent agreement is
 308 observed between the three model solutions (Figures 3c-3e). These figures show the
 309 correctness of the newly developed DG-DFN model.



310 **Figure 3.** Results for Example 1: distributions of the dimensionless concentration with
 311 DG (a) and FE (b) methods. Time variation of the concentrations at O_1 (c), at O_2 (d) and
 312 concentration distribution along the fracture F_1 (e) with the DG, FV and FE methods.

313 **4.2 Advection-dominated transport in a DFN (Example 2)**

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3 314 This example aims at investigating the advantages of the developed DG-DFN model in the
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5 315 case where the transport is advection dominated because of the fast fluid flow inside the
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8 316 fractures. The example is similar to Example 1, but we assume very little diffusion and no
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10 317 dispersion. The input parameters for Example 2 are given in Table 1.

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13 318 A mesh sensitivity analysis is performed with the DG, FV and FE methods. It is observed that
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16 319 with the DG scheme, a mesh-independent solution is obtained with a relatively coarse mesh
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18 320 (272 nodes). The time variation of concentration at the observation point O₂ obtained with DG
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21 321 using a coarse mesh formed by 272 nodes is compared to the solution of FE and FV using two
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23 322 meshes: a coarse mesh formed by 272 nodes and a fine mesh formed by 1145 nodes. The
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26 323 results of FE on the coarse mesh show strong unphysical oscillations (note plotted). When the
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28 324 mesh is refined, the FE solution improves but still contains important unphysical oscillations
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31 325 in the vicinity of the sharp concentration fronts (Figure 4a). The FV solution with the coarse
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33 326 mesh shows large numerical diffusion smearing the concentration front. When the mesh is
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36 327 refined, the FV solution yields smaller numerical diffusion and the FV results converge
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38 328 toward the DG results.

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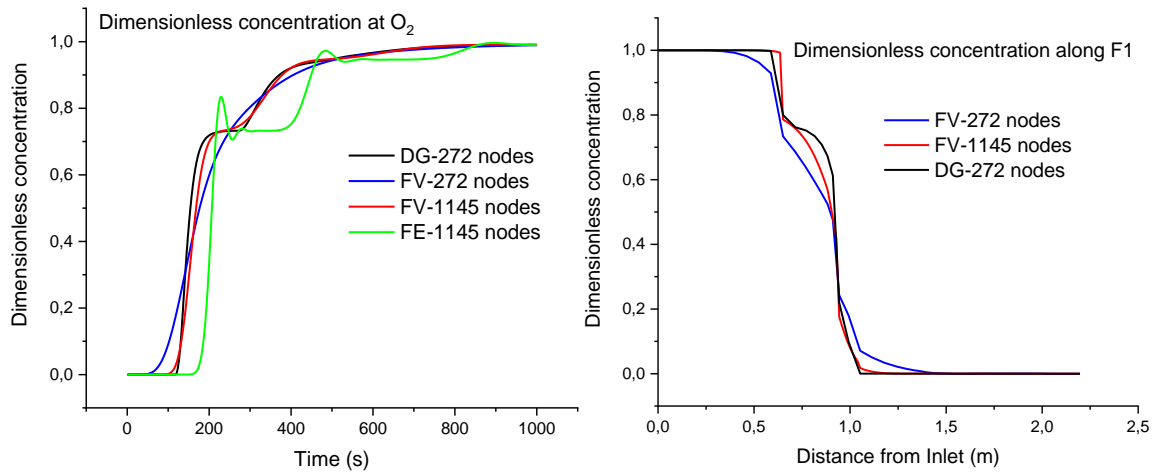


Figure 4. Comparison between DG, FE and FV for Example 2: (a) Time variation of the dimensionless concentration at the observation point O₂ and (b) distribution of the dimensionless concentration at t=200s along line F₁.

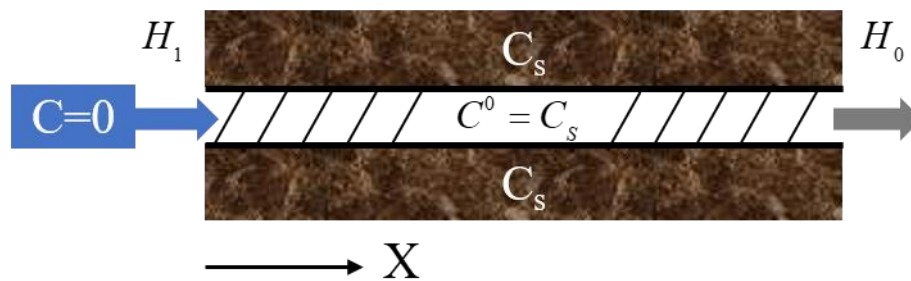
The convergence of FV results toward DG results by refining the mesh is also observed in the concentration profile along F₁ which is plotted at the time t=200 seconds in Figure 4b. This figure shows that the FV concentration profile along line F₁ is spread when using 272 nodes due to numerical diffusion. The use of a mesh formed by 1145 nodes improves the FV results, although they remain less accurate than the DG results with only 272 nodes. These results confirm the high accuracy and efficiency of the DG model for advection dominant transport in DFNs since it avoids the unphysical oscillations observed with FE and the large numerical diffusion observed with FV.

4.3 Coupled flow, transport and dissolution in a single fracture with high diffusion

(Example 3)

The simulated examples 1 and 2 allowed for the verification of the new DG code in the case of flow and transport in fractures with constant apertures (i.e., without dissolution). However, one of the challenges of the DG-DFN model is the accurate simulation of coupled flow-transport-dissolution processes with fracture evolution. Simulating these processes in a DFN is not possible with COMSOL. Hence, in this example, we consider flow and transport in a variable aperture single fracture (Figure 5). The fracture of 1m length is initially saturated by

351 the dissolving species. Freshwater is injected into the fracture from the left boundary and
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 2 352 leaves from the right. The COMSOL model has been developed by using 1D geometry and
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 4 353 coupling the interfaces ‘Darcy’s Law $-dl$ ’ for flow and ‘Transport of Diluted Species in
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 7 354 Porous media $-tds$ ’ for transport. Reaction term has been included in the transport with the
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 9 355 module “Reaction”. Equation (7) describing the evaluation of the fracture aperture due to
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 12 356 dissolution processes is considered in COMSOL using the interface “Domain ODEs and
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 14 357 DAEs” with the module “Distributed ODE”. All the equations are solved simultaneously in
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 16 358 COMSOL with the fully coupled approach.



359
 360 **Figure 5.** Conceptual model for dissolution in a single fracture.

361 The input parameters for Example 3 are shown in Table 1. The head difference $\Delta H = 0.01\text{m}$
 362 corresponds to an initial velocity of 0.00833 m/s (720 m/day). The problem is simulated with
 363 a high diffusion value $D_m = 10^{-7}\text{ m}^2.\text{s}^{-1}$. This high value enables a fair comparison between
 364 the FE, FV and DG models since it allows reducing the unphysical oscillations observed with
 365 the FE solution and the numerical diffusion generated by the upwind FV scheme. The three
 366 numerical models (FE, FV and DG) are employed to calculate the concentration of the solute
 367 species in the domain as well as the evolution of the thickness of the fracture due to the
 368 dissolution process. The temporal and spatial variation of the dimensionless concentration and
 369 of the fracture thickness are plotted in Figure 6. The dimensionless concentration is calculated
 370 as the ratio of the concentration to the initial concentration in the domain.

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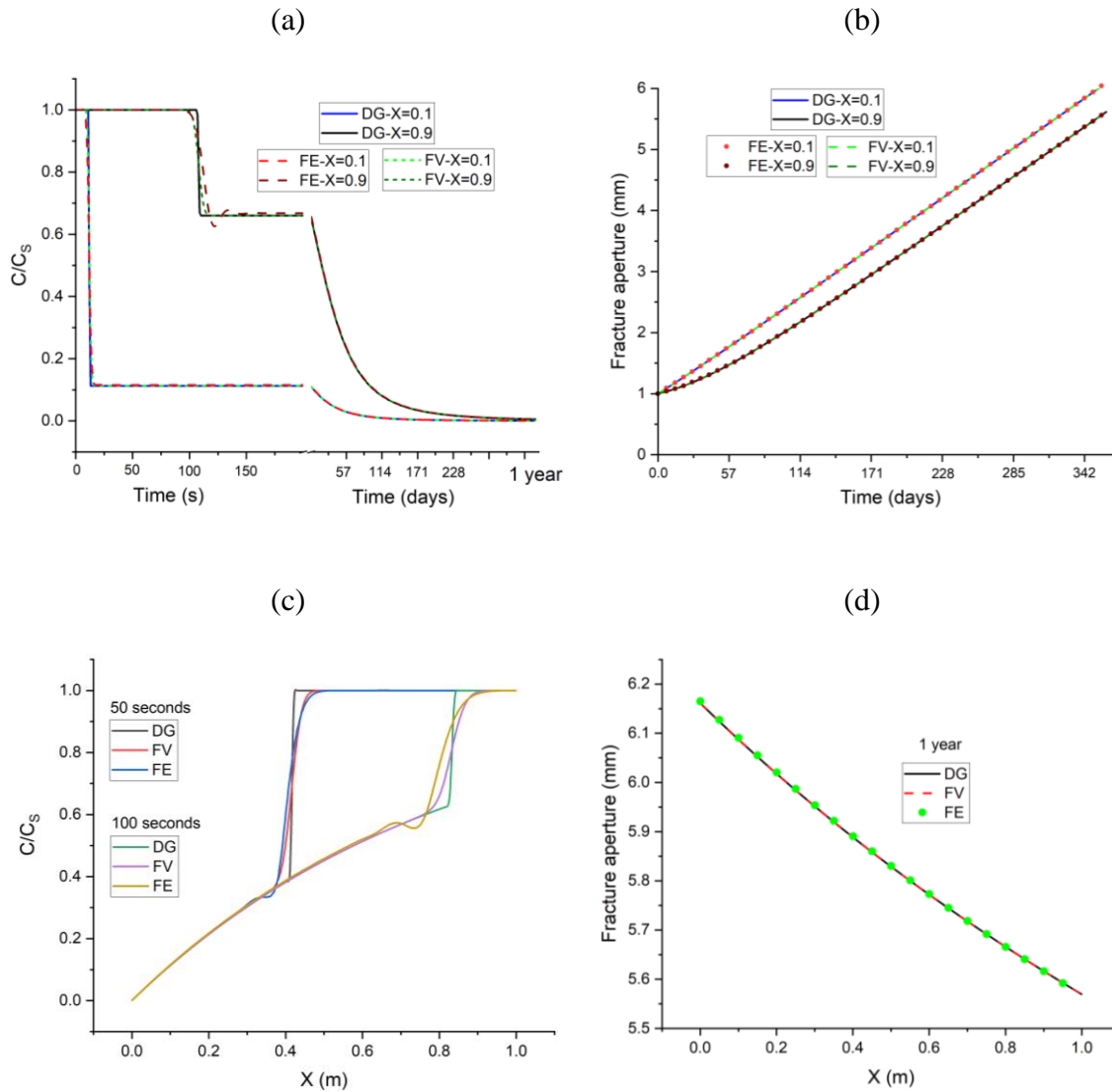


Figure 6. Example 3: Comparison between the FE, FV and DG results for dissolution in a single fracture: (a) time variation of the dimensionless concentration, (b) time variation of fracture aperture, (c) distribution of the dimensionless concentration along the fracture after 50 and 100 seconds and (d) aperture of the fracture after one year.

Figures 6a and 6b show the time variation of the dimensionless concentration as well as the thickness at two different points of the domain ($x=0.1$ m and 0.9 m). The dimensionless concentration at $t=50$ s and 100 s and the thickness after one year are also plotted along the fracture in Figures 6c and 6d, respectively. Although we have used a high diffusion value, the results show numerical oscillations in the FE solution, especially near the outlet (Figures 6a and 6c). These oscillations are notably observed at short times and disappear in the mid and

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383 long-term results. Indeed, in short times, advection is the dominant process that derives the
384 freshwater/saltwater front towards the outlet. At long times, dissolution and diffusion become
385 significant processes affecting thickness and concentration evolutions. Despite the differences
386 observed between the short-time concentrations of the three schemes, the long-term results are
387 almost similar, and the three numerical models yield equivalent results for the concentration
388 distribution and the aperture evolution. These results show the correctness of the DG-DFN
389 model for coupled flow-transport-dissolutions problems.

390 **5. Effect of the dissolution rate on the flow, transport and aperture** 391 **evolution**

392 In this section, the DG-DFN model is used to investigate the effect of the dissolution rate on
393 the flow, transport and aperture evolution in the case of a single fracture and in the case of a
394 DFN.

395 ***4.1 The case of a single fracture***

396 The investigated test case is similar to Example 3 (Figure 5), but we consider a more realistic
397 problem with a reduced diffusion coefficient $D_m = 10^{-9} \text{ m}^2 \cdot \text{s}^{-1}$ and use the new DG model to
398 assess the effect of the dissolution rate on the fracture aperture evolution. The problem is
399 simulated using low ($K_c = 10^{-5} \text{ m/s}$) and high ($K_c = 10^{-4} \text{ m/s}$) dissolution rates.

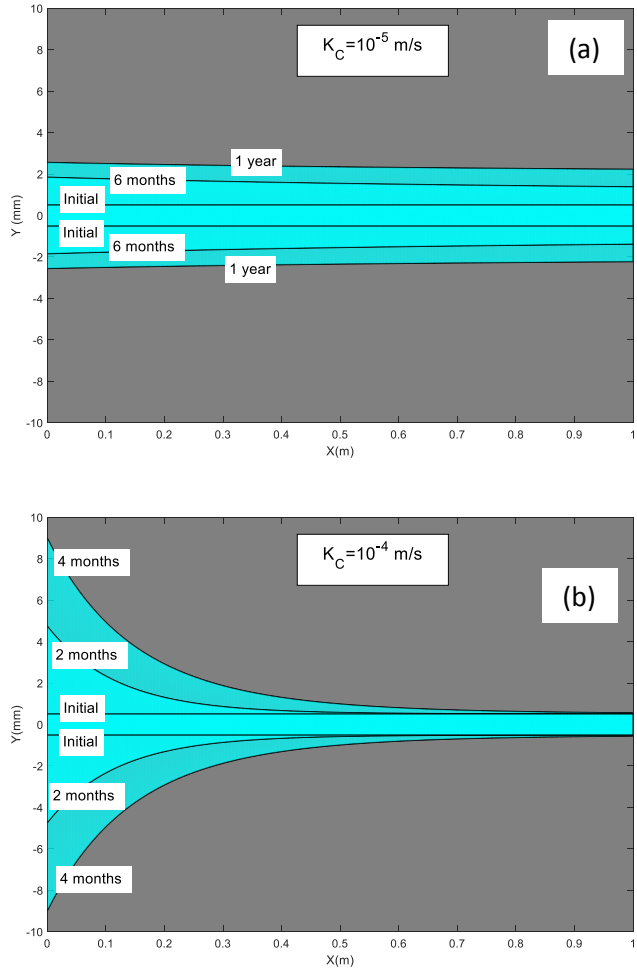


Figure 7. Fracture aperture evolution: (a) for low ($K_C = 10^{-5}$ m/s) and (b) for high ($K_C = 10^{-4}$ m/s) dissolution rates.

For the case of low dissolution, Figure 7a shows an almost uniform enlargement of the fracture both at 6 months and 1 year. In this situation, the dissolution is very slow while the fluid velocity is relatively high. Thus, the crossing fluid remains weakly contaminated (the fluid is never saturated by the dissolved species during its travel). The enlargement of the aperture (Figure 7a) is a little more pronounced near the inlet, where the concentration is zero, than near the outlet, where the fluid is slightly contaminated. Figures 8a, b and c depict, respectively the concentration, the aperture and the velocity distribution along the fracture at $t = 6$ months.

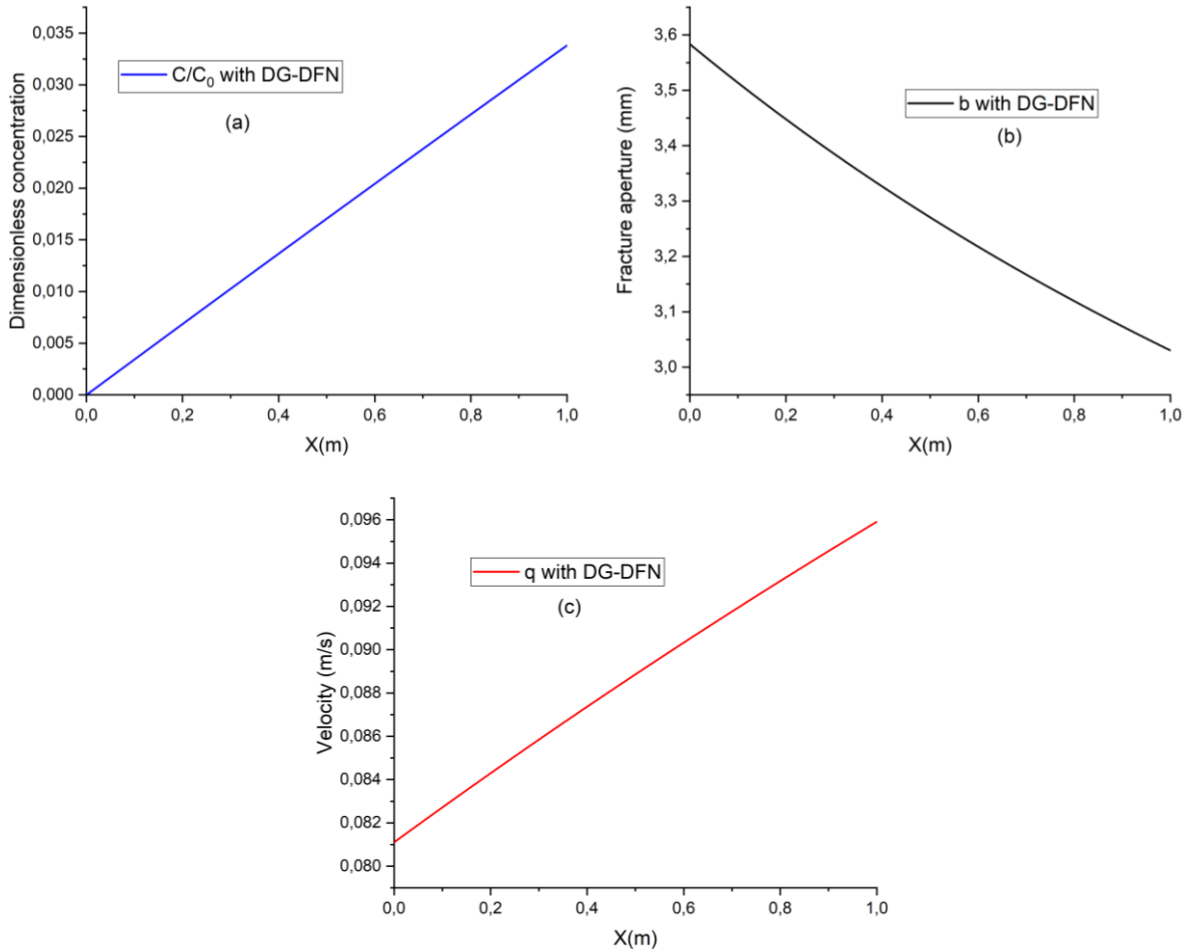


Figure 8. Dimensionless concentration (a), aperture (b) and velocity (c) along the fracture at $t = 6$ months for the low dissolution case.

In Figure 8a, the concentration increases linearly inside the fracture from the left boundary, where the concentration is zero (freshwater boundary condition) to the right outlet boundary. During its travel, the fluid is contaminated by the dissolved species, which explains the linear increase of the concentration along the fracture (Figure 8a). Because of the low dissolution rate, the concentration of the fluid remains very small, reaching a maximum dimensionless concentration of only 0.035 near the outlet. Hence, the dissolved quantity along the fracture, which is proportional to $(C_s - C)$, is almost constant. Figure 8b depicts the enlargement along the fracture at $t = 6$ months. The aperture is around 15% more significant near the upstream (where the concentration is fixed to zero) than near the downstream. On the other hand,

1
2 423 because of mass conservation, a linear increase of the velocity is observed along the fracture
3 424 (Figure 8c) while the aperture decreases linearly (figure 8b).

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5 425 Figure 9 depicts the time evolution of the concentration, the aperture, and the velocity at the
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7 426 center ($x=0.5m$) of the fracture. The dimensionless concentration (figure 9a) decreases
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9 427 quickly from 1 (initially, the fluid inside the fracture is saturated by the dissolution species) to
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11 428 0.4 due to fast arrival of the advection front. Then, the concentration decreases with time due
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13 429 to the enlargement of the fracture aperture. After 1 month, the concentration is very small,
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15 430 thus the rate of dissolution, which is proportional to $(C_s - C)$ is almost constant. This explains
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17 431 the linear increase of the fracture aperture with time, as shown in Figure 9c. The velocity
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19 432 inside the fracture evolves more significantly during time (Figure 9b) since it is proportional
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21 433 to the square of the aperture (See Eq.(2)).
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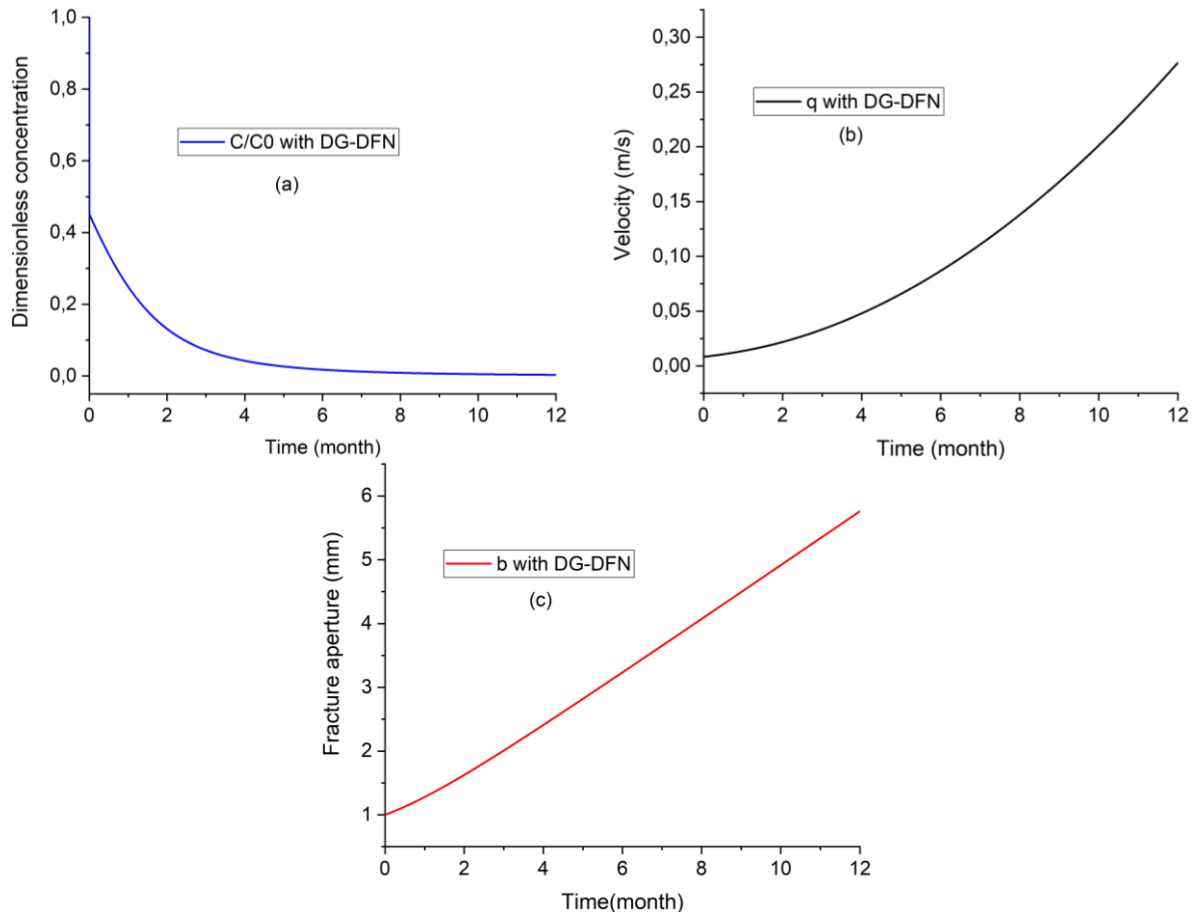
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440 **Figure 9.** Time evolution of the dimensionless concentration (a), velocity (b) and aperture (c)
 441 at $x=0.5\text{m}$ the low dissolution case.

442 For the case of a high dissolution rate, Figure 7b shows a funnel-shaped enlargement of the
 443 fracture with a strong increase of the aperture near the upstream, whereas the downstream
 444 remains unchanged. This result is coherent with the experimental results obtained in Li et al.
 445 2021. Figures 10a, b and c depict, respectively the concentration, the aperture, and the
 446 velocity distribution along the fracture at $t = 2\text{months}$.

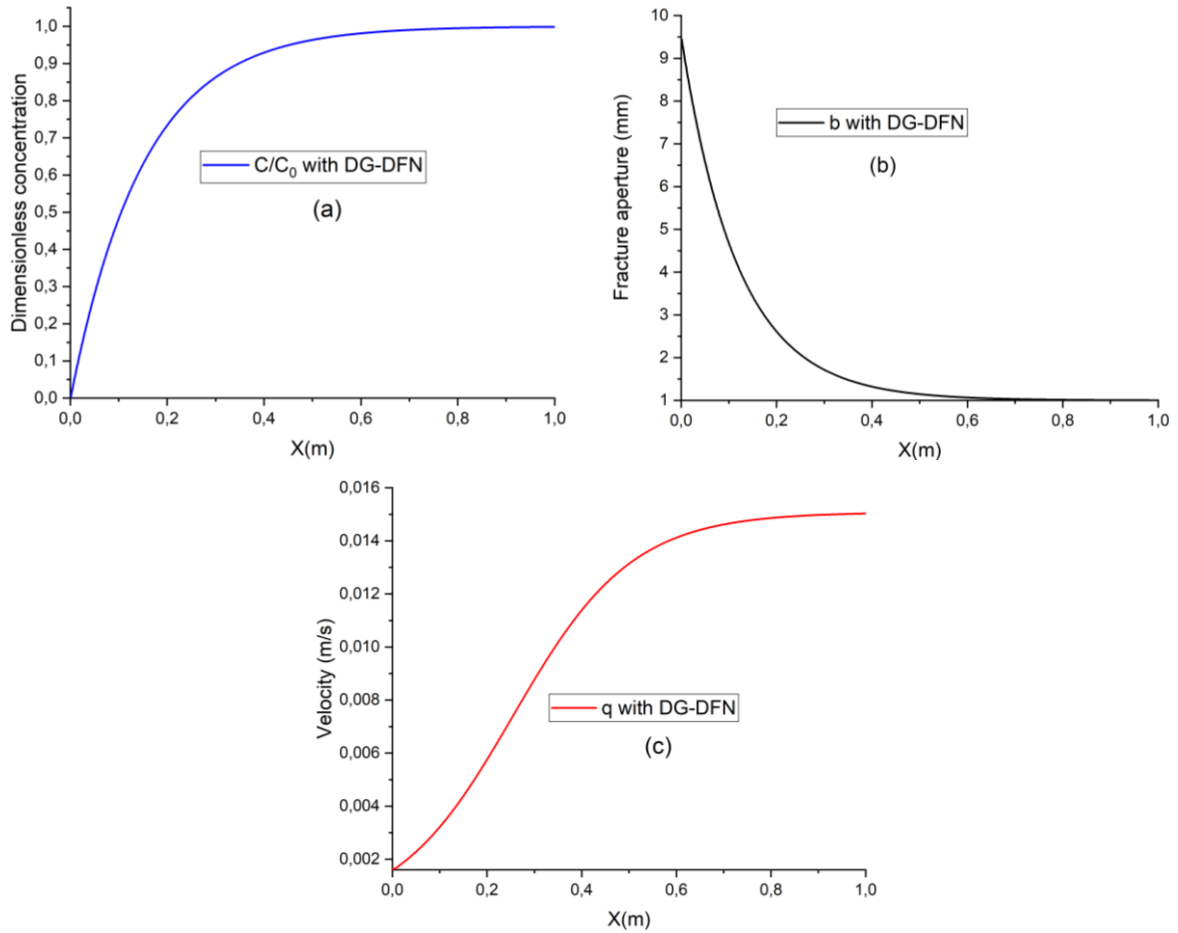


Figure 10. Dimensionless concentration (a), aperture (b) and velocity (c) along the fracture at $t = 2$ months for the high dissolution case.

Because of the high dissolution rate, the concentration of the fluid crossing the fracture increases significantly from zero (at the entry of the fracture) to reach 90% of C_s after only 0.3m (Figure 10a) and the fluid is almost saturated by the dissolved species at around 0.5m. As a consequence, the fracture evolution, which is proportional to $(C_s - C)$, is very high near the inlet, but quickly decreases and becomes negligible for the second half of the fracture (Figure 10b). Further, due to mass conservation, the fluid velocity along the fracture (Figure 10c) starts with a very weak value at the left (because of the high enlargement at the entry), then, it increases until around the center of the fracture (because of the decrease of the aperture) and remains almost constant for the second half of the fracture for which the aperture was unchanged.

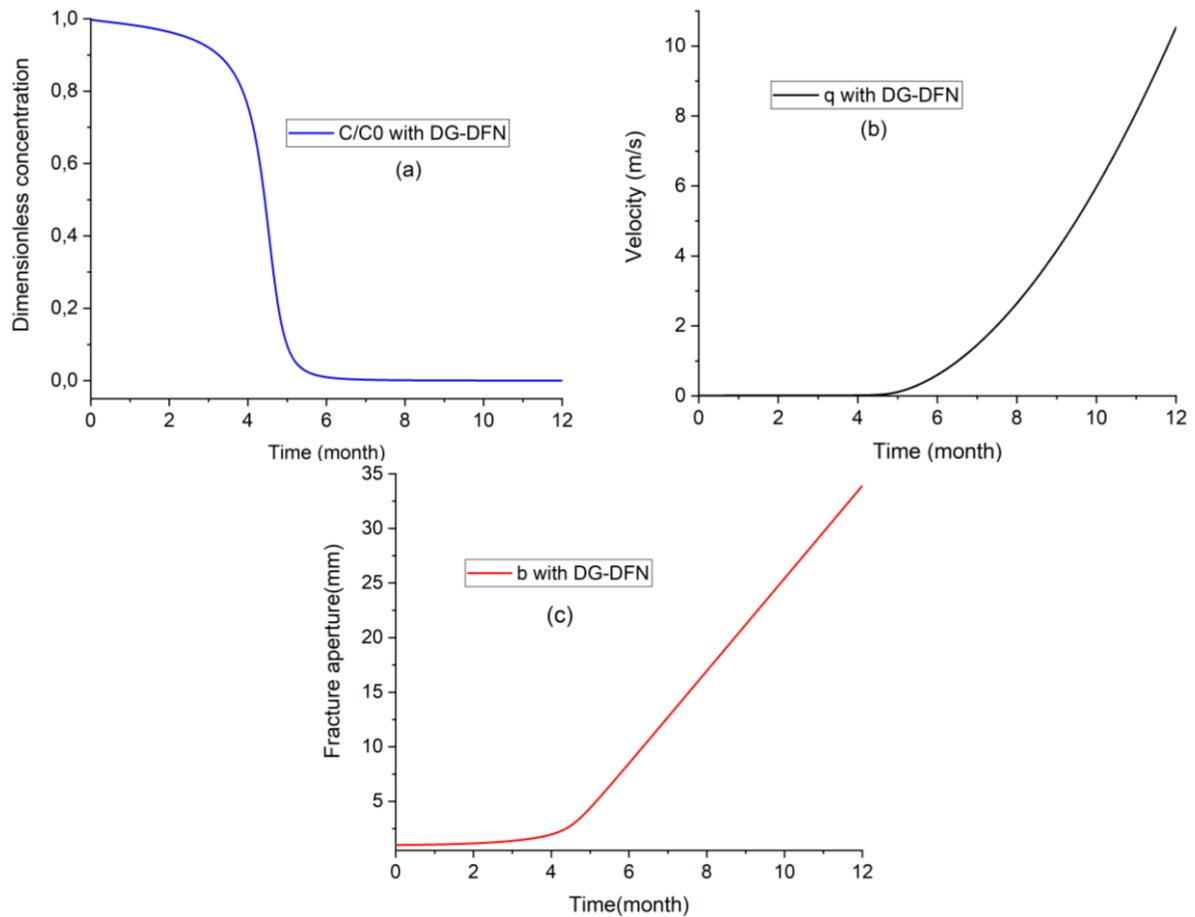


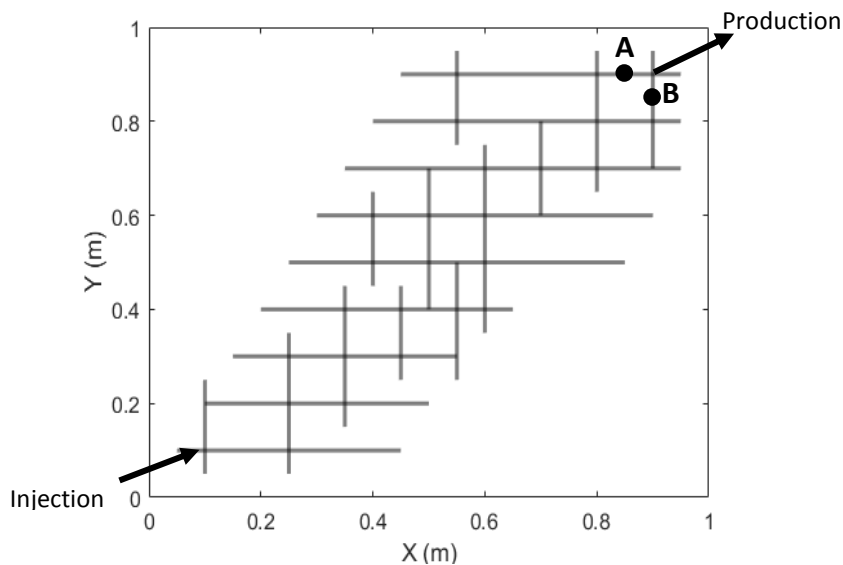
Figure 11. Time evolution of dimensionless concentration (a), velocity (b) and aperture (c) at $x=0.5\text{m}$ for the high dissolution case.

Figure 11 depicts the time evolution of the concentration, velocity and aperture at the center of the fracture ($x = 0.5\text{m}$) for the high dissolution case. The velocity (Figure 11b) remains very small inside the fracture until a time $t^* \approx 5$ months, then it starts to considerably increase with time. Indeed, during the first period ($t \leq t^*$), the funnel aperture distribution (Figure 7b) yields a permeability (proportional to the square of the aperture) distribution which is very high near the entry (large aperture), and very low for the right part of the fracture with the unchanged small aperture. For such a situation (low and high permeabilities in series), the fluid velocity is mainly controlled by the low permeability. Thus, the velocity remains small until the enlargement reaches the right end of the fracture which occurs at around t^* . After that time, all the fracture enlarges (Figure 11c) and the equivalent permeability of the fracture strongly increases, yielding a highly increasing velocity (Figure 11b). As a consequence, the

473 concentration in the fracture (Figure 11a) strongly reduces after t^* due to the high advection
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3 474 flux of freshwater arriving from the left boundary.

475 *4.2 Effect of the dissolution rate in the case of a DFN*

476 This section extends the discussion of the previous section to a DFN. We consider a network
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11 477 of connected horizontal and vertical fractures, as in Figure 12.



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35 480 **Figure 12.** Conceptual model for flow, dissolution and transport in a network of fractures.

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39 482 The domain is a horizontal plane, initially saturated with a solute species and as shown in the
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41 483 Figure 12, the freshwater is injected at the left-bottom corner. Contaminated water, due to the
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44 484 dissolved species, is pumped out at the right-top corner. The production rate is fixed to 10^{-4}
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46 485 m^2/s and is similar to the injection rate. Two dissolution values are considered, corresponding
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49 486 to low ($K_c = 10^{-5} \text{ m/s}$) and high ($K_c = 10^{-4} \text{ m/s}$) dissolution rates. For the rest of the input
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52 487 parameters, the same values are considered as in the previous case of the single fracture.

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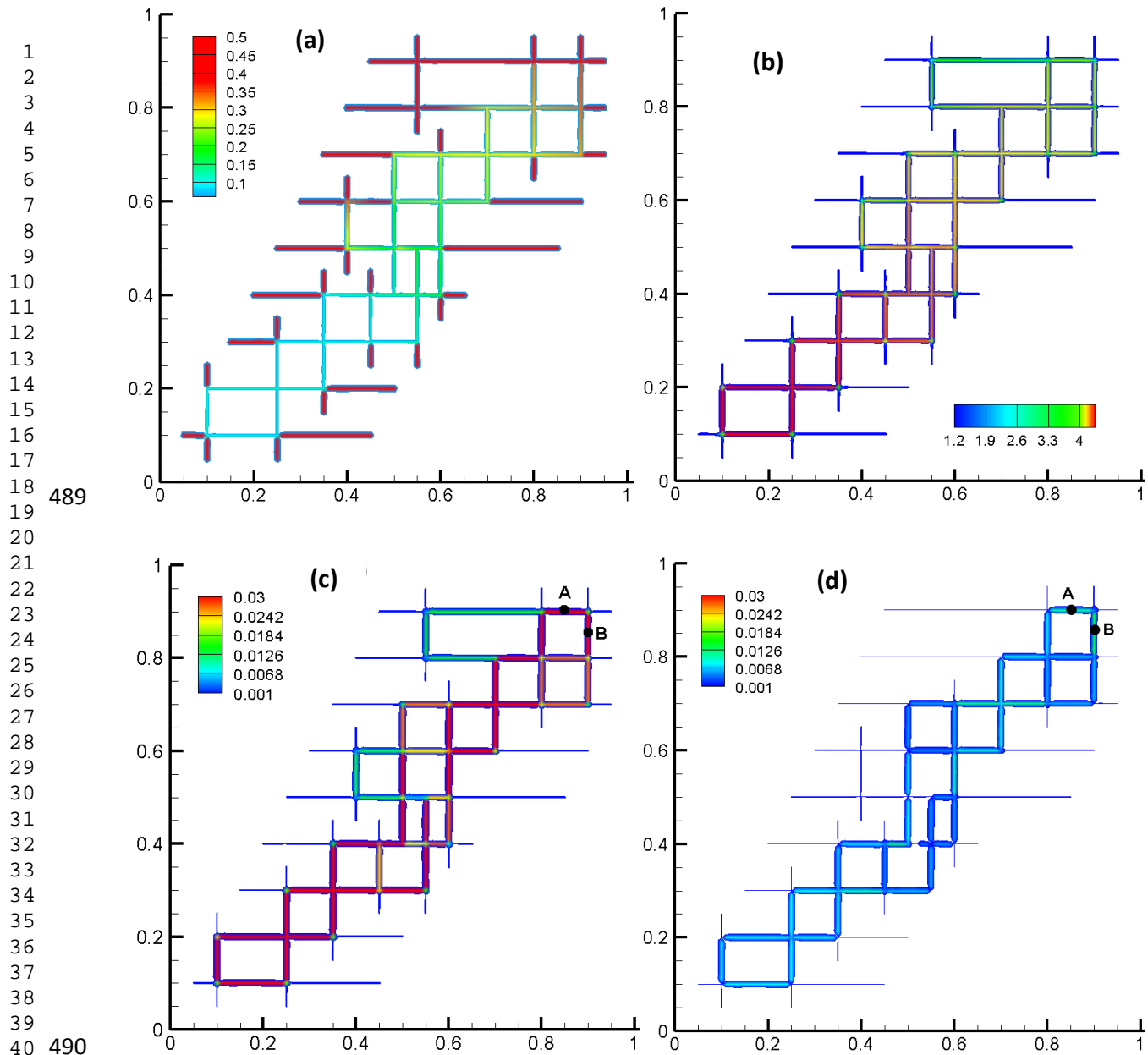


Figure 13. Results for the low dissolution rate ($K_c = 10^{-5}$ m/s): (a) distribution of the dimensionless concentration after one year, (b) aperture (mm) distribution after 1 year, (c) and (d) are the distributions of the velocity magnitude (m/s) after 100s and 1 year, respectively.

Figure 13a shows the distribution of the dimensionless concentration after one year. Initially, all fractures are filled by the dissolved species, and no dissolution occurs. During its travel, the injected freshwater is contaminated by the dissolution process. As a consequence, the concentration inside the fractures increases from the injection well to the production well as can be observed in the Figure 13a. This figure also shows that isolated fracture branches remain contaminated by the dissolution species as the injected water cannot go through these

500 branches since they are plugged at their extremities. Figure 13b shows the fracture aperture
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2 501 distribution in the DFN after one year. Since dissolution is proportional to the difference
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5 502 ($C-C_s$), the aperture is maximum near the injection well, reaching 6mm and decreases
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8 503 almost linearly when going to the production well in the vicinity of which it reaches 4mm.
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10 504 This uniform decrease is almost similar to the one observed for the single fracture in the case
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12 505 of the small dissolution rate (Figure 7a). Figure 13c and 13d show the velocity distribution,
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15 506 respectively, after 100s and 1 year. At $t=100s$, the velocity is almost significant in all
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17 507 connected fractures (it is zero for plugged fractures). Due to the enlargement of fractures
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20 508 aperture, the velocity inside the fractures decreases with time as can be observed after 1 year
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22 509 in Figure 13d. Note that this phenomenon has not been observed for the single fracture for
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25 510 which we obtained an increase of the velocity with time for both low (Figure 9b) and high
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27 511 (figure 11b) dissolution rates. The difference between the previous single fracture
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30 512 configuration and the current DFN configuration is due to the difference of the boundary
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32 513 conditions between these two configurations. Indeed, for the single fracture, we had fixed
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35 514 hydraulic heads in upstream and downstream. Thus, the total flow rate crossing the fracture, is
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37 515 controlled by the difference between the upstream and downstream heads as well as by the
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40 516 permeability of the fracture. The enlargement of the fracture, due to dissolution, induces a
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42 517 significant increase of the flow rate because of the increase of the permeability, which is
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44 518 proportional to the square of the aperture. Hence, due to dissolution, the velocity inside the
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47 519 fractures increases with time (see Figures 9b and 11b). For the current DFN configuration, the
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49 520 total flow rate is fixed and corresponds to the injected flow rate. Thus, dissolution induces an
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52 521 enlargement of the fracture aperture, which induces a decrease in the velocity because of mass
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54 522 conservation principle. Note that for the DFN configuration, although the total flow rate is
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57 523 fixed, the flow rate inside each fracture varies with time since the distribution of the aperture
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59 524 of fracture branches can vary due to dissolution. As a consequence, during simulation, the

525 distribution of the permeability in the DFN can vary, which can induce a modification of the
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2 526 flowrate repartition between the fractures. For instance, Figure 14 shows the aperture, velocity
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5 527 and flow rate evolution for the two last fractures A and B in the vicinity of the pumping well
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7 528 (see location in Figure 12). For both fractures A and B, the aperture increases linearly with
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10 529 time (Figures 14a and 14d). The velocity inside the two fractures decreases with time (Figures
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12 530 14b and 14e). The flow rate inside the two fractures is not constant and has a different
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15 531 monotonic behavior during time. The flow rate decreases with time in the fracture A (Figure
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17 532 14c), whereas it increases in the fracture B (Figure 14f). The sum of the two flowrates in the
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19 533 fractures A and B is constant and is equal to the pumped flowrate since A and B are the only
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22 534 non-plugged fractures connected to the pumping well.
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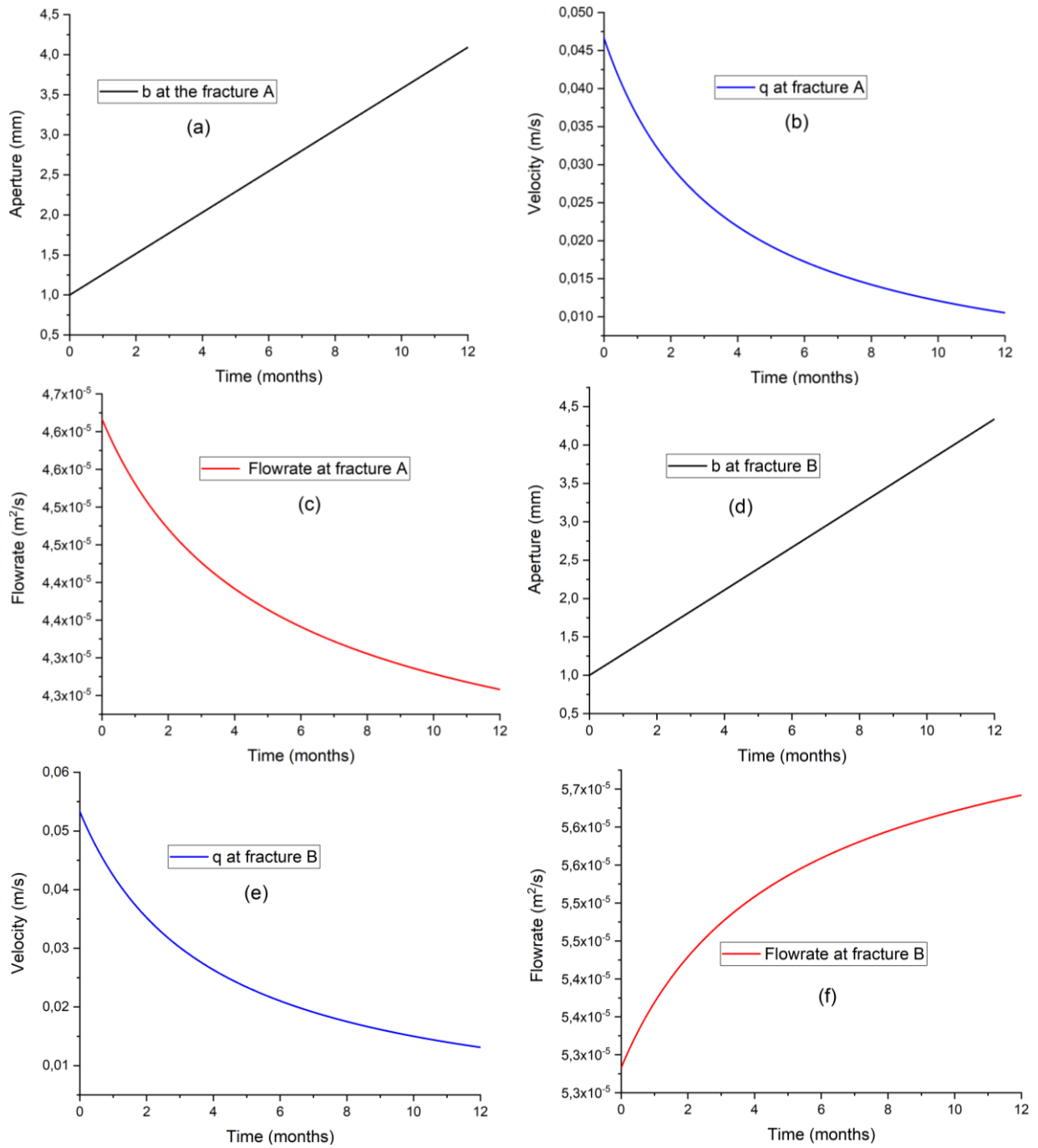


Figure 14. Aperture (a), velocity (b) and flowrate (c) evolutions in the fracture A and aperture (d), velocity (e) and flow rate (f) evolutions in the fracture B for the low dissolution case ($K_c = 10^{-5}$ m/s).

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548 Figure 15 shows the concentration, aperture and velocity distribution in the case of high
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3 549 dissolution ($K_C = 10^{-4}$ m/s). After one year, the concentration reduces only near the injection
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5 550 well (Figure 15a). Indeed, due to the high dissolution rate, during its travel, the fluid becomes
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7 551 quickly saturated by the dissolution species. As in the single fracture configuration (Figure
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9 552 7b), a very high enlargement is observed near the injection well, with an aperture reaching a
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11 553 value of 50 mm, whereas the aperture remains almost at its initial value near the pumping well
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13 554 (Figure 15b). The velocity distribution after 100s shows a high velocity for almost all
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15 555 connected fractures (Figure 15c), whereas after 1 year, it has strongly reduced (due to the high
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17 556 enlargement) near the injection point and remains high close to the pumping well (Figure
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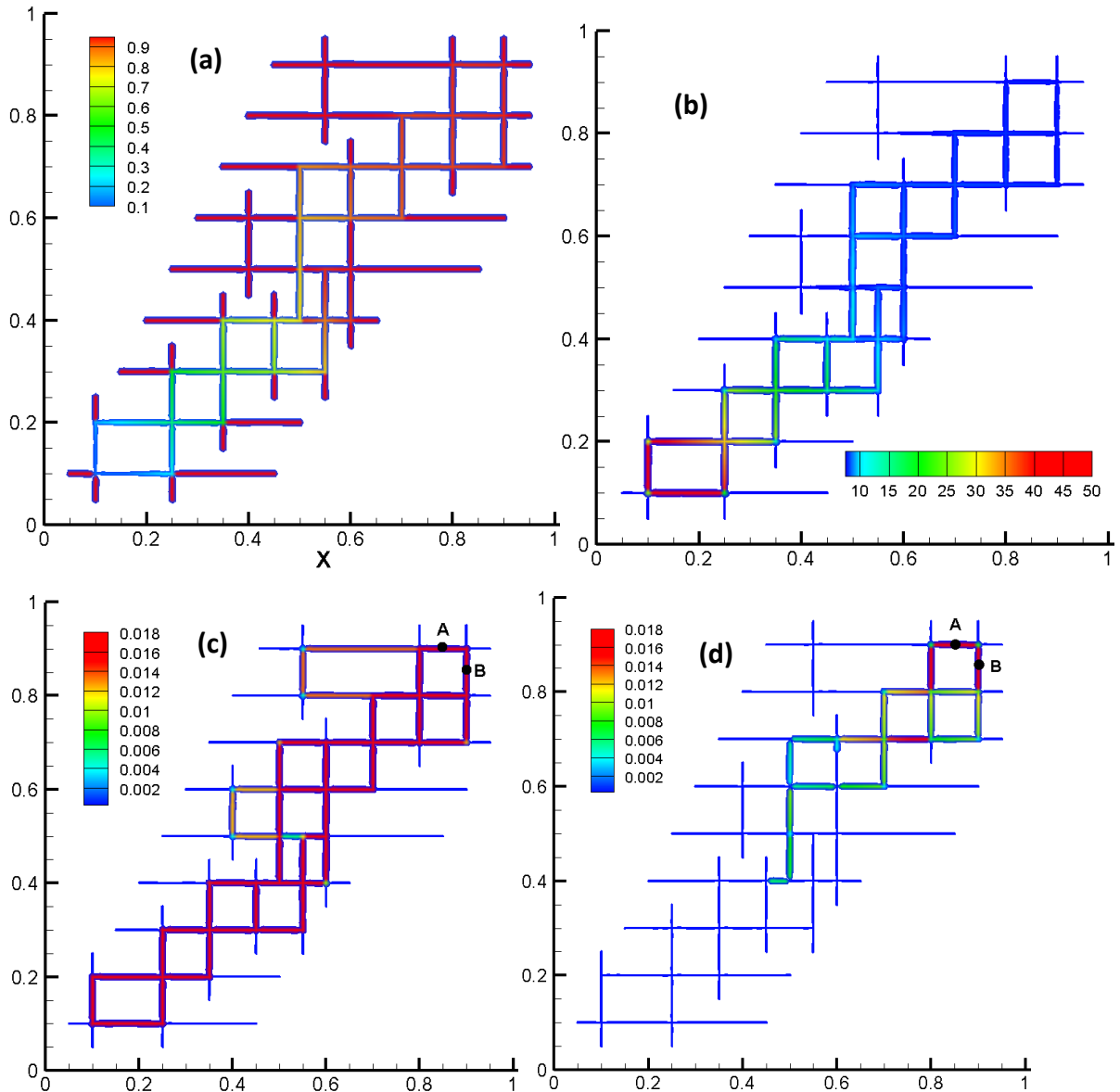
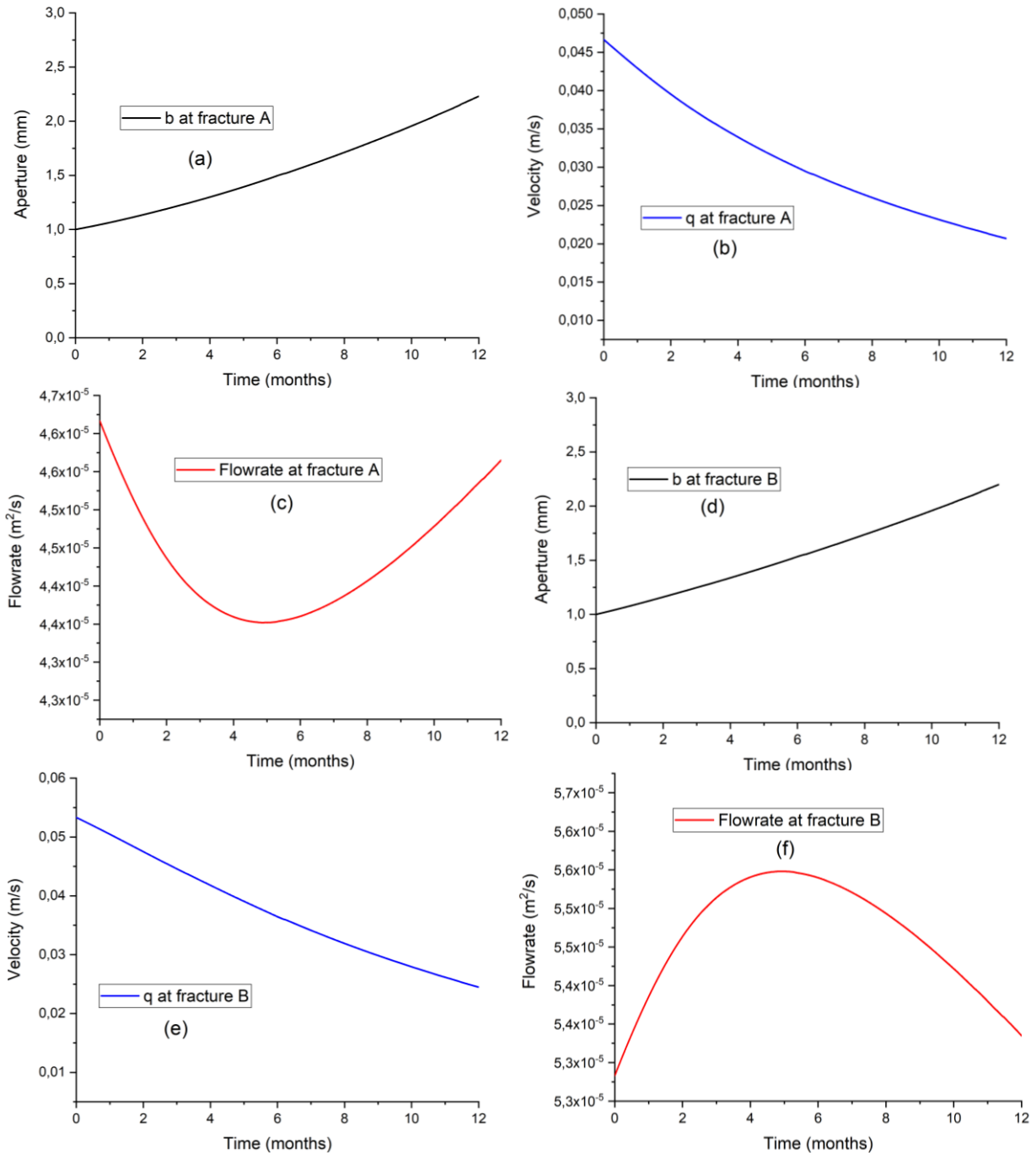


Figure 15: Results for the high dissolution rate ($K_c = 10^{-4}$ m/s): Distribution of the dimensionless concentration (-) (a) and aperture (mm) (b) after 1 year and magnitude of the velocity (m/s) after 100s (c) and 1 year (d).

Figure 16 depicts the evolution of aperture, velocity and flow rate in the two fractures A and B, sharing the pumping well for the high dissolution case. For both fractures A and B, the aperture (Figures 16a and 16d) shows an almost slight linear evolution with time which is less significant than for the low dissolution case (Figure 14). According to this aperture increase, the velocity in both fractures shows an almost linear decrease with time (Figures 16b and 16e). However, contrarily to the low dissolution case, the flow rate in the two fractures shows a non-monotonic evolution with time (Figures 16c and 16e). Indeed, the flow rate in the

578 fracture A shows a decrease until around 5 months and an increase after that, whereas the
 579 flow rate in the fracture B depicts the opposite behavior, thus the sum of the two flow rates
 580 remains constant. This parabolic evolution of the flow rate is probably related to the change of
 581 the permeability distribution during the simulation due to dissolution in the DFN.



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 583 **Figure 16:** Aperture (a), velocity (b) and flowrate (c) evolutions in the fracture A and aperture
 584 (d), velocity (e) and flowrate (f) evolutions in the fracture B for the high dissolution rate (
 585 $K_C = 10^{-4}$ m/s).

6. Conclusion

Modeling dissolution processes in a DFN is a computationally challenging problem because of the nonlinear coupling between flow, transport and reactive processes, introduced by the impact of dissolution on the fracture aperture. The challenge is accentuated by the fact that in the fractures, the transport is advection dominated. In such a case, standard FE formulations introduce unphysical oscillations that can lead to convergence issues. The upwind FV formulation give stable results but introduces a large amount of numerical diffusion. In this work, we developed a new model based on advanced numerical schemes to simulate dissolution processes in DFNs. The model is based on coupling the MFE method for flow with the DG scheme for transport. The DG method is known to be suitable for advection-dominated problems, as it avoids unphysical oscillations and reduces numerical diffusion. A new formulation of the DG was proposed for the Riemann solver at the intersection of fractures. The newly developed scheme calculates the upstream concentration based on the average of the nodal concentration of all fractures having an inflow at the intersection node, weighted by the water fluxes through these fractures. The dispersion term is calculated with the MFE method which allows to ensure the continuity of the dispersive flux at the intersection of several fractures having different apertures. The time integration of the obtained nonlinear coupled flow-transport-dissolution equations is improved by using an advanced solver of ODEs via the MOL.

The developed DG model is first verified for linear transport in a DFN by comparison against a standard FE solution obtained with COMSOL Multiphysics and an upwind FV model. Good agreement is obtained between the three models in the case of high diffusion. For advection-dominated transport configurations, often encountered because of the rapid flow in fractures, the DG-DFN model avoids the unphysical oscillations observed with FE results and allows for capturing sharp concentration fronts with a relatively coarse mesh. The upwind FV

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2 612 method fails to capture these fronts because of numerical diffusion, but its solution converges
3 towards the DG solution when a very fine mesh is employed.

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5 613 The new DG model was then verified in the case of coupled flow, transport and aperture
6 evolution due to dissolution for a single fracture. Although, we have used a high diffusion
7 value, the FE solution showed numerical oscillations at short times near the outlet. The
8 overall long-term results of FE, FV and DG models are in very good agreement in terms of
9 concentration distribution and aperture evolution.

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11 614 The new DG model was then used to investigate the effect of the dissolution rate on flow,
12 transport and aperture evolutions in the case of a single fracture and in the case of a DFN. In
13 the case of low dissolution, the fluid remains weakly contaminated (the fluid is never
14 saturated by the dissolved species during its travel). A linear increase of the fracture aperture
15 with time is observed. The velocity inside the fractures increases over time because of the
16 increase of permeability (proportional to the square of the aperture) in the case of flow
17 induced by fixed hydraulic heads at the upstream and downstream boundaries. In the case
18 where the total flow rate is fixed, the velocity inside fractures decreases when the aperture
19 increases due to dissolution, because of fluid mass conservation.

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21 618 In the case of a high dissolution rate, the fluid becomes quickly saturated by the dissolution
22 species during its travel. As a consequence, a very high enlargement is observed near the
23 injection, whereas the aperture remains almost at its initial value away from the injection. The
24 velocity is strongly reduced (due to the high enlargement) near the injection point and remains
25 high away from the injection.

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27 619 In the case of a DFN, although the total flow rate is fixed, the flowrate through each fracture
28 can vary with time since the distribution of the permeability of the fractures can vary with
29 time because of dissolution, which can induce a variation of the flowrate repartition between
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635 the fractures. For the case of a low dissolution rate, we observed a monotonic evolution of the
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2 636 flow rate inside fractures, whereas, in the case of a high dissolution rate, we observed a
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5 637 parabolic evolution of the flowrate through fractures, related to the change of the permeability
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7 638 distribution during the simulation due to the dissolution in the DFN.
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10 639 The model considered in this study is limited to dissolution processes, but the numerical
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12 640 scheme can be generalized to dissolution/precipitation processes. Precipitation processes may
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15 641 occur and could be important under non isothermal conditions. Thus, a future extension of
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17 642 this work could be including precipitation and thermal processes.
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