Reactive Transport Modeling of Dissolution/Precipitation in Fractured Porous Media

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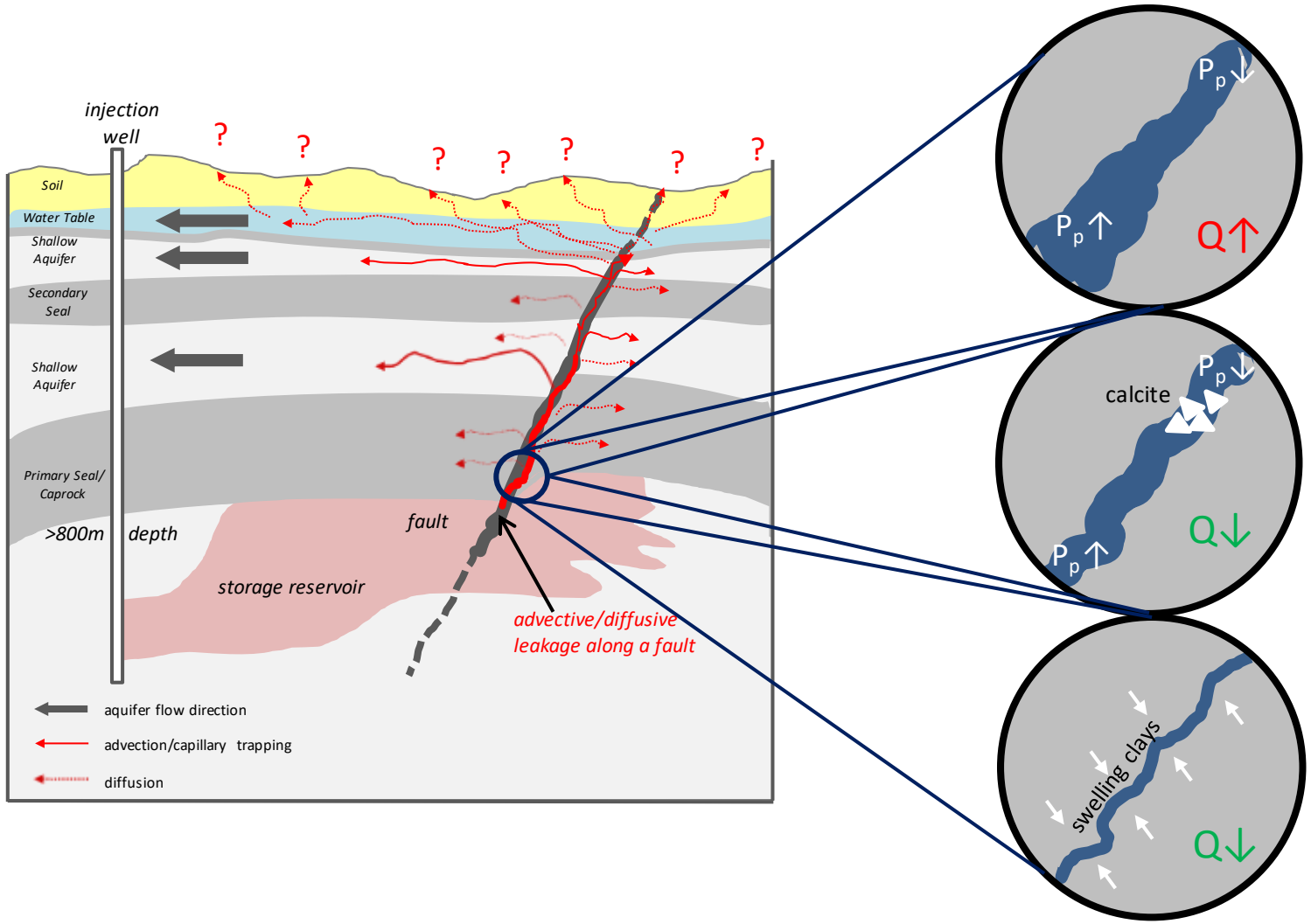
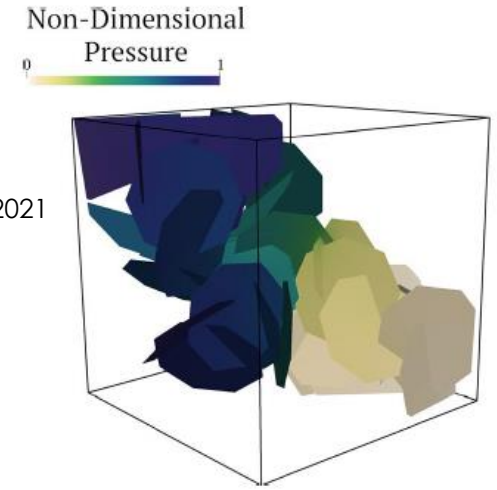
The final goal is to simulate the mineralization in fracture network of a fractured caprock

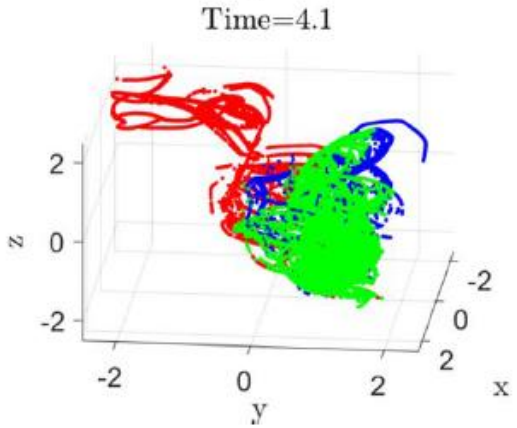


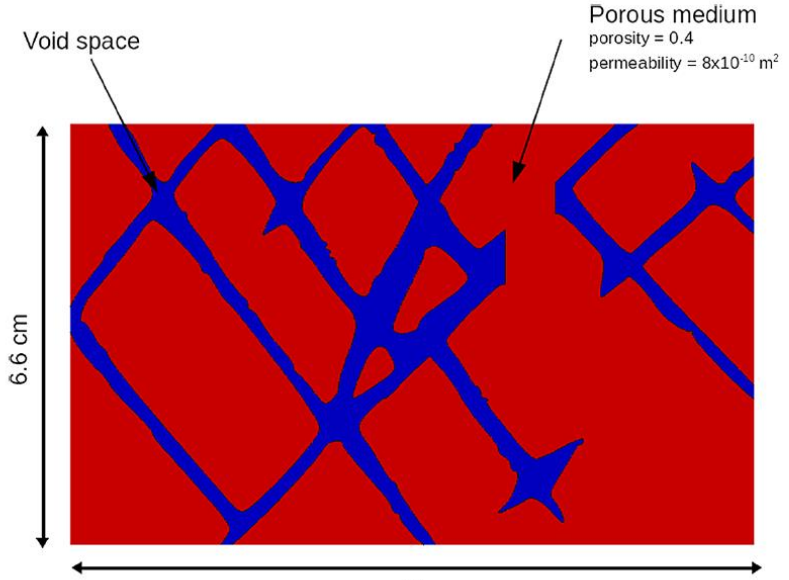
Examples of different approaches for reactive transport modelling in fractured media

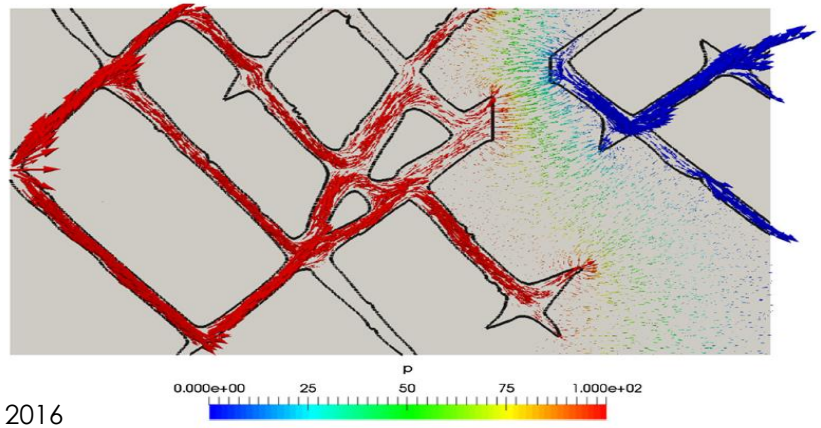
Reactive transport using DFN



Sherman et al., 2021





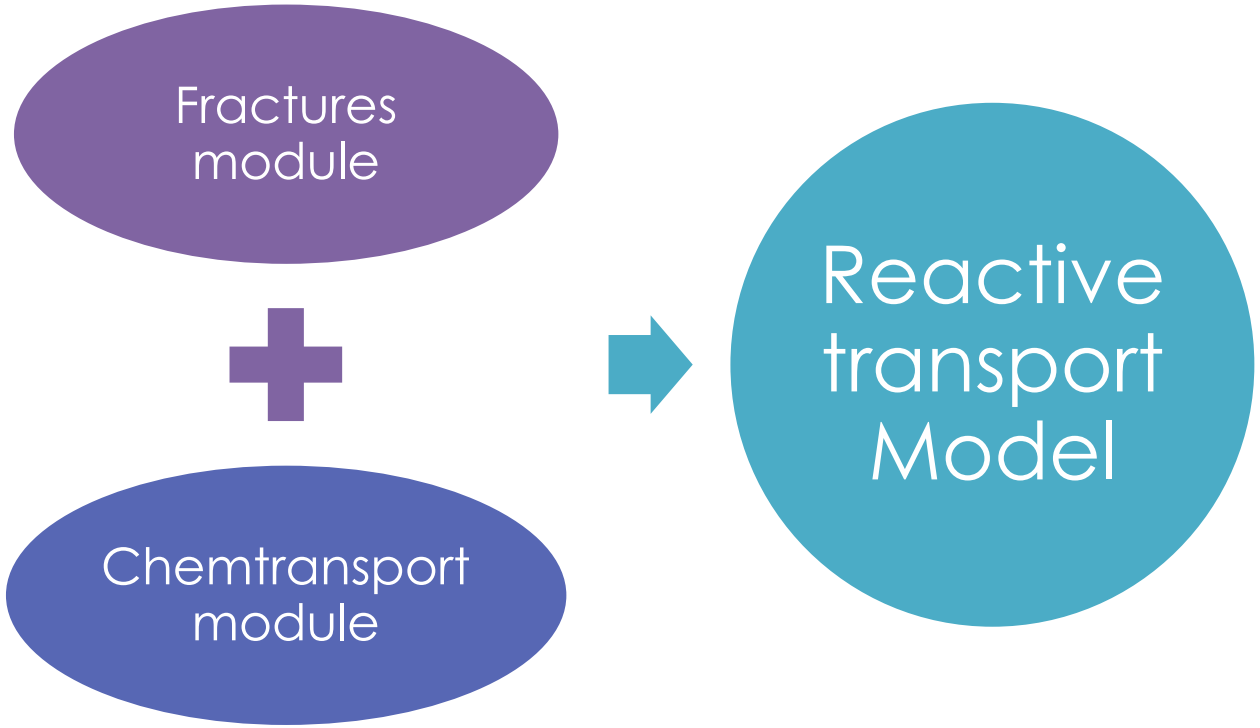


Soulaine et al., 2016

Reactive transport using micro-continuum methods

The reactive transport model

- The reactive transport model used in our work is developed within MRST (https://bitbucket.org/mrst).



Single-phase reactive transport equation

The single-phase reactive transport equation of chemical species (involved in equilibrium reactions) can be written in terms of element concentration:

$$\frac{\partial X}{\partial t} + \nabla.(uX) = 0$$

$$X = C^t x$$

$$A\hat{x} = \hat{K} \text{ (the law of mass action)}$$

Where X is element concentration, u is fluid phase velocity, x is species concentration, A is the matrix containing the stochiometric coefficients in set of $k = 1, \dots, n_e$ reactions which involve $i = 1, \dots, n_c$ species: $A_{k,1}C_1 + A_{k,2}C_2 + \cdots + A_{k,n_C}C_{n_c} \leftrightarrow 0$. $\hat{K}$ is the logarithm of K, which is the equilibrium constant of the reaction, and C is a constitutive matrix where $AC = 0$ and it contains the amount of atoms in each species.

Discrete Fracture and matrix (DFM) model

- The fluid phase velocity in the reactive transport equation is solved for using the DFM approach where it represents the discontinuities explicitly.
- The flux of phase α, across the face is

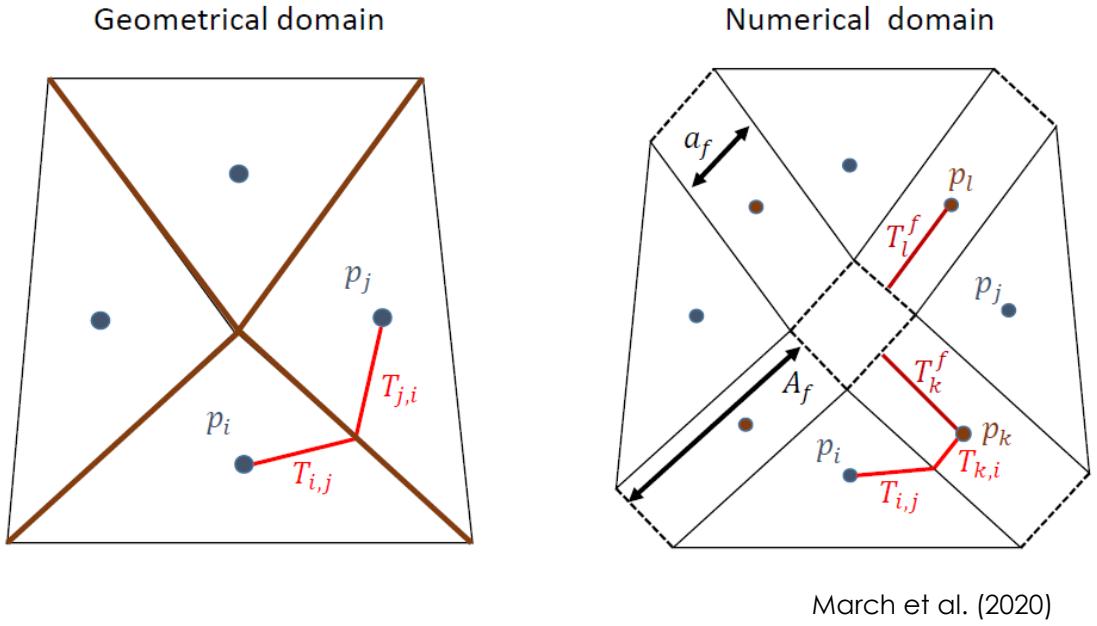
$$v_{\alpha ij} = \left(T_{i,j}^{-1} + T_{j,i}^{-1}\right)^{-1} \lambda_\alpha \left(p_{\alpha i} - p_{\alpha j}\right) = T_{ij} \lambda_\alpha \left(p_{\alpha i} - p_{\alpha j}\right)$$

- The transmissibilities between fracture and matrix are calculated as:

$$T_{ik} = \left(T_{i,j}^{-1} + T_{k,i}^{-1}\right)^{-1} \; where \; T_{k,i} = \frac{2A_k k_k}{a_k}$$

- The transmissibility between two fracture edges is computed as:

$$T_{kl}^f = \frac{T_k^f T_l^f}{\sum_{i=1}^{n} T_i^f} \; , T_i^f = \frac{2a_i k_i}{A_i}$$



March et al. (2020)

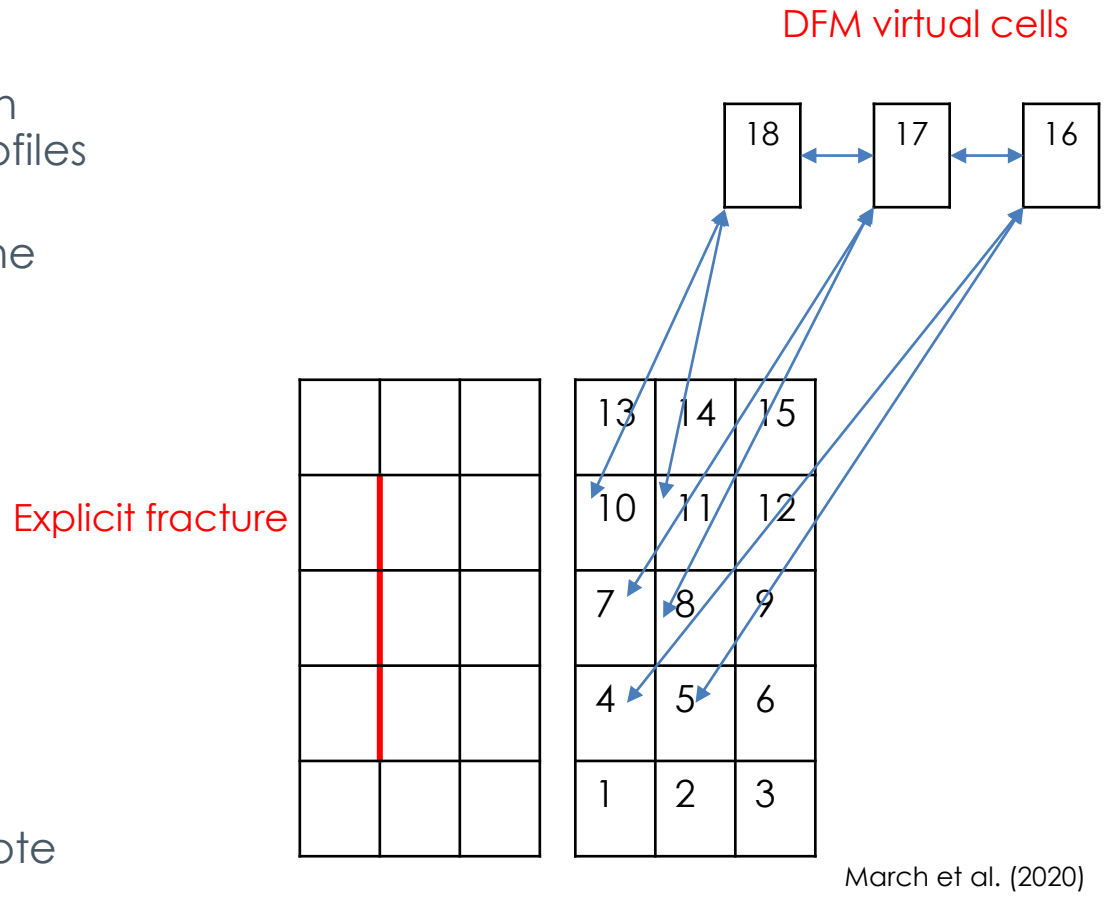
Discrete Fracture and matrix (DFM) model

- The flux of the phase α is used in the mass conservation equation in order to solve for pressure and velocity profiles inside the domain.
- The finite-volume method involves the integration of the mass-conservation equations in a grid volume:

$$\frac{\partial}{\partial t}\int_{\Omega_e}\varphi\rho_\alpha S_\alpha dV + \oint_{\partial\Omega_e}\rho_\alpha \boldsymbol{q_\alpha}.\boldsymbol{dS} = \int_{\Omega_e}\rho_\alpha I_\alpha dV$$

Ω_e: the domain of a cell element.

- The second term of equation accounts for the mass flowrate of phase α across the faces of element Ω_e. Note that these faces can be faces in the geometrical grid (between geometrical cells), virtual faces that connect geometrical and virtual cells, and virtual faces that connect virtual cells



March et al. (2020)

Chemistry and transport coupling

- Equations to be solved in every timestep:
  - ✓ $\frac{\partial X}{\partial t} + \nabla.(uX) = 0$
  - ✓ $A\hat{x} = \hat{K}$
  - ✓ $X = C^t x$
  - ✓ $\frac{\partial(\rho\varphi)}{\partial t} + \nabla.(\rho \boldsymbol{v}) = \rho q, \boldsymbol{v} = -k\lambda\nabla p$

- Timestep iteration in the reactive transport simulation:

$$R_p(x, X, p) = 0$$
$$R_c(x, X, p) = 0$$
$$R_t(x, X, p) = 0$$

Fully coupled Newton step:

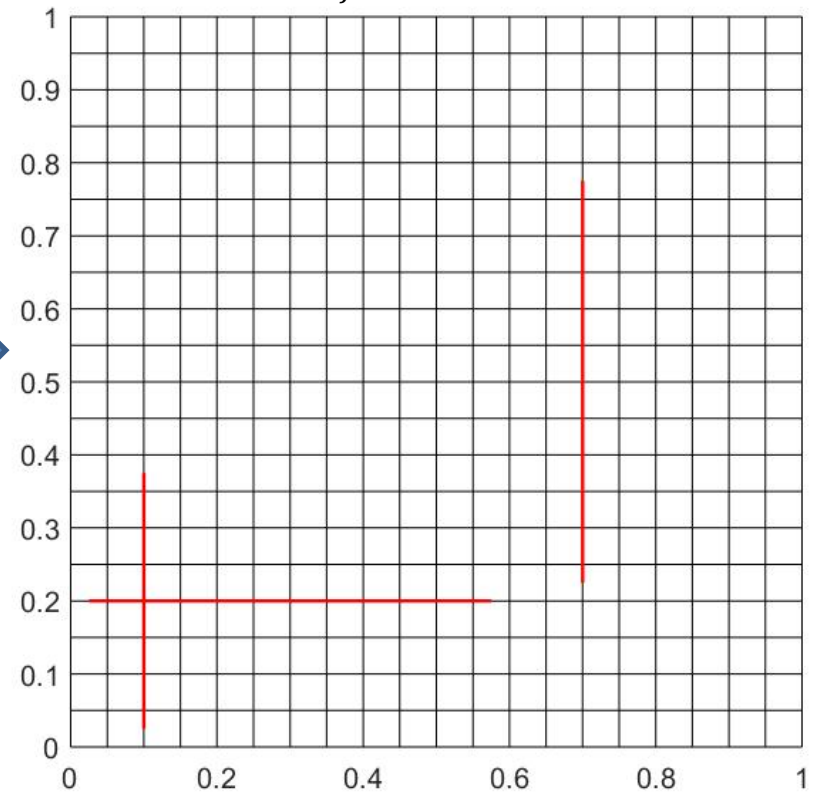
$$\left(\hat{x}^{n+1}, \hat{X}^{n+1}, p^{n+1}\right) = \left(\hat{x}^n, \hat{X}^n, p^n\right) - \partial R^{-1} R$$

Example I: Dissolution and precipitation in a 2D fractured domain

$$\varphi_m = 0.2$$
$$k_m = 0.01\ mD$$
$$a_f = 1\ mm$$

Injected solution

Influx rate: $0.01\ m^3/day$



Constant pressure boundary

species	Initial solution $(mol.L^{-1})$	Injected solution $(mol.L^{-1})$
H^+	10^{-12}	1×10^{-2}
OH^-	10^{-2}	1×10^{-12}
H_2O	1	1
Ca^{2+}	5.15×10^{-5}	10^{-5}
CO_3^{2-}	6.43×10^{-5}	2.08×10^{-18}
HCO_3^-	1.37×10^{-6}	4.45×10^{-10}
H_2CO_3	3.08×10^{-12}	10^{-6}
$CaCO_3(s)$	1×10^{-2}	–

$$H_2O = OH^- + H^+$$
$$CaCO_3(s) = Ca^{2+} + CO_3^{2-}$$
$$CO_3^{2-} + H^+ = HCO_3^-$$
$$HCO_3^- + H^+ = H_2CO_3$$

Example I

- Species:

$H_2O, OH^-, H^+, CaCO_3(s), Ca^{2+}, CO_3^{2-}, HCO_3^-, H_2CO_3$

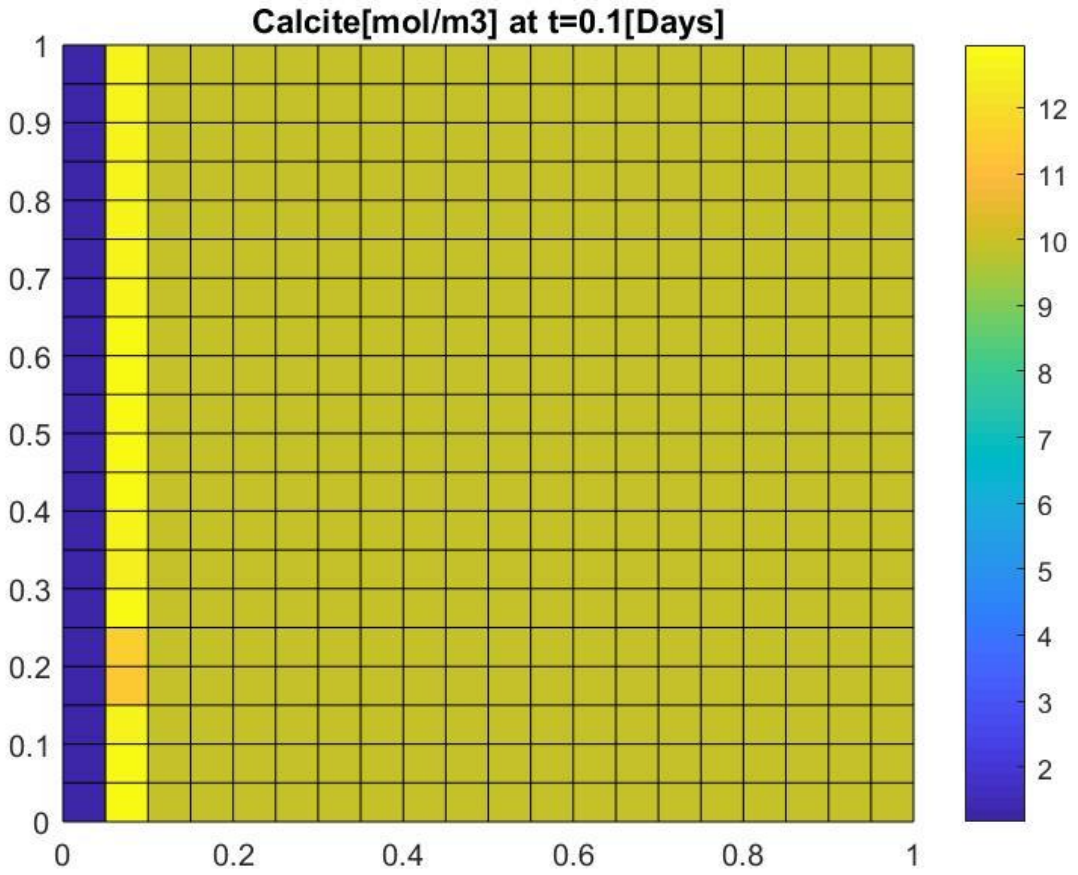
- Elements:

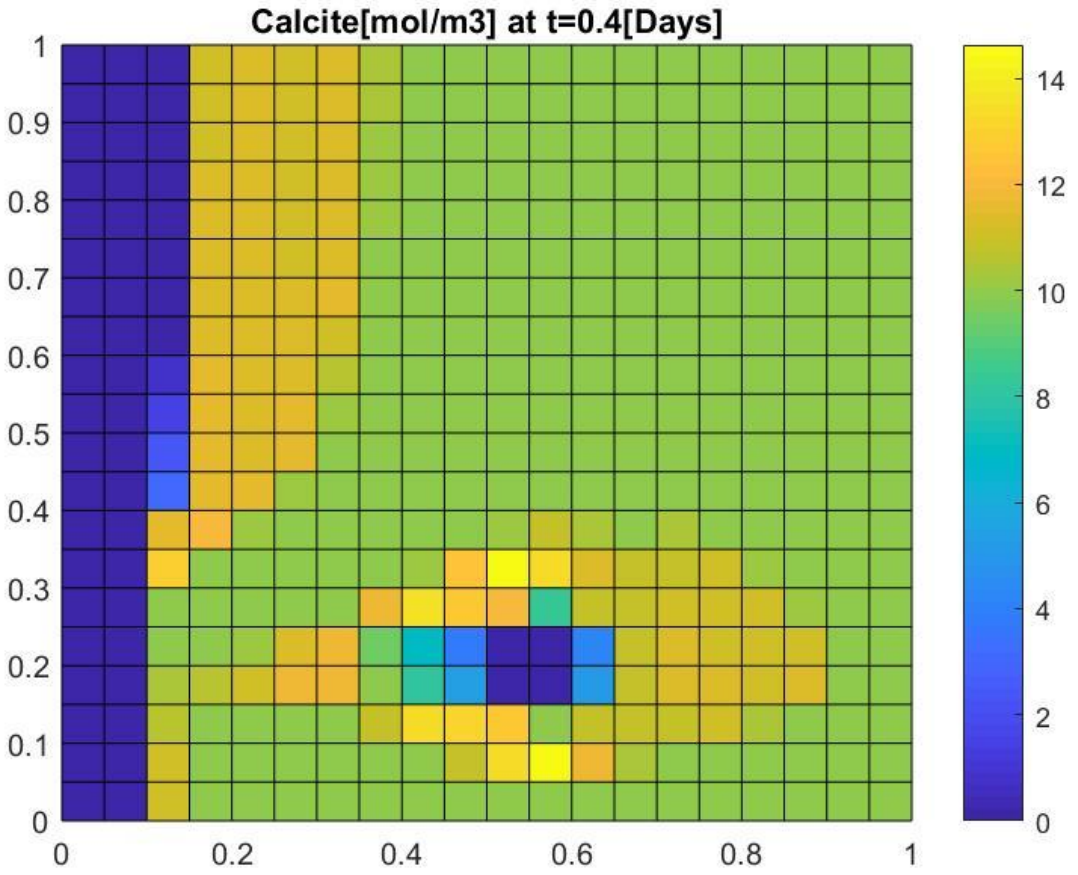
O, H, Ca, CO_3

- Equations to be solved:
  - ✓ Four transport equations for transport of O, H, Ca, CO_3 $(\frac{\partial X}{\partial t} + \nabla.(uX) = 0)$
  - ✓ Four mass action equations for four reactions $(A\hat{x} = \hat{K})$
  - ✓ Four equations relating elements and species concentrations $(X = C^t x)$
  - ✓ 1 flow equation $(\frac{\partial(\rho\varphi)}{\partial t} + \nabla.(\rho \boldsymbol{v}) = \rho q, \boldsymbol{v} = -k\lambda\nabla p)$

Example I

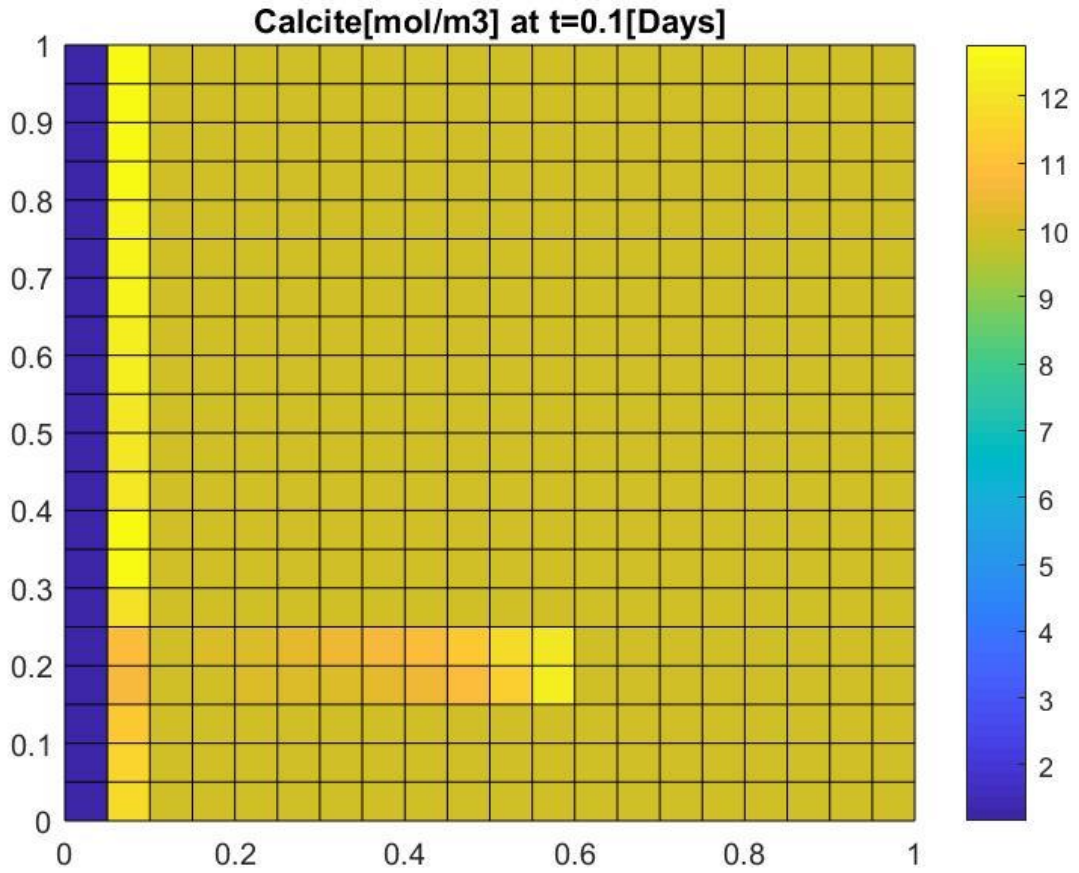
Permeability is constant

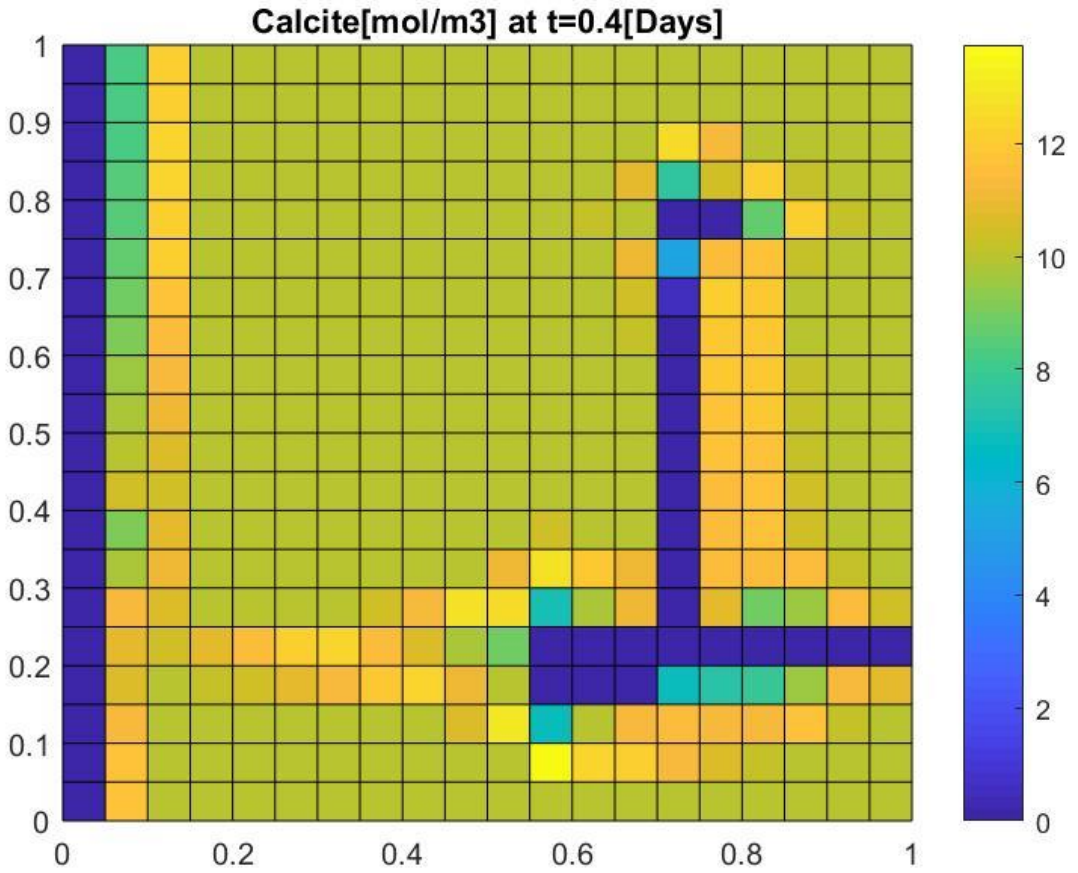




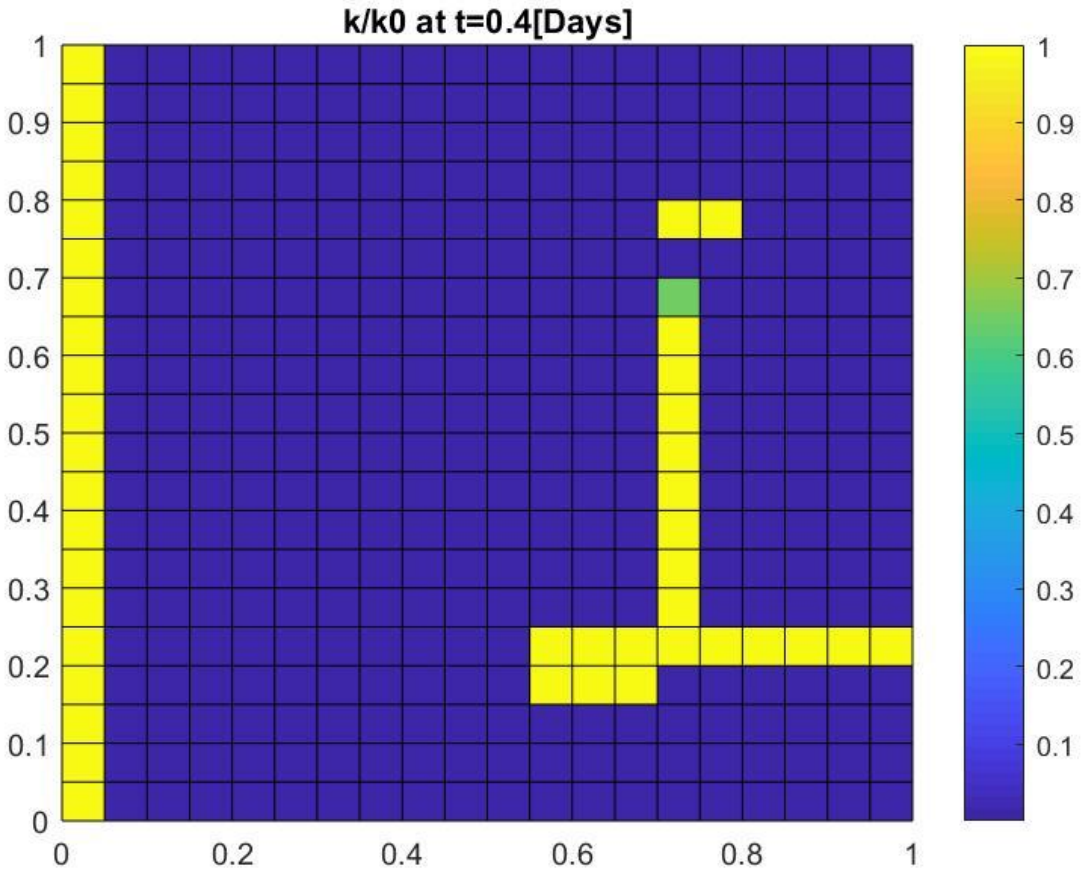
Example I

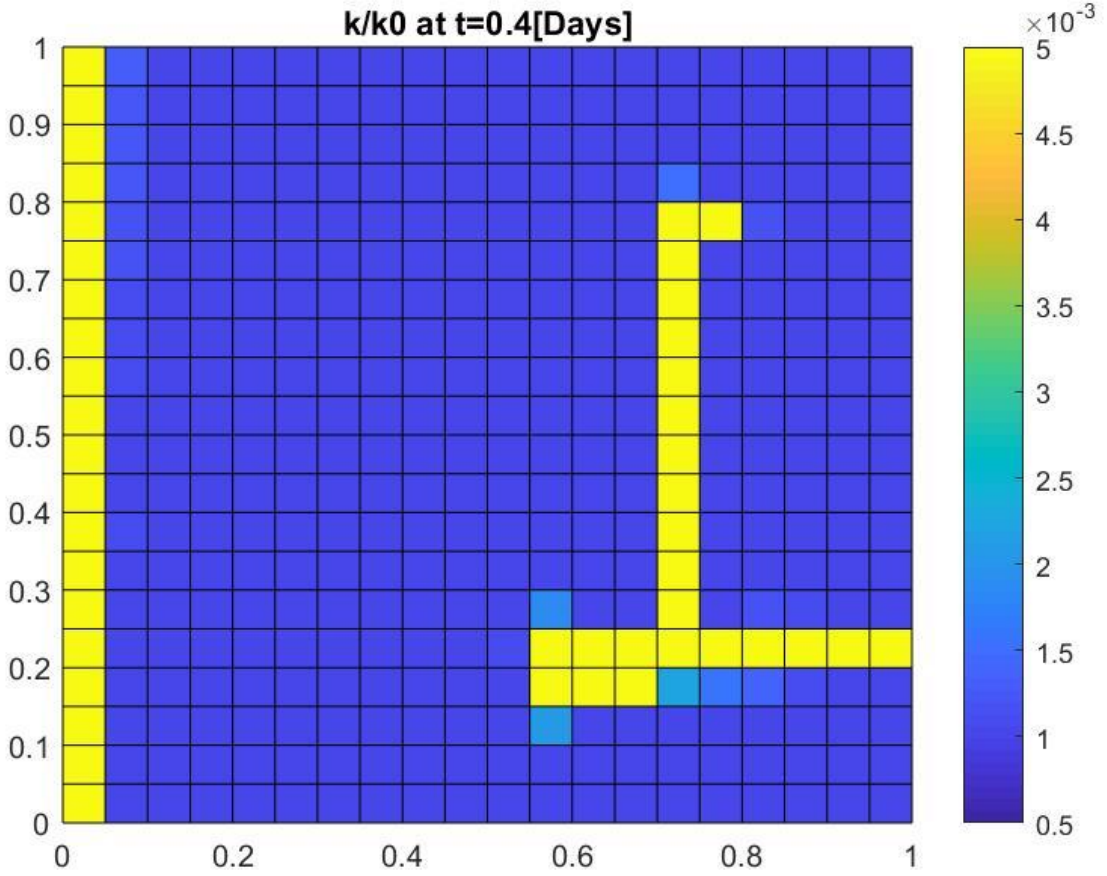
$$k = k_0 e^{-\alpha[CaCO_3(s)]}$$



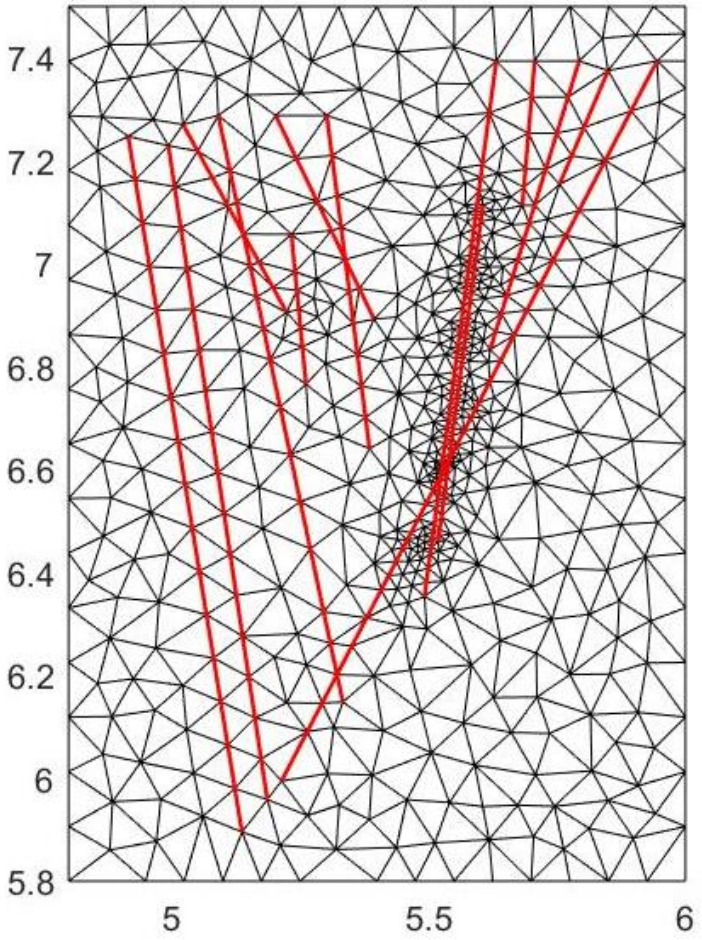


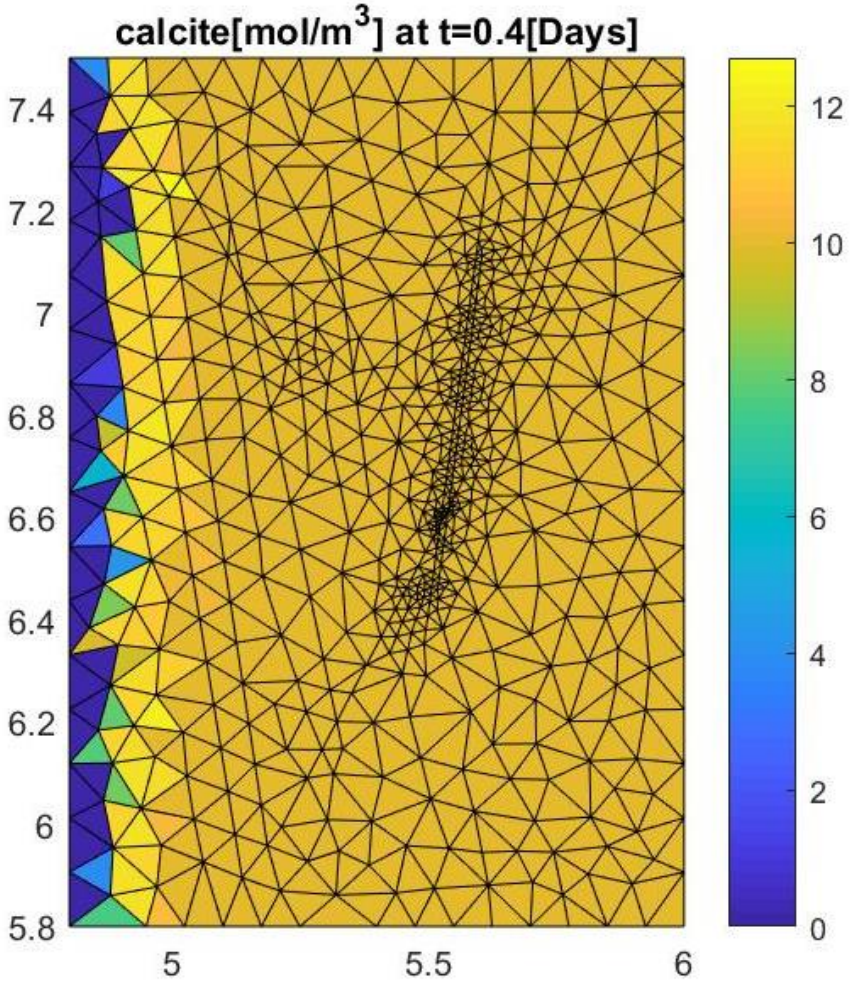
Example I


k/k0 at t=0.4[Days]
1
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
0
0.2
0.4
0.6
0.8
1


k/k0 at t=0.4[Days]
×10^-3
5
4.5
4
3.5
3
2.5
2
1.5
1
0.5
1
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
0
0.2
0.4
0.6
0.8
1

Example II


7.4
7.2
7
6.8
6.6
6.4
6.2
6
5.8
5
5.5
6


calcite[mol/m3] at t=0.4[Days]
7.4
7.2
7
6.8
6.6
6.4
6.2
6
5.8
5
5.5
6
12
10
8
6
4
2
0

Summary

- A single-phase reactive transport solver for equilibrium reactions has been developed for flow and transport in fractured media (combining MRST modules, *chemtransport* and *fractures*)
- The AD framework of MRST allows for drastic permeability changes (factor of 1000).
- Dissolution potentially connect fractures, where DFM approach can capture this effect.