Thermal-compositional-reactive simulation with DARTS

Denis Voskov

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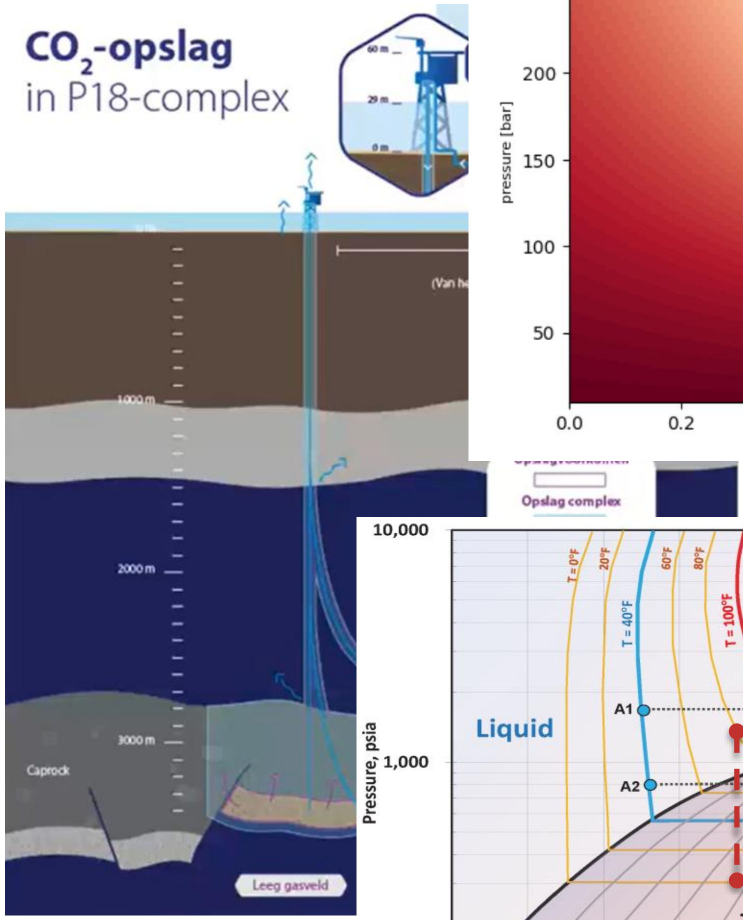
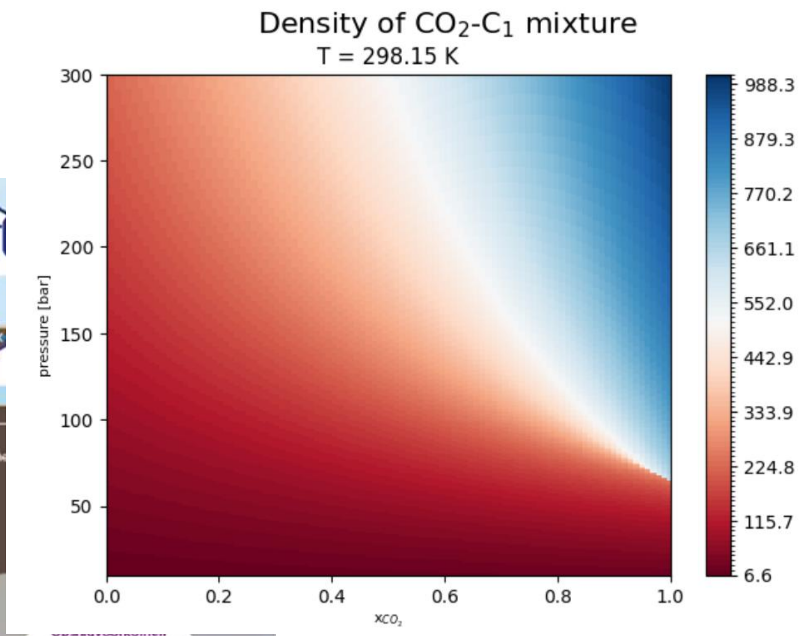
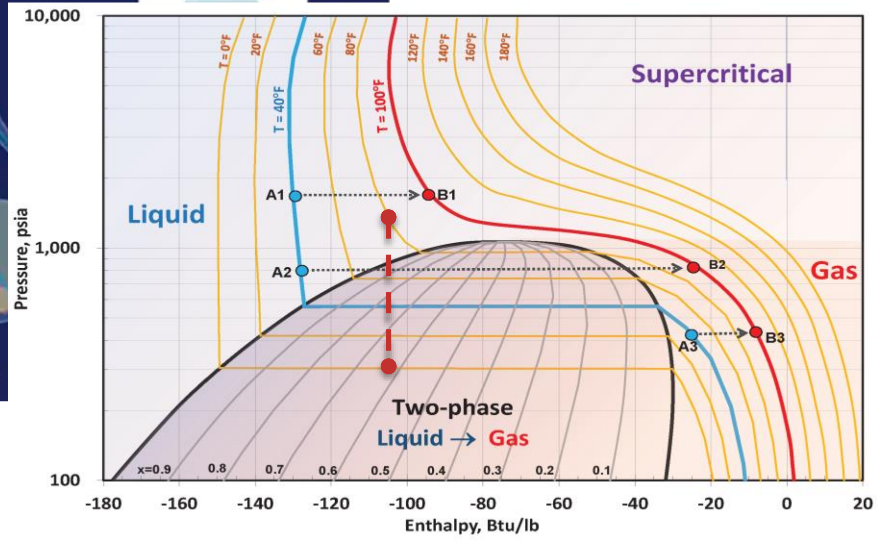
CO_2 injection into depleted fields

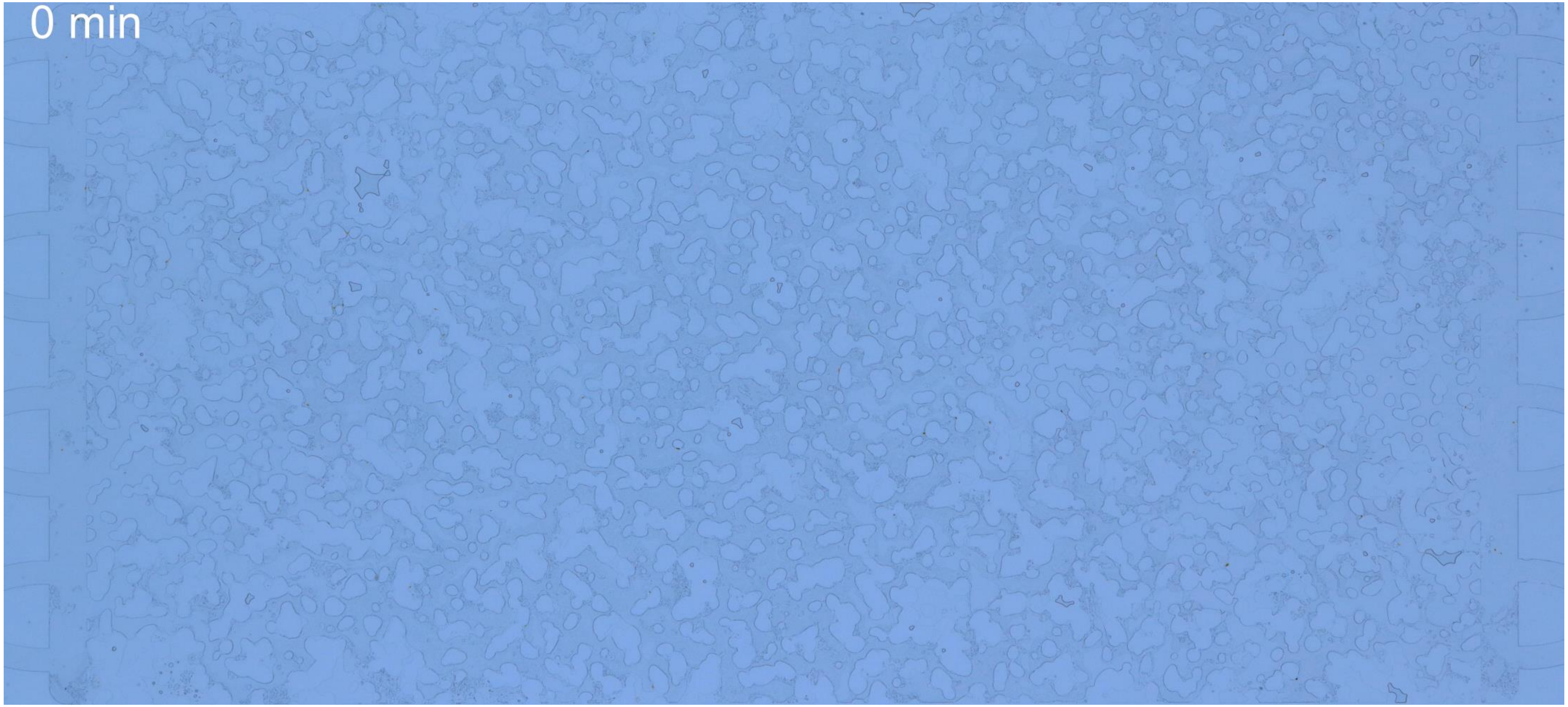
- Injection into depletion reservoir
  - Pressurizing reservoir
  - Joule-Thomson cooling
    - Hydrate formation
  - Salt precipitation
    - Capillary backflow
- Injectivity interruption
  - Pressure depletion
  - Two-phase cooling
    - Hydrate formation





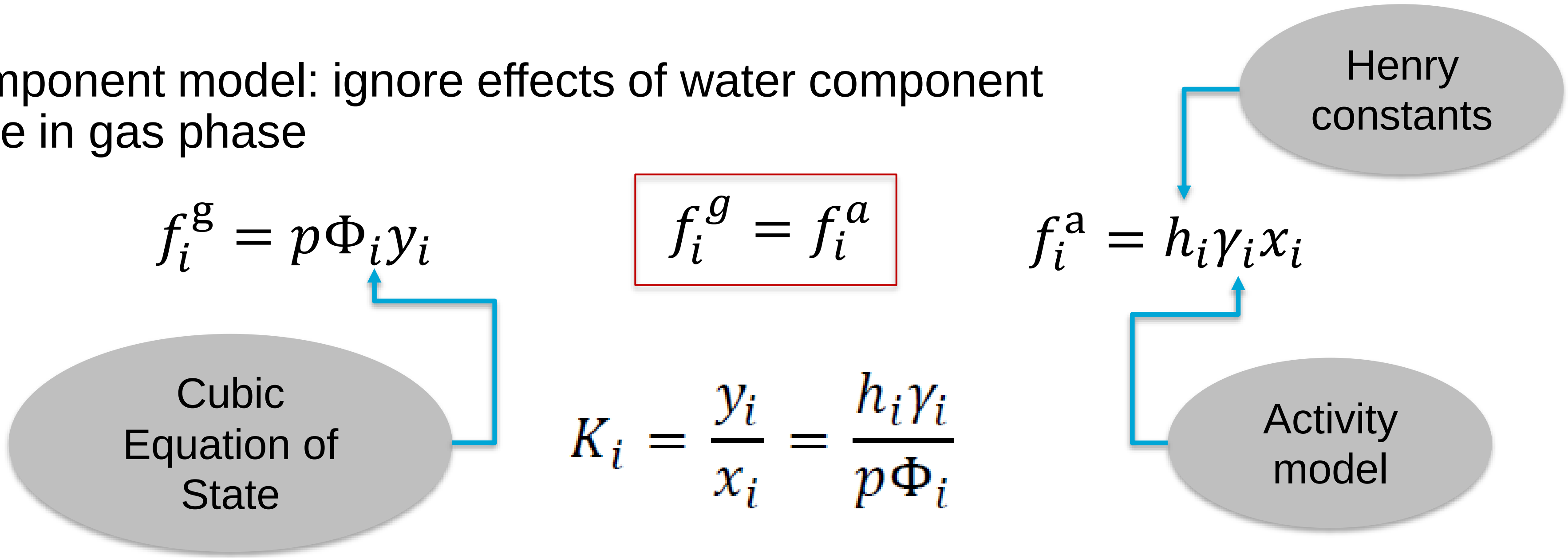


Challenge with CO2 expansion



Thermodynamics of CO_2-gas-brine

- Gas component model: ignore effects of water component presence in gas phase



$$f_i^{\mathrm{g}} = p\Phi_i y_i$$

$$f_i^{g} = f_i^{a}$$

$$f_i^{\mathrm{a}} = h_i \gamma_i x_i$$

$$K_i = \frac{y_i}{x_i} = \frac{h_i \gamma_i}{p\Phi_i}$$

- Derive enthalpy directly from EoS for gas, liquid and aqueous phases

$$h^g = -RT^2 \sum n_i \left(\frac{\partial \ln \phi_i}{\partial T}\right), \quad h^a = -RT^2 \sum n_i \left(\frac{\partial \ln \gamma_i}{\partial T}\right)$$

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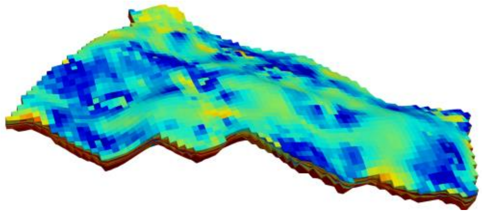
Wapperom et al., SPE RSC, 2023

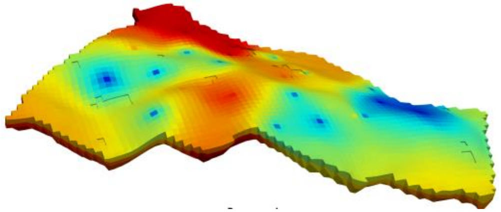
PDE for Energy Transition applications

$$\boldsymbol{g}(\boldsymbol{\omega}) = \boldsymbol{a}_t(\boldsymbol{\omega}, \boldsymbol{\xi}) + \nabla \cdot \boldsymbol{b}(\boldsymbol{\omega}, \boldsymbol{\xi}) + \Delta \boldsymbol{c}(\boldsymbol{\omega}, \boldsymbol{\xi}) + \boldsymbol{d}(\boldsymbol{\omega}, \boldsymbol{\xi}) = 0$$

$\boldsymbol{\xi} = \{\boldsymbol{G}, \boldsymbol{\phi}_0, \boldsymbol{K}_0\}$



$\boldsymbol{\omega} = \{\boldsymbol{p}, \boldsymbol{H}, \boldsymbol{z}\}$



Compressibility, phase change and convection

$$\boldsymbol{g}(\boldsymbol{\omega}) = \frac{\phi_0 V}{\Delta t}[\boldsymbol{\alpha}(\boldsymbol{\omega}) - \boldsymbol{\alpha}(\boldsymbol{\omega}_n)] + \sum_l v_t^l \boldsymbol{\beta}(\boldsymbol{\omega}) = \mathbf{0}$$

$$\alpha_c(\boldsymbol{\omega}) = c(\boldsymbol{\omega})\, z_c \sum_{j=1}^{n_p} \rho_j s_j,$$

$$\beta_c(\boldsymbol{\omega}) = \frac{1}{\Lambda} \sum_{j=1}^{n_p} x_{cj}^l \rho_j^l \frac{k_{rj}^l}{\mu_j^l}$$

$$\boldsymbol{g}(\boldsymbol{\omega}) = \frac{\phi_0 V}{\Delta t}[\boldsymbol{\alpha}(\boldsymbol{\omega}) - \boldsymbol{\alpha}(\boldsymbol{\omega}_n)] + \sum_l v_t^l \boldsymbol{\beta}(\boldsymbol{\omega}) + \sum_l \mathbf{D}^l(\chi^l - \chi)\boldsymbol{\gamma}(\boldsymbol{\omega}) + \mathrm{V}\boldsymbol{\delta}(\boldsymbol{\omega}) = \mathbf{0}$$

+ diffusion and reactions

$$\gamma_c(\boldsymbol{\omega}) = c(p) \sum_{j=1}^{n_p} x_{cj} \rho_j s_j d_{cj}, \qquad \delta_c(\boldsymbol{\omega}) = \sum_{\mathrm{k}=1}^{n_k} \nu_{ck} r_k$$

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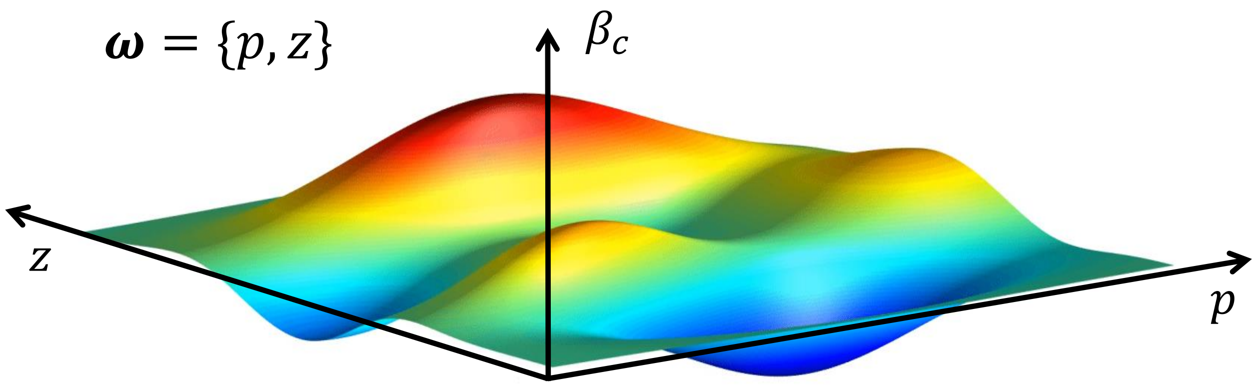
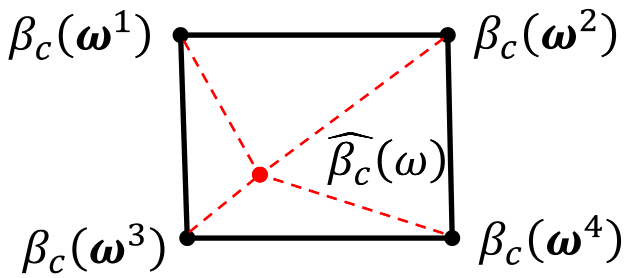
Operator-Based Linearization

$$\beta_c(\boldsymbol{\omega}) = \frac{1}{\Lambda}\sum_{j=1}^{n_p} x_{cj}^{l}\rho_j^{l}\frac{k_{rj}^{l}}{\mu_j^{l}}$$





$$\frac{\partial \boldsymbol{g}}{\partial \boldsymbol{\omega}} = \frac{\partial \boldsymbol{\alpha}}{\partial \boldsymbol{\omega}}\bar{\boldsymbol{a}}(\boldsymbol{\omega},\boldsymbol{\xi}) + \frac{\partial \boldsymbol{\beta}}{\partial \boldsymbol{\omega}}\bar{\boldsymbol{b}}(\boldsymbol{\omega},\boldsymbol{\xi}) + \frac{\partial \boldsymbol{\gamma}}{\partial \boldsymbol{\omega}}\bar{\boldsymbol{c}}(\boldsymbol{\omega},\boldsymbol{\xi}) + \frac{\partial \boldsymbol{\delta}}{\partial \boldsymbol{\omega}}\bar{\boldsymbol{d}}(\boldsymbol{\omega},\boldsymbol{\xi}) + \bar{\boldsymbol{f}}(\boldsymbol{\omega},\boldsymbol{\xi})$$

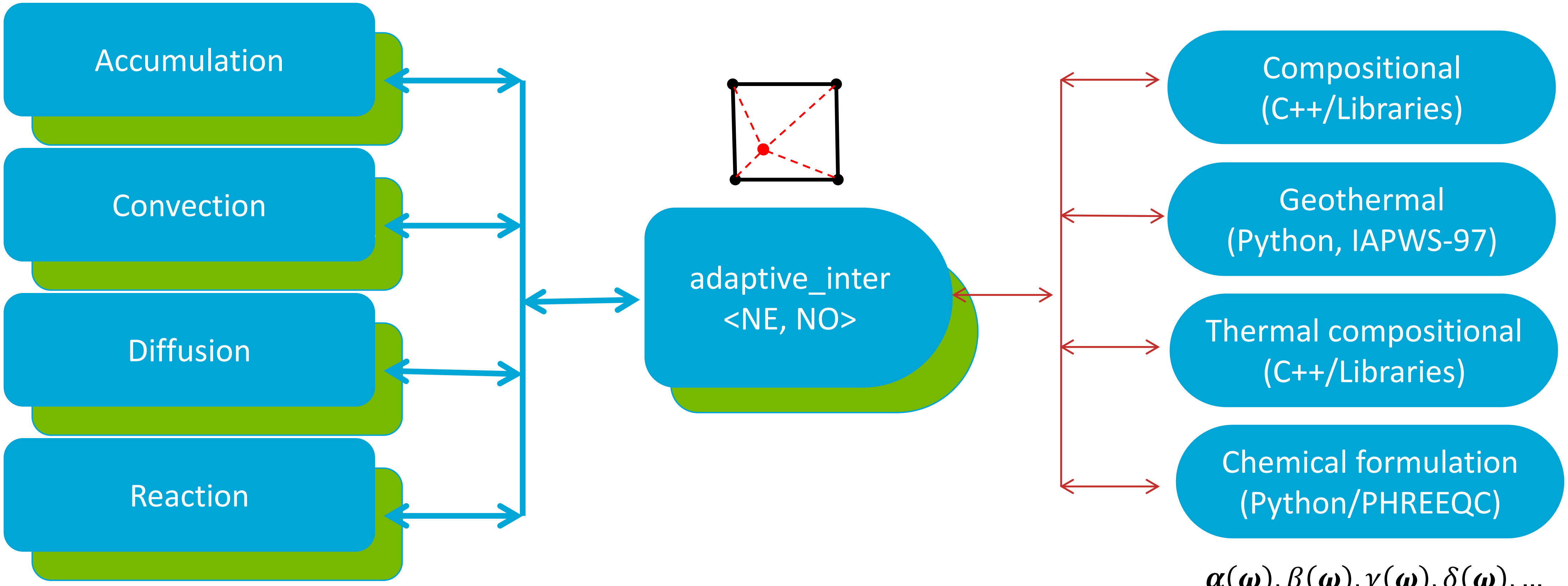
$$\left|\widehat{\beta_c} - \beta_c\right| \le cA^2 \sup_{\omega}\left|\nabla^2 \beta_c\right|$$

Voskov, JCP 2017

Open Delft Advanced Research Terra Simulator

DARTS-engine: C++ & CUDA

DARTS-physics: hybrid



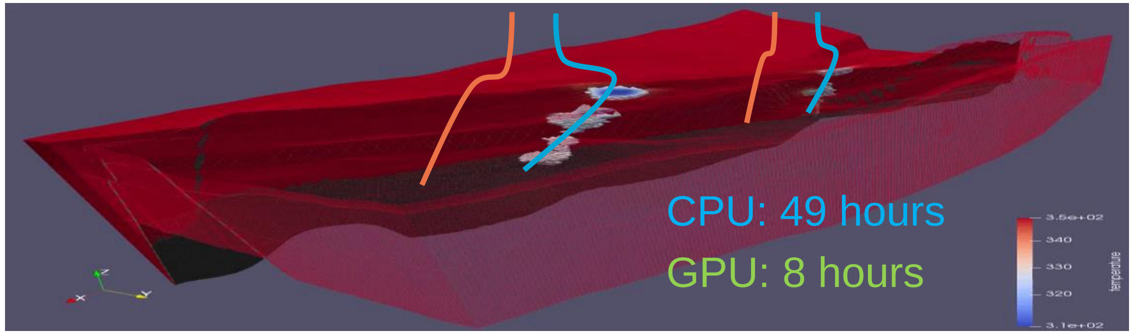
$$\boldsymbol{\alpha}(\boldsymbol{\omega}), \boldsymbol{\beta}(\boldsymbol{\omega}), \boldsymbol{\gamma}(\boldsymbol{\omega}), \boldsymbol{\delta}(\boldsymbol{\omega}), \ldots$$

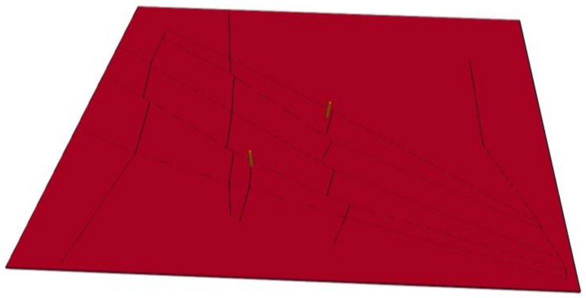
$$\boldsymbol{a}_t(\boldsymbol{\omega}, \boldsymbol{\xi}) + \nabla \cdot \boldsymbol{b}(\boldsymbol{\omega}, \boldsymbol{\xi}) + \Delta \boldsymbol{c}(\boldsymbol{\omega}, \boldsymbol{\xi}) + \boldsymbol{d}(\boldsymbol{\omega}, \boldsymbol{\xi}) = \boldsymbol{0}$$

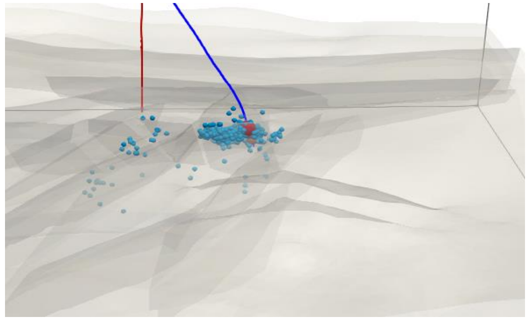
Voskov et al., JOSS, 2024

Delft Advanced Research Terra Simulator

- Operator-Based Linearization
  - Parametrization of thermodynamics
  - Adaptivity in parametrization
- Flexibility and performance
  - Flexible nonlinear physics
  - Implementation at GPU
  - Adjoint capabilities
- Complex thermodynamics
  - Thermal-compositional formulation
  - Fully coupled chemistry
  - Fully coupled geomechanics







Lyu et al., IJGGC, 2021; Wang et al., RENE, 2021

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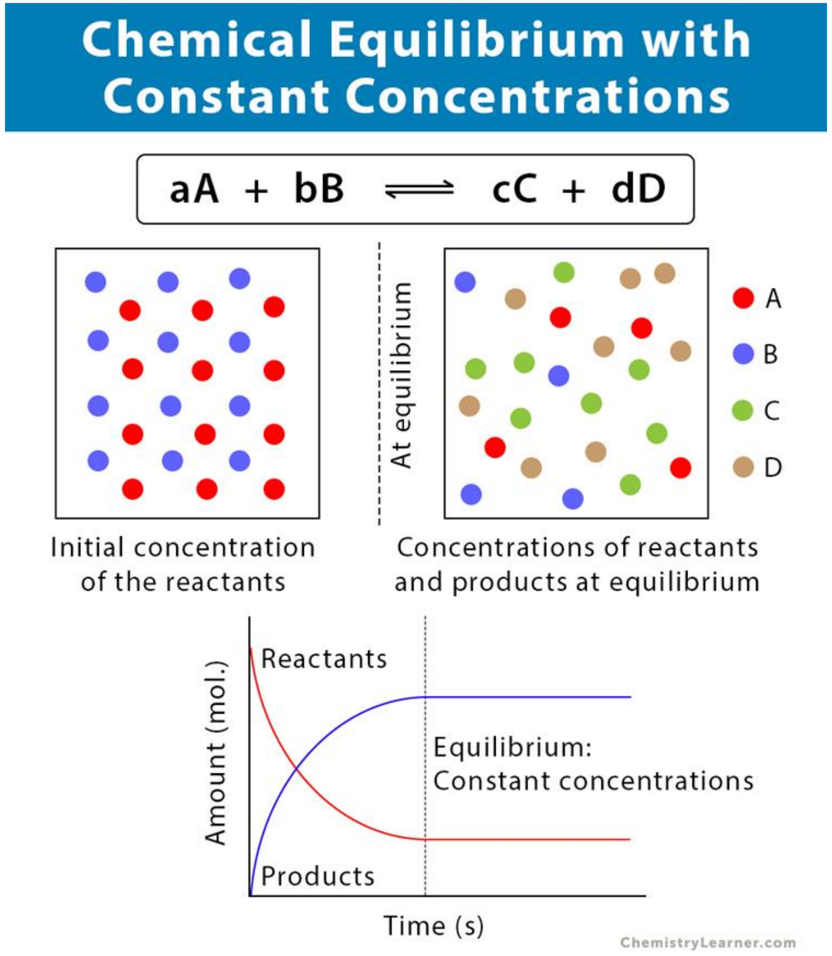
Equilibrium chemical reactions



Component mass balance:

$$\frac{\partial}{\partial t}(\phi \rho_j z_c) + \nabla \cdot (\rho_j z_c \boldsymbol{u}) = \sum_{r=1}^{n_r} v_{c,r} r_r$$

$$\boldsymbol{a_c} \quad + \quad \boldsymbol{l_c} \quad = \quad \boldsymbol{Vr}$$

$$\boldsymbol{a_c} + \boldsymbol{l_c} = \boldsymbol{Vr} \times \boldsymbol{E} \quad \Rightarrow \quad \boldsymbol{a_e} + \boldsymbol{l_e} = \boldsymbol{0}$$

Chemical reactions:

$K_{sp} - Q_{sp} = 0$ equilibrium

$a_c^k + l_c^k = \boldsymbol{vr}^k$ kinetic

Equilibrium reactions in brine-CO_2 system

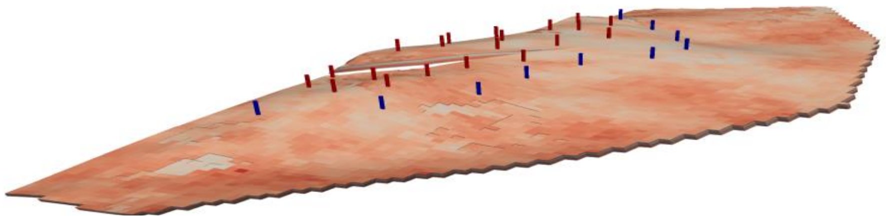
$$\boldsymbol{E} \times \frac{\partial}{\partial t}(\phi \rho_t z_c) + \mathrm{div}(l_c) = \sum_{q=1}^{n_q} v_{cq} r_q$$

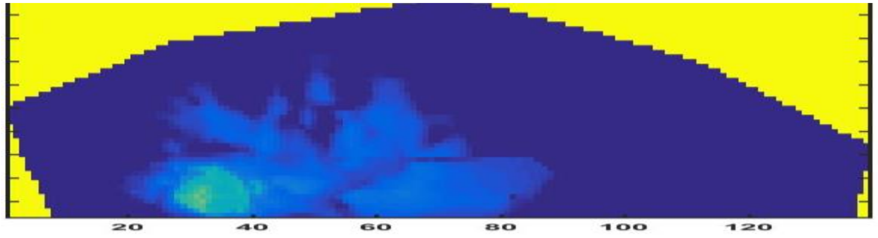
$$f_i^g = f_i^l$$

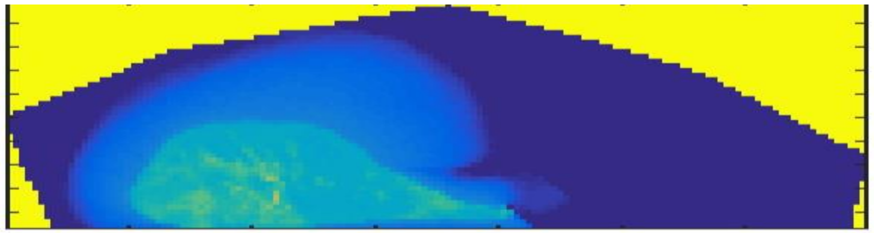
$$\frac{\partial}{\partial t}\left(\phi^T \rho_t^E z_i^E\right) + \mathrm{div}(\boldsymbol{e_i l}) = 0$$

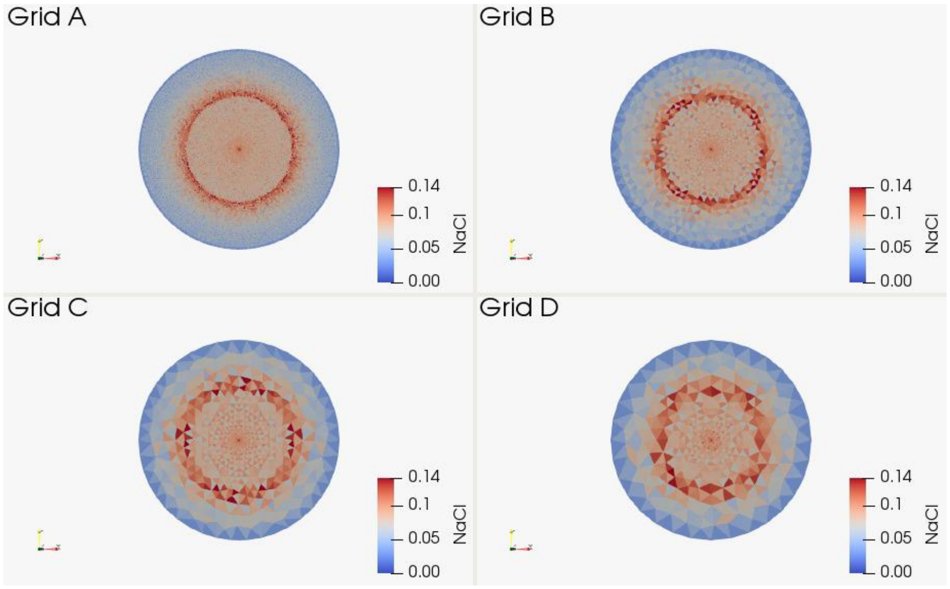
$$f_i^g = f_i^l \qquad \rho_t^E = \rho_t \textstyle\sum_{i=1}^{n_e} \boldsymbol{e_i z}$$

$$\prod_{c=1}^{n_c} a_c^{v_{cq}} - K_q = 0 \qquad \boldsymbol{z^E} \textstyle\sum_{i=1}^{n_e} \boldsymbol{e_i z} - \boldsymbol{Ez} = 0$$









Kala and Voskov, Comput. Geosci, 2020

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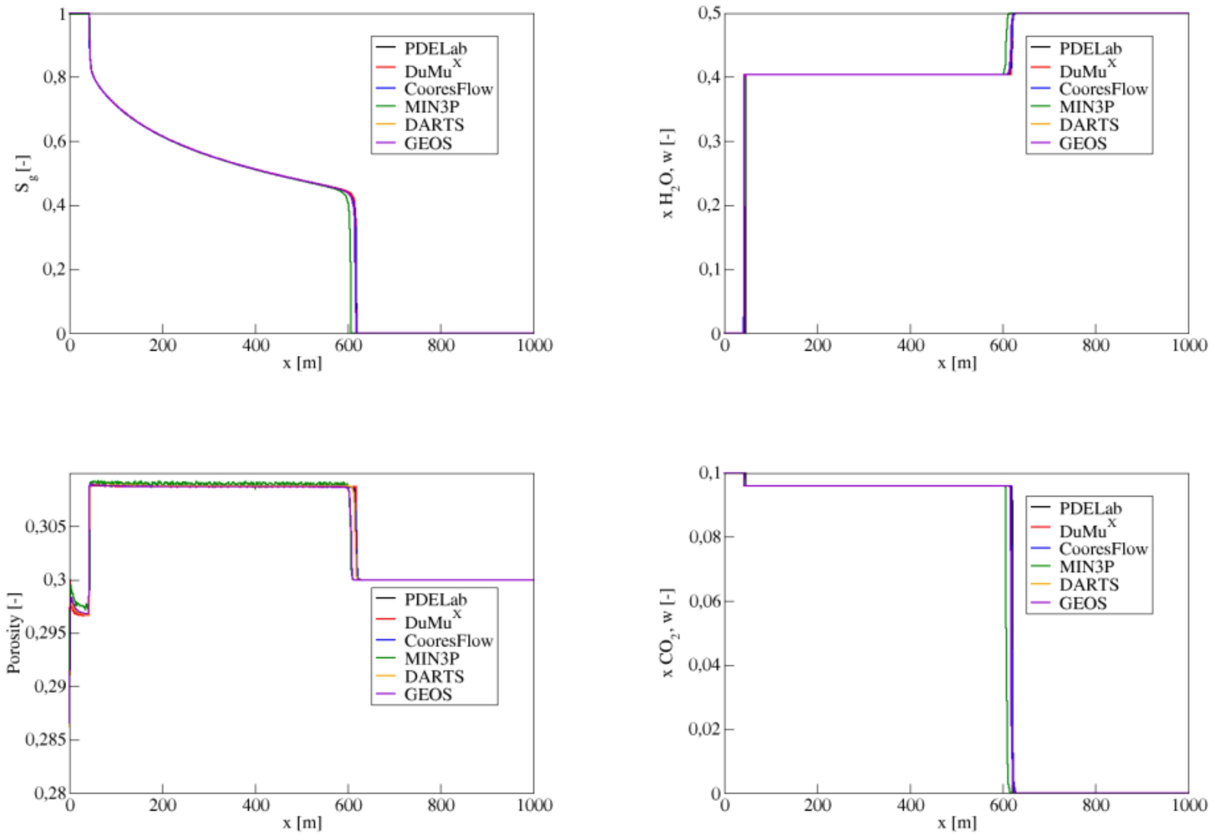
Multiphase flow with reactions (1D benchmark)

$$\frac{\partial n_c}{\partial t} + l_c + q_c = \sum_{k=1}^{K} v_{ck} r_k^K + \sum_{q=1}^{Q} v_{cq} r_q^Q, \quad c = 1, \ldots, C + M,$$

$$\phi = \phi^T \left(1 - \sum_{m=1}^{M} \hat{s}_m\right) \quad k = k_0 \left(\frac{\phi}{\phi_0}\right)^A$$

$$H_2O \rightleftharpoons H^+ + OH^-,$$
$$CO_2 + H_2O \rightleftharpoons HCO_3^- + H^+,$$
$$HCO_3^- \rightleftharpoons H^+ + CO_3^{-2},$$
$$CaCO_3 + H^+ \rightleftharpoons Ca^{+2} + HCO_3^-.$$



de Hoop et al., Comput. Geosci, 2024; Ahusborde et al., Comput. Geosci, 2024

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CO2 injection into calcite core

- Carbon dioxide dissolution:

$$CO_{2(g)} \rightleftharpoons CO_{2(aq)},$$

- Acid formation:

$$CO_{2(aq)} + H_2O \rightleftharpoons H_2CO_3,$$

$$H_2CO_3 \rightleftharpoons H^+ + HCO_3^-,$$

$$HCO_3^- \rightleftharpoons H^+ + CO_3^{2-},$$

- Calcite dissolution:

$$CaCO_3 + H^+ \rightarrow Ca^{2+} + HCO_3^-,$$

$$CaCO_3 + H_2CO_3^* \rightarrow Ca^{2+} + 2HCO_3^-,$$

$$CaCO_3 \rightarrow Ca^{2+} + CO_3^{2-}.$$

- Other aqueous reactions considered:

$$H_2O \rightleftharpoons H^+ + OH^-,$$

$$2H_2O \rightleftharpoons O_2 + 4H^+ + 4e^-,$$

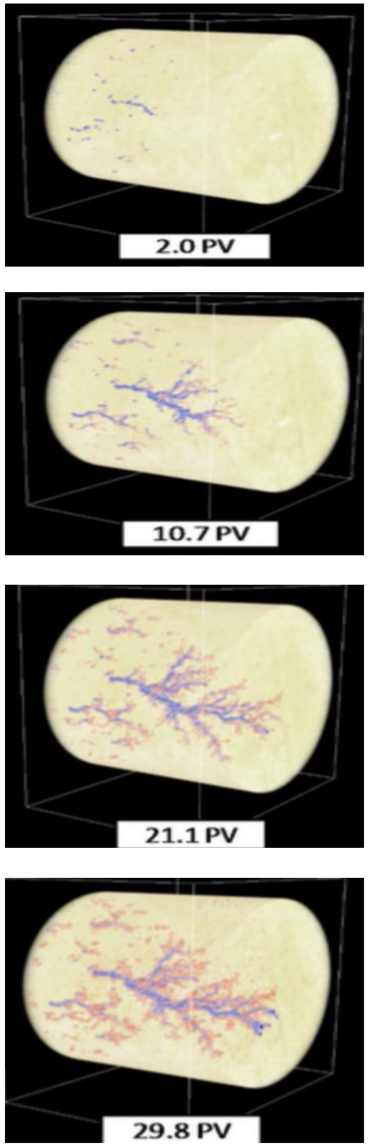
$$2H^+ + 2e^- \rightleftharpoons H_2,$$

$$CO_3^{-2} + 2H^+ \rightleftharpoons CO_2 + H_2O,$$

$$CO_3^{-2} + 10H^+ + 8e^- \rightleftharpoons CH_4 + 3H_2O,$$

$$Ca^{2+} + H_2O \rightleftharpoons CaOH^+ + H^+,$$

$$Ca^{2+} + CO_3^{-2} + H^+ \rightleftharpoons CaHCO_3^+.$$



Snippe et al., IJGGC, 2019

Use PHREEQC for equilibrium chemistry calculation.

Element balance reduction

$$\mathbf{S} = \begin{array}{l} \\ H_2O \\ H^+ \\ OH^- \\ CO_2 \\ HCO_3^- \\ CO_3^{2-} \\ CaCO_3 \\ Ca^{2+} \\ CaOH^+ \\ CaHCO_3^+ \\ CaCO_{3,solid} \end{array} \begin{array}{c} \begin{array}{ccccccc} q_1 & q_2 & q_3 & q_4 & q_5 & q_6 & k_1 \end{array} \\ \begin{bmatrix} -1 & 0 & 1 & -1 & 0 & 0 & 0 \\ 1 & -1 & -2 & 1 & 0 & -1 & 0 \\ 1 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 & 0 & 0 \\ 0 & -1 & -1 & 0 & -1 & -1 & 1 \\ 0 & 0 & 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & -1 & -1 & -1 & 1 \\ 0 & 0 & 0 & 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & -1 \end{bmatrix} \end{array}$$

$$\mathbf{S}_{C\times R} = \left[\begin{array}{c|c} \mathbf{Q}_{Q\times K} & \text{-}\mathbf{I}_{1,\,Q\times Q} \\ \text{-}\mathbf{I}_{2,\,K\times K} & \mathbf{S}_{3,\,K\times Q} \\ \hline \mathbf{S}_{1,\,(C-R)\times K} & \mathbf{S}_{2,\,(C-R)\times Q}, \end{array}\right]$$

$$\mathbf{E}_{1(E\times C)} = \begin{bmatrix} e_{11} & e_{12} & \dots & e_{1C} \\ e_{21} & e_{22} & \dots & e_{2C} \\ \dots & \dots & \ddots & \dots \\ e_{E1} & e_{E2} & \dots & e_{EC} \end{bmatrix}$$

$$\mathbf{E}_{2(K\times C)} = \begin{bmatrix} \text{-}\mathbf{S}_{1,\,K\times Q} & \text{-}\mathbf{I}_{2,\,K\times K} & \mathbf{0}_{K\times(C-R)} \end{bmatrix}$$

$$\mathbf{E} = \begin{array}{l} \\ H \\ O \\ C \\ Ca \\ CaCO_{3,solid} \end{array} \begin{array}{c} \begin{array}{cccccccc} H_2O & H^+ & OH^- & CO_2 & HCO_3^- & CO_3^{2-} & \dots & CaCO_{3,solid} \end{array} \\ \begin{bmatrix} 2 & 1 & 1 & 0 & 1 & 0 & \dots & 0 \\ 1 & 0 & 1 & 2 & 3 & 3 & \dots & 3 \\ 0 & 0 & 0 & 1 & 1 & 1 & \dots & 1 \\ 0 & 0 & 0 & 0 & 0 & 0 & \dots & 1 \\ 0 & 0 & 0 & 0 & 0 & 0 & \dots & 1 \end{bmatrix} \end{array}$$

$$\frac{\partial \mathbf{n}}{\partial t} + \mathbf{l} = \mathbf{Vr},$$

$$\frac{\partial\,(\mathbf{En})}{\partial t} + \mathbf{El} = \mathbf{ESr} = \begin{bmatrix} \mathbf{E}_1\mathbf{Sr} \\ \mathbf{E}_2\mathbf{Sr} \end{bmatrix} = \begin{bmatrix} 0 \\ \mathbf{r}_k \end{bmatrix}$$

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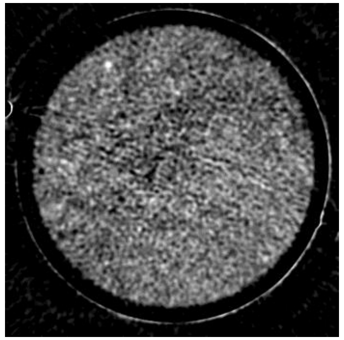
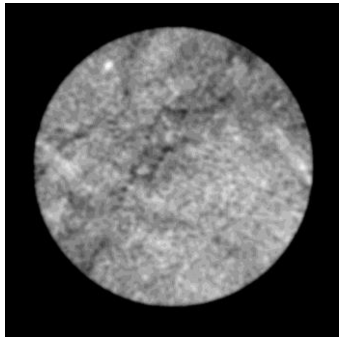
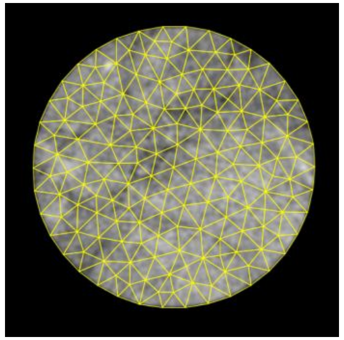
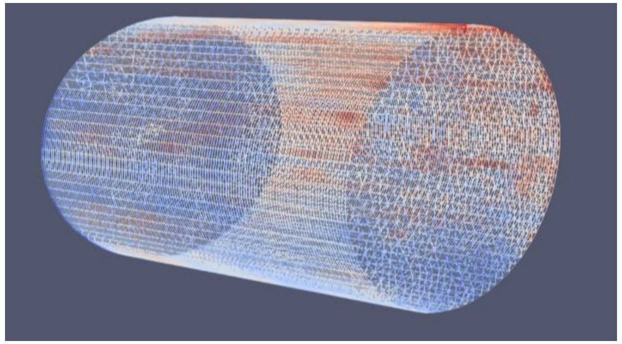
Modeling of dissolution at core scale

Step 1: porosity interpretation (image subtraction, filtering, gridding)

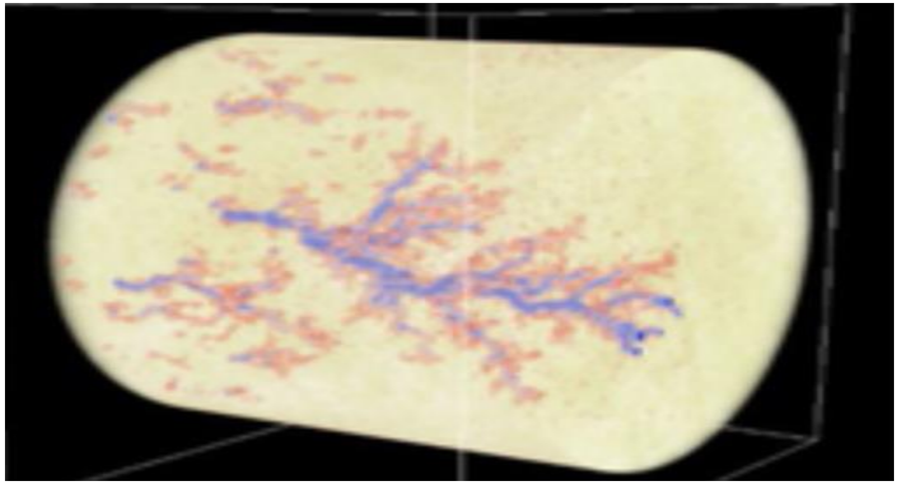


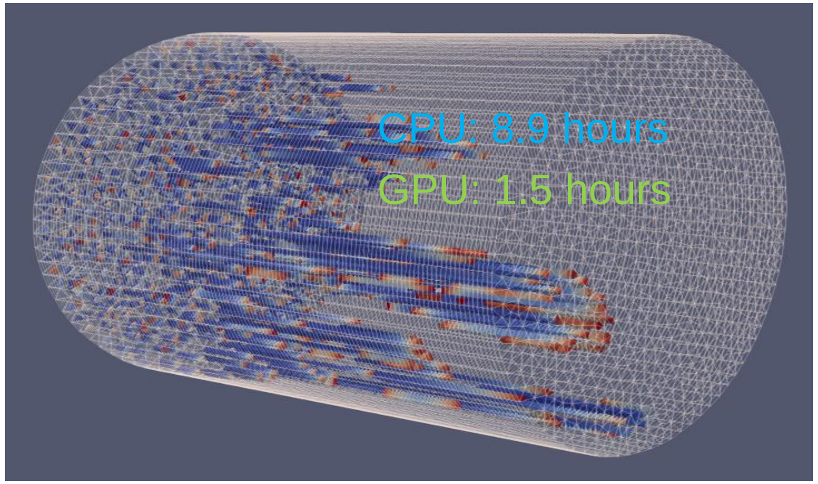






Step 2: modeling of dissolution (combination of DARTS + PHREEQC)





Run time:

8.9 hours (CPU engine)

PHREEQC call: 12 min

Points generated:

26068 (<0.01%)

Interpolations: 2.8e9

Margert, MSc thesis, 2019

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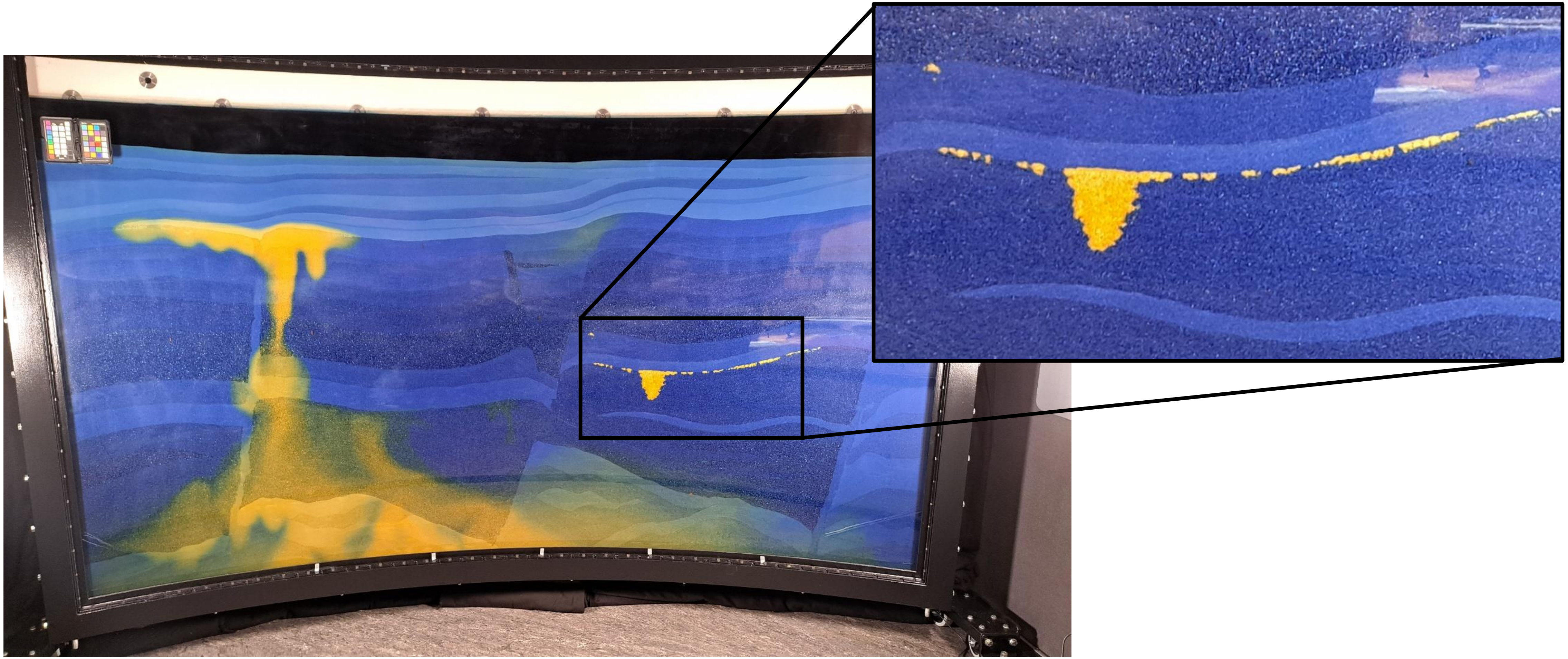
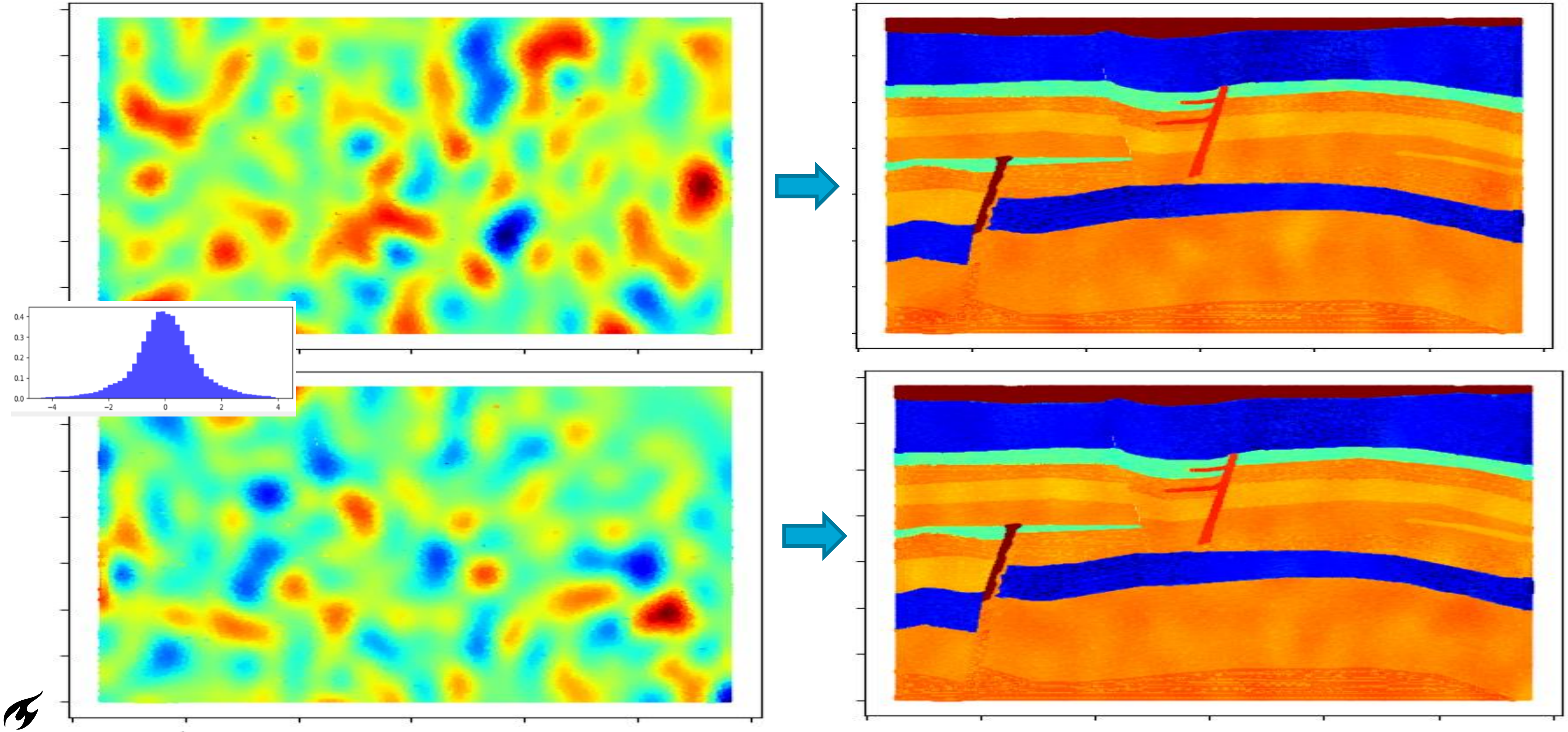
FluidFlower benchmark



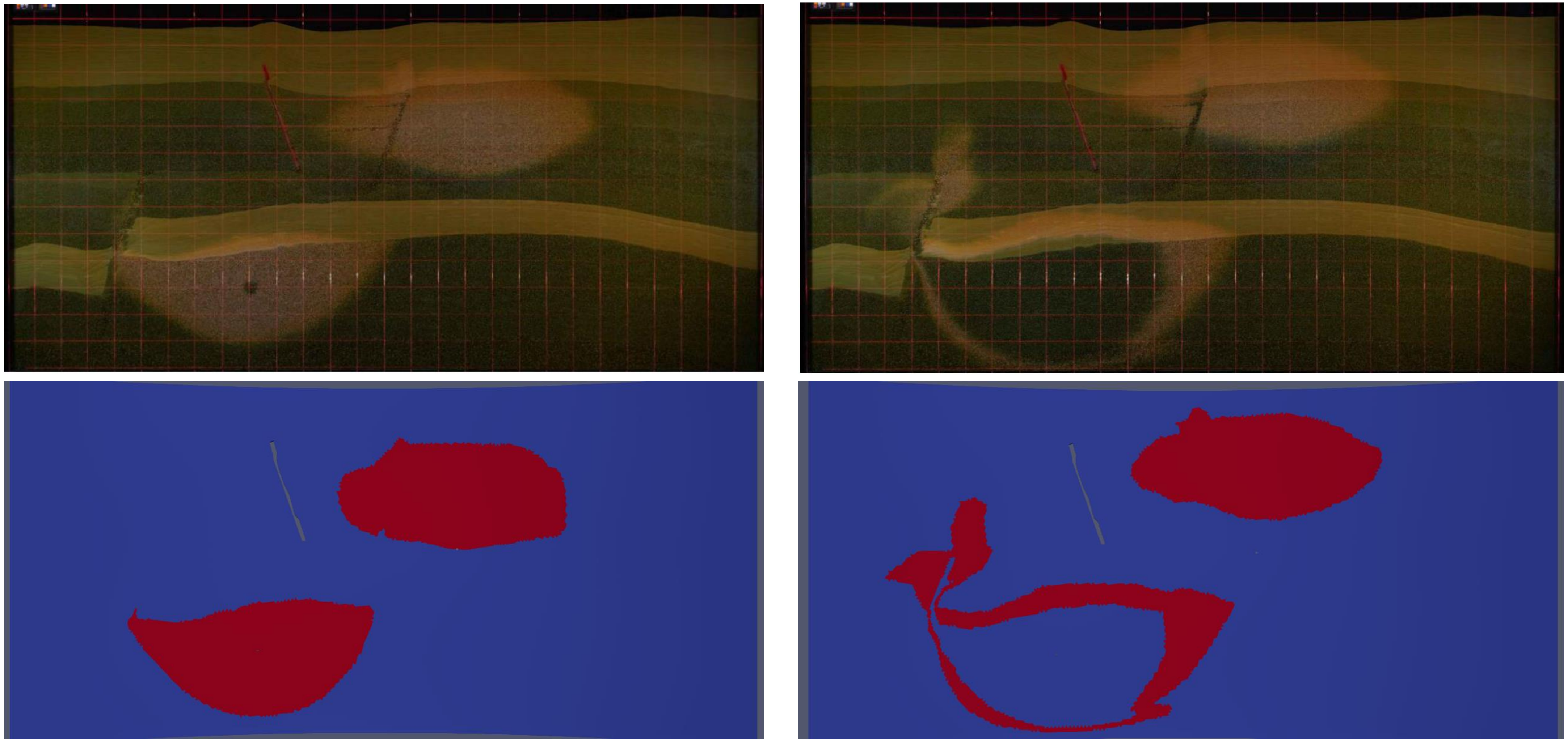
Flemish et al., TIPM, 2024

Generation of prior realizations



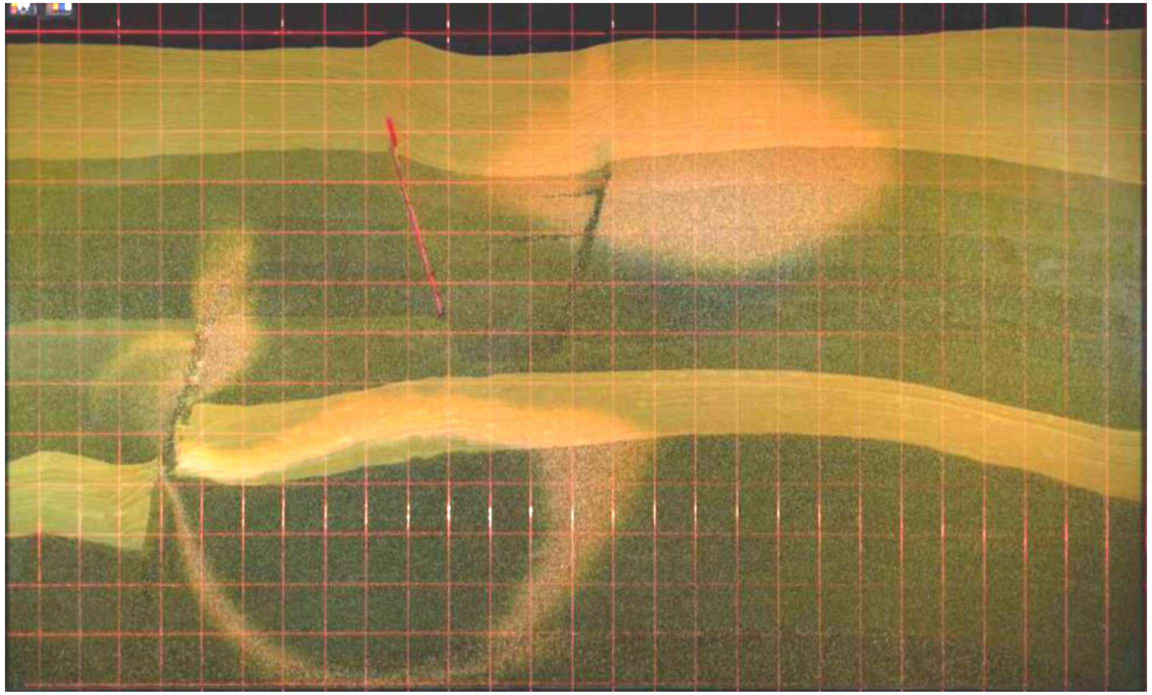
Tian et al., TIPM, 2024

Digitizing of tracer test

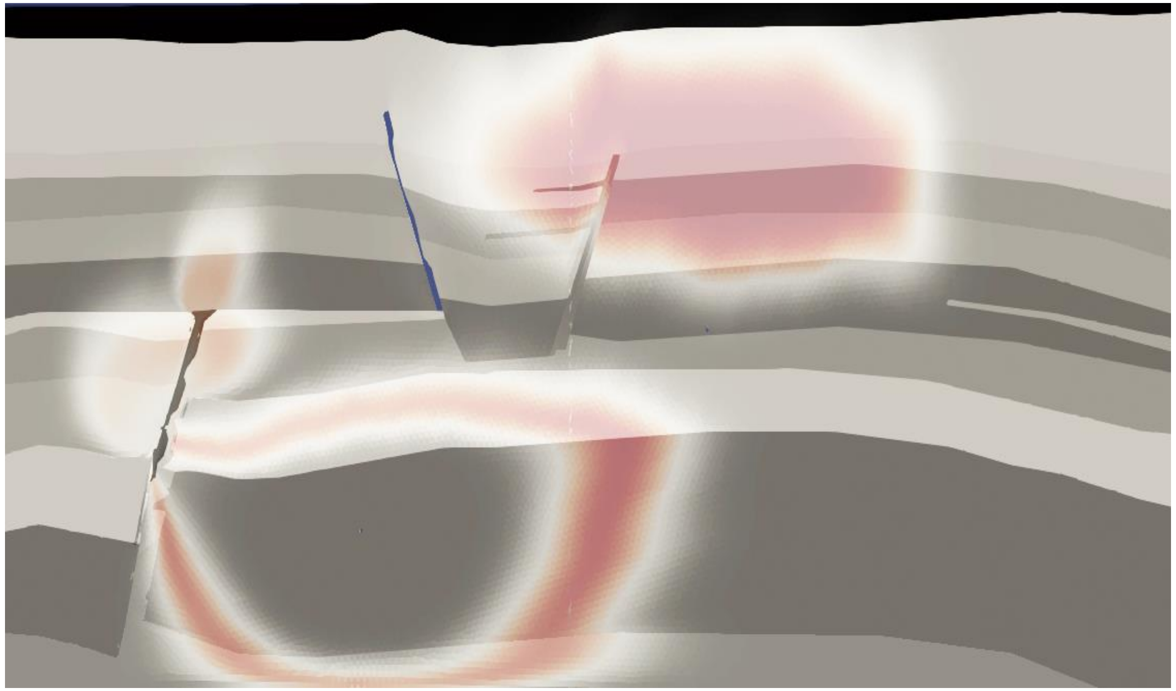


History matching using RML (single realization)

Tracer observations (high resolution images)



Inversed model (RML, 18,278 forward runs for 100 priors)



$$E(\boldsymbol{u}) = \frac{1}{2}(\boldsymbol{u} - \boldsymbol{u}_{\mathrm{ref}})^{\mathrm{T}} C_M^{-1} (\boldsymbol{u} - \boldsymbol{u}_{\mathrm{ref}}) + \frac{1}{2}(G(\boldsymbol{u}) - \mathbf{d}_{\mathrm{obs}} + \boldsymbol{\epsilon})^{\mathrm{T}} C_D^{-1} (G(\boldsymbol{u}) - \mathbf{d}_{\mathrm{obs}} + \boldsymbol{\epsilon})$$

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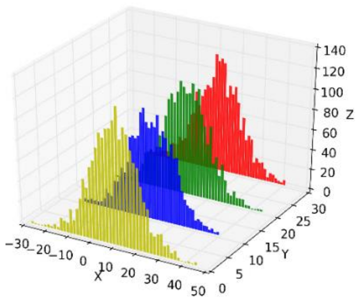
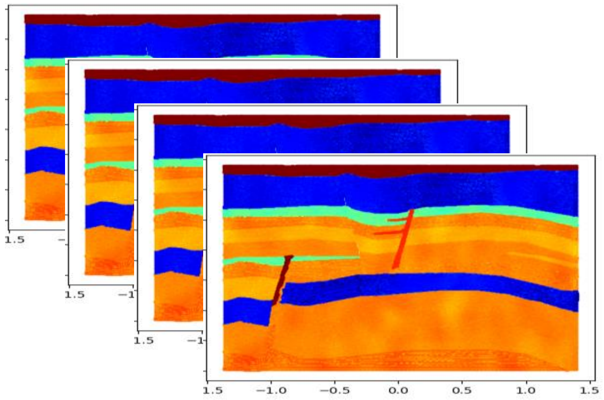
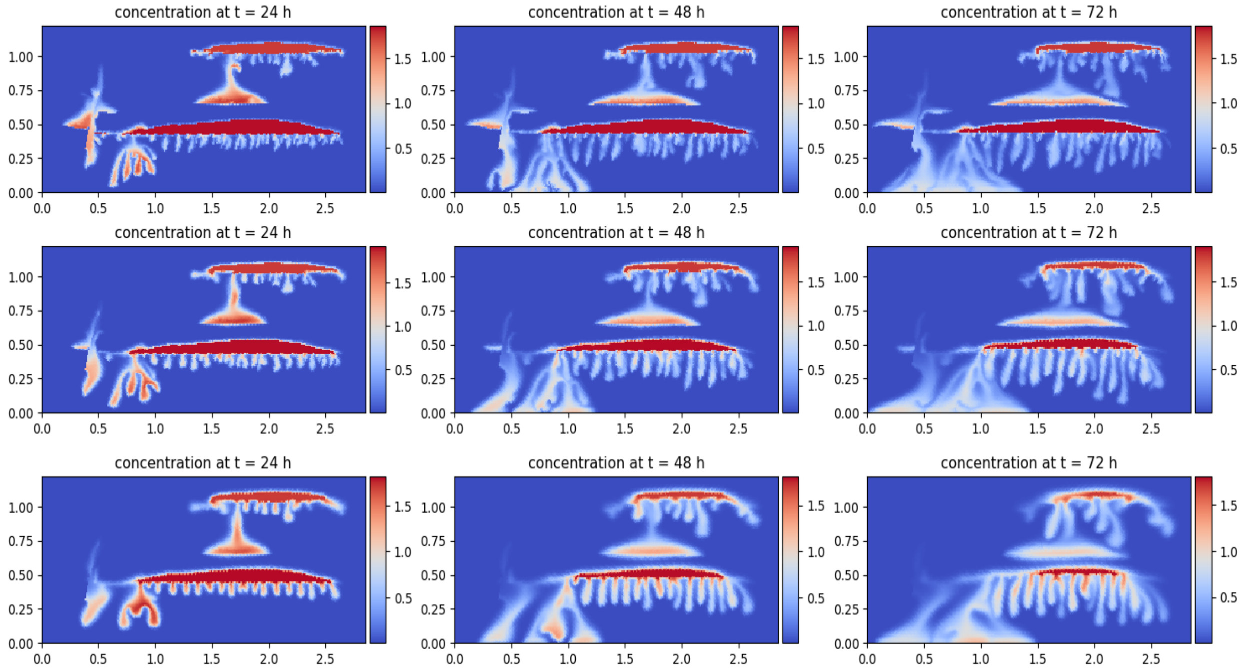
Data Assimilation for FluidFlower experiments

- Models: 100 history matched permeability maps
- Temperature: $23°C \pm 2$, normal distribution
- Diffusion: $2 \cdot 10^{-10} - 2 \cdot 10^{-9}$, log-normal distribution
- Two phase: Corey parameters with std from 5 to 50%, normal distribution

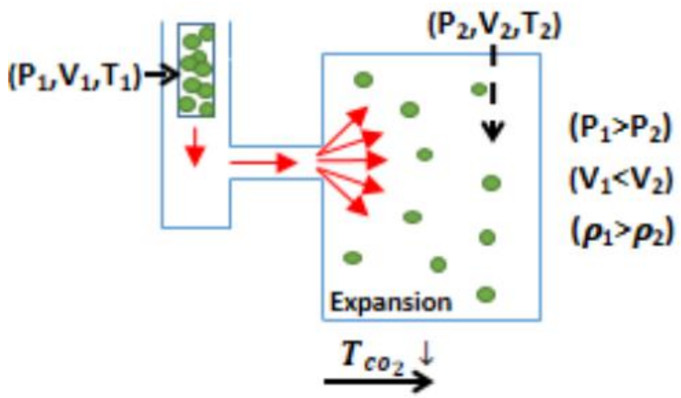


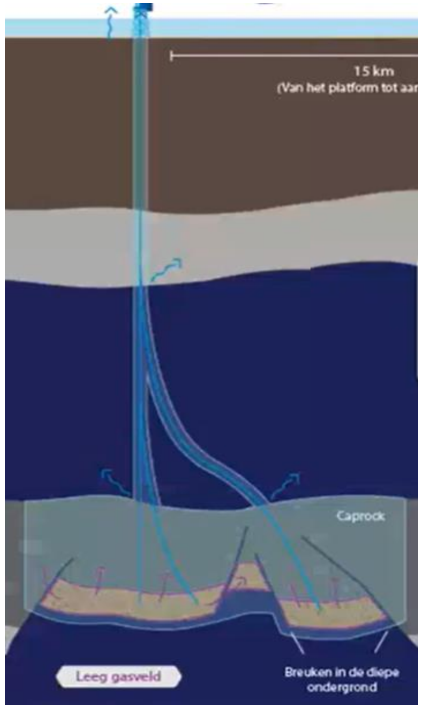




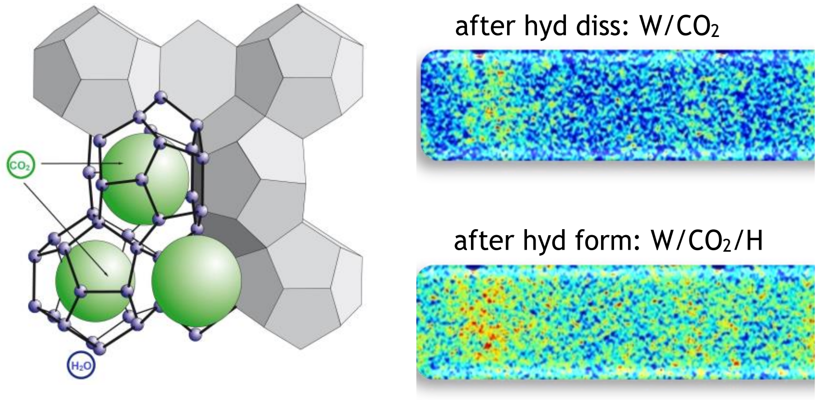
CCS in depleted fields: Joule-Thomson cooling

Isenthalpic cooling

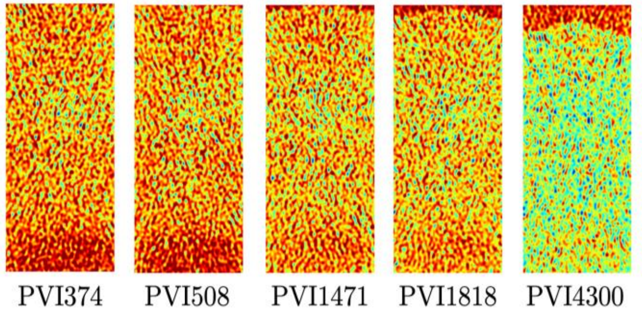


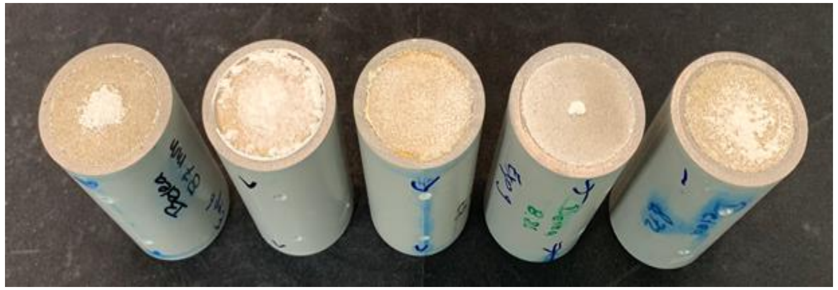


Hydrate formation



Salt precipitation





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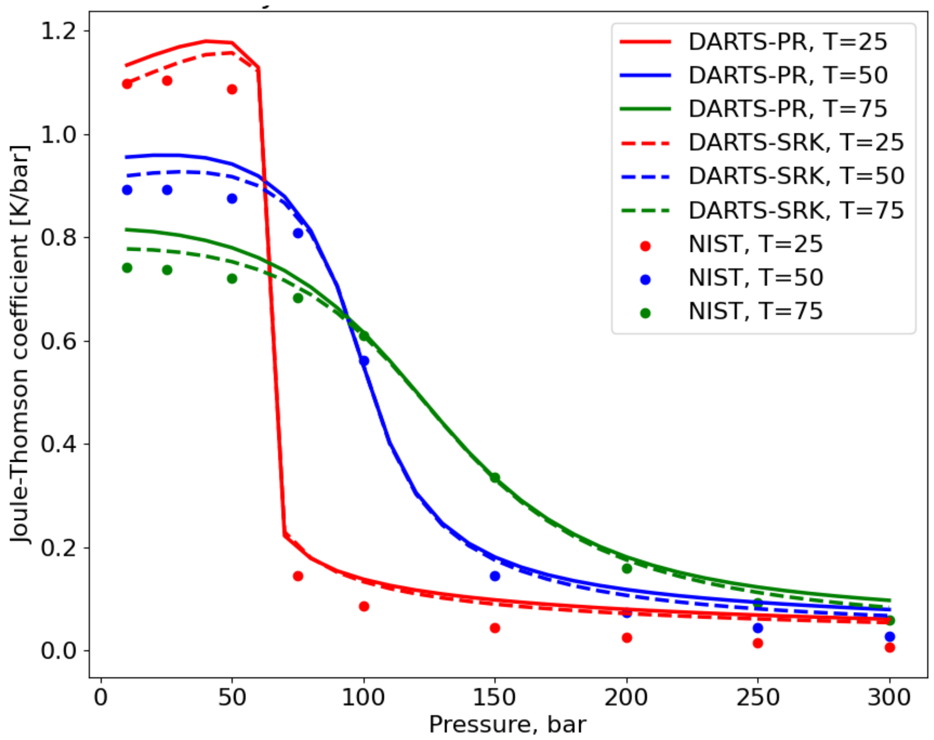
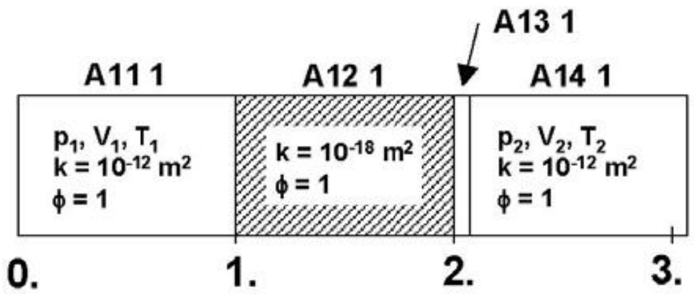
JT-cooling validation

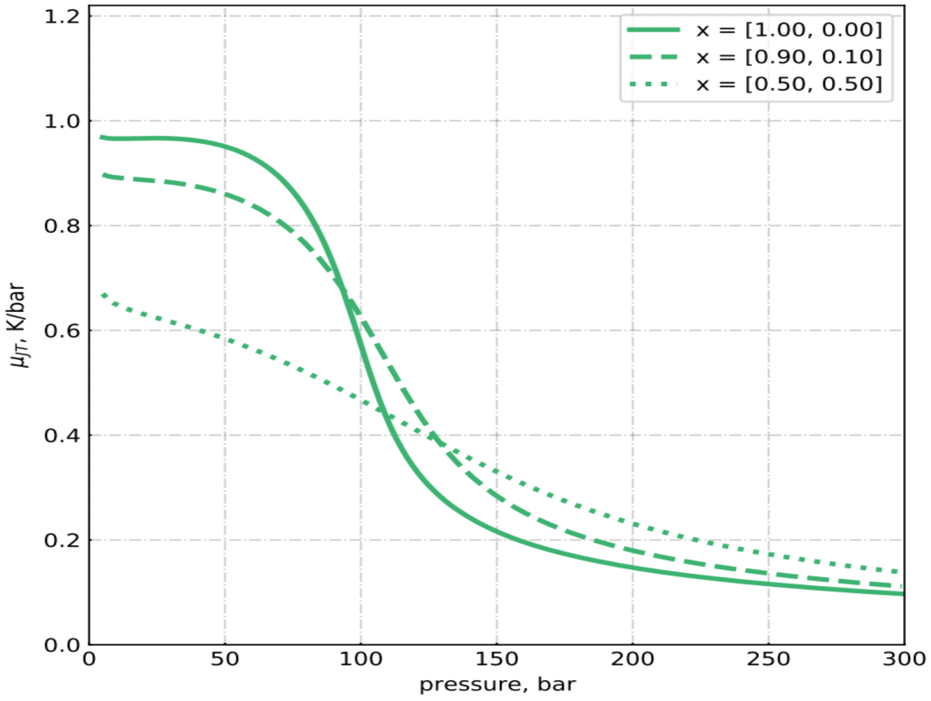
Cooling for Pure CO_2





Oldenburg, 2006

Cooling for Mixtures CO_2-CH_4



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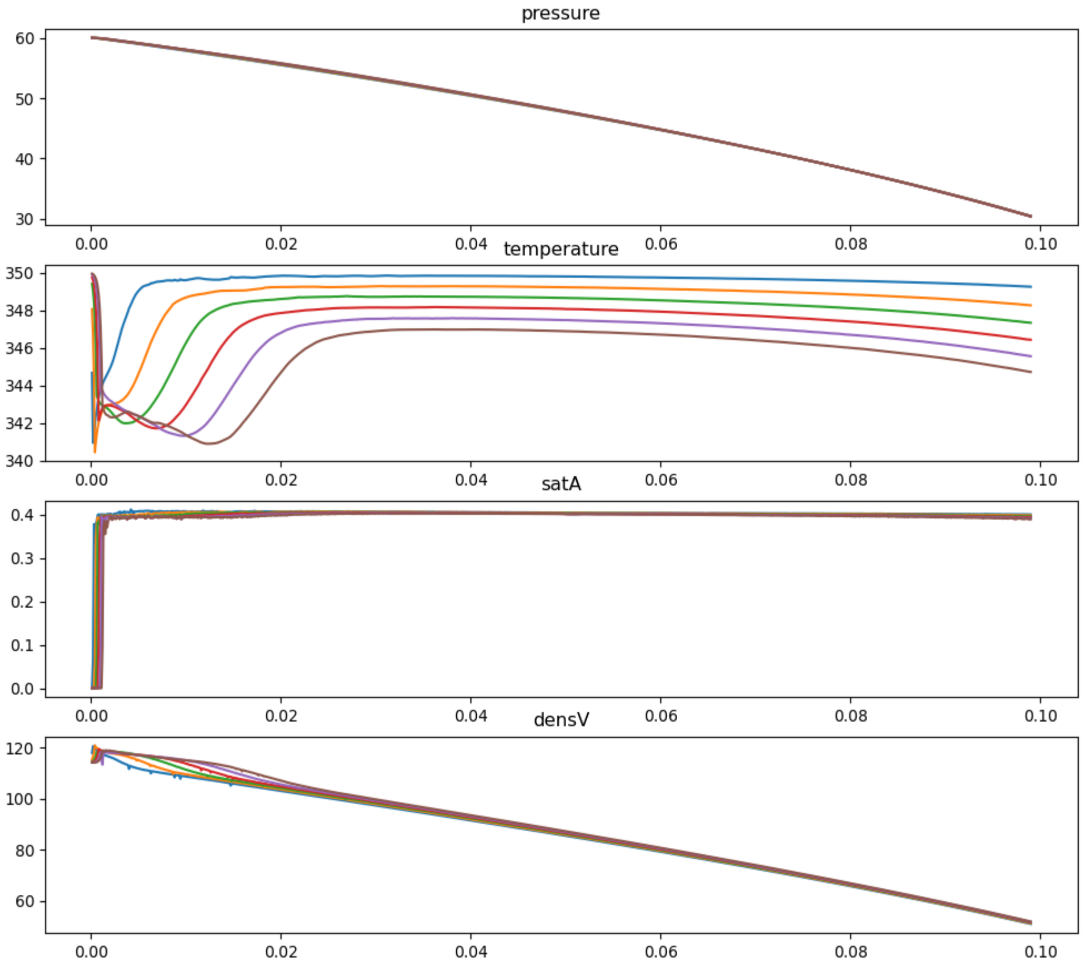
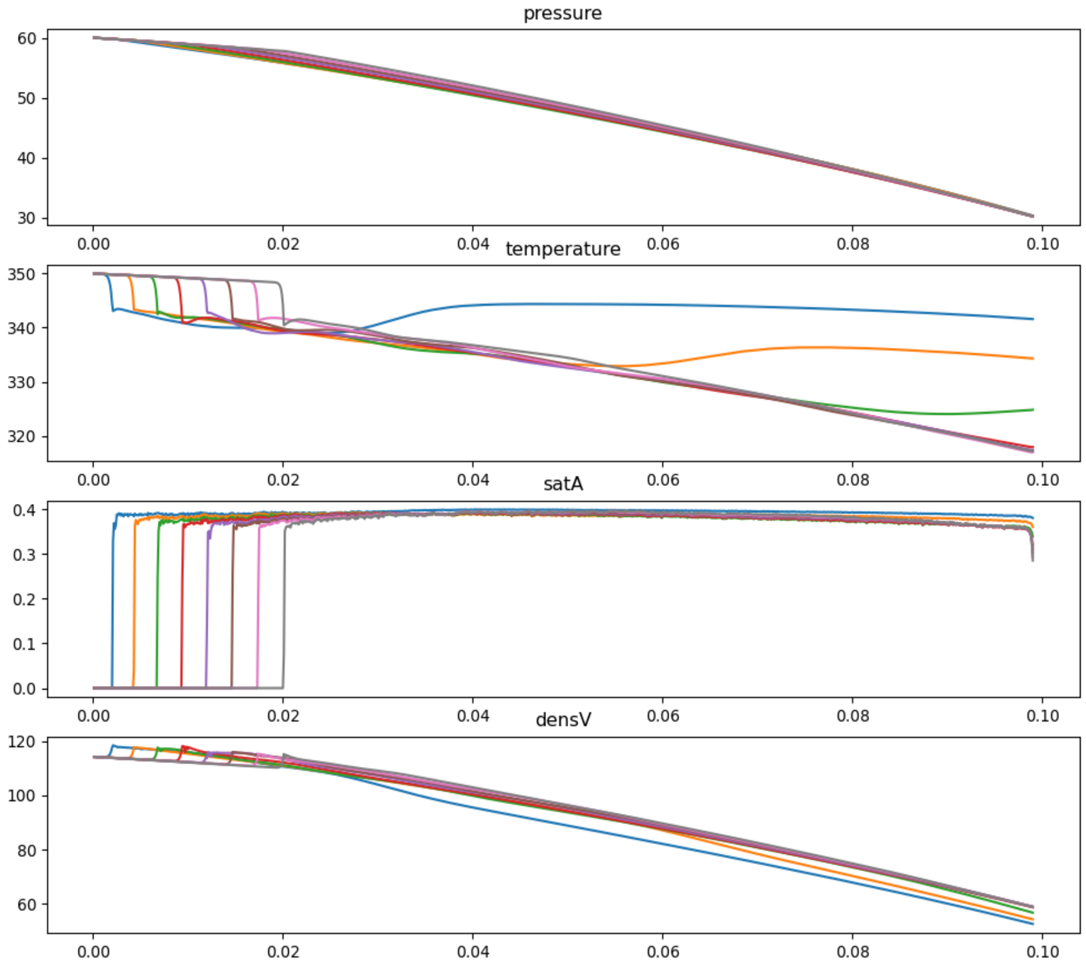
JT effect at various scales

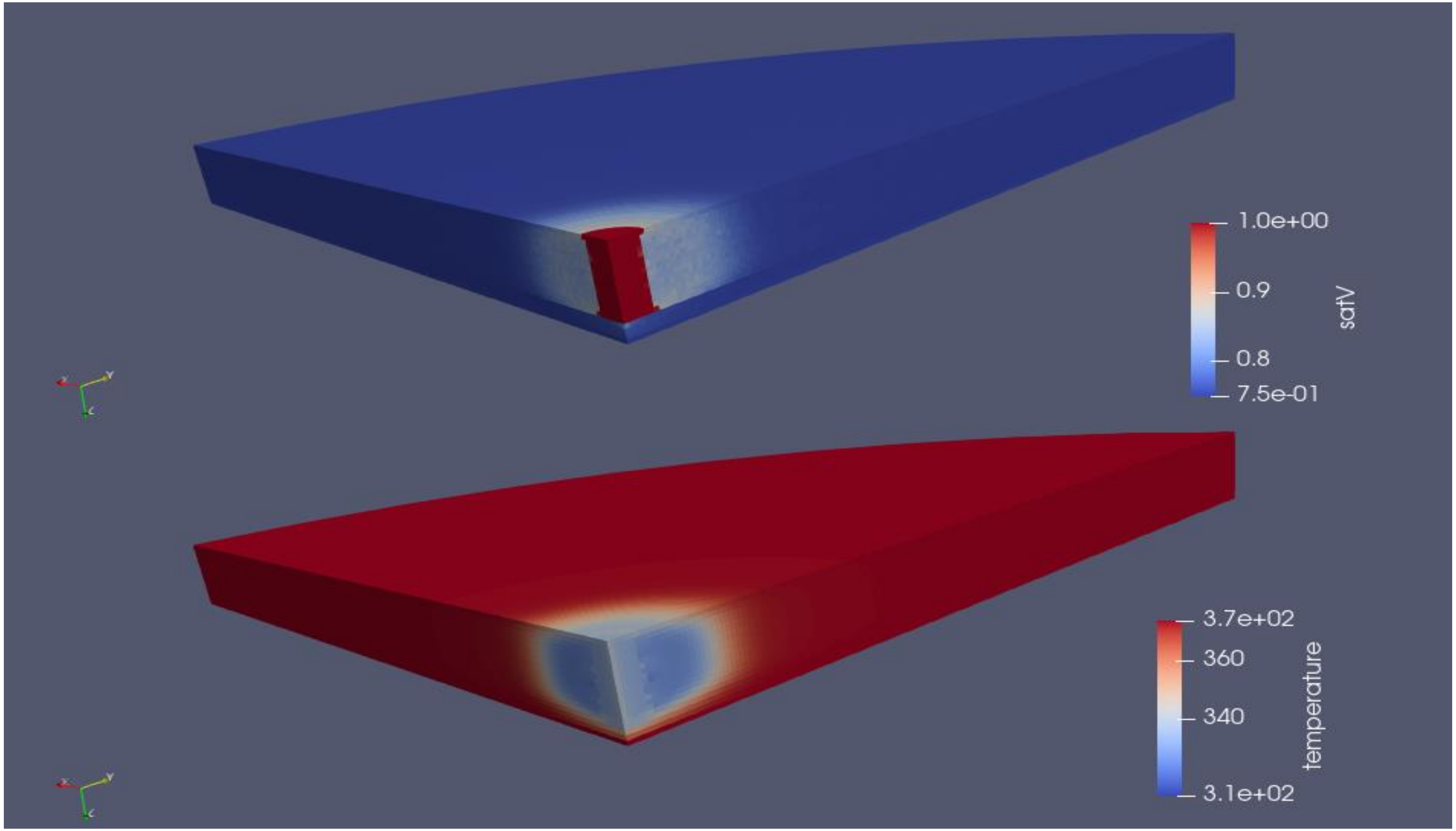
JT in CO2-CH4-H2O system at experimental scale, 1 sec



JT in CO2-CH4-H2O system at experimental scale, 10 sec

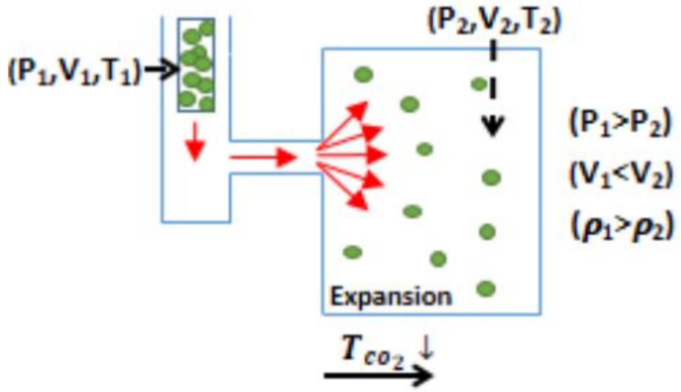


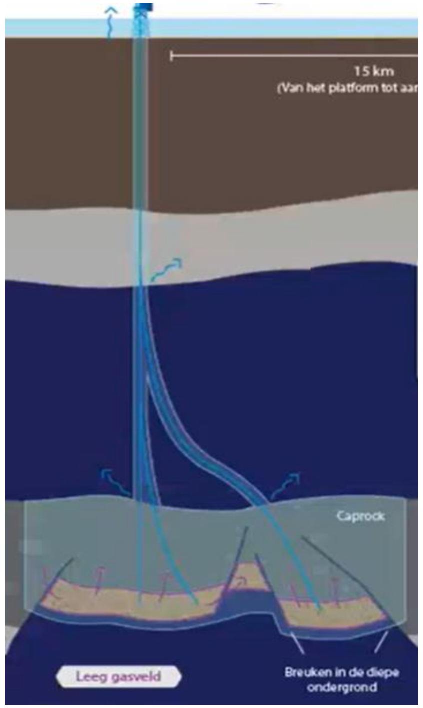
Radial model results



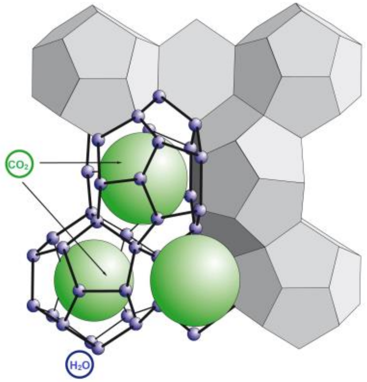
CCS in depleted fields: impact of salt precipitation

Isenthalpic cooling

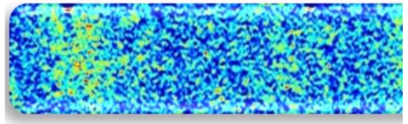




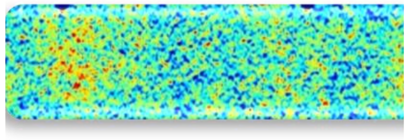
Hydrate formation



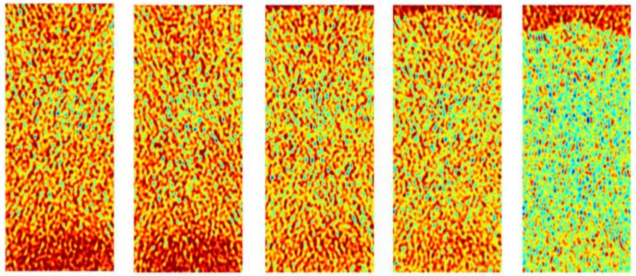
after hyd diss: W/CO_2



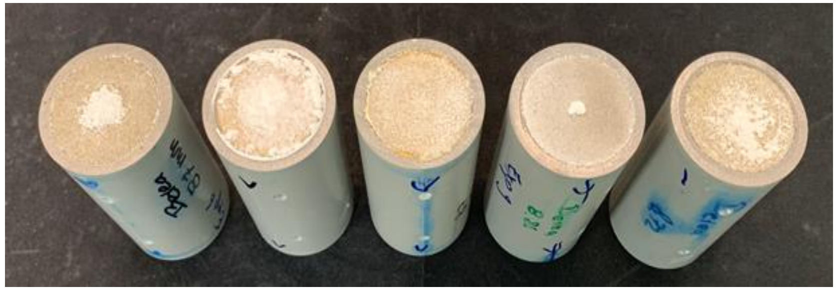
after hyd form: W/CO_2/H



Salt precipitation



PVI374 PVI508 PVI1471 PVI1818 PVI4300



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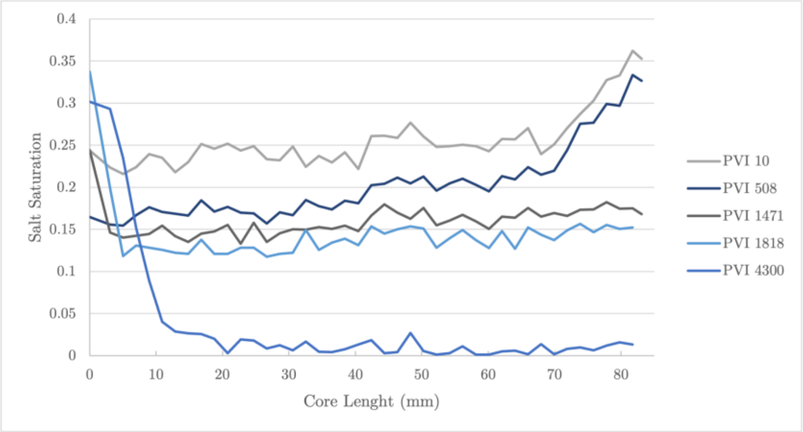
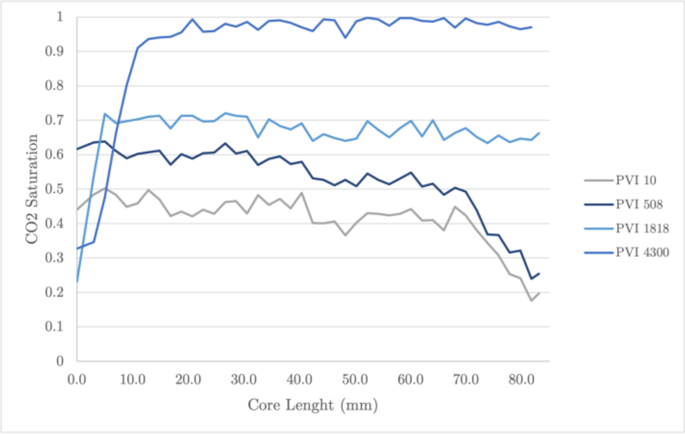
Salt precipitation: core flood experiments

Gas and salt saturations in a homogeneous core





3D reconstruction of salt in Bentheimer core

Permeability reduction in a homogeneous core

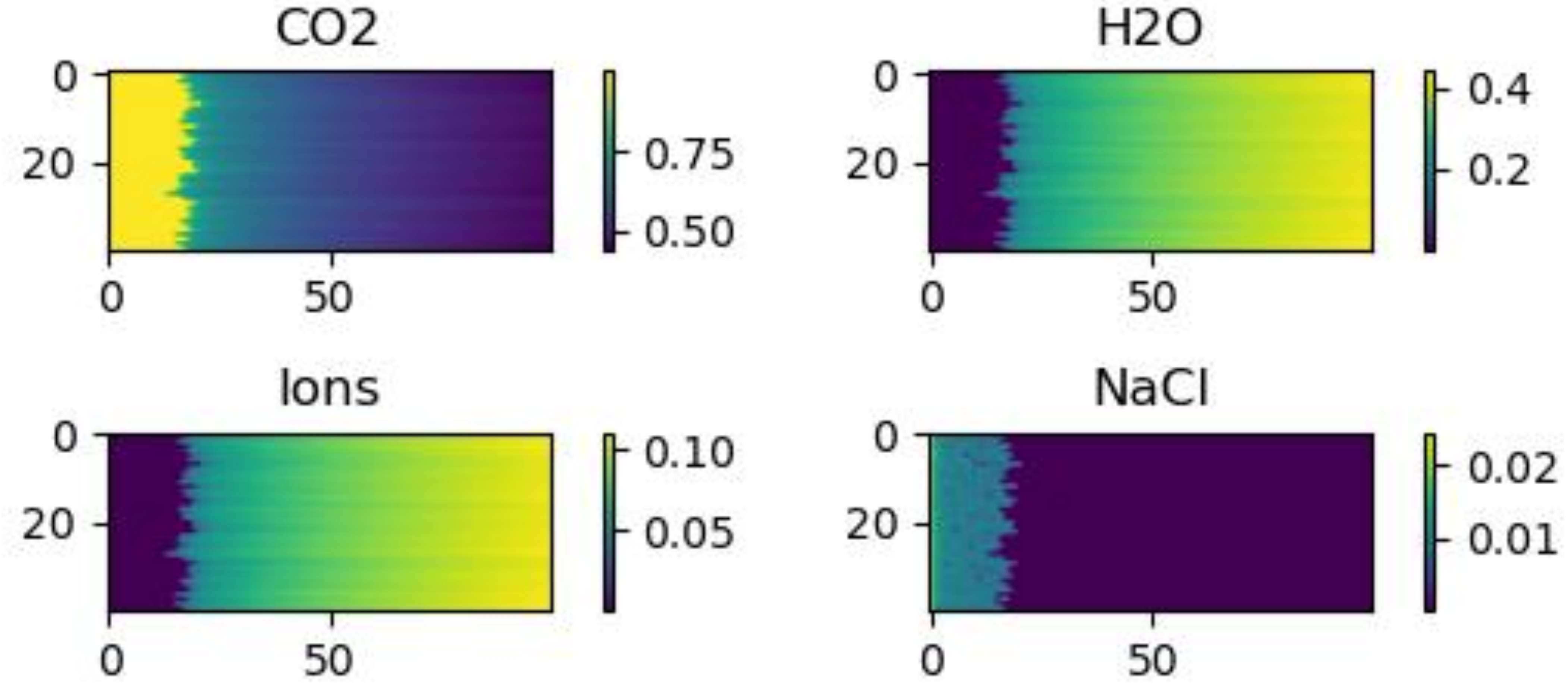
Core type	1st section of core			2nd section of core		
	K initial (mD)	K final (mD)	K reduction	K initial (mD)	K final (mD)	K reduction
Bentheimer	2164	24	99%	1707	170	90%
Berea	164	0.8	99%	168	148	12%
Fontainebleau	164	18	99%	187	170	9%

Salt mainly precipitated at inlet regions and permeability had a huge reduction in it

TU Delft

Yan et al., in prep.

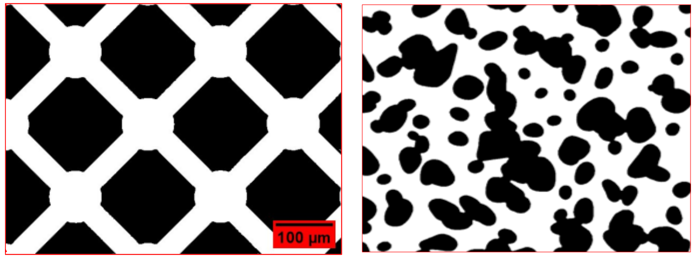
Simulation results with DARTS

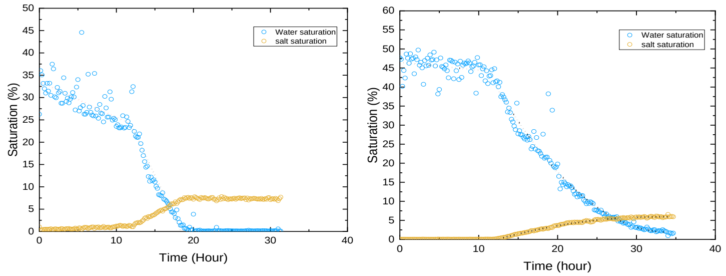


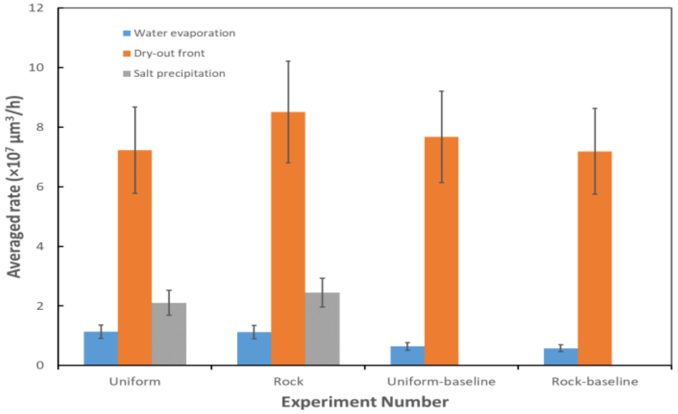
TU Delft

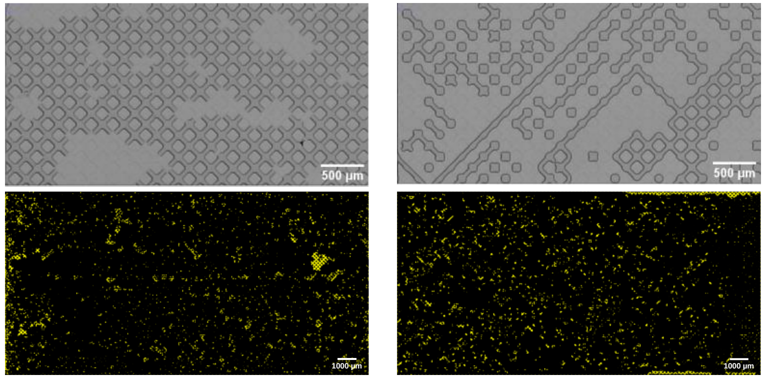
Wapperom et al., ECMOR, 2023

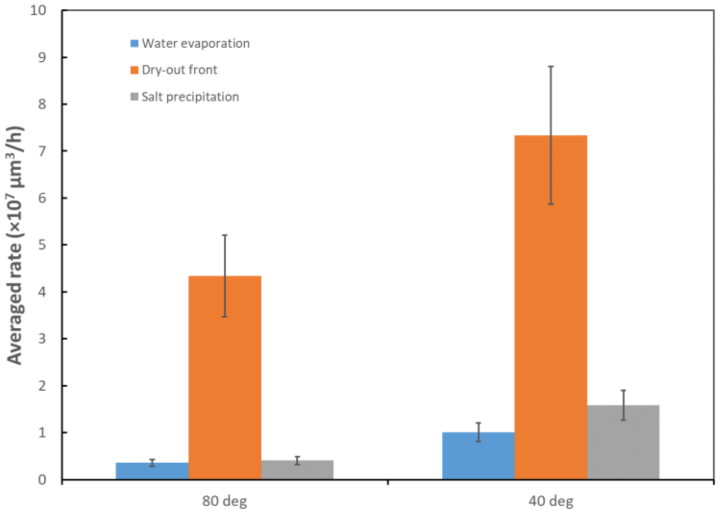
Impact of pore structure, wettability and direction





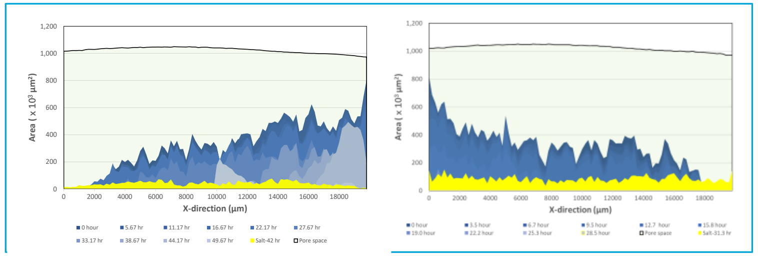




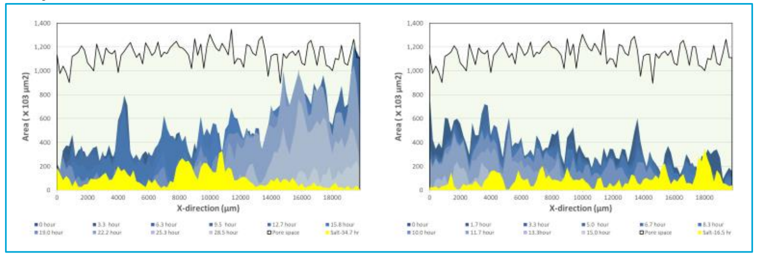


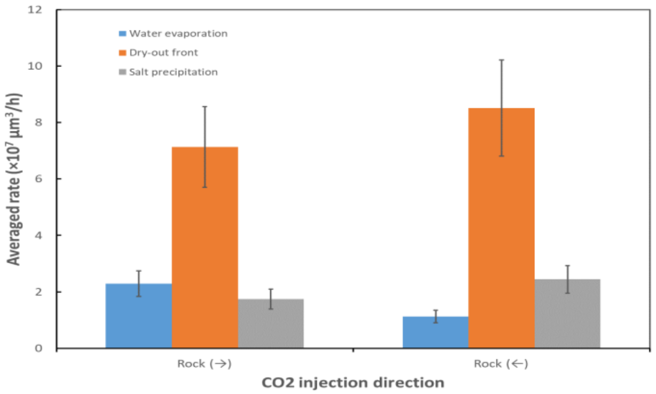
CO2 injection → CO2 injection ←

Uniform structure



Physical rock structure





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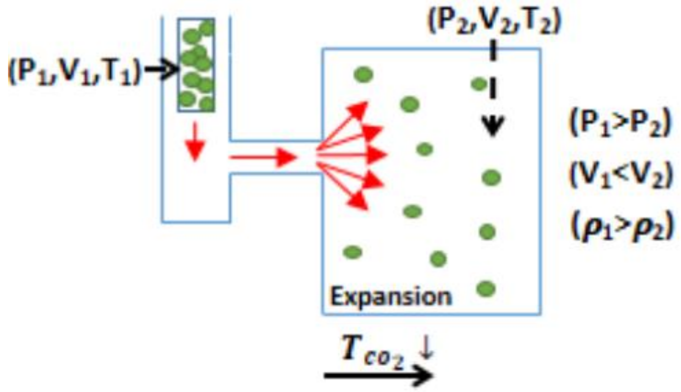
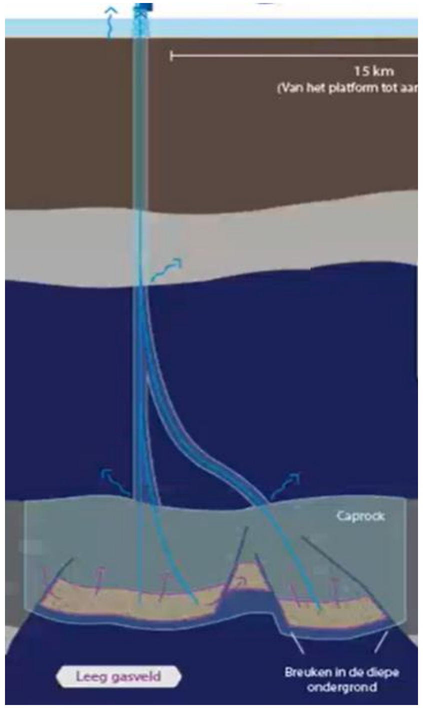
Yan et al., JCIS, 2025

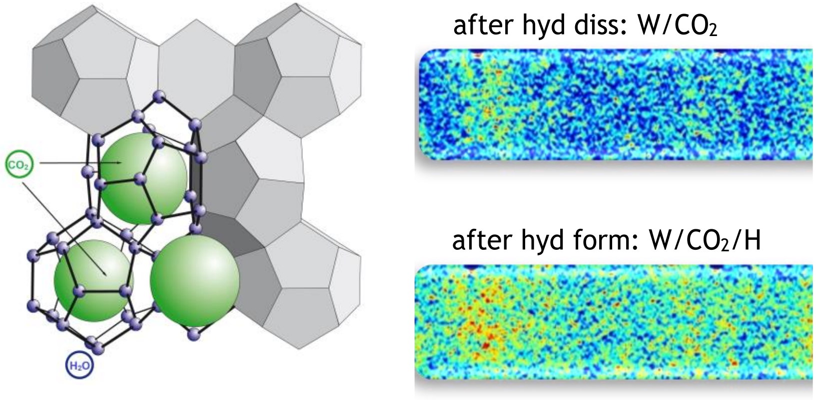
CCS in depleted fields: risk of hydrate formation

Isenthalpic cooling

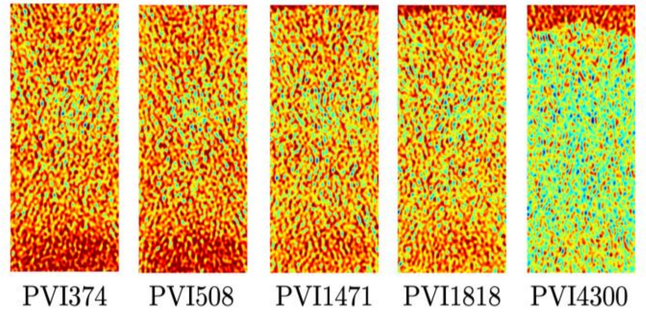


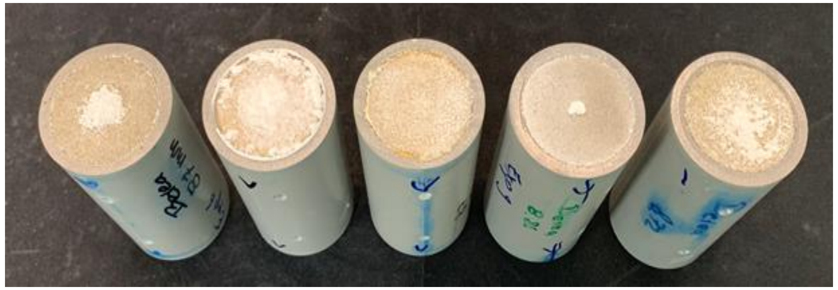


Hydrate formation



Salt precipitation





Thermodynamic models for hydrates

- Van der Waals and Platteeuw (1958)
  - Chemical potential change upon cage filling

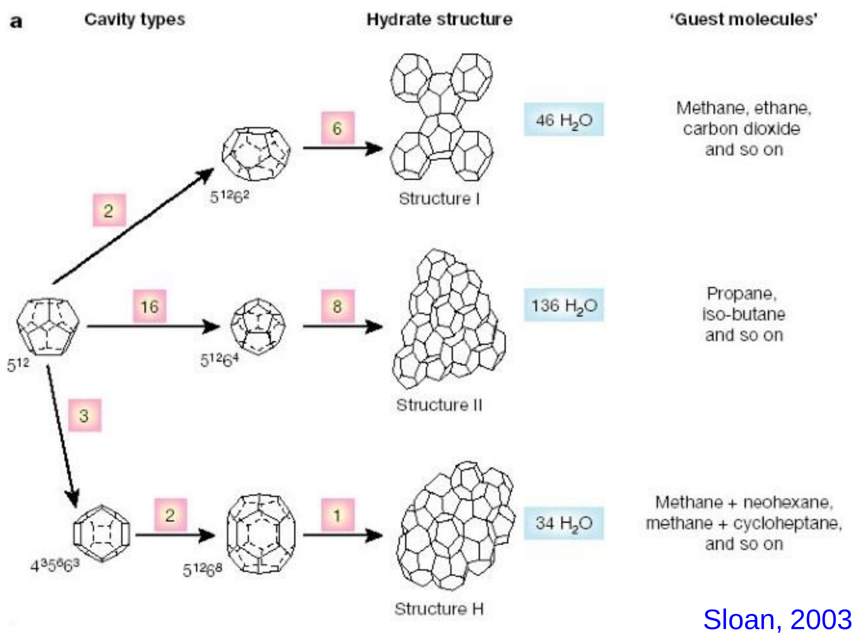
$$\frac{\Delta\mu_{w,H}}{RT} = \frac{\mu_{w,H}}{RT} - \frac{g_{w,\beta}}{RT} = \sum_m \nu_m \ln\left(1 - \sum_j \theta_{jm}\right)$$

  - Fugacity related to reference phase

$$f_{w,H} = f_{w,A} \exp\left[\frac{\Delta\mu_{w,H} - \Delta\mu_{w,A}}{RT}\right]$$

  - Langmuir 'adsorption'

$$\theta_{jm} = \frac{C_{jm} f_j}{1 + \sum_k C_{km} f_k}$$



Sloan, 2003

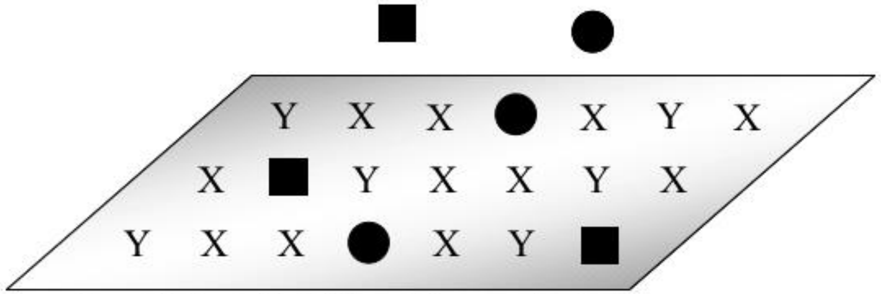


Figure 4.1 Visual of multi-site, multi-component adsorption

Ballard, 2002

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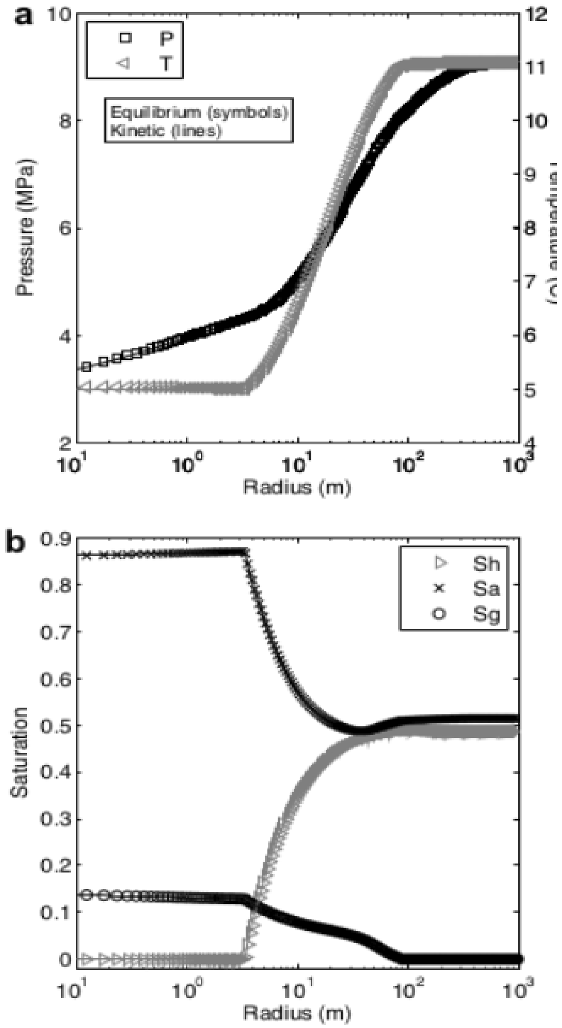
Hydrate dissociation

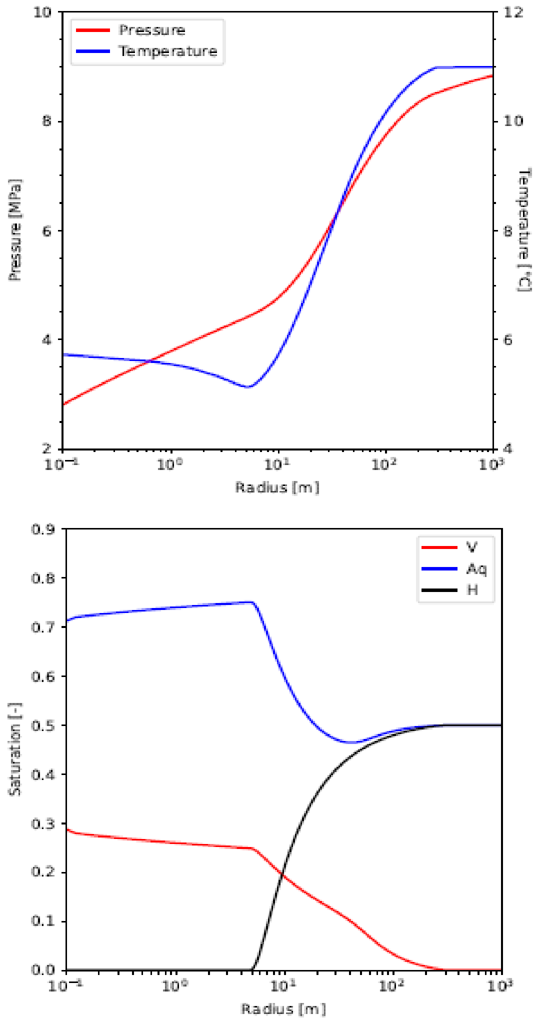
- Class 3 hydrate deposit
  - Radial reservoir
  - Partially saturated with CH_4-hydrate
  - Pressure at well 27 bar

TOUGH+Hydrate



Kowalsky & Moridis, 2007

DARTS



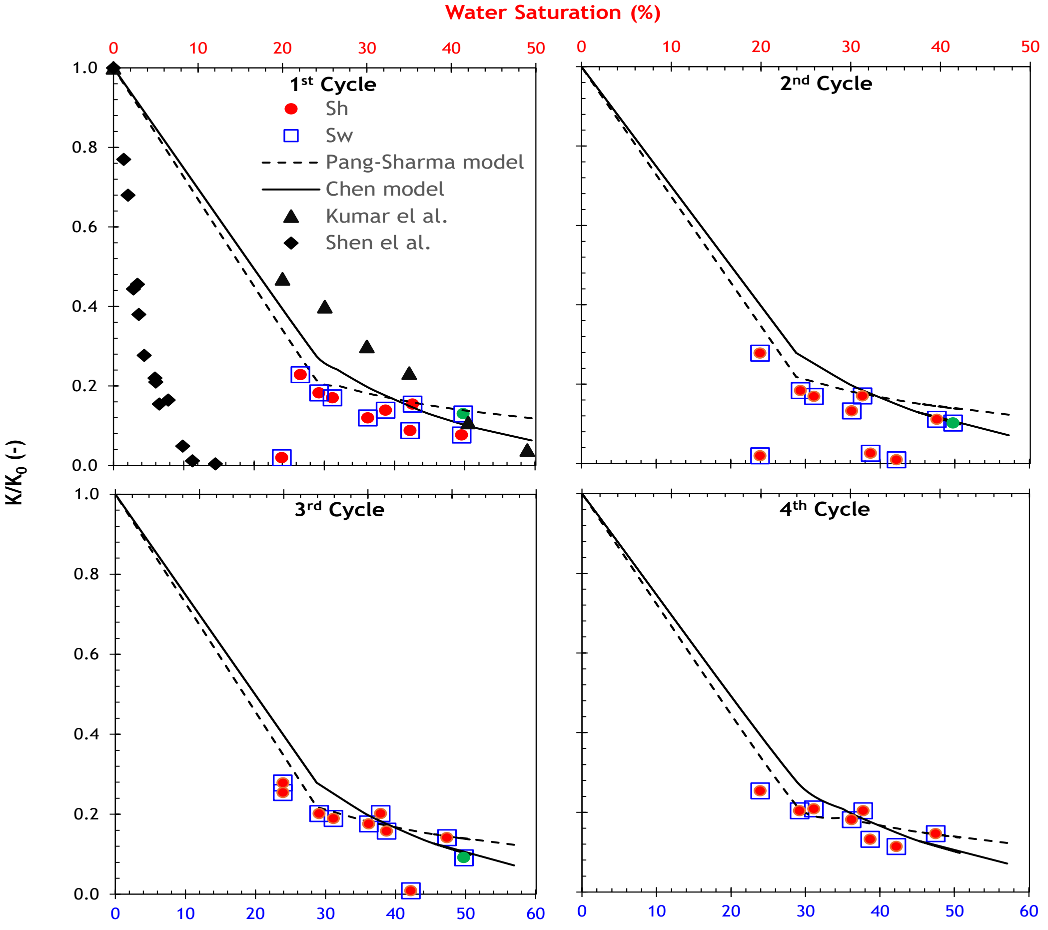
Wapperom & Voskov, in prep.

Impact of water saturation on permeability

Exp	Core	Salt type/concentration
1-9	Bentheimer	1wt% NaCl
10	Bentheimer	1wt% NaCl

Note: In exp 10, for the first 2 cycles, the normalized permeability was calculated based on the differential pressure, while for the third cycle brine permeability test was performed in the presence of hydrate to validate the previous calculation.

Permeability is directly influenced by hydrate saturation, which is, in turn, dependent on the water saturation level.



Aghajanloo et al., in prep.

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- Students and collegues: Dan Nichita, Juan Diego Heringer, Andrey Margert, Michel Kern, Brahim Amaziane, Jan Norbotten, Bernd Flemisch, James Gunning

open-DARTS

https://darts.citg.tudelft.nl

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Time for Questions and Answers