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# Sequential upscaling of multiphase dispersion in porous media

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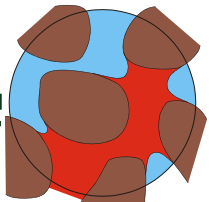
# Sequential upscaling of multiphase dispersion in porous media

J. Guo<sup>1</sup>, F. Laouafa<sup>2</sup> and M. Quintard<sup>3</sup>

<sup>1</sup> Southwest Jiaotong University, P.R. China

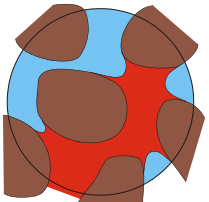
<sup>2</sup> INERIS, France

<sup>3</sup> D.R.C.E. CNRS Emeritus - Université de Toulouse (Institut de Mécanique des Fluides de Toulouse), France - [quintard@imft.fr](mailto:quintard@imft.fr)



# Outline

- Introduction to dissolution applications, Multi-scale aspects: coupling reaction and multiphase transport?
- Pore to Darcy-scale upscaling:
  - Introduction
  - Various models
  - Effective properties
- Darcy-scale behavior
- Large-Scale upscaling
- Conclusions



# Applications...

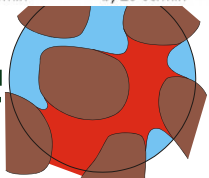
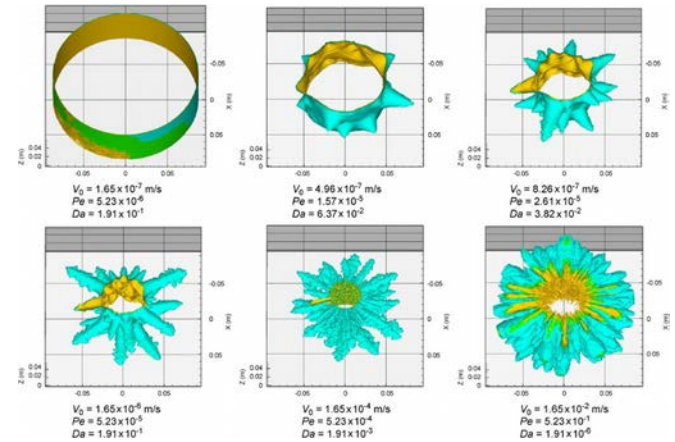
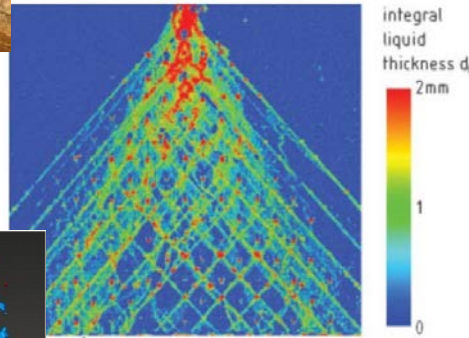
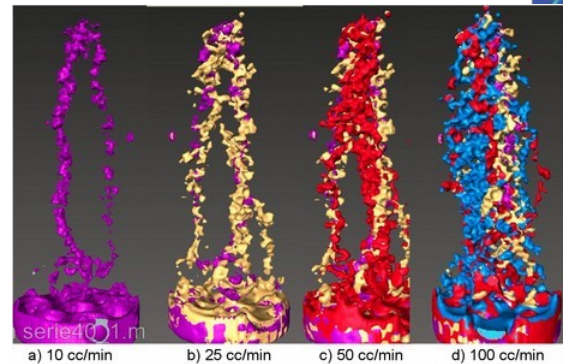


Karsts



Pet engng: CO2 storage, acid injection, etc...

## Chal engng



# Generic Problems: 2-phase flow

$$\nabla \cdot \mathbf{v}_\alpha = 0$$

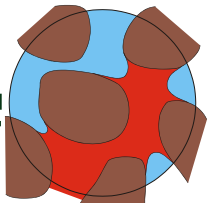
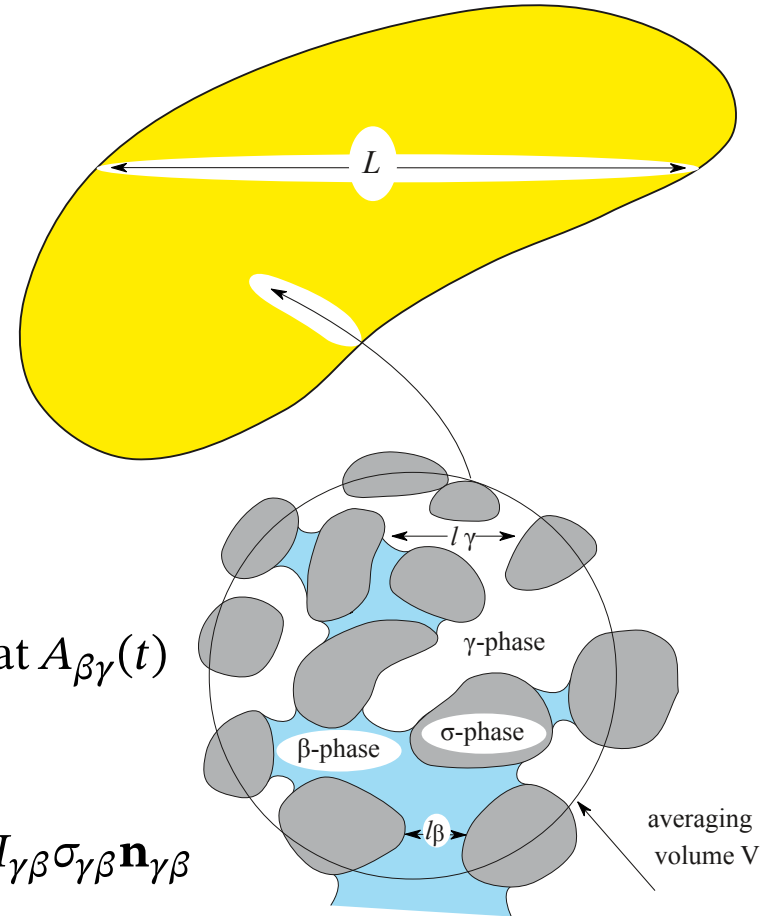
$$\frac{\partial \rho_\alpha \mathbf{v}_\alpha}{\partial t} + \nabla \cdot (\rho_\alpha \mathbf{v}_\alpha \mathbf{v}_\alpha) = -\nabla p_\alpha + \rho_\alpha \mathbf{g} + \mu_\alpha \nabla^2 \mathbf{v}_\alpha \text{ in } V_\alpha \quad \alpha = \beta, \gamma$$

$$\text{B.C. 1 } \mathbf{v}_\beta = 0 \text{ at } A_{\beta\sigma}(t)$$

$$\text{B.C. 2 } \mathbf{v}_\gamma = 0 \text{ at } A_{\gamma\sigma}(t)$$

$$\text{B.C. 3 } \mathbf{v}_\beta - \mathbf{v}_\beta \cdot \mathbf{nn} = \mathbf{v}_\gamma - \mathbf{v}_\gamma \cdot \mathbf{nn} \quad \text{instead of } \mathbf{v}_\beta = \mathbf{v}_\gamma \text{ at } A_{\beta\gamma}(t)$$

$$\text{B.C. 4 } \mathbf{n}_{\gamma\beta} \cdot (\rho_\gamma \mathbf{v}_\gamma (\mathbf{v}_\gamma - \mathbf{w}_{\gamma\beta}) + p_\gamma \mathbf{I} - \tau_\gamma) = \mathbf{n}_{\gamma\beta} \cdot (\rho_\beta \mathbf{v}_\beta (\mathbf{v}_\beta - \mathbf{w}_{\gamma\beta}) + p_\beta \mathbf{I} - \tau_\beta) - 2H_{\gamma\beta} \sigma_{\gamma\beta} \mathbf{n}_{\gamma\beta}$$



# Generic Problems: Reactive transport

Homogeneous reaction

$$\frac{\partial \rho_\beta \omega_\beta}{\partial t} + \nabla \cdot (\rho_\beta \omega_\beta \mathbf{v}_\beta) = \nabla \cdot (\rho_\beta D_\beta \nabla \omega_\beta) + \overset{\text{Homogeneous reaction}}{\downarrow} r_\beta \quad \text{in } V_\beta$$

Heterogeneous reaction:

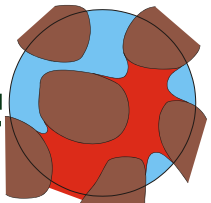
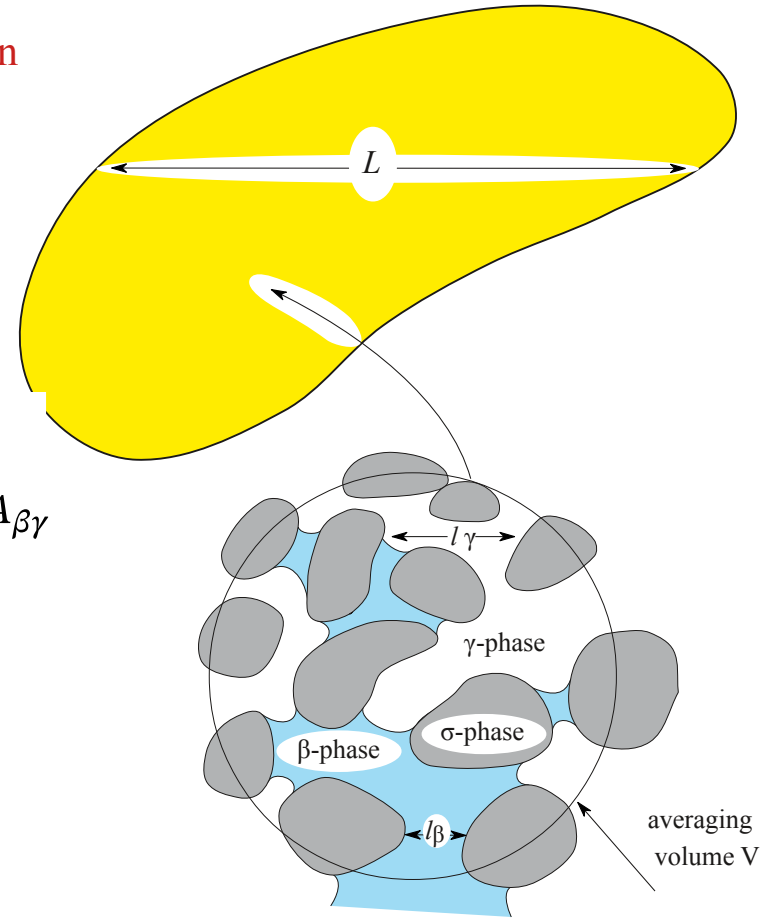
$$\text{B.C.1} \quad \mathbf{n}_{ls} \cdot (\rho_l \omega_l (\mathbf{v}_l - \mathbf{w}_{sl}) - \rho_l D_l \nabla \omega_l) = -M_{Ca} k_s \left(1 - \frac{\omega_l}{\omega_{eq}}\right)^n \quad \text{at } A_{\beta\gamma}$$

Local Equilibrium:

$$\text{B.C.1} \quad \omega_{eq} = \omega_l \quad \text{at } A_{\beta\gamma}$$

or more complex eqs.

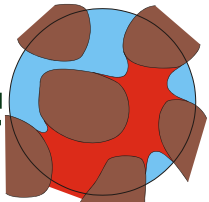
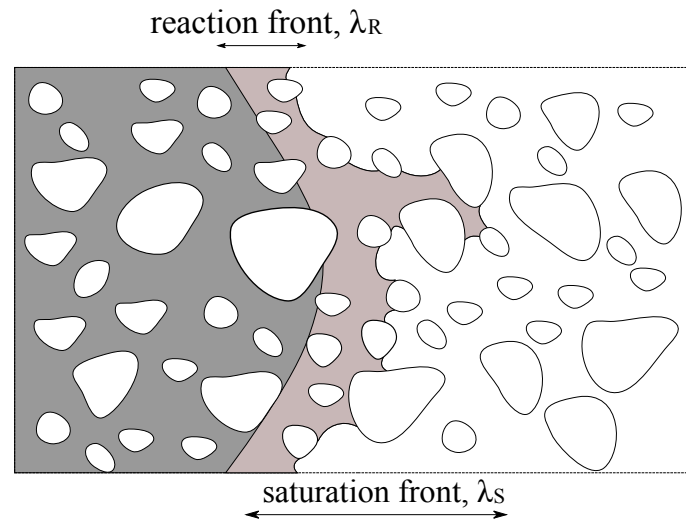
+ other mass balance equations



# Upscaling: momentum equations

- Decoupling between two-phase flow and reaction?
  - Need to neglect terms involving  $w_{\beta\gamma}$
  - If  $\rho$ ,  $\mu$  and  $\sigma$  depends on concentration: need

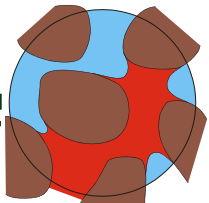
$$\lambda_r \gg \lambda_s \quad ?$$





# Decoupled momentum transfer: various models

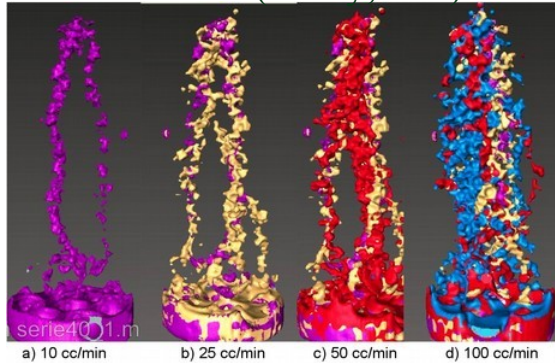
Model	PDEs
Quasi-static, heuristic (Muskat)	$\frac{\partial \varepsilon S_\alpha}{\partial t} + \nabla \cdot \mathbf{V}_\alpha = 0 ; \mathbf{V}_\alpha = -\frac{1}{\mu_\alpha} \mathbf{K}_\alpha \cdot (\nabla P_\alpha - \rho_\alpha \mathbf{g}) \quad \alpha = \beta, \gamma$
Quasi-static, low Re, with cross terms	$\mathbf{V}_\alpha = -\frac{1}{\mu_\alpha} \mathbf{K}_\alpha \cdot (\nabla P_\alpha - \rho_\alpha \mathbf{g}) + \mathbf{K}_{\alpha\kappa} \cdot \mathbf{V}_\kappa \quad \alpha, \kappa = \beta, \gamma \quad \alpha \neq \kappa$
Quasi-static, inertia effects, with cross terms	$\mathbf{V}_\alpha = -\frac{1}{\mu_\alpha} \mathbf{K}_\alpha \cdot (\nabla P_\alpha - \rho_\alpha \mathbf{g}) - \mathbf{F}_{\alpha\alpha} \cdot \mathbf{V}_\alpha + \mathbf{K}_{\alpha\kappa} \cdot \mathbf{V}_\kappa - \mathbf{F}_{\alpha\kappa} \cdot \mathbf{V}_\kappa \quad \alpha, \kappa = \beta, \gamma \quad \alpha \neq \kappa$
More dynamic models (transient terms, “pseudo-functions”, ...)	$p_c = \mathcal{F} \left( S_\beta, (\rho_\gamma - \rho_\beta) \mathbf{g}, \nabla P_\beta, \frac{\partial \varepsilon S_\beta}{\partial t}, \dots \right), P_\gamma - P_\beta = p_c - L_1 \frac{\partial \varepsilon S_\beta}{\partial t}, \text{ etc...}$



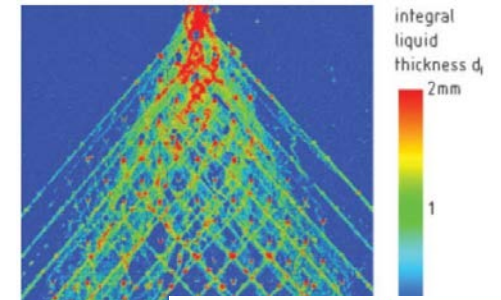
# ...cont.: hybrid models, N-eqs models

PNM with dynamic laws!

Trickle Bed (X-ray, IFP)

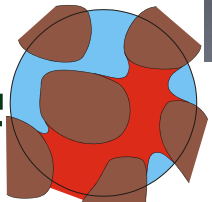
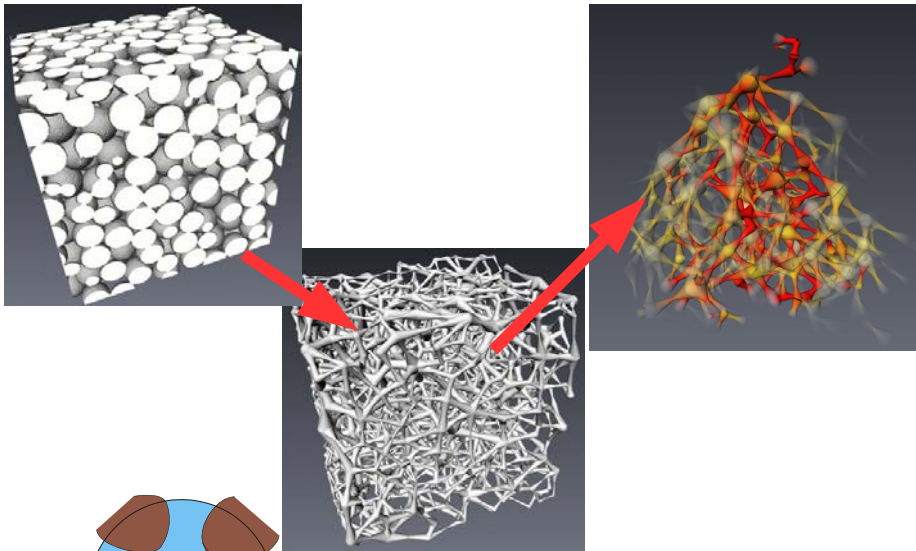
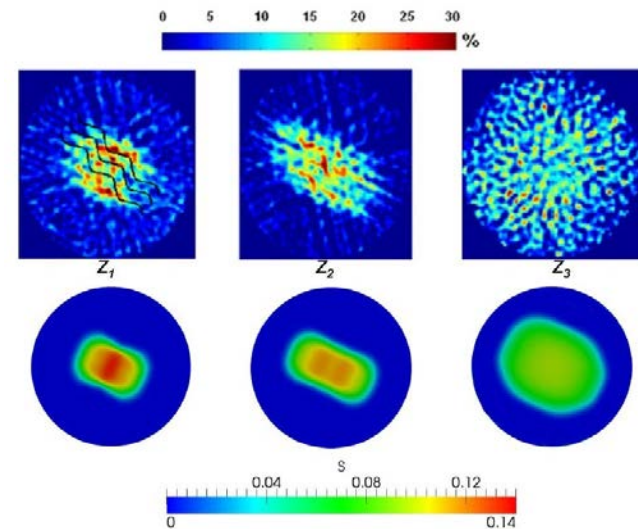
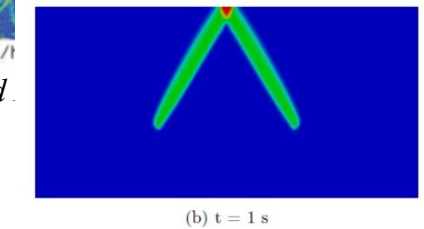


Phase “splitting”  
→ N-eqs

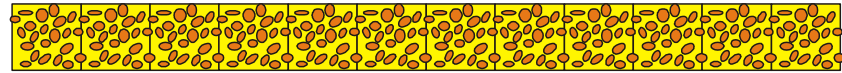


Mahr and

(Soulaire et al., 2014;  
Pasquier, 2018)



# Upscaling Dispersion → various models!



DNS



meso-scale Network model



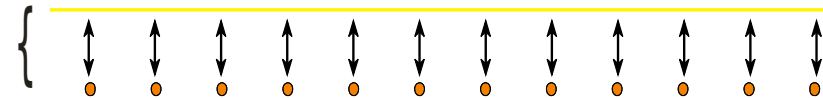
1-eq local equilibrium



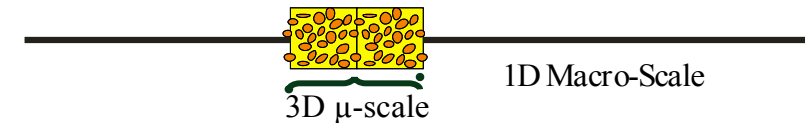
1-eq non-eq: convolution, asympt. 2-eq, frac. deriv., wave eq., CTRW, ...



2-equation, N-equation (multi-rate or MRMT, ...)



Mixed or Hybrid models

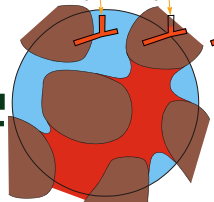


Mixed or Hybrid models for front problems



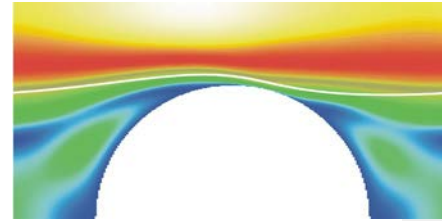
Mixed or Hybrid Network model (PNM+VOF)

Classical dispersion

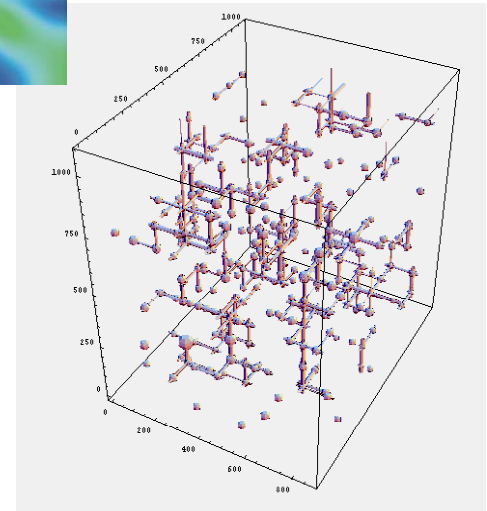


# Active Dispersion: specific aspects

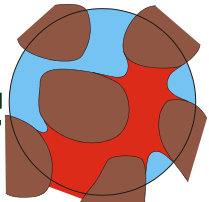
- Impact of n.w
- Tortuosity and dispersion  $\neq$  from passive dispersion
- Effective reaction rate
- Convective correction (“drift”)
- Importance of non-local effects



- 2- $\phi$  VOF, etc..
- PNM
- Large-scale



This talk  $\rightarrow$  mainly trapped phase



# Fully coupled micro-macro model with “n.w” terms

$$\Omega_l = \langle \omega_l \rangle^l = \frac{1}{V_l} \int_{V_l} \omega_l dV$$

+

$$\omega_l = \Omega_l + \tilde{\omega}_l$$

$$\text{I. } \frac{\partial \varepsilon_l \rho_l}{\partial t} + \nabla \cdot (\rho_l \mathbf{V}_l) = -\dot{m}$$

$$\text{II. } \frac{\partial \varepsilon_s \rho_s}{\partial t} = \dot{m}$$

$$\text{III. } \dot{m} = \frac{1}{V} \int_{A_{ls}} \mathbf{n}_{ls} \cdot (-\rho_s \mathbf{w}_{sl}) dA$$

$$\text{IV. } \frac{\partial \varepsilon_l \rho_l \Omega_l}{\partial t} + \nabla \cdot (\varepsilon_l \rho_l \Omega_l \mathbf{U}_l) + \nabla \cdot (\rho_l \langle \tilde{\omega}_l \tilde{\mathbf{v}}_l \rangle) =$$

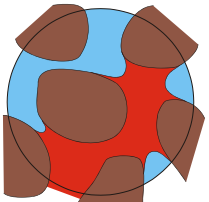
→ dispersion  
→ tortuosity

$$\nabla \cdot \left( \varepsilon_l \rho_l D_l \nabla \Omega_l + \frac{1}{V} \int_{A_{ls}} \mathbf{n}_{ls} \rho_l D_l \tilde{\omega}_l dA + \frac{1}{V} \int_{A_{li}} \mathbf{n}_{li} \rho_l D_l \tilde{\omega}_l dA \right) - \dot{m}_l$$

$$\text{V. } \dot{m}_l = \frac{1}{V} \int_{A_{ls}} \mathbf{n}_{ls} \cdot (\rho_l \omega_l (\mathbf{v}_l - \mathbf{w}_{sl}) - \rho_l D_l \nabla \omega_l) dA = \frac{M_{Ca}}{M_g} \dot{m} = -a_{vl} M_{Ca} k_s \left\langle \left( 1 - \frac{\omega_l}{\omega_{eq}} \right)^n \right\rangle_{ls}$$

$$\text{specific area: } a_{vl} = \frac{1}{V} \int_{A_{ls}} dA$$

(see Soulaire et al., 2011; Guo et al., 2015)



# Macro-scale model and effective parameters (simplified closure, no n.w)

$$\frac{\partial \varepsilon_l \rho_l \Omega_l}{\partial t} + \nabla \cdot (\varepsilon_l \rho_l \Omega_l \mathbf{U}_l + \varepsilon_l \rho_l (\Omega_l - \omega_{eq}) \mathbf{U}_l^*) = \nabla \cdot (\varepsilon_l \rho_l \mathbf{D}_l^* \cdot \nabla \Omega_l) - \dot{m}_l$$

Additional convective terms:

$$\mathbf{U}_l^* = \langle \tilde{\mathbf{v}}_l s_l \rangle^l - \frac{1}{V_l} \int_{A_{lc}} \mathbf{n}_{ls} D_l s_l dA - \frac{1}{V_l} \int_{A_{li}} \mathbf{n}_{ls} D_l s_l dA$$

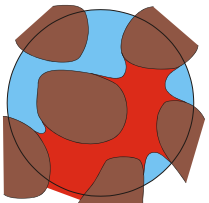
Dispersion tensor  $\neq$  from passive dispersion:  $\mathbf{D}_l^* = D_l \left( \mathbf{I} + \frac{1}{V_l} \int_{A_{ls}} \mathbf{n}_{ls} \mathbf{b}_l dA + \frac{1}{V_l} \int_{A_{li}} \mathbf{n}_{ls} \mathbf{b}_l dA \right) - \langle \tilde{\mathbf{v}}_l \mathbf{b}_l \rangle^l$

Mass exchange term:  $\dot{m}_l = -a_{vl} M_{Ca} \left( 1 - \frac{\Omega_l}{\omega_{eq}} \right)^n k_{s,eff} + a_{vl} M_{Ca} \left( 1 - \frac{\Omega_l}{\omega_{eq}} \right)^{n-1} \mathbf{h}_l^* \cdot \nabla \Omega_l$

$$\text{with } \mathbf{h}_l^* = n \frac{k_s}{\omega_{eq}} \langle \mathbf{b}_l \rangle_{ls}$$

“Effective reaction”:

$$k_{s,eff} = k_s (1 + n \langle s_l \rangle_{ls})$$



# Effective “reaction rate” (ex.: linear reaction rate)

Pore-scale Damkhöler number:

$$Da = \frac{M_{Ca}}{\rho_l \omega_{eq}} \frac{l_r k_s}{D_l}$$

strat. system:  $k_{s,eff}/k_s = \frac{1}{1 + \frac{1}{6}\epsilon_l Da}$

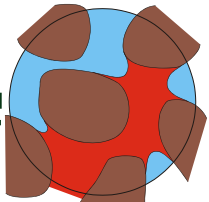
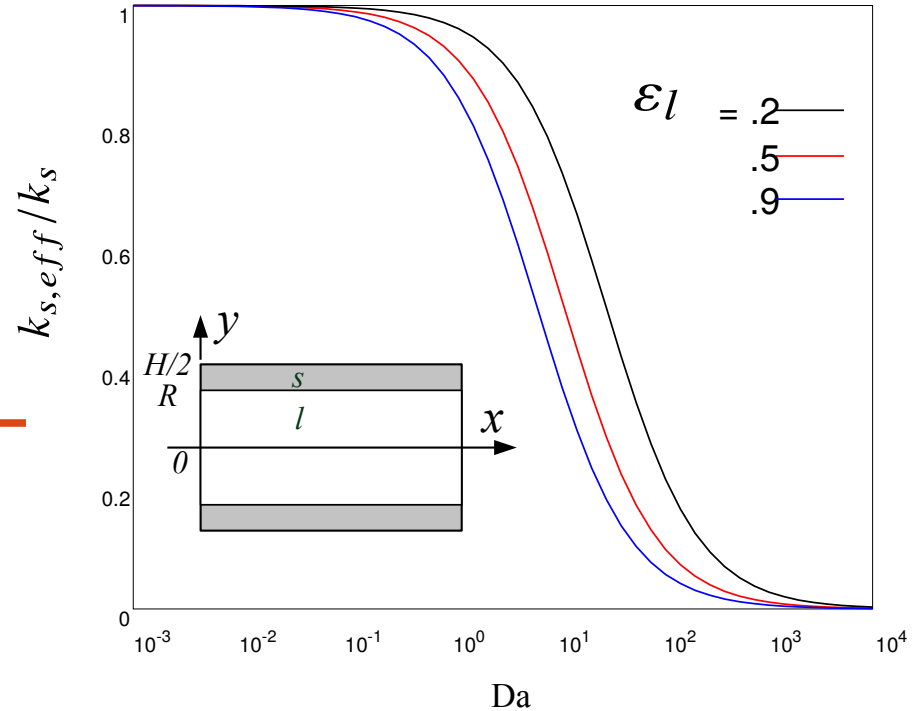
Note: if  $Da \rightarrow \infty$

Purely transport limited =

**Local Non-Equilibrium Model**

$$\dot{m}_l = \omega_{eq} \dot{m} + \rho_l \alpha_l (\Omega_l - \omega_{eq}) + \rho_l \mathbf{h}_l \cdot \nabla \Omega_l$$

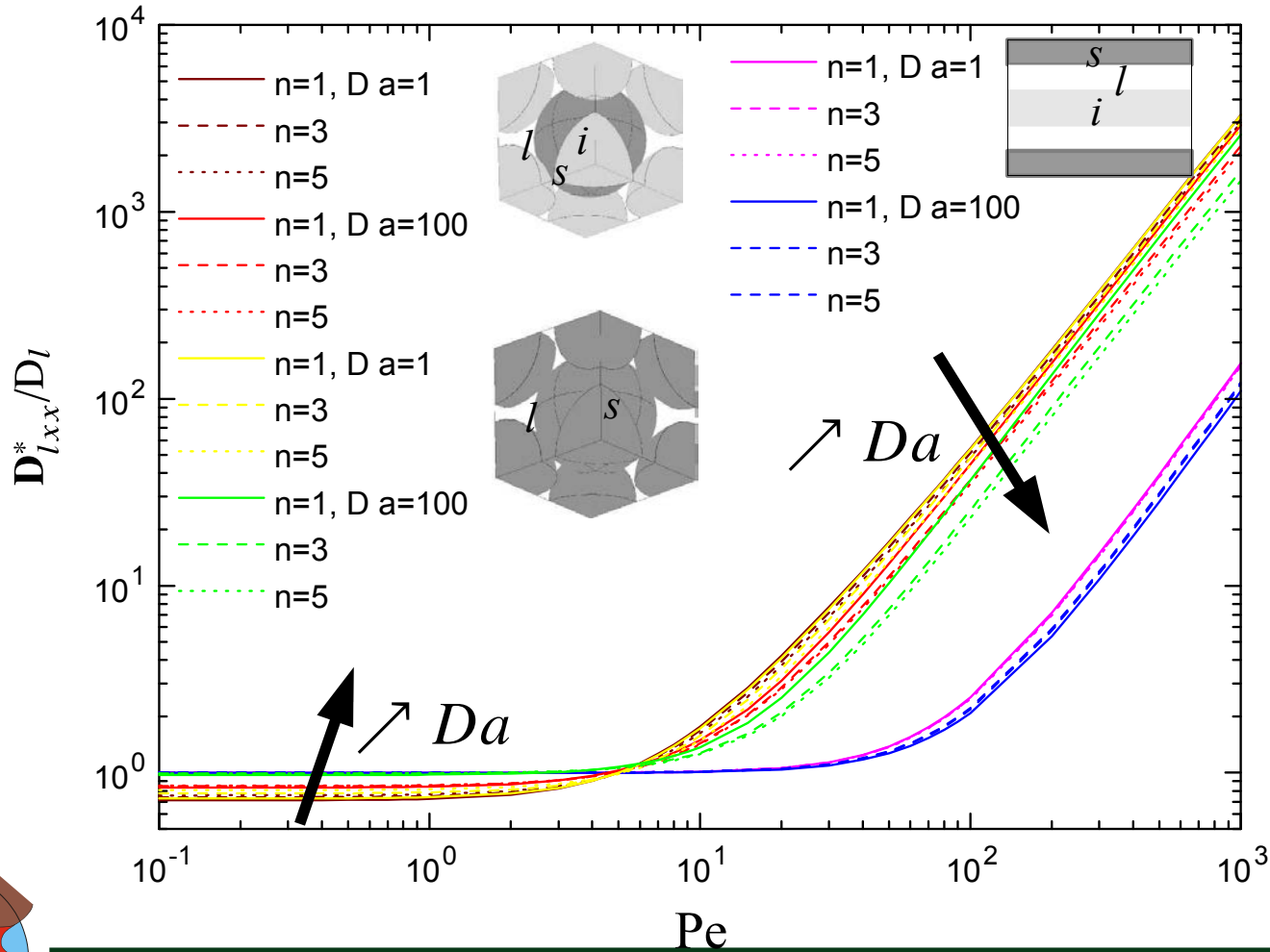
strat. system:  $\frac{\alpha_l H^2}{D_l} = \frac{12}{\epsilon_l}$



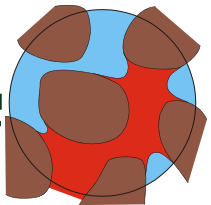
# Dispersion

$Da=0 \rightarrow$  passive case

$Da \rightarrow \infty \rightarrow$  uniform equil. conc. at  $A_{\beta\sigma}$



Guo et al.,  
2015





# Importance of non-local effects and drift

Hyp.:  $Re \sim 0 \rightarrow$  Darcy ,  $Ra=0$

Comparison with 1D averaged model  
Note: need additional “convective” terms

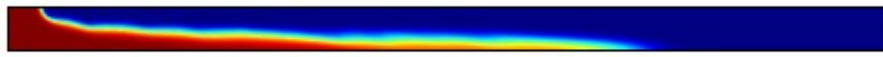
DNS



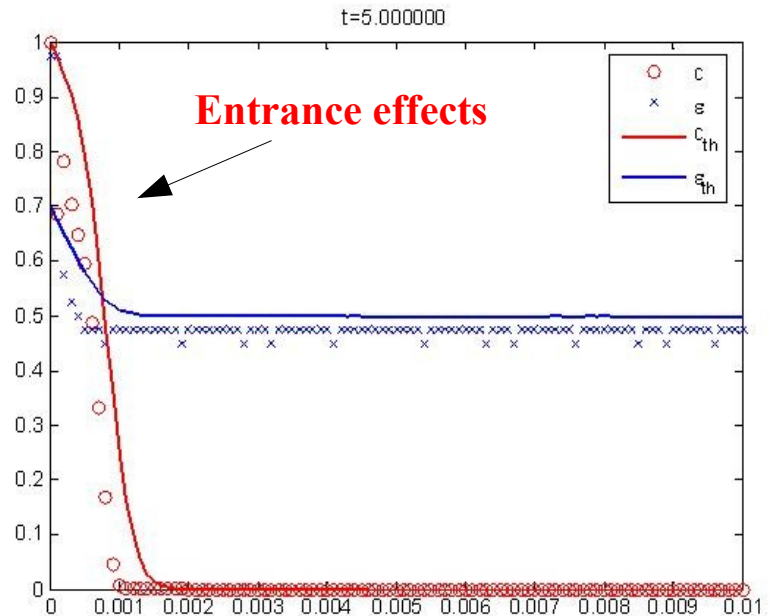
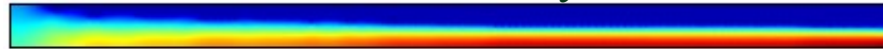
fluid/solid repartition



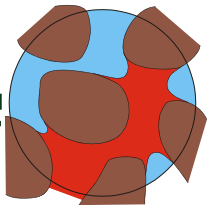
fluid concentration



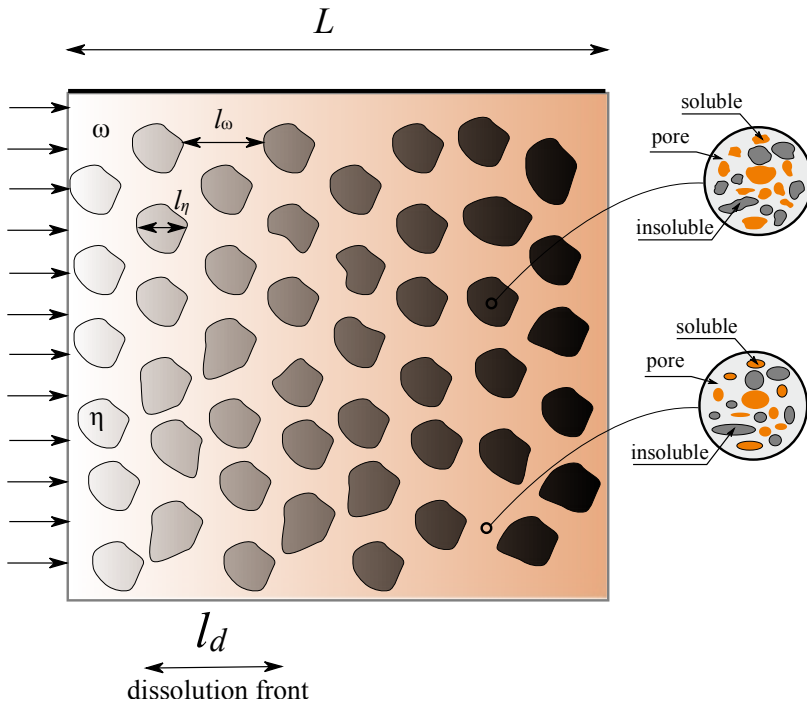
fluid x-velocity



Improvements: use of non local effective parameters...  
...or hybrid formulations!



# Darcy-Scale $\rightarrow$ Large-Scale



$\alpha = s, i, l$

$s$  : soluble phase

$i$  : insoluble material

$l$  : liquid phase (water + dissolved species)

Pore-scale model

1<sup>st</sup> upscaling

Darcy-scale model

DNS

$l_\omega, l_\eta \ll l_d \sim L$

2<sup>nd</sup> upscaling

Large-scale model

?

$l_\alpha \ll l_\omega, l_\eta \ll L$

$l_\omega, l_\eta \ll l_d \ll L$



# Darcy-scale model (ex.: gypsum)

$$\frac{\partial \varepsilon_s}{\partial t'} = \frac{\rho_r}{\rho_s} \frac{M_{salt}}{M_a} \omega_{eq} Da_M \alpha' (\Omega'_l - 1)$$

$$\frac{\partial (\rho'_l \varepsilon_l)}{\partial t'} + Pe_M \nabla' \cdot (\rho'_l \mathbf{V}'_l) = -\frac{M_{salt}}{M_a} \omega_{eq} Da_M \alpha' (\Omega'_l - 1)$$

$$\frac{\partial (\rho'_l \Omega'_l \varepsilon_l)}{\partial t'} + Pe_M \nabla' \cdot (\rho'_l \Omega'_l U_r \mathbf{V}'_l) = \nabla' \cdot (\rho'_l \varepsilon_l \mathbf{D}'_l \cdot \nabla' \Omega'_l) - Da_M \alpha' (\Omega'_l - 1)$$

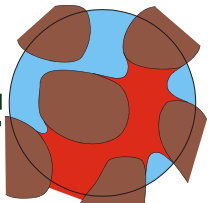
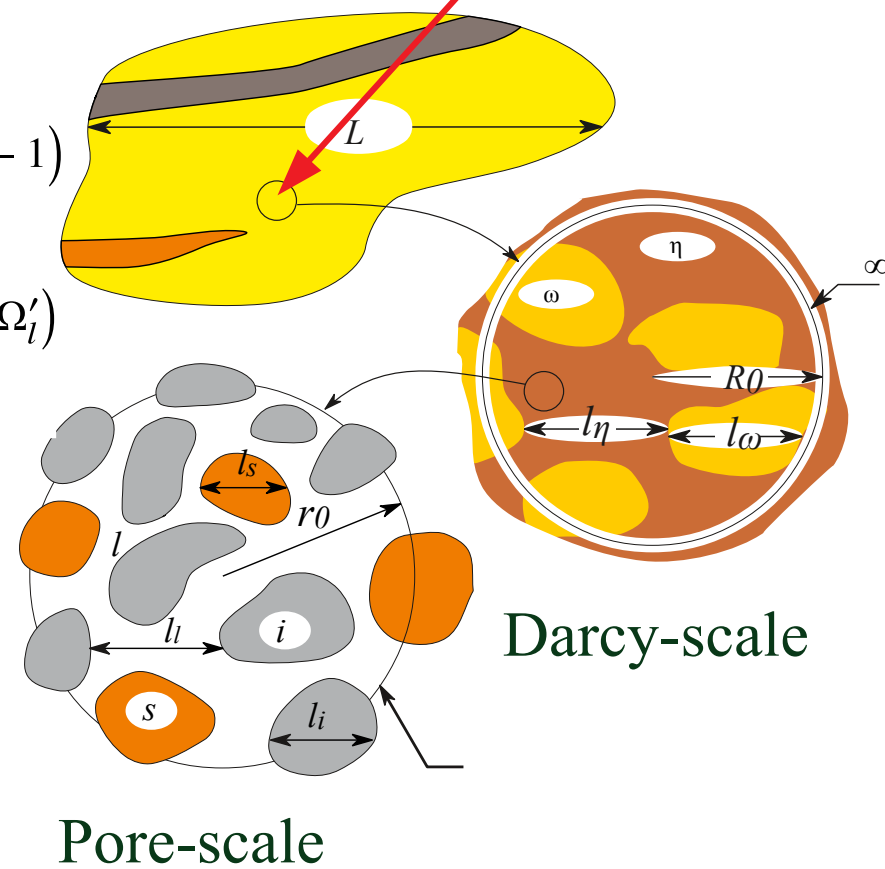
$$0 = -\nabla' P'_l + \frac{1}{Re_M} Gr_M \omega'_l \frac{\mathbf{g}}{\|\mathbf{g}\|} - N_K^{-1} \mu'_l (\mathbf{K}')^{-1} \cdot \mathbf{V}'_l$$

$$Da_M = \frac{\alpha_0 L_r^2}{D_l}$$

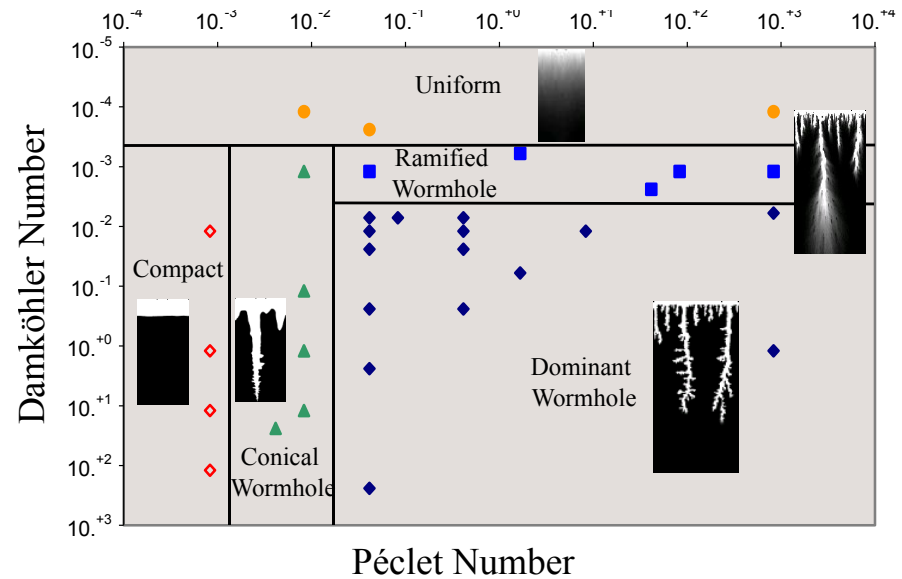
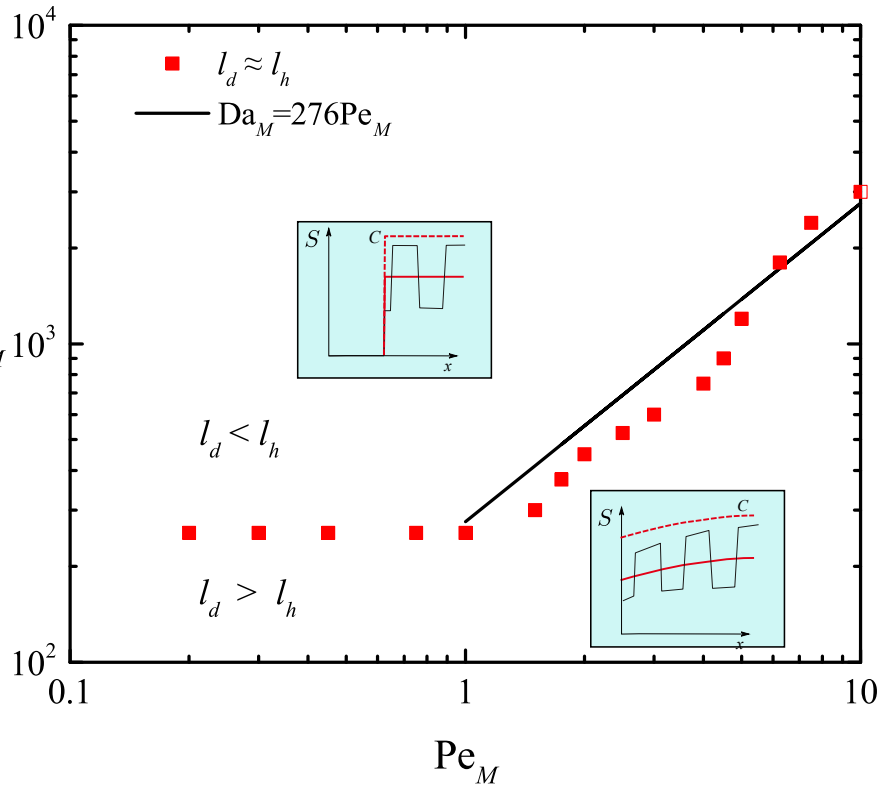
**Damköhler number**

$$\alpha = \alpha_0 \alpha' (\varepsilon_l, \dots)$$

**Large-scale?**



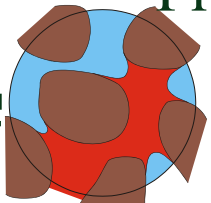
# Dissolution of heterogeneous systems: scale separation?



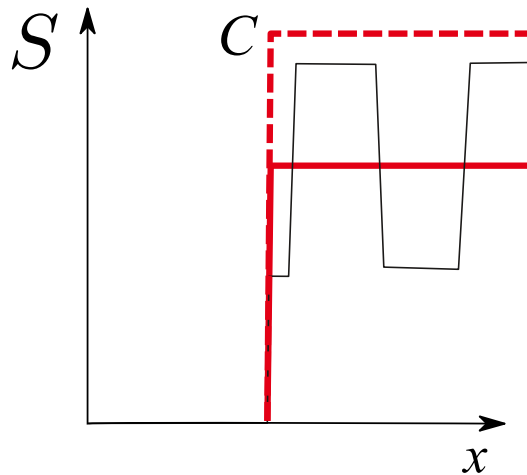
after Golfier et al., 2000

Front Thickness?

Dissolution instabilities?



# Heterogeneous systems: properties of the Darcy-Scale fields (cont.)



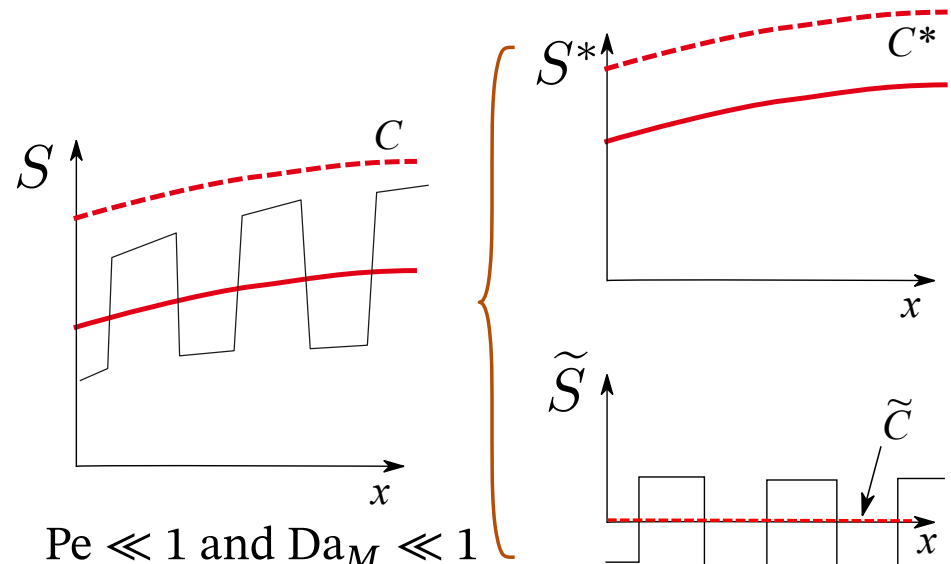
$$Pe \ll 1 \text{ and } Da_M \gg 1$$

or

$$Pe \gg 1 \text{ and } \frac{Da_M}{Pe} \gg 1$$

**local equilibrium dissolution**

→ sharp front



$$Pe \ll 1 \text{ and } Da_M \ll 1$$

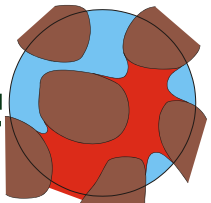
or

$$Pe \gg 1 \text{ and } \frac{Da_M}{Pe} \ll 1$$

**non-local equilibrium dissolution**

→ **diffused front!**

$$l_\omega, l_\eta \ll l_d \ll L$$



# L-S Upscaling of a simple dissolution model, small “Damköhlers” (case $Pe > 1$ )

$$\nabla \cdot (\varepsilon_T (1 - S) \mathbf{U}_l) = 0 \quad \frac{\partial (\varepsilon_T (1 - S) C_l)}{\partial t} + \nabla \cdot (\varepsilon_T (1 - S) \mathbf{U}_l C_l) = -\alpha(S) (C_l - C_{eq})$$

$$\varepsilon_T \frac{\partial S}{\partial t} = \frac{\rho_l}{\rho_s} \alpha(S) (C_l - C_{eq})$$

$$\mathbf{V}_l = \varepsilon_T (1 - S) \mathbf{U}_l = -\frac{1}{\mu_l} \mathbf{K}_l \cdot (\nabla P_l - \rho_l \mathbf{g})$$

$$\mathbf{K}_l = \mathbf{K} k_{rl}(S)$$

## Definitions for large-scale averages:

$$\{\varepsilon_T\} = m * \varepsilon_T \approx \frac{1}{V_\infty} \int_{V_\infty} \varepsilon_T dV$$



$$\{\varepsilon_T S\} = \{\varepsilon_T\} S^*$$

$$\{\varepsilon_T (1 - S) C_l\} = \{\varepsilon_T\} (1 - S^*) C_l^*$$



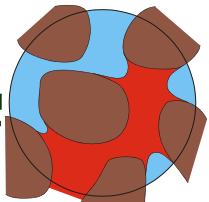
$$\{\varepsilon_T (1 - S) \mathbf{U}_l\} = \{\varepsilon_T\} (1 - S^*) \mathbf{U}_l^*$$

## Deviations:

$$C_l = C_l^* + \tilde{C}_l$$

$$S = S^* + \tilde{S}$$

Small Damköhlers  $\rightarrow$   
 $\tilde{C}_l \approx 0$  over UC!

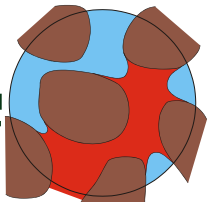


# Coupled Darcy-Scale and Large-Scale problem (case $Pe > 1$ )

$$\begin{array}{l}
 \text{L.-S.} \\
 \left\{ \begin{array}{l}
 \frac{\partial}{\partial t} (\{\varepsilon_T\} (1 - S^*) C_l^*) + \nabla \cdot (\{\varepsilon_T\} (1 - S^*) \mathbf{U}_l^* C_l^*) + \nabla \cdot \{\varepsilon_T (1 - S) \mathbf{U}_l \tilde{C}_l\} = -\alpha^* (C_l^* - C_{eq}) \\
 \frac{\partial}{\partial t} (\{\varepsilon_T\} S^*) = \frac{\rho_l}{\rho_s} \alpha^* (C_l^* - C_{eq})
 \end{array} \right. \\
 \\
 \text{D.-S.} \\
 \left\{ \begin{array}{l}
 \varepsilon_T (1 - S) \frac{\partial \tilde{C}_l}{\partial t} + \varepsilon_T (1 - S) \tilde{\mathbf{U}}_l \cdot \nabla C_l^* + \varepsilon_T (1 - S) \mathbf{U}_l \cdot \nabla \tilde{C}_l = - \left( \alpha - \frac{\varepsilon_T (1 - S) \alpha^*}{\{\varepsilon_T\} (1 - S^*)} \right) (C_l^* - C_{eq}) \\
 \varepsilon_T \tilde{S} = \varepsilon_T \tilde{S}_0 + \frac{\rho_l}{\rho_s} \int_{u=0}^{u=t} \left( \alpha - \frac{\varepsilon_T \alpha^*}{\{\varepsilon_T\}} \right) (C_l^* - C_{eq}) du
 \end{array} \right.
 \end{array}$$

**Dissolution history!**

+ problem for Darcy's law with heterogeneous permeability  
(induced by variation of soluble material saturation!)

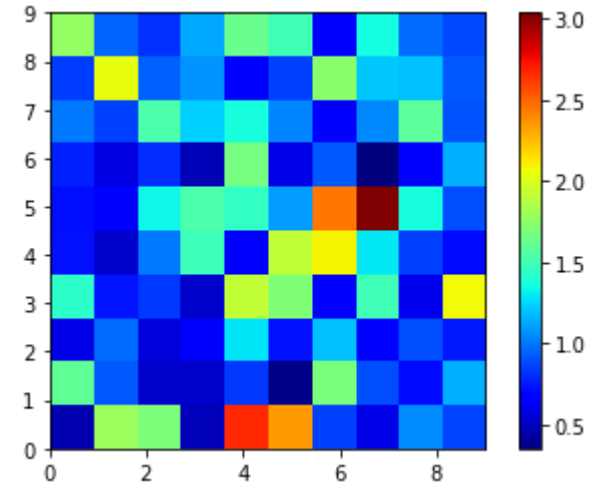


# Large-Scale properties: Preliminary calculations of effective coefficients

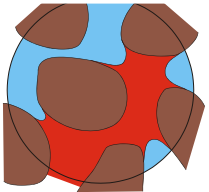
1.  $S^*$ ,  $S$  at  $t$
2. compute  $S$  spatial distribution at  $t + \delta t$ 

$$\frac{\partial S}{\partial t} = \frac{\{\varepsilon_T\} \alpha}{\varepsilon_T \alpha^*} \frac{\partial S^*}{\partial t}$$

key:  $C_l^* - C_{eq}$  constant over REV
3. compute  $\alpha(S)$
4. compute  $\alpha^*(S^*)$
5. estimate  $\mathbf{K}(S)$
6. compute  $\mathbf{K}^*(S^*)$   
see closure Quintard and Whitaker (1987)
7. iterate by going at Step 1

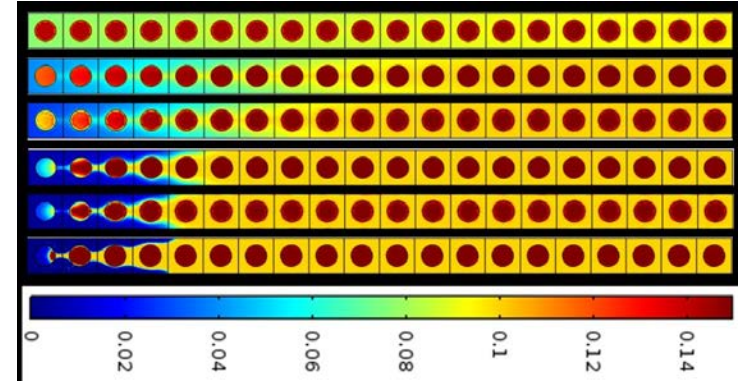
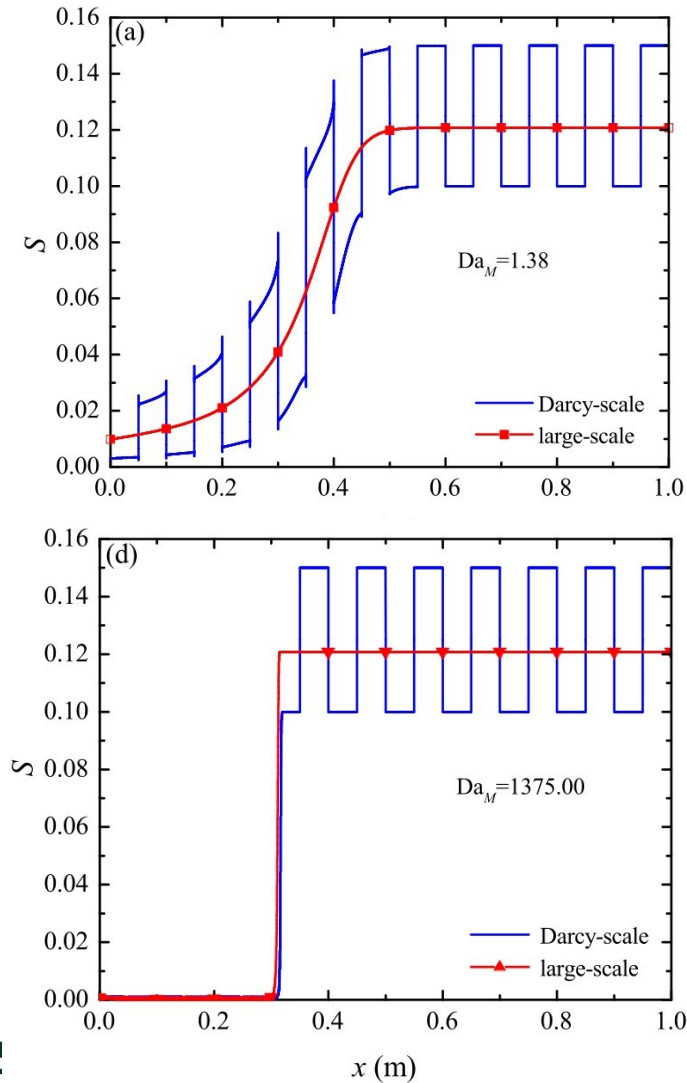


Tools developed  
for spatially  
distributed Darcy-  
scale parameters!

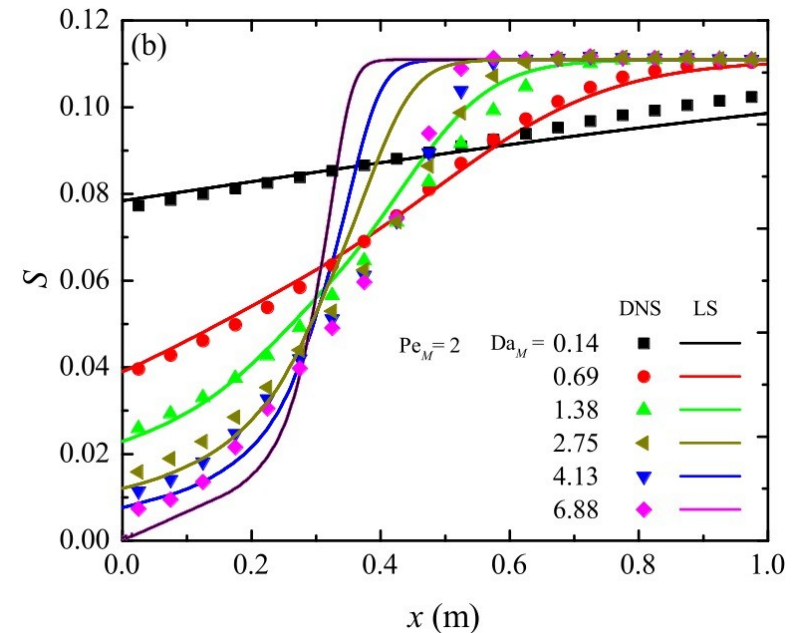




# Comparison DNS $\leftrightarrow$ Theory



**Robust theory!**



# Conclusions

## Mostly about OPEN PROBLEMS!

- **N-phase flow:**
  - Coupling has been marginally studied
  - Classical generalized Darcy's law mostly used. What about models with cross terms, dynamic models, etc...?
- **Multicomponent, reactive**
  - Complex chemistry and/or multicomponent thermodynamics
  - Instabilities
- **Sequential upscaling:**
  - Limited homogenization results for low Da numbers
  - Large Da?
- **History and memory effects**
- **Coupling with heat transfer (combustion, pyrolysis), geomechanics, ...**

