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Sequential upscaling of multiphase dispersion in porous media

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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...

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

Karsts

Chal engng

Mahr and Mewes (2007)
Generic Problems: 2-phase flow

\[ \nabla \cdot \mathbf{v}_\alpha = 0 \]

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

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

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

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

B.C. 4 \( \mathbf{n}_\gamma \breve{\beta} \cdot (\rho_\gamma \mathbf{v}_\gamma (\mathbf{v}_\gamma - \mathbf{w}_\gamma \beta) + p_\gamma \mathbf{I} - \tau_\gamma) = \mathbf{n}_\gamma \breve{\beta} \cdot (\rho_\beta \mathbf{v}_\beta (\mathbf{v}_\beta - \mathbf{w}_\gamma \beta) + p_\beta \mathbf{I} - \tau_\beta) - 2H_{\gamma \beta} \sigma_{\gamma \beta} \mathbf{n}_\gamma \breve{\beta} \)
Generic Problems: Reactive transport

\[
\frac{\partial \rho_\beta \omega_\beta}{\partial t} + \nabla \cdot (\rho_\beta \omega_\beta \mathbf{v}_\beta) = \nabla \cdot (\rho_\beta D_\beta \nabla \omega_\beta) + r_\beta \quad \text{in } V_\beta
\]

Homogeneous reaction

Heterogeneous reaction:

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

Local Equilibrium:

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

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

<table>
<thead>
<tr>
<th>Model</th>
<th>PDEs</th>
</tr>
</thead>
<tbody>
<tr>
<td>Quasi-static, heuristic (Muskat)</td>
<td>$\frac{d\varepsilon S_\alpha}{dt} + \nabla \cdot \mathbf{V}<em>\alpha = 0$ ; $\mathbf{V}</em>\alpha = -\frac{1}{\mu_\alpha} \mathbf{K}<em>\alpha \cdot (\nabla P</em>\alpha - \rho_\alpha \mathbf{g})$ $\alpha = \beta, \gamma$</td>
</tr>
<tr>
<td>Quasi-static, low Re, with cross terms</td>
<td>$\mathbf{V}<em>\alpha = -\frac{1}{\mu</em>\alpha} \mathbf{K}<em>\alpha \cdot (\nabla P</em>\alpha - \rho_\alpha \mathbf{g}) + \mathbf{K}<em>{\alpha \kappa} \cdot \mathbf{V}</em>\kappa$ $\alpha, \kappa = \beta, \gamma$ $\alpha \neq \kappa$</td>
</tr>
<tr>
<td>Quasi-static, inertia effects, with cross terms</td>
<td>$\mathbf{V}<em>\alpha = -\frac{1}{\mu</em>\alpha} \mathbf{K}<em>\alpha \cdot (\nabla P</em>\alpha - \rho_\alpha \mathbf{g}) - \mathbf{F}<em>{\alpha \alpha} \cdot \mathbf{V}</em>\alpha$ $\alpha, \kappa = \beta, \gamma$ $\alpha \neq \kappa$</td>
</tr>
<tr>
<td>More dynamic models (transient terms, “pseudo-functions”, ...)</td>
<td>$p_c = \mathcal{F} \left( S_\beta, (\rho_\gamma - \rho_\beta) \mathbf{g}, \nabla P_\beta, \frac{\partial \varepsilon S_\beta}{\partial t}, ... \right)$, $P_\gamma - P_\beta = p_c - L_1 \frac{\partial \varepsilon S_\beta}{\partial t}$, etc...</td>
</tr>
</tbody>
</table>
...cont.: hybrid models, N-eqs

models

PNM with dynamic laws!

Phase “splitting” → N-eqs

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

Trickle Bed (X-ray, IFP)

Mahr and Mewes (2007)
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)
Active Dispersion: specific aspects

- Impact of n.w
- Tortuosity and dispersion ≠ from passive dispersion
- Effective reaction rate
- Convective correction ("drift")
- Importance of non-local effects

This talk → mainly trapped phase
Fully coupled micro-macro model with “n.w” terms

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

II. \( \frac{\partial \varepsilon_s \rho_s}{\partial t} = \dot{m} \)

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

IV. \( \frac{\partial \varepsilon_l \rho_l \Omega_l}{\partial t} + \nabla \cdot (\varepsilon_l \rho_l \Omega_l \mathbf{U}_l) + \nabla \cdot (\rho_l \langle \tilde{\omega}_l \mathbf{v}_l \rangle) = \nabla \cdot \left( \varepsilon_l \rho_l D_l \nabla \Omega_l + \frac{1}{V} \int_{A_{ls}} \mathbf{n}_{ls} \rho_l D_l \tilde{\omega}_l \, dA + \frac{1}{V} \int_{A_{li}} \mathbf{n}_{li} \rho_l D_l \tilde{\omega}_l \, dA \right) - \dot{m}_l \)

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

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

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

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

\[
\frac{\partial \varepsilon l \rho l \Omega l}{\partial t} + \nabla \cdot \left( \varepsilon l \rho l \Omega l U l + \varepsilon l \rho l \left( \Omega l - \omega_{eq} \right) U^*_l \right) = \nabla \cdot \left( \varepsilon l \rho l D^*_l \cdot \nabla \Omega l \right) - m_l
\]

Additional convective terms:

\[ U^*_l = \langle \bar{v}_l s_l \rangle^l - \frac{1}{V_l} \int_{A_{ls}} n_{ls} D_l s_l dA - \frac{1}{V_l} \int_{A_{nl}} n_{ls} D_l s_l dA \]

Dispersion tensor \( \neq \) from passive dispersion:

\[ D^*_l = D_l \left( I + \frac{1}{V_l} \int_{A_{ls}} n_{ls} b_l dA + \frac{1}{V_l} \int_{A_{nl}} n_{ls} b_l dA \right) - \langle \bar{v}_l b_l \rangle^l \]

Mass exchange term:

\[ m_l = -a_{vl} M_{Ca} \left( 1 - \frac{\Omega l}{\omega_{eq}} \right)^n k_{s, eff} + a_{vl} M_{Ca} \left( 1 - \frac{\Omega l}{\omega_{eq}} \right)^{n-1} h^*_l \cdot \nabla \Omega l \]

"Effective reaction":

\[ k_{s, eff} = k_s \left( 1 + n \langle s_l \rangle_{ls} \right) \]
**Effective “reaction rate” (ex.: linear reaction rate)**

*Pore-scale* Damköhler number:

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

strat. system: \( k_{s,eff}/k_s = \frac{1}{1 + \frac{1}{6} \varepsilon_l Da} \)

**Note:** if \( Da \to \infty \)

Purely transport limited = *Local Non-Equilibrium Model*

\[
\dot{m}_l = \omega_{eq} \dot{m} + \rho_l \alpha_l (\Omega_l - \omega_{eq}) + \rho_l h_l \cdot \nabla \Omega_l
\]

strat. system: \( \frac{\alpha_l H^2}{D_l} = \frac{12}{\varepsilon_l} \)
**Dispersion**

Da = 0 → passive case
Da → ∞ → 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

Entrance effects

Improved: use of non-local effective parameters...
...or hybrid formulations!
Darcy-Scale → Large-Scale

$\omega \quad l_\omega$

$\eta \quad l_\eta$

$d$ dissolution front

$\alpha = s, i, l$

$s$: soluble phase

$i$: insoluble material

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

Pore-scale model

$1^{st}$ upscaling

$\omega, \eta \ll d \sim L$

DNS

Darcy-scale model

$2^{nd}$ upscaling

$\omega, \eta \ll d \ll L$

Large-scale model

？
Darcy-scale model (ex.: gypsum)

\[
\frac{\partial \varepsilon_s}{\partial t'} = \frac{\rho_r}{\rho_s} \frac{M_{salt}}{M_a} \omega_{eq} D_{M} \alpha' (\Omega'_l - 1)
\]

\[
\frac{\partial \left( \rho'_l \varepsilon_l \right)}{\partial t'} + Pe_M \nabla' \cdot \left( \rho'_l \nabla' \right) = -\frac{M_{salt}}{M_a} \omega_{eq} D_{M} \alpha' (\Omega'_l - 1)
\]

\[
\frac{\partial \left( \rho'_l \Omega'_l \varepsilon_l \right)}{\partial t'} + Pe_M \nabla' \cdot \left( \rho'_l \Omega'_l U_r \nabla' \right) = \nabla' \cdot \left( \rho'_l \varepsilon_l D_{l}^{*} \cdot \nabla' \Omega'_l \right) - D_{M} \alpha' (\Omega'_l - 1)
\]

\[
0 = -\nabla' P'_l + \frac{1}{Re_M} Gr_M \omega'_l \frac{g}{\|g\|} - N^{-1} \mu'_l (K')^{-1} . V'_l
\]

\[
D_{M} = \frac{\alpha_0 L_r^2}{D_l} \quad \text{Damköhler number}
\]

\[
\alpha = \alpha_0 \alpha' (\varepsilon_l, \ldots)
\]
Dissolution of heterogeneous systems: scale separation?

\[ \text{Péclet Number} \]

Front Thickness?

Dissolution instabilities?

after Golfier et al., 2000

\[ l_d \approx l_h \]

\[ Da_M = 276 Pe_M \]

\[ l_d < l_h \]

\[ l_d > l_h \]
Heterogeneous systems: properties of the Darcy-Scale fields (cont.)

Local equilibrium dissolution → sharp front

- \( \text{Pe} \ll 1 \) and \( \text{Da}_M \ll 1 \)
- \( \text{Pe} \gg 1 \) and \( \frac{\text{Da}_M}{\text{Pe}} \gg 1 \)

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)

\[ \nabla \cdot (\varepsilon_T (1 - S) \mathbf{U}_l) = 0 \]

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

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

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

\[ \mathbf{K}_l = K kr_l(S) \]

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 \( \rightarrow \)

\[ \tilde{C}_l \approx 0 \text{ over UC!} \]
Coupled Darcy-Scale and Large-Scale problem (case $Pe > 1$)

\[
\frac{\partial}{\partial t} \left\{ \begin{array}{c}
\varepsilon_T (1 - S^*) \, C_l^* \\
\{ \varepsilon_T \} (1 - S^*) \, U_l \, C_l^* \\
\end{array} \right\} + \nabla \cdot \left\{ \begin{array}{c}
\varepsilon_T (1 - S^*) \, U_l \, \tilde{C}_l \\
\{ \varepsilon_T \} (1 - S) \, U_l \, \tilde{C}_l \\
\end{array} \right\} = -\alpha^* \left( C_l^* - C_{eq} \right)
\]

\[
\frac{\partial}{\partial t} \{ \varepsilon_T \} \, S^* = \frac{\rho_l}{\rho_s} \alpha^* \left( C_l^* - C_{eq} \right)
\]

\[
\varepsilon_T (1 - S) \frac{\partial \tilde{C}_l}{\partial t} + \varepsilon_T (1 - S) U_l \cdot \nabla C_l^* + \varepsilon_T (1 - S) U_l \cdot \nabla \tilde{C}_l = - \left( \alpha - \frac{\varepsilon_T (1 - S) \alpha^*}{\{ \varepsilon_T \} (1 - S^*)} \right) \left( C_l^* - C_{eq} \right)
\]

\[
\varepsilon_T \tilde{S} = \varepsilon_T \tilde{S}_0 + \frac{\rho_l}{\rho_s} \int_{u=0}^{u=t} \left( \alpha - \frac{\varepsilon_T \alpha^*}{\{ \varepsilon_T \}} \right) \left( C_l^* - C_{eq} \right) \, du
\]

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 $K(S)$

6. compute $K^*(S^*)$

   see closure Quintard and Whitaker (1987)

7. iterate by going at Step 1

Tools developed for spatially distributed Darcy-scale parameters!
Comparison DNS ↔ 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, ...