Experimental measurements and multi-scale modelling of the relative gas permeability of a clay-rich tight carbonate

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Outline

Industrial context

Experimental characterization

Multi-scale modelling

Conclusion
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Caprock for underground gas storage

- Studied caprock: clay-rich tight carbonate

- Expected confinement properties
  - static barrier: threshold capillary pressure
  - dynamic barrier: low permeability
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Two porosity measures: water and gas (Argon)

- with water:
  \[ \phi = \frac{\text{wet mass} - \text{dry mass}}{\text{volume} \times \text{water density}} \]
  
  vacum pump 24h

- with gas:
  \[ \phi V = \frac{P_c}{P_t} V_t \]

Measured porosities \( \phi(\%) \) at various confining pressures:

<table>
<thead>
<tr>
<th>sample</th>
<th>water, 0 MPa</th>
<th>gas, 2 MPa</th>
<th>gas, 9 MPa</th>
<th>gas, 2 MPa</th>
</tr>
</thead>
<tbody>
<tr>
<td>MIL-13</td>
<td>9.6</td>
<td>11.8</td>
<td>10.7</td>
<td>11.5</td>
</tr>
<tr>
<td>MIL-14</td>
<td>8.9</td>
<td>12.4</td>
<td>11.7</td>
<td>12.2</td>
</tr>
<tr>
<td>MIL-22</td>
<td>10.2</td>
<td>12.7</td>
<td>11.8</td>
<td>12.5</td>
</tr>
<tr>
<td>MIL-23</td>
<td>9.0</td>
<td>11.7</td>
<td>10.8</td>
<td>11.5</td>
</tr>
<tr>
<td>MIL-24</td>
<td>9.1</td>
<td>11.8</td>
<td>11.1</td>
<td>11.8</td>
</tr>
</tbody>
</table>
Water sorption/desorption isotherms

Capillary pressure

\[ P_{\text{cap}} = P_{\text{gaz}} - P_{\text{liq}} \]

- Kelvin’s law
  \[ P_{\text{cap}} \propto - \ln RH \]
  \( RH = \text{relative humidity} \)

- Laplace’s law
  \[ P_{\text{cap}} \propto \frac{1}{e} \]
  \( e = \text{pore size} \)
Capillary pressure curve - pore size distribution
Capillary pressure curve - pore size distribution

Kelvin-Laplace’s flat pore aperture $e$ (nm)

Water saturation $S_w$

$e_{min} = 0.62$

$e_{max} = 368$

Desorption

Sorption

Log-uniform law fit
Gas permeability measurements

- Steady-state Argon permeameter

\[ P_{\text{out}} = P_{\text{atm}} \]

\[ q_{\text{out}} \]

- Dry samples, in-situ confining pressure:
  - Effective gas permeability:
    \[ K^{\text{eff}} = 1.25 \pm 0.15 \times 10^{-18} \text{m}^2 \]
    at \( P_{\text{in}} = 1 \text{ MPa} \)
  - Intrinsic permeability:
    \[ K^{\text{int}} = 2.4 \pm 1.0 \times 10^{-19} \text{m}^2 \]

with Klinkenberg’s law
\[ K^{\text{eff}} = K^{\text{int}} \left( 1 + \frac{\beta}{P_{\text{m}}} \right) \]

- Klinkenberg’s coefficient:
  \( \beta \) from 1.0 to 3.4 MPa
  \[ \rightarrow \text{pore size } e \approx 15 \text{ to } 30 \text{ nm} \]
  (slip flow from kinetic theory of gas)
Gas relative permeability

Confining pressure = 2 MPa

![Graph showing gas relative permeability vs. water saturation for different MILs with markers for MIL-12, MIL-13, MIL-14, MIL-22, MIL-23, and MIL-24.]
Gas relative permeability

Confining pressure $= 9$ MPa ($\approx$ as in-situ)

![Graph showing relative gas permeability vs. water saturation for different MIL materials](image)
Gas relative permeability

Confining pressure = 9 MPa (≈ as in-situ)

- Capillary breakthrough pressure:
  - sample MIL-14: 5.8 MPa
  - sample HAU-1: 2.9 MPa

- Effective gas permeability at breakthrough:
  - sample MIL-14: $8.4 \times 10^{-23} \text{m}^2$
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Permeability upscaling

- **Two scales of flow description**
  - Engineer scale: Darcy law
  - Pore scale: Stokes equations

- **Ideally**: solve Stokes equations on a 3D representative volume element of this material...

... out of reach! ⇒ simplification
Equivalent heterogeneous Darcy medium - dry case

- Actual microstructure
  - canaliculi
  - clay particles
  - carbonate aggregate
  - high porosity region

- Simplified microstructure
  - grain
  - interface
  - high porosity region
  - Interface dominated flow: $e \ll R$

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Equivalent heterogeneous Darcy medium - dry case

- Simplified microstructure

- Interface dominated flow: $e \ll R$

- Interface equivalent permeability $k^{\text{int}} = \frac{e^2}{12}$ (Poiseuille flow)

$\Rightarrow$ grain + interface $\Leftrightarrow$ homogeneous medium with $k^{\text{eq}} = \frac{e^2}{12} \times \frac{e}{R}$
Equivalent heterogeneous Darcy medium - wet case

- Log-uniform interface aperture distribution

\[ e^* = \frac{2\gamma}{P_{\text{cap}}} \] = critical interface aperture

\[ s = \text{fraction of water saturated interfaces} \]

\[ S_w = \text{water saturation} \]

\[ f = \text{fraction of high porosity regions} \]

- Involved Eshelby problems

\[ \begin{align*}
  \text{gas:} & \quad \mathbf{p} = \nabla P_0 \cdot \mathbf{z} \\
  \text{water:} & \quad k = 0 \\
  \text{high porosity regions:} & \quad k \gg k_{\text{hom}} \\
  \text{grains:} & \quad k = 0 \\
  \text{interfaces:} & \quad k_{\text{hom}} \gg k \\
  \text{gas, water:} & \quad \bar{e} > e^* \\
  \text{gas, water:} & \quad \bar{e} < e^*
\end{align*} \]

- Homogenized gas permeability

\[ K_{\text{hom}} = \frac{e_{\text{med}}^3}{24 R} \left( \frac{\rho_{1-f}}{1-f} \right)^{\frac{3}{2}} \left( \frac{1-3f(1-S_w)}{1-f} \right) + \frac{3}{2} (2s-1) \]

with

\[ \begin{cases}
  e_{\text{med}} = \sqrt{e_{\text{min}} e_{\text{max}}} \\
  \rho = e_{\text{max}} / e_{\text{min}}
\end{cases} \]
Comparison to experimental data

![Graph showing relative gas permeability vs. water saturation for different MIL samples.]

- Percolation threshold at critical water saturation $S_{wc}$
- With $s = S_w$ and $f = \phi = 11\%$

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- Experimental characterization on caprocks has evidenced:
  - a static barrier due to capillary breakthrough pressure
  - a dynamic barrier due to very low effective gas permeability

- Multi-scale modelling allows to retrieve:
  - the general trend of relative gas permeability curve
  - a critical water saturation for breakthrough

Thank you for your attention