Gas-solid Interaction in ECBM

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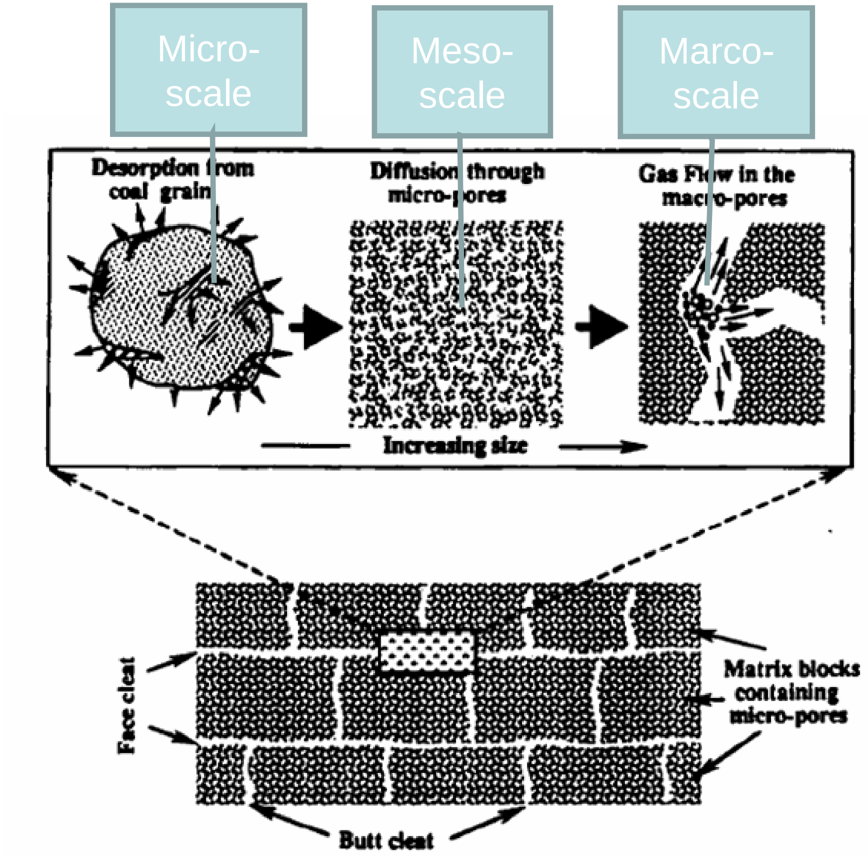
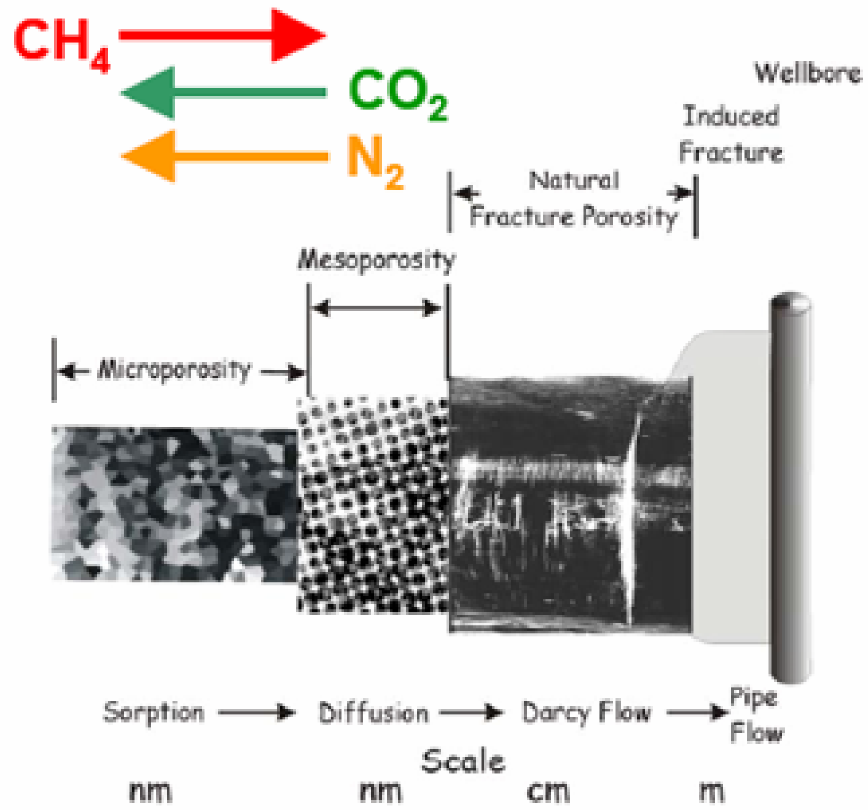
Outline

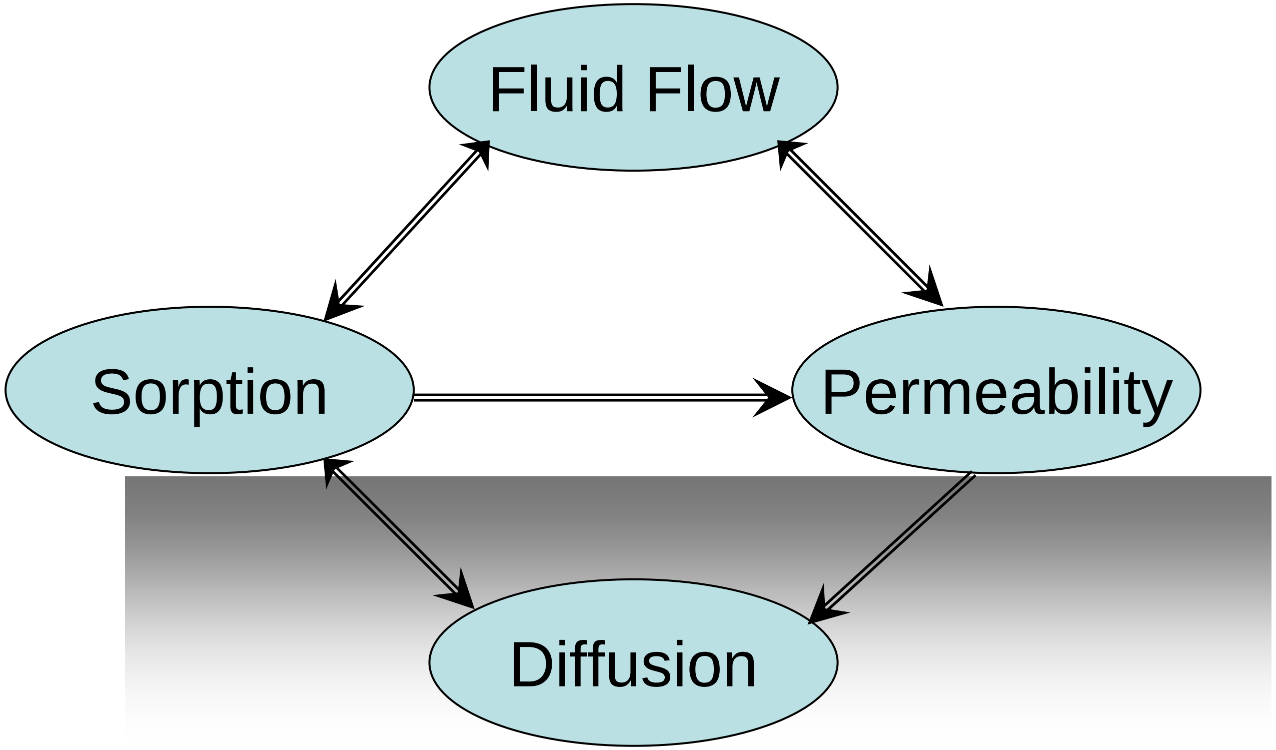
- Overall process
  - Gas transport in coal
  - Multi-scale and multi-component coupling
- Gas sorption and diffusion in coal
  - Measurements of gas adsorption on coals
  - Gas adsorption/desorption in coal
  - Gas diffusion - sorption kinetics
  - Transport properties of coal and their roles
- Sorption-induced swelling/shrinkage of coal
  - Measurement of sorption-induced stain in coal
  - Improved modeling with the sorption-induced stain data
  - Influence on CBM/ECBM process
- Supercritical CO_2 and coal
  - Reaction of ScCO_2 with coal
  - Structural evolution and element migration in coal with ScCO_2

Gas transport in coal





Overall ECBM process



CBM/ECBM deals with coupling processes in a multi-component system (gas) under multi-scale porous media (solid).

Coupling processes in CBM/ECBM

- Key transport behaviors include:
  - Gas sorption equilibrium
  - Gas diffusion
  - Gas flow
  - Dynamic permeability
- Interactions:
  - Initially receives the pressure drop determined by the initial reservoir pressure and wellbore pressure
  - Forms the pressure fields necessary for desorption of gas from coal.
  - Desorbed gas driven by gas concentration gradient enters cleats mainly through surface diffusion in microspores and Knudsen and molecular diffusion in meso-macropores, forming the gas flow in coal.
  - The gas flow alter the pressure fields and hence the adsorption/desorption equilibrium which will further affect gas diffusion.
  - Meanwhile, the gas flow would be dynamically changed as permeability changes due to the sorption-induced matrix shrinkage/swelling in coal.
  - The latter will alter pore structure to some extent and hence the gas diffusion in coal.

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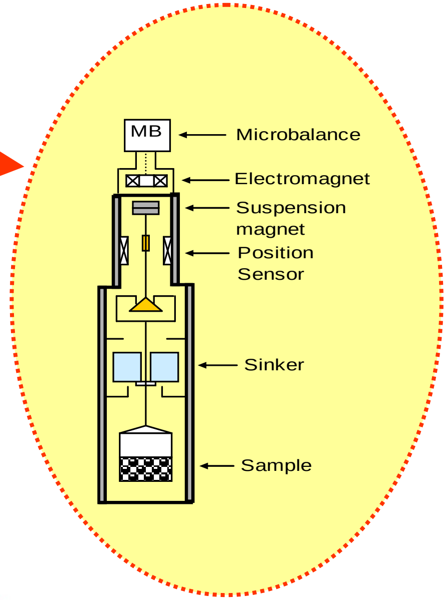
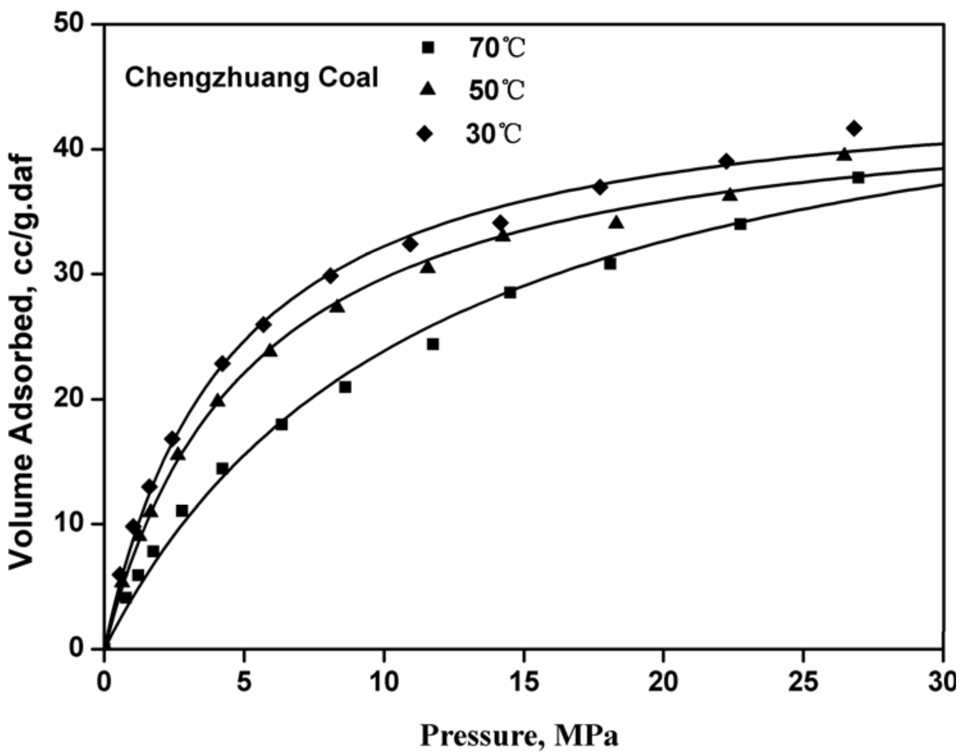
Measurements of gas adsorption on coals

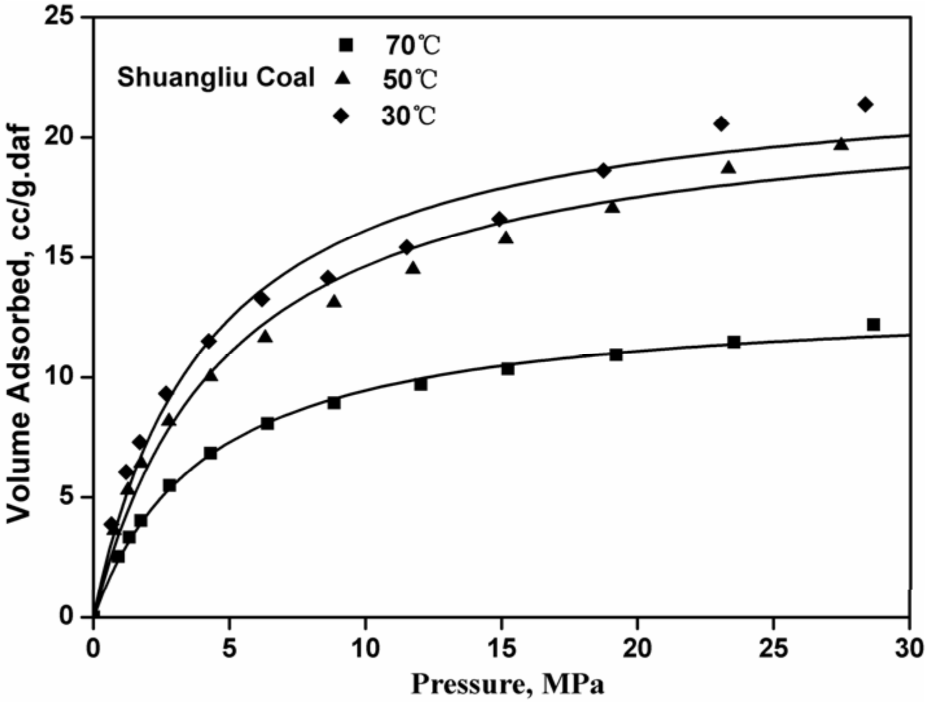
- Effective probes to characterize coal samples: PSD, SSA,…
- Representative coal particle size: crushing, grinding,…
- Proper experimental setups: volumetric, gravimetric,…LP, HP,…
- Analysis: Equilibrium and Kinetics on the basis of dry or ME



HP CH_4 adsorption isotherms

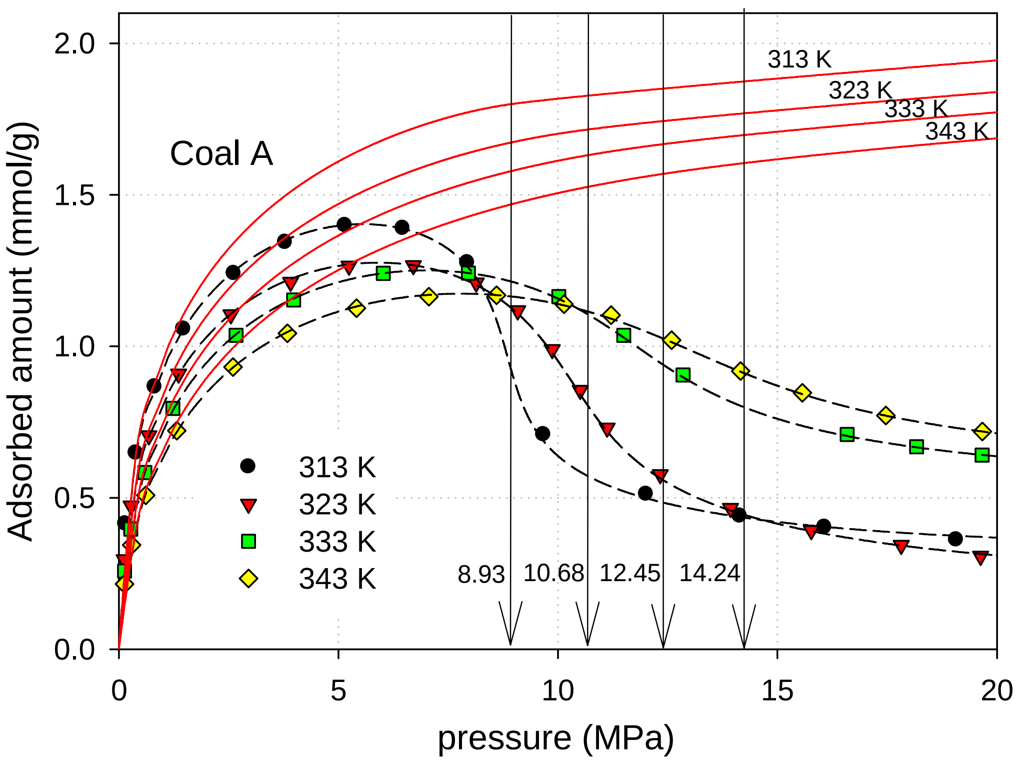
HP sorption means adsorption/desorption at higher pressures (>20 MPa)





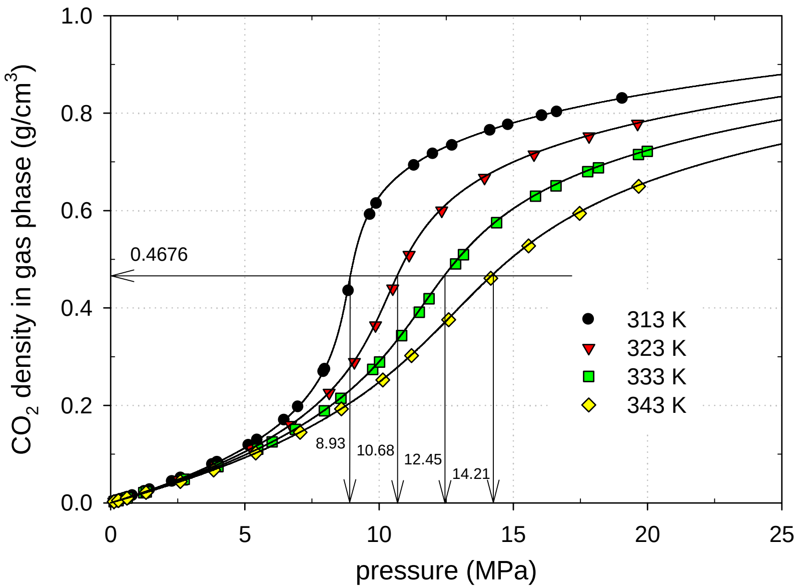
HP CO_2 adsorption isotherms

High-pressure CO_2 isotherms on coal A



$$N_{exe} = (\rho_{adsorbed} - \rho_{gas}) \cdot V_{pore}$$

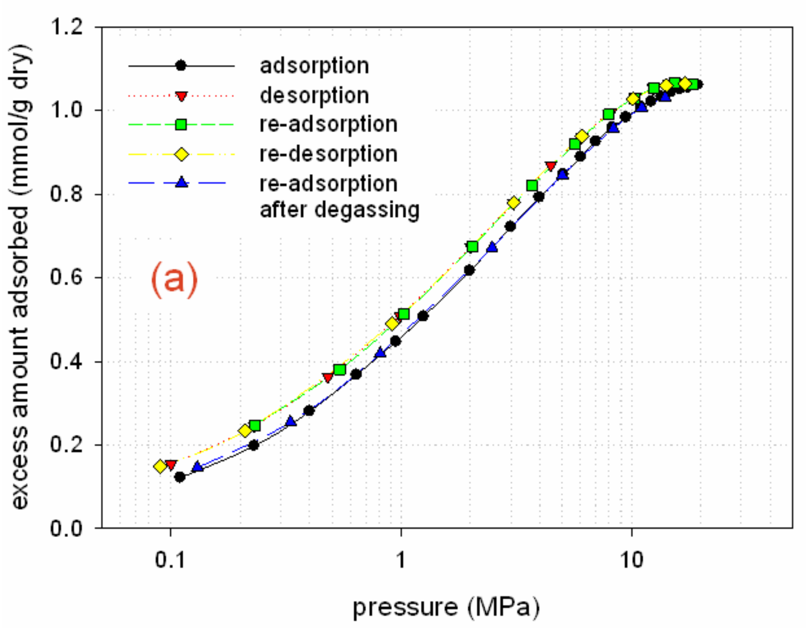
$$\frac{dN_{exe}}{dP} = \left(\frac{d\rho_{adsorbed}}{dP} - \frac{d\rho_{gas}}{dP} \right) \cdot V_{pore}$$



Ref.: J.-S. Bae and S. Bhatia (2006): High-Pressure Adsorption of Methane and Carbon Dioxide on Coal. Energy Fuels 20 (6), 2599–2607.

CO_2 density against gas pressure

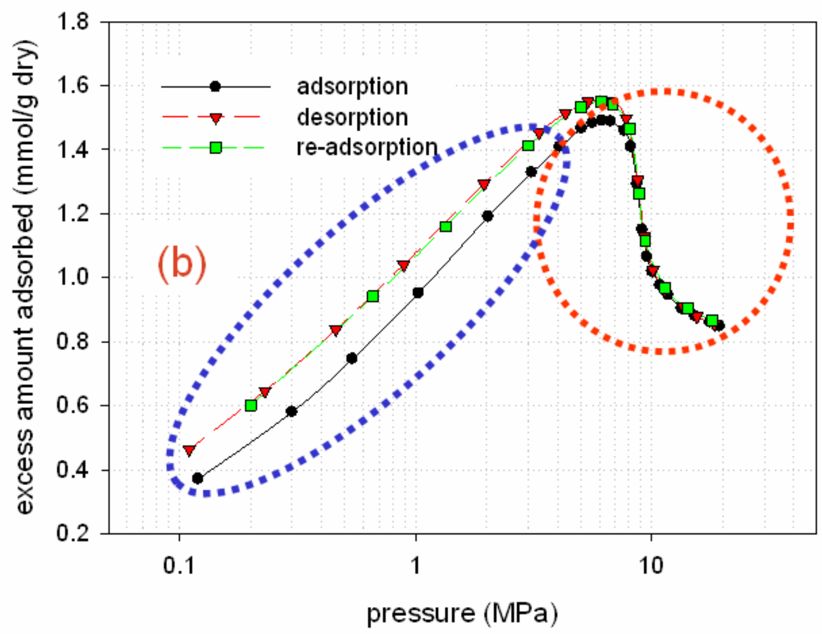
Excess sorption & open hysteresis



Open Hysteresis Phenomena

$$N_{exe} = (\rho_{adsorbed} - \rho_{gas}) \cdot V_{pore}$$

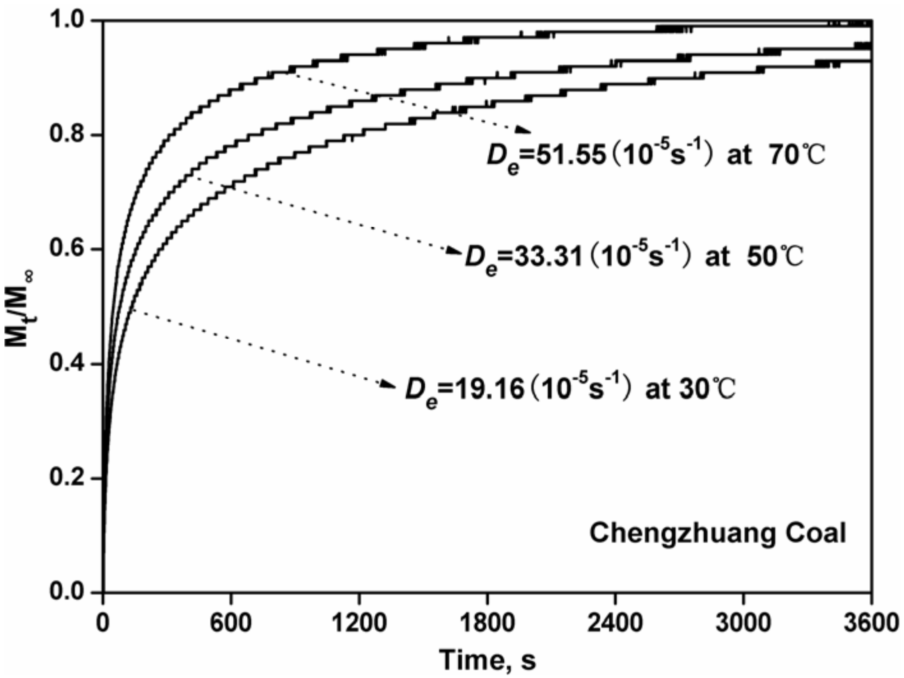
$$\frac{dN_{exe}}{dP} = \left(\frac{d\rho_{adsorbed}}{dP} - \frac{d\rho_{gas}}{dP} \right) \cdot V_{pore}$$

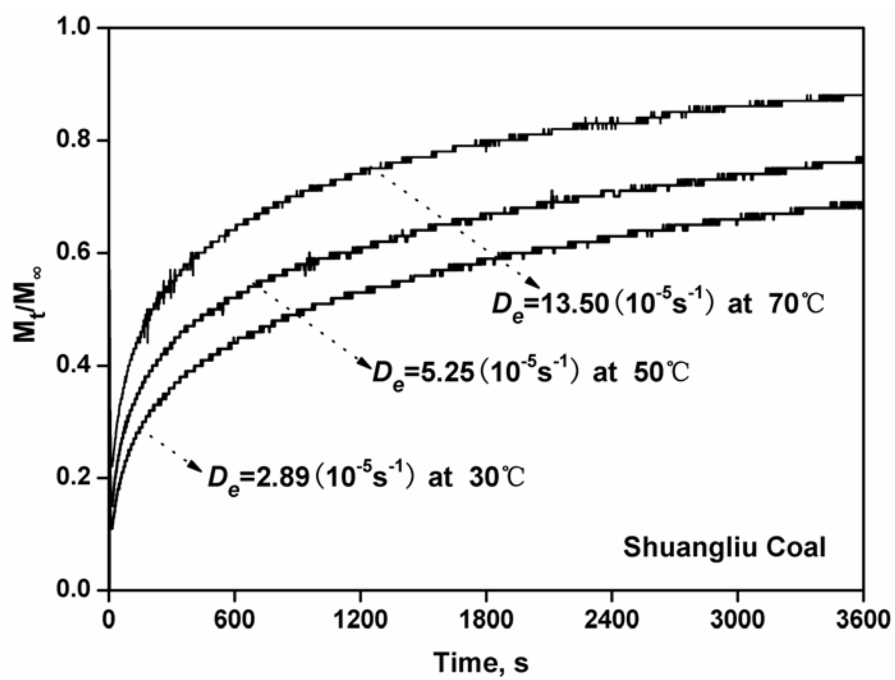


Adsorption/desorption isotherms of (a) CH_4 and (b) CO_2 on coal A at 313 K

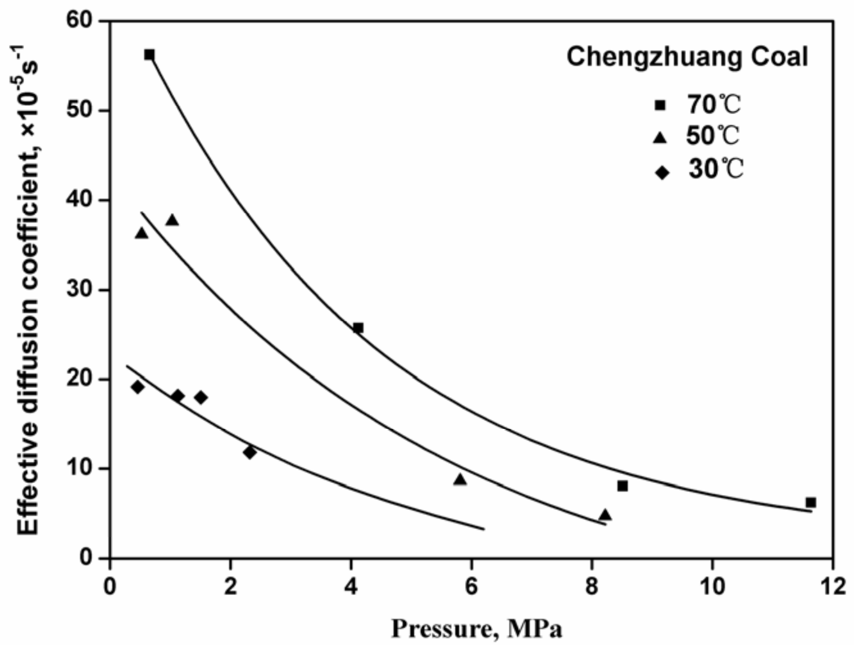
Ref.: J.-S. Bae, S. Bhatia, V. Rudolph and P. Massarotto (2009): Pore Accessibility of Methane and Carbon Dioxide in Coals. Energy Fuels 23 (6), 3319–3327.

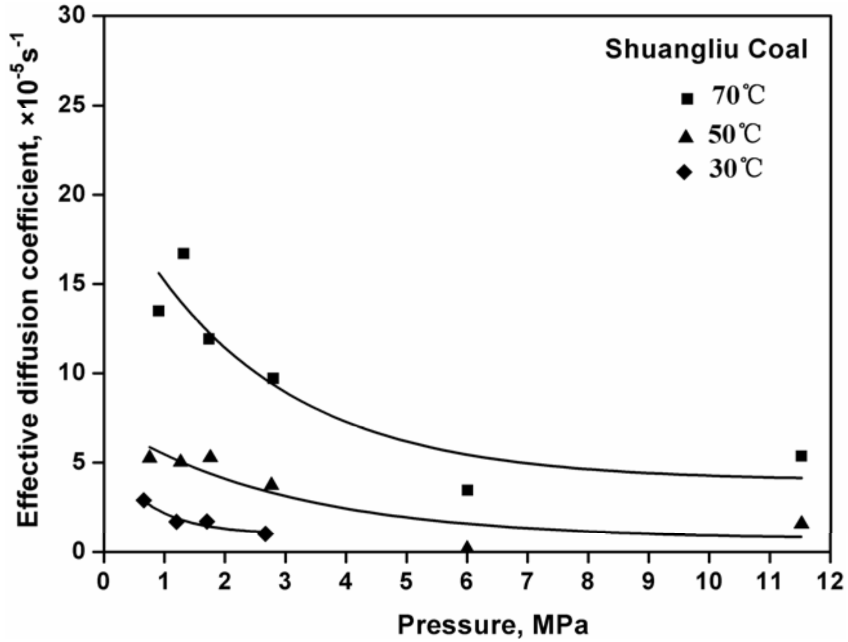
Gas sorption kinetics analysis





Gas diffusion in coal





Roles of transport properties

Table Transport properties of coal and their roles in CBM recovery

Process	Parameter	Role
Desorption	Langmuir pressure, P_L (MPa)	Determination of gas content or adsorption/desorption capacity of CBM reservoir
	Langmuir volume, V_L (cm^3/g)	
	Sorption time, τ (day)	
Diffusion	Diffusivity, D (cm^2/s)	Controlling microspore transport, i.e. migration of gas molecules in microspores of coal
	Micro-permeability, k (md)	
Darcy flow	Permeability, k (md)	Dominating gas transport in cleats/features of coal

- V_L - maximum storage capacity
- P_L - the pressure at which gas storage capacity equals one half of V_L
- τ - the time required to desorb 63.2% of the initial gas volume
- *D* - diffusion property of coal
- *K* - index how well gases pass through coal

- To make a good ECBM producer:
  - ✓High net effective coal thickness
  - ✓Laterally extensive coals
  - ✓High gas content/saturation
  - ✓High permeability confined to coal seams
  - ✓Shallow depth (low cost to drill)
  - ✓Low CO_2 content

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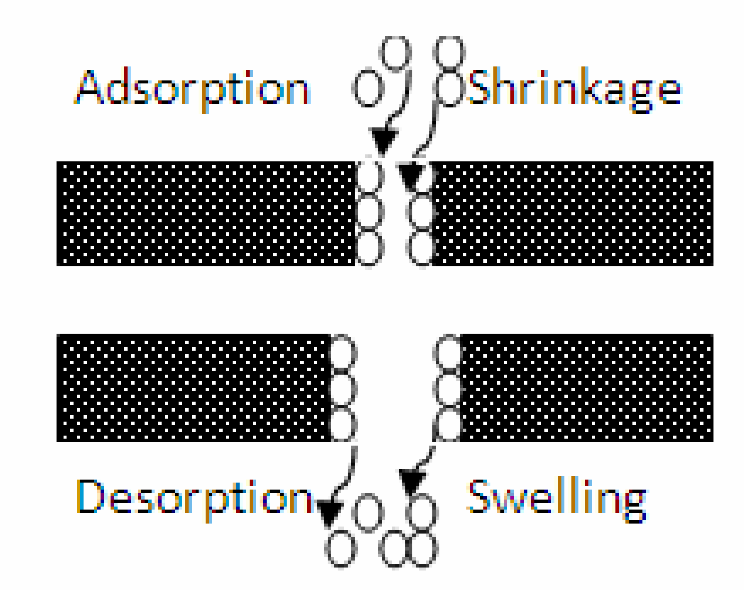
Swelling/shrinkage of coal



- The swelling and shrinkage that occurs in coal as gas is adsorbed or desorbed are well-known phenomena, referred to as sorption-induced strain. The sorption-induced strain of the coal matrix leads to a change in the width of cleats or fractures and hence has a significant effect on permeability, and hence the gas flow in coal.

As much as 90% of the change in reservoir permeability may be due to sorption-induced strain!

Sorption-induced stain

- Sorption-induced strain represents

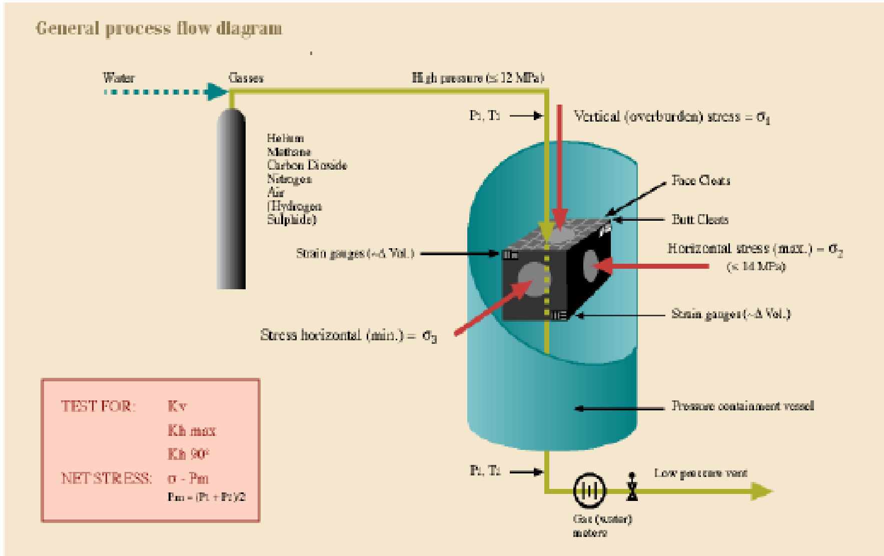
  - Strain:

$$S = \begin{cases} \dfrac{\Delta V}{V}(Volumetric) \\ \dfrac{\Delta L}{L}(Longitudinal) \end{cases}$$

  - Swelling or shrinkage caused by gas desorption/adsorption

- Permeability is strong function of sorption-induced strain

Measurement of sorption-induced stain in coal



<u>Main activities</u>

Stressed anisotropic permeability

Stressed sorption

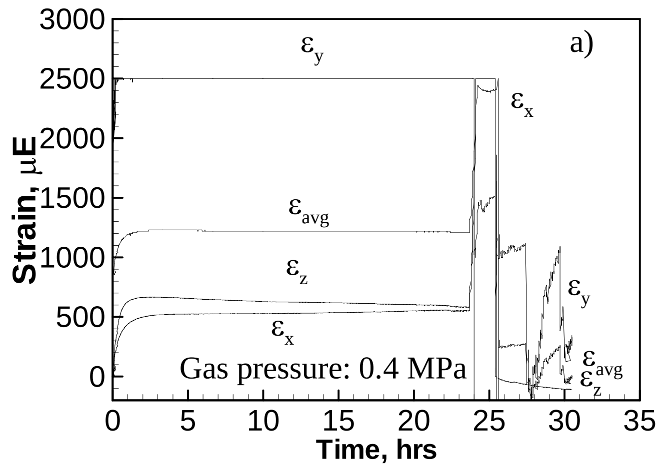
Competitive sorption/diffusion

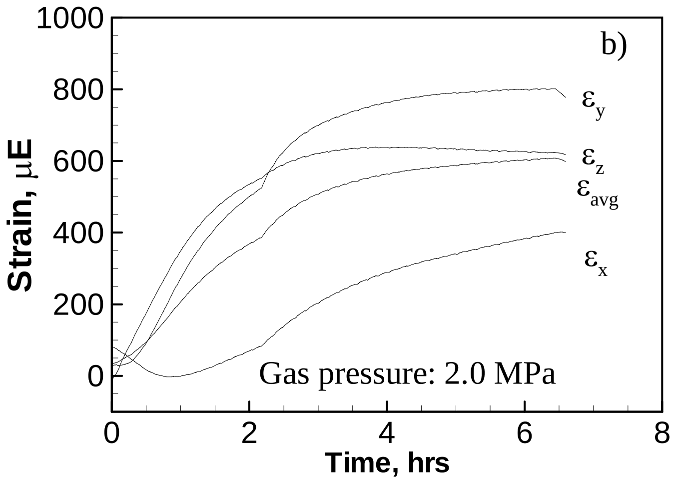
Water behavior

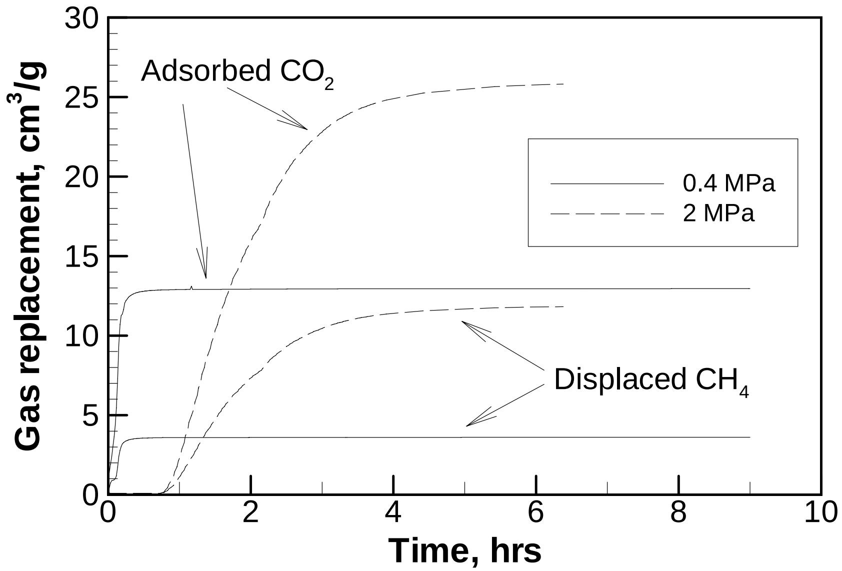
Long term interactions

Open source CO2ECBM reservoir model

Strain changes and gas displacement during CO_2 flush (sequestration)







Ref.: G.X. Wang, X.R. Wei, K. Wang, P. Massarotto and V. Rudolph (2010): Sorption-induced swelling/shrinkage and permeability of coal under stressed adsorption/desorption conditions. IJCG 83, 46-54.

Dynamic permeability during stressed adsorption/desorption

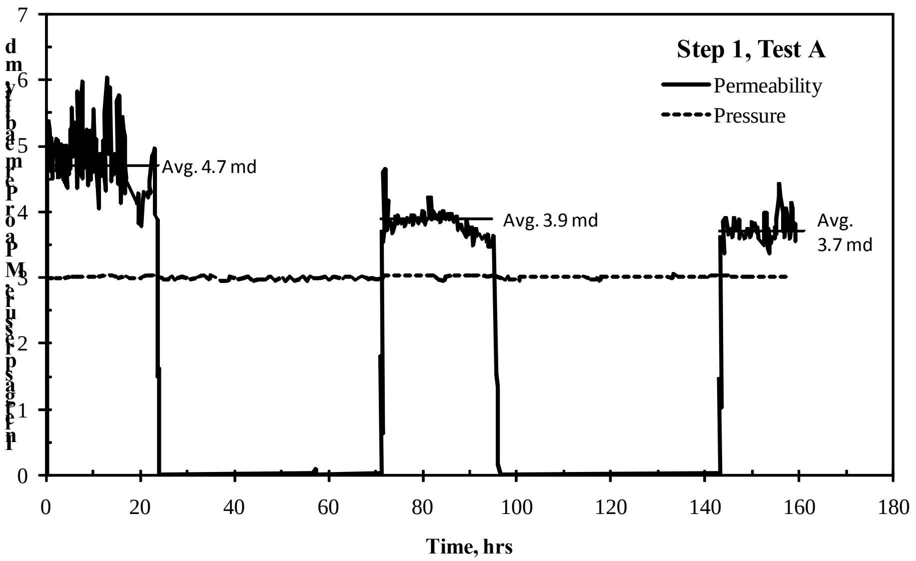


Fig 2 Changes of permeability during CH_4 pre-adsorption at Step 1 of Test A.

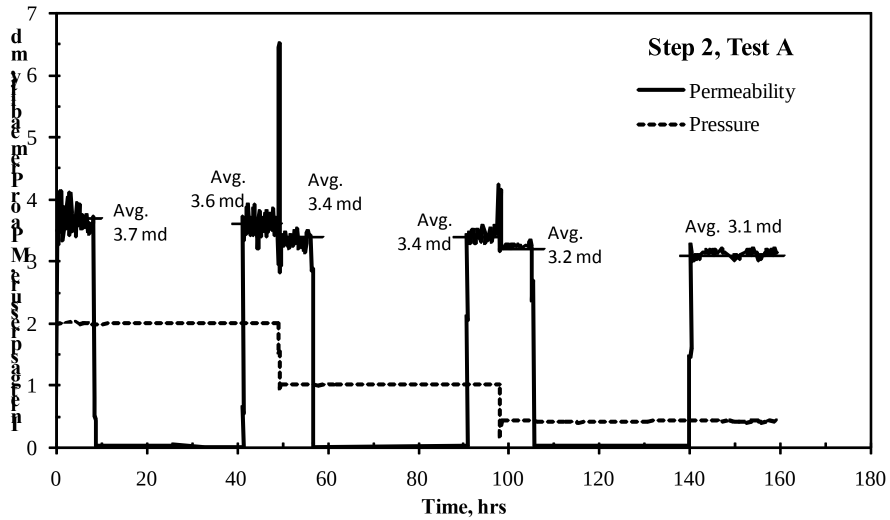
Step 1

CH_4 pre-adsorption for one week by changing helium to CH_4 gas at 3 MPa to approach the CH_4 adsorption equilibrium

Step 2

CH_4 desorption by reducing the gas pressure to 2, 1 and 0.4 MPa step by step, 48 hrs each step

Fig3 CH_4 dynamic permeability and adsorption at Step 2 of Test A.



Ref.: G.X. Wang, X.R. Wei, K. Wang, P. Massarotto and V. Rudolph (2010): Sorption-induced swelling/shrinkage and permeability of coal under stressed adsorption/desorption conditions. IJCG 83, 46-54.

Some sorption-induced stain data

Table 4 Sorption induced strain data of some coals as reported in literature.

Coal	Sorbed gas	Sorption-induced strain: Type	Sorption-induced strain: Value, %	Pressure*	Reference
Various	CH_4	Volumetric	0.2 - 1.6	~15.2 MPa	Moffat & Weale (1955)[83]
Kentucky coals	CO_2	Volumetric	1.31	20 psi	Reucroft & Patel (1986)[84]
Unclear	CH_4 CO_2	Longitudinal Longitudinal	0.06 1.0	800 psi 800 psi	Gray (1987) [29]
Unclear	CH_4	Volumetric	0.6	1000 psi	Harpalani & Schraufnagel (1990)[27]
Bituminous	CO_2	Volumetric	0.36 - 1.31	0.41 MPa	Harpalani & Chen (1992)[82]
Unclear	CH_4 CO_2	Longitudinal Longitudinal	0.1 0.8	1000 psi 800 psi	Seidle and Hutti (1995)[36]
High-volatile bituminous	CH_4 CO_2	Longitudinal Longitudinal	0.2 0.5	1000 psi 750 psi	Levine (1996)[35]
Sub-bituminous	CO_2	Longitudinal	0.00182/MPa		St. George & Barakat (2001)[41]
Unclear	CH_4 CO_2	Volumetric Volumetric	0.5 1.1	1000 psi 750 psi	Zutshi & Harpalani (2004)[80]
Unclear	CH_4 CO_2	Volumetric Volumetric	0.49 2.41	1000 psi 800 psi	Chikatamarla & Bustin (2004)[76]
Bituminous Sub-bituminous	CO_2	Longitudinal Volumetric	0.2-0.3 0.3-5.0	101.325kPa	Fry etc. (2009) [79]
Bituminous	CH_4 CO_2 Ar	Volumetric	0.65 0.37-0.92 0.63	~8.1MPa	Van Bergen etc. (2009) [78]
Sub-bituminous	CO_2	Volumetric	1.05-1.49	~8.1MPa	Van Bergen etc. (2009) [78]

* 1 MPa= 145.038 psi

Improved modeling by including sorption-induced stain

$$k_z = k_{z0}[\zeta_x(1-\varepsilon_x^+)^n + \zeta_y(1-\varepsilon_y^+)^n]$$

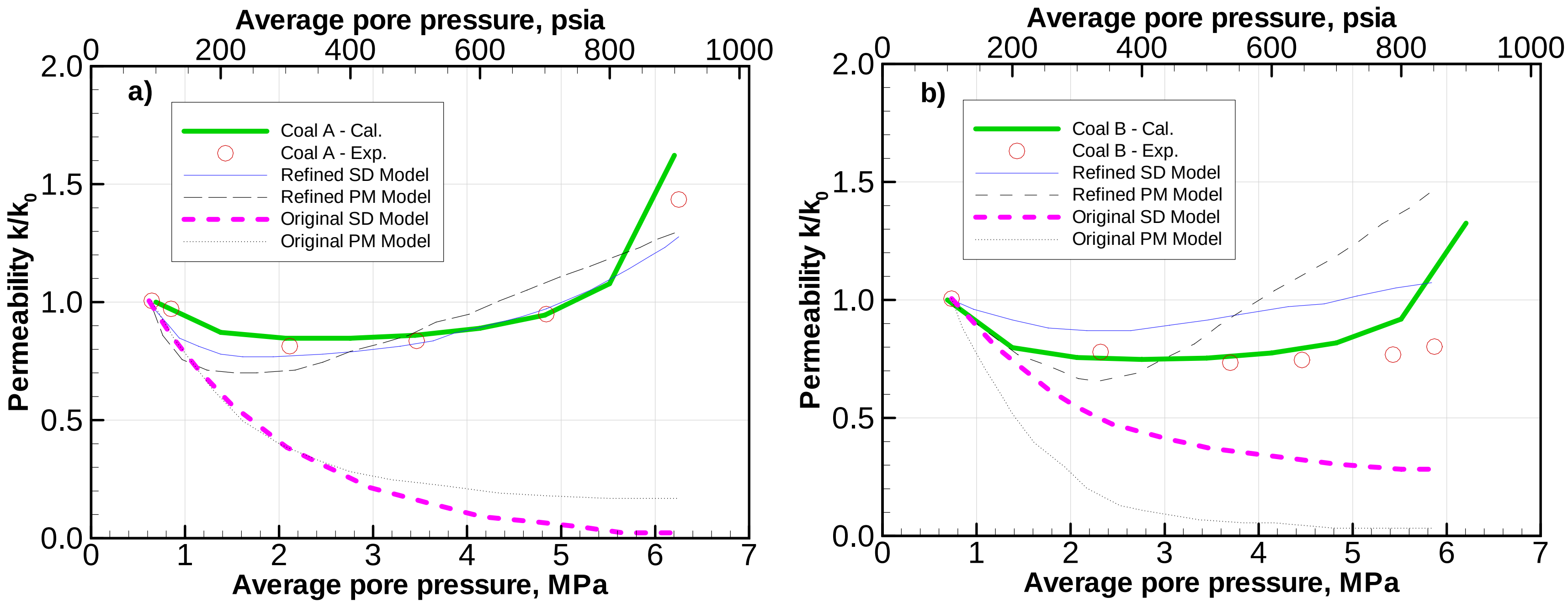
$$\zeta_i = \zeta_{i0}\exp[-D_z E_z \varepsilon_z^+] \quad with \quad \zeta_{x0} = \frac{l_x}{l_x + l_y}; \quad \zeta_{y0} = \frac{l_y}{l_x + l_y}$$

$$\varepsilon_i^+ = \frac{\sigma_i - \sigma_{i0}}{E_i} - \sum_{j=x}^{z}[(1-\delta_{ij})\nu_{ji}\frac{(\sigma_j - \sigma_{j0})}{E_j}] + \lambda\sum_{n=1}^{n_c}\alpha_{sn}(q_{\mu n} - q_{\mu_0 n}) \quad (i = x, y, z) \quad (\varepsilon \sim \sigma)$$

Ref.: G.X. Wang, P. Massarotto and V. Rudolph (2009): An improved permeability model of coal for coalbed methane recovery and CO2 geosequestration. IJCG 77, 127-136.

Improved modeling with sorption-induced stain data

Predictions *vs.* literature data (for a gas mixture: 51%N_2+49%CO_2)



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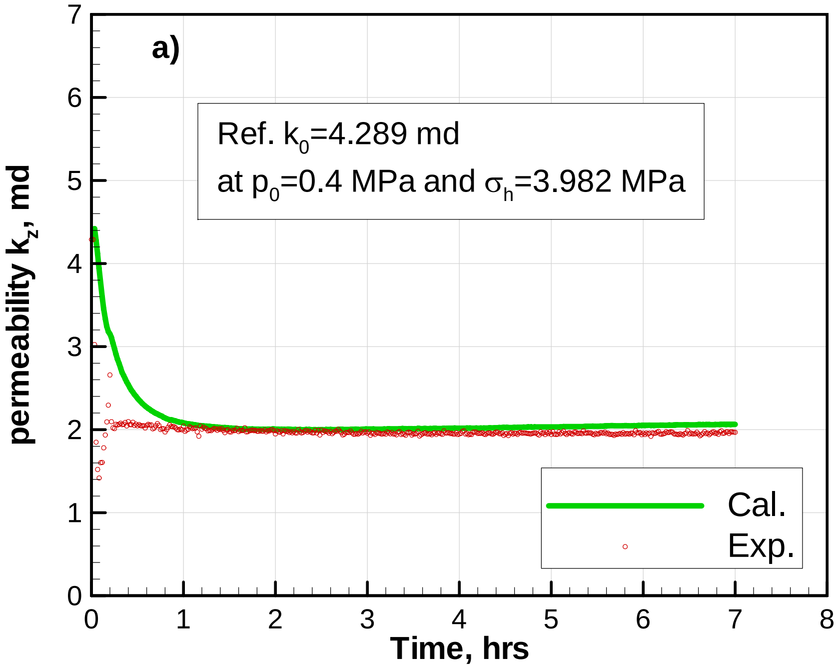
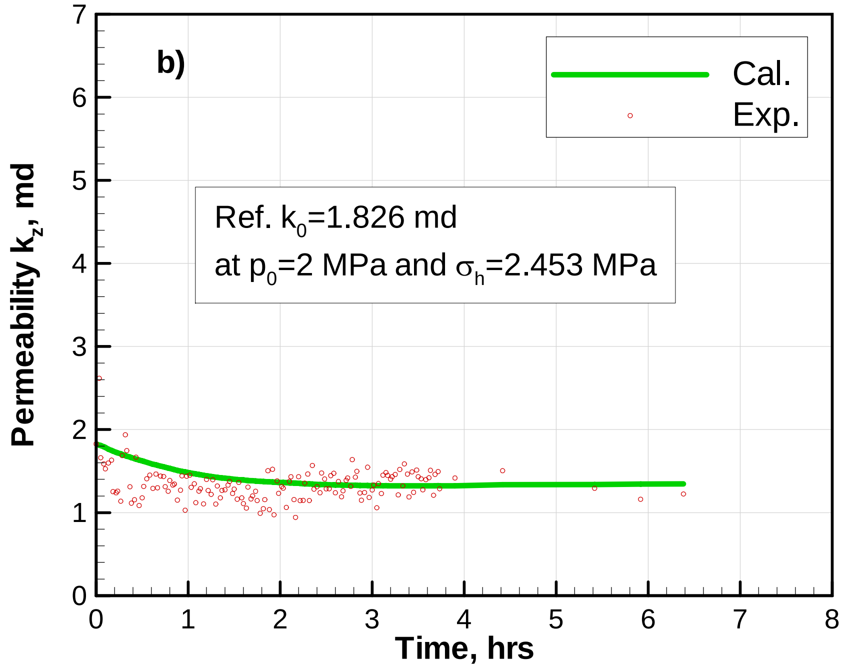
Comparison with UQ TTSCP data





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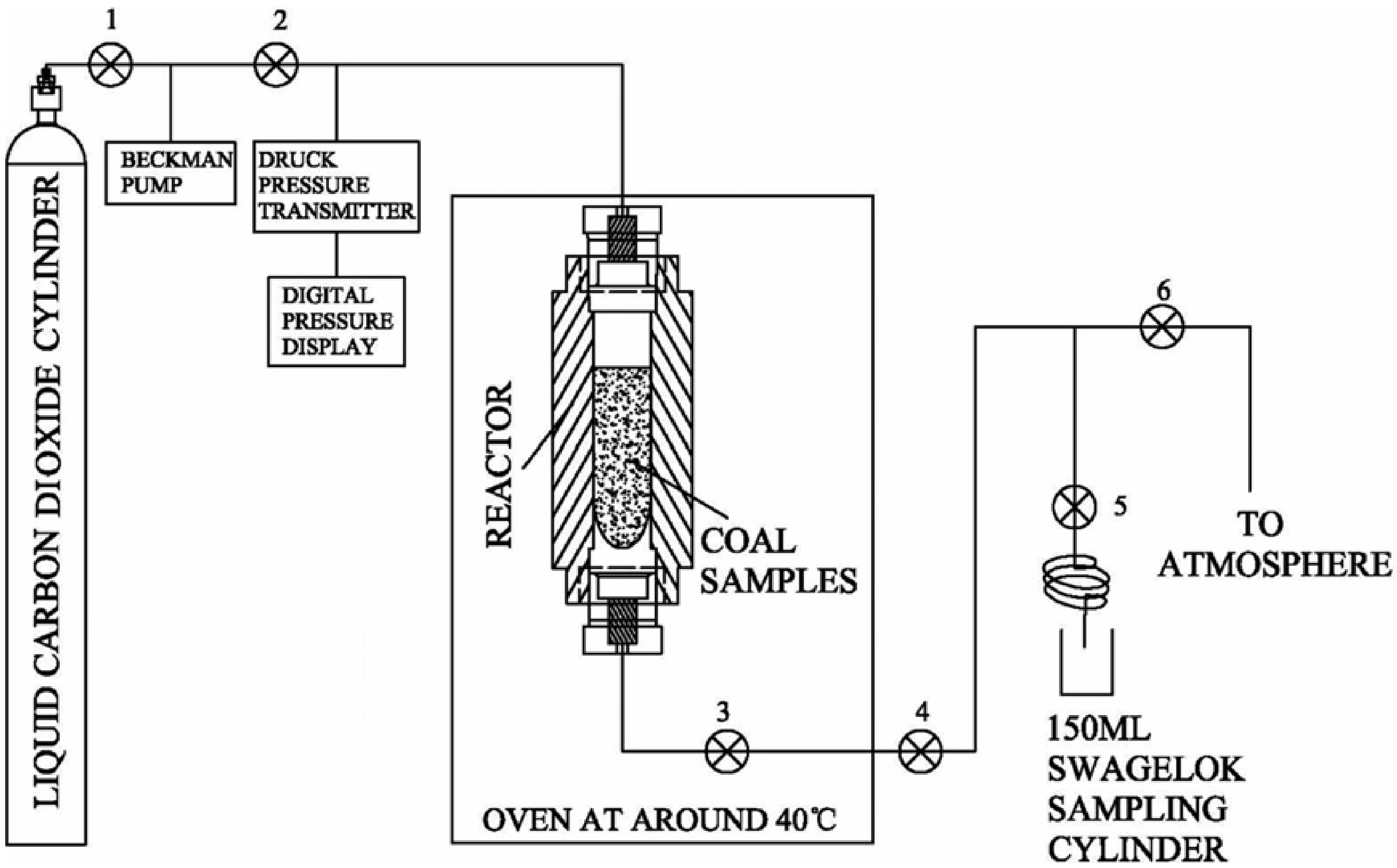
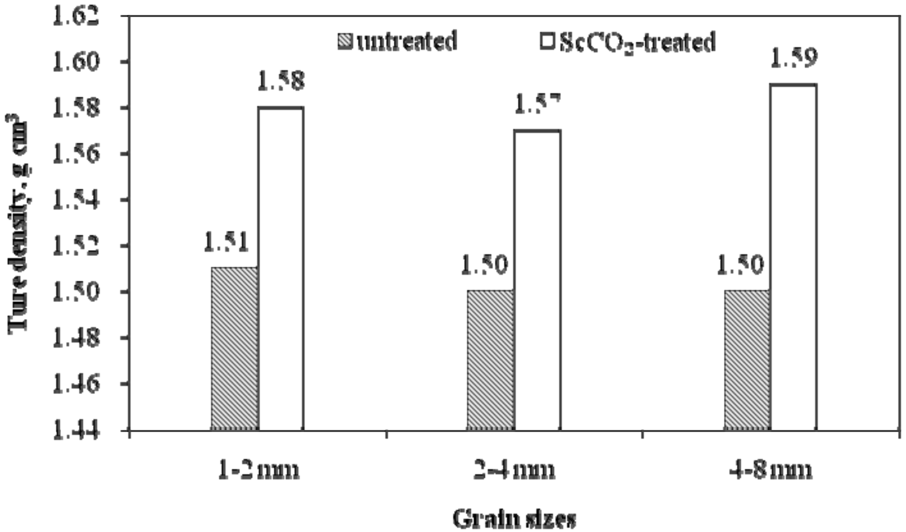
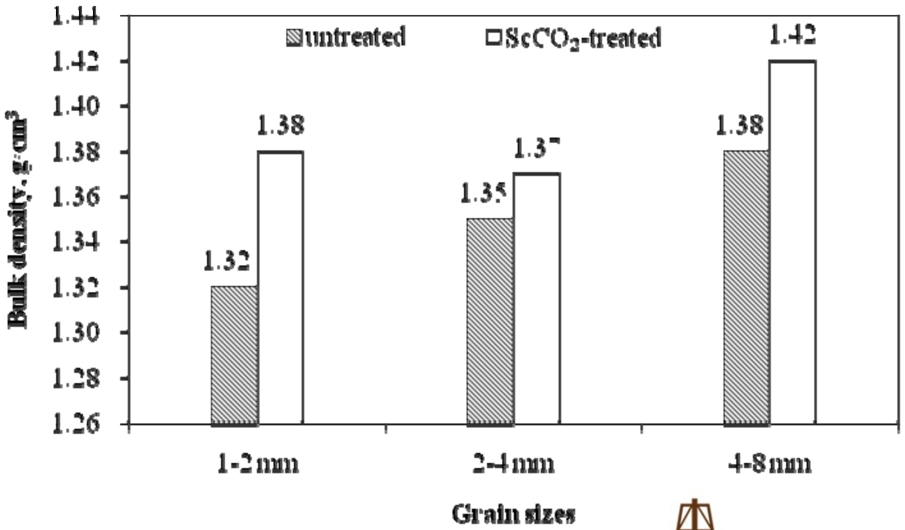
HP-ScCO$_2$ geochemical reactor



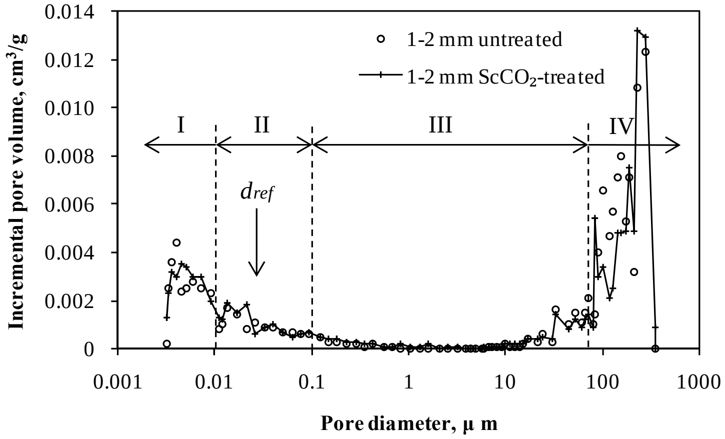
Density changes with ScCO2 treatment

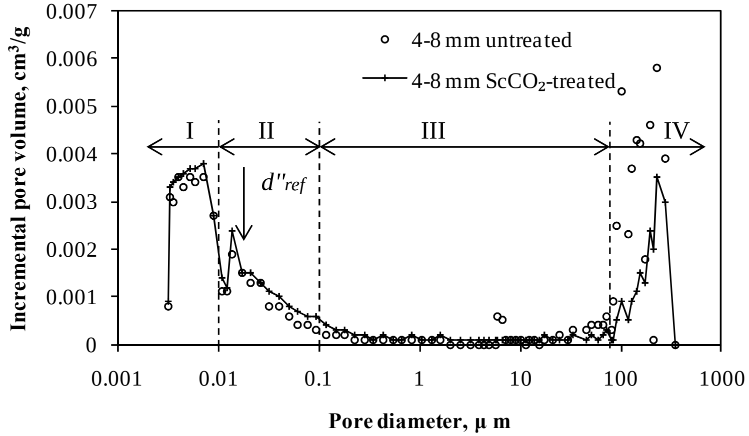


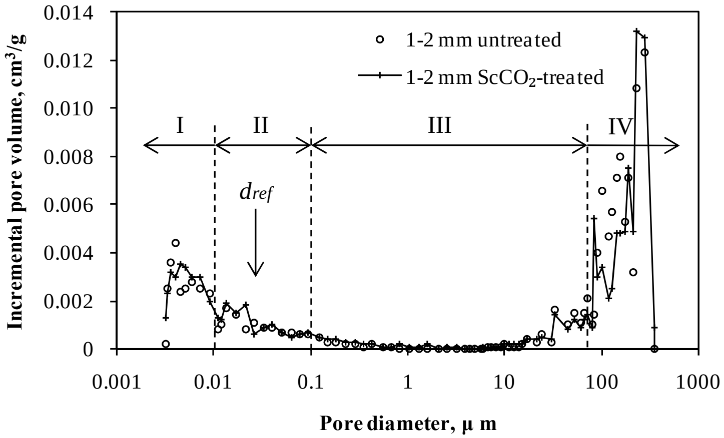


Ref.: C.J. Liu, G.X. Wang, S.X. Sang and V. Rudolph (2010): Changes in pore structure of anthracite coal associated with CO_2 sequestration process. Fuel 89(10), 2665-2672.

Incremental pore volumes



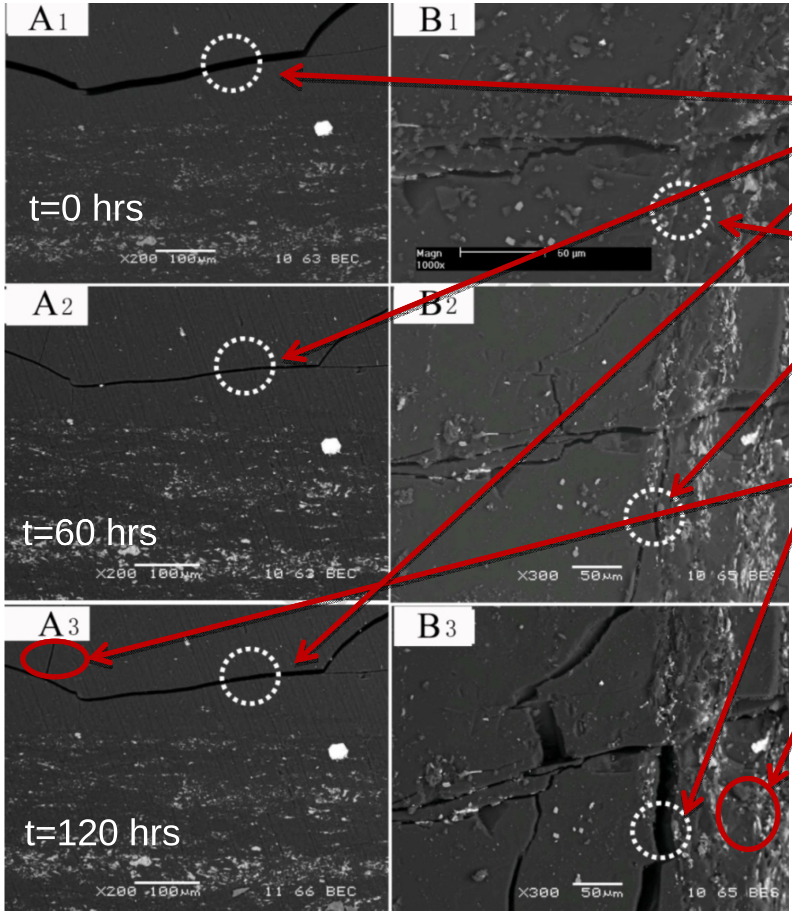




I: micropores (<10 nm)
II: mesopores (10-100 nm)
III: macropores (>100 nm)
IV: interpores

Ref.: C.J. Liu, G.X. Wang, S.X. Sang and V. Rudolph (2010): Changes in pore structure of anthracite coal associated with CO_2 sequestration process. Fuel 89(10), 2665-2672.

Structural deformation with $ScCO_2$



Coal A

Coal B

Aperture sizes of the same cracks

$A_1 \rightarrow A_2 \rightarrow A_3$: 1.07→0.36→0.54 μm

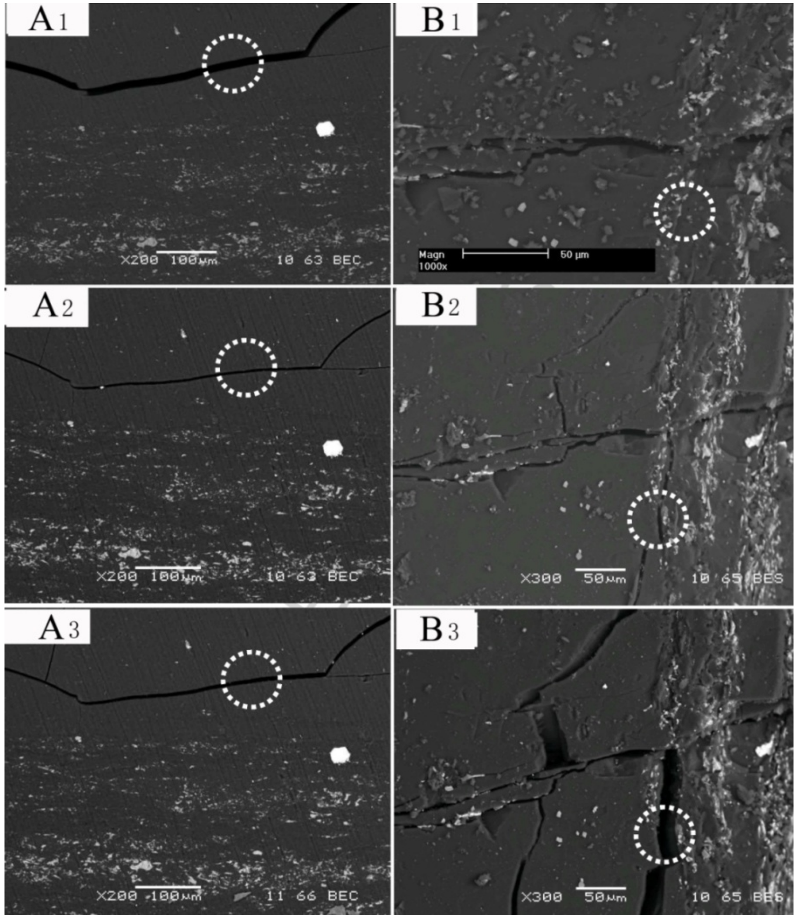
$B_1 \rightarrow B_2 \rightarrow B_3$: 0→0.03→1.75 μm

New cracks

Increasing fluid pressure increases extension of cracks

Ref.: P. Massarotto, S.D. Golding, J.-S. Bae, R. Iyer and V. Rudolph (2010): Changes in reservoir properties from injection of supercritical CO_2 into coal seams — A laboratory study. IJCG 82(3-4), 269-279.

Permeability changes with $ScCO_2$



$$K=\frac{1.013\times10^{9}b^{3}}{12s}$$

According to Levine）

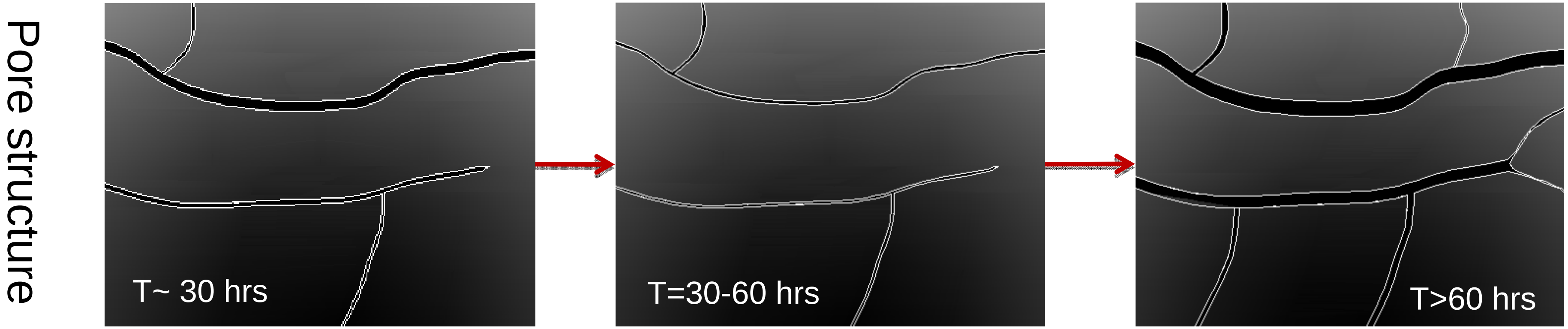
Permeability decreases Initially

Impermanent stabilization

Permeability increases gradually

Final stabilization

Structural evolution & element migration



Na	+++	++	++++
Mg	++++	++	+++
Al	++	++	++++
Si	+	+	++++
K	+	+	++
Ca	++++	++++	+++
Fe	++	++++	+
Ti	+	+	++++
S	+	++++	++

Summary

- ✓ CO_2 sequestration can be thought as an attractive technology to reduce GHG emission and enhance CBM recovery from deep coals.
- ✓ CBM/ECBM involves complex and coupling processes such as adsorption kinetics, diffusion and gas flow, which are highly related to the gas-solid interactions.
- ✓ Three key transport properties of coal, i.e. adsorption isotherm, diffusivity and permeability, have different behaviors and play different roles which have discussed.
- ✓ Permeability, a key factor that controls CBM/ECBM recovery and CCS process, should specifically investigated by experiments. Simulation with a proper model is important.

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