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coal cores exposed to CO₂: Adsorption, swelling strains,
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Measurement and modeling of adsorptive-poromechanical properties of bituminous coal cores exposed to CO₂: adsorption, swelling strains, swelling stresses and impact on fracture permeability

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Abstract

The reservoir response of unmineable coal seams to primary and enhanced natural gas recovery is strongly affected by the gas sorption and swelling properties of the coal reservoir rock. In-depth understanding of the process of gas sorption/desorption in the coal matrix, induced deformation and measurement of relevant physical parameters are critical for predictive reservoir management. Models used in industry practice are based on swelling strains measured in “free” swelling coal or on empirical correlations between strain and adsorption, and predict permeability changes based on changes of porosity or stress calculated assuming an analogy with thermoelasticity. However, not only coal seams are subjected to in-situ stresses and geometrical boundary conditions but also sorption and strain are strongly coupled. Representative experiments and a truly coupled model for coal seams are needed in challenging applications. We present a set of triaxial testing mea-

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9 surements on 38 mm diameter fractured sub-bituminous/bituminous coal
10 cores exposed to CO₂. Testing includes the measurement of fluid uptake,
11 adsorption-induced strains and stresses, and the impact on simultaneously
12 measured permeability. Noteworthy, we measured increases in effective stress
13 of up to 29 MPa when injecting CO₂ at 5 MPa and preventing the coal core
14 to swell. The results are analyzed with a poromechanical model in which
15 coal matrix microporosity and adsorption-induced phenomena are embed-
16 ded into a fractured reservoir rock with transverse isotropic properties. The
17 adsorption-mechanical coupling in the coal matrix is integrated through an
18 adsorption stress function and fractured coal permeability is estimated as
19 a function of Terzaghi's effective stresses (parallel and perpendicular to the
20 bedding plane). The experimental results and model predictions help iden-
21 tify the characteristic response of coal microporosity and cleat macroporosity
22 on the poromechanical response of coal cores, and suggest that the order of
23 magnitude change of reservoir permeability observed in the field are linked
24 to sorption-induced change on Terzaghi's effective horizontal stress under
25 laterally constrained displacement condition. Together, the modeling and
26 experimental characterization offer unprecedented insights into the mechan-
27 ics of coal.
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45 *Keywords:* sorption, CBM, unconventional, natural fractures,
46 nanoporosity, chemo-mechanical coupling
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51 **1. Introduction**
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54 2 Currently, natural gas accounts for roughly 21% of the World's energy
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56 3 supply, being coal-bed methane an important contributor in countries such
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9 as Australia, USA, Canada, and China (IEA, 2013; EIA, 2013). Natural
10 gas production from coal beds is expected to increase as more reservoirs are
11 discovered and technology enables enhanced production. Coal beds may also
12 contribute to storing and sequestering geologically CO₂, conceivable up to
13 20 GtCO₂ (Gale, 2004).
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19 Various characteristics make coal beds a unique geomaterial, showing
20 poromechanical properties notably different from other reservoir rocks:

21 (1) Coal seams are naturally fractured reservoirs. Diagenetic processes lead
22 to opening mode fractures predominantly oriented perpendicular to the bed-
23 ding plane, called cleats (Laubach et al., 1998). Cleats compose most of
24 the macroporosity, where fluid advection takes place (Mazumder et al., 2006;
25 Pan and Connell, 2007). The presence of cleats induces hydro-mechanical
26 anisotropy to the rock properties (Hu et al., 2010).
27

28 (2) At the smallest scale, coal seams are constituted by a microporous disor-
29 dered organic continuum, which we call from now on the coal matrix. Micro-
30 pores sized in the order of 10⁻⁹ m compose the coal microporosity. The coal
31 matrix can also host mesopores sized in the order of 10⁻⁸ m. The coal matrix
32 is capable of adsorbing various gases including CO₂, CH₄, and N₂, which lead
33 to coal matrix volumetric swelling in the order of few percents upon adsorp-
34 tion (Reucroft and Sethuraman, 1987; Ceglarska-Stefanska and Czaplinski,
35 1993; Levine, 1996; Mazumder et al., 2006; Pan and Connell, 2007; Pini,
36 2009). Conversely, desorption leads to coal matrix shrinkage. Overburden
37 and bedding processes add anisotropy to the coal matrix properties (Cody
38 et al., 1988; Hol and Spiers, 2012; Espinoza et al., 2013). Within the coal ma-
39 trix, fluid transport occurs by a combination of diffusion and slow advection
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9 (Ceglarska-Stefanska and Zarebska, 2002).

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11 The variety of pore sizes in coal seams yields various types of fluid pore
12 habit. Macropores host fluids mostly in bulk conditions. Mesopores host
13 fluids in adsorbed state near solid walls and in bulk state far from solid
14 walls. Micropores contain fluids just in adsorbed state, sometimes called
15 “dissolved” fluid in the solid matrix. Additional fluids may be present as
16 solutes in the water phase.
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22 Sorption-induced swelling has an important effect on coal seam permeabil-
23 ity (Pan and Connell, 2012). Changes of more than two orders of magnitude
24 in permeability have been observed (1) during primary methane production –
25 permeability increase due to desorption-induced shrinkage and cleat opening
26 (Palmer and Mansoori, 1998; Scott et al., 2012) and (2) during CO₂ injection
27 – permeability decrease due to adsorption-induced swelling and cleat closing
28 (Pekot and Reeves, 2002; Oudinot et al., 2011). Such a unique behavior of
29 coal has led to the development of semi-empirical models and poromechanical
30 models to account for the adsorption-strain coupling developed in microp-
31 orous coal matrix and predict its impact on seam permeability (Palmer and
32 Mansoori, 1998; Shi and Durucan, 2004; Cui et al., 2007; Wu et al., 2010;
33 Vandamme et al., 2010; Pijaudier-Cabot et al., 2011; Brochard et al., 2012;
34 Nikoosokhan et al., 2012, 2014).
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49 Coal seam coupled reservoir simulation seeks to predict the evolution of
50 permeability as a function of bottomhole pressure. The relevant parameters
51 include cleat compressibility, mechanical moduli and a measurement that
52 can account for the swelling properties of coal. Current industry codes use
53 swelling strains (making an analogy with thermoelasticity) in order to couple
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10 54 the effects of sorption on the variation of in-situ stress and porosity with
11 55 depletion. These parameters are usually obtained from laboratory testing
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13 56 and field data history matching.

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15 57 Studies of adsorption in coal cores and ensuing change in permeability
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17 58 are scarce because of several reasons including difficulty in coring fractured
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19 59 coal and obtaining representative coal samples, long time required to reach
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21 60 adsorption thermodynamic equilibrium, influence of confining stress on per-
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23 61 meability as a rate-limiting factor for CO₂ delivery into coal matrix micro-
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25 62 pores, and need for special non-adsorbing membranes and sealing o-rings.
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27 63 Salient observations from selected experimental studies follow (Czaplinski
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29 64 and Holda, 1982; Levine, 1996; Chikatamarla et al., 2004; Mazumder et al.,
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31 65 2006; Viete and Ranjith, 2006; Day et al., 2008; Pini, 2009; Pone et al., 2009;
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33 66 Hagin and Zoback, 2010; Pan et al., 2010; Hol et al., 2012; Masoudian et al.,
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35 67 2013, 2014): (1) CO₂ equilibration times increase as the specimen size in-
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37 68 creases and vary from about 1 hour to more than 20 days, (2) swelling along
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39 69 the direction perpendicular to the bedding plane is generally higher –up to
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41 70 60%– than along the direction parallel to the bedding plane, (3) absolute
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43 71 permeability decreases with Terzaghi’s effective stress at a given fluid pres-
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45 72 sure, however, the variation of permeability as a function of pressure of the
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47 73 adsorbate fluid at constant Terzaghi’s effective stress is not clear, and (4)
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49 74 CO₂ sorption in coal lowers –up to 25%– the Young’s modulus and shear
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51 75 strength and enhances creep, compared to dry coal or coal saturated with
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53 76 helium.

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54 77 The objective of this study is to measure the coupled poromechanical
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56 78 response and permeability of coal cores injected with an adsorbate under
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9 different loading and strain boundary conditions, interpret the results in
10 the context of a double porosity model for transverse isotropic fractured
11 microporous solids (fully coupled regarding fluid mass, fluid pressure, and
12 solid stress), and give insight into the evolution of permeability of the coal
13 seam in-situ during primary and secondary methane recovery.
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20 **2. Double porosity model for transverse isotropic fractured coal**

21 *2.1. Poromechanical model*

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25 We present a poromechanical model for double porosity transverse isotropic
26 reservoirs rocks. An isotropic version of this model is found elsewhere (Nikoosokhan
27 et al., 2012, 2014). The double porosity system is composed by (1) the
28 cleat macroporosity where the fluid can be modeled as in bulk conditions
29 with a proper equation of state, and (2) the microporosity where adsorption
30 takes place, fluid behavior deviates from bulk conditions, and adsorptive-
31 mechanical coupling develops (swelling or shrinkage). The influence of cleat
32 compressibility (natural fractures) on the behavior of the fractured coal is
33 modeled with conventional anisotropic poroelasticity (Cheng, 1997; Coussy,
34 2004; Cowin, 2004) and the effect of adsorption-induced phenomena on the
35 coal matrix is modeled with the theory of poromechanics for microporous
36 media developed by Brochard et al. (2012). Appendix A details the model
37 derivation. The following set summarizes the equilibrium, porosity, and mass
38 balance equations for a cartesian representative elementary volume (REV)
39 of fractured coal with transverse isotropy around axis 3 (See Figure 1):
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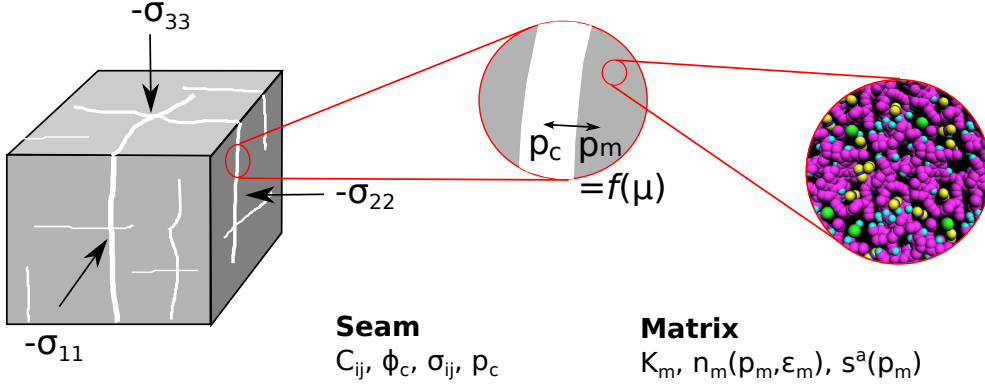


Figure 1: Representative elementary volume of fractured coal and relevant modeling parameters. Fracture zoom sketches the interplay between pressure p_c of bulk fluid in the cleats, and thermodynamic pressure p_m of fluid in the coal matrix. Coal matrix zoom shows a molecular representation of coal and adsorbed fluid molecules (Courtesy: L. Brochard).

$$\left\{ \begin{array}{l}
 \sigma_{11} = C_{11}\epsilon_{11} + C_{12}\epsilon_{22} + C_{13}\epsilon_{33} - b_1 p_c - (1 - b_1)s^a(p_m) \\
 \sigma_{22} = C_{12}\epsilon_{11} + C_{11}\epsilon_{22} + C_{13}\epsilon_{33} - b_1 p_c - (1 - b_1)s^a(p_m) \\
 \sigma_{33} = C_{13}\epsilon_{11} + C_{13}\epsilon_{22} + C_{33}\epsilon_{33} - b_3 p_c - (1 - b_3)s^a(p_m) \\
 \sigma_{23} = 2C_{44}\epsilon_{23} \\
 \sigma_{31} = 2C_{44}\epsilon_{31} \\
 \sigma_{12} = 2\frac{C_{11}-C_{12}}{2}\epsilon_{12} \\
 \phi_c - \phi_{c0} = [p_c - s_a(p_m)]/N + b_1(\epsilon_{11} + \epsilon_{22}) + b_3\epsilon_{33} \\
 n_T = (1 - \phi_{c0})n_m(p_m, \epsilon_m) + \phi_c \rho_b
 \end{array} \right. \quad (1)$$

101 The first six equations capture the fractured coal stress tensor includ-
 102 ing fracture poroelastic effects and adsorption stresses from the coal matrix.
 103 Equations indicate that the fractured coal mechanical deformational behavior
 104 depends on the five independent stiffness coefficients C_{11} , C_{12} , C_{13} , C_{33} , C_{44}
 105 of the fractured coal (include effect of fracture compressibility), the porome-
 106 chanical coupling depends on the fracture-induced Biot coefficients (b_1 , b_3) of
 107 the fractured coal (Equation A.20) and the adsorptive-mechanical coupling

108 depends on the pressure-dependant adsorption stress developed by the coal
 109 matrix $s^a(p_m)$. The adsorption stress quantifies the swelling potential of the
 110 coal matrix, it is considered isotropic here and it is equivalent to the stress
 111 that should be applied to a specimen of coal matrix exposed to an adsor-
 112 bate at pressure p_m not letting it develop swelling strains. The adsorption
 113 stress can be measured experimentally at the scale of the tested specimen.
 114 Our model predicts its value based on the amount of adsorbed fluid n_m ,
 115 the adsorption-strain coupling coefficient $c(p_m)$, and the coal matrix bulk
 116 modulus K_m (Eq. A.10). The seventh equation allows calculating the cleat
 117 porosity ϕ_c as a function of the initial porosity ϕ_{c0} , the poroelastic coeffi-
 118 cient N (Eq. A.20), the strains of fractured coal ε_{ij} , cleat pressure p_c , and
 119 adsorption stress $s^a(p_m)$. The eighth equation permits obtaining the total
 120 amount of fluid in the fractured coal per unit volume of fractured coal n_T
 121 as sum of the amount of fluid sorbed in the matrix n_m (as a function of
 122 pressure p_m and volumetric strain ε_m of the coal matrix – first term) and
 123 the amount of fluid in the cleats (which depends on cleat porosity ϕ_c and
 124 bulk fluid molar density ρ_b –second term). The coal matrix strain is given
 125 by Equation (A.5). The total amount of sorption in the coal matrix n_m is
 126 modeled assuming a Langmuir isotherm (parameters n_0^{max} , p_{L0}), and a first-
 127 order expansion respect to coal matrix strain with a proportionality factor
 128 termed adsorption-strain coupling coefficient $c(p_m)$ (Equation A.3). The coal
 129 matrix is assumed isotropic with bulk modulus K_m (the origin of anisotropy
 130 is discussed in Section 5.2.1). Thus, the full double porosity model has 10
 131 independent parameters, six of them defined at the fractured coal scale and
 132 four at the coal matrix scale.

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133 Non-sorbing and non-swelling rocks have negligible adsorption $n_m(p_m, \epsilon_m) =$
134 0 and adsorption stress $s^a(p_m) = 0$. In this case Equation (1) simplifies to
135 conventional transverse poroelasticity (Equation A.19). Coal is able to sorb
136 different gases with ensuing swelling. The value of $n_m(p_m, \epsilon_m)$ is in the order
137 of 1 mol/L and adsorption stresses s^a reach several tens of MPa (Brochard
138 et al., 2012; Espinoza et al., 2013).

139 *2.2. Stress-permeability empirical relationship*

140 Stress-based models correlate the logarithm of permeability to stresses,
141 e.g., the mean Terzaghi's effective stress $\sigma' = -(\sigma + p)$, or to the horizontal
142 Terzaghi's effective stress $\sigma'_h = -(\sigma_h + p)$ (Somerton et al., 1975; Shi and
143 Durucan, 2004; Cui et al., 2007; Pan and Connell, 2012). In the line of
144 previous work (Seidle et al., 1992; Connell et al., 2010; Wu et al., 2010),
145 we adopt a permeability which depends on Terzaghi's effective stress and
146 considers independently parallel to bedding and perpendicular to bedding
147 stresses through permeability fracture compressibility coefficients α_{\parallel} and α_{\perp}
148 respectively, such that,

$$k = k_0 \exp [\alpha_{\parallel}(\sigma'_{\parallel} - \sigma'_{\parallel 0}) + \alpha_{\perp}(\sigma'_{\perp} - \sigma'_{\perp 0})] \quad (2)$$

149 where k_0 is the reference permeability at a reference state $(\sigma_{\parallel 0}, \sigma_{\perp 0}, p_{c0}, s^a(p_{m0}))$.
150 This equation only accounts for the stress-sensitive part of fracture perme-
151 ability and disregards horizontal stress anisotropy and shear-enhanced per-
152 meability.

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10 **3. Coal core triaxial testing**

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12 *3.1. Coal characterization*

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14 The tested coal comes from Forzando mine in South Africa, at an original
15 depth of ~ 500 m. The recovered coal block (about $\sim 1\text{m}^3$) shows high de-
16 gree of fracturing with predominant orthogonally intersecting cleats, many
17 of them filled with calcite. Thin layers of clay mixed with the coal matrix are
18 visible in several sections. Vitrinite reflectance equal to 0.57% classify this
19 coal as sub-bituminous A/high volatile C bituminous (ASTM D 388). Sev-
20 eral cores were drilled in directions perpendicular and parallel to the bedding
21 plane (Figure 2). Specimens showed little damage during coring. Selected
22 specimens have been subjected to X-ray microtomography to investigate the
23 degree of specimen fracturing and fractures orientation. Figure 2 shows to-
24 mographic cross sections obtained in direction perpendicular to the bedding
25 plane. Empty cleats are seen as black regions while calcite-filled cleats are
26 imaged in white. Some empty cleats may be artifacts due to sample recovery
27 and coring.
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41 The bulk density of cores ranges from 1318 to 1356 kg/m^3 (See Table
42 1). The mass density of the coal skeleton is about $\rho_m = 1510 \text{ kg}/\text{m}^3$ as
43 measured by the accessible helium void volume. The combination of these
44 two measurements indicates specimen total helium porosity (fracture and
45 microporosity) ranging from ~ 11.3 to 13.9%. SEM imaging performed on 1
46 cm diameter coal cores shows the presence of microfractures ranging from 50
47 μm to less than 1 μm . Figure 3-a shows a calcite-filled fracture with total
48 aperture 50 μm being intercepted at $\sim 90^\circ$ by a smaller 10 μm empty frac-
49 ture. We measured pore size distribution with a mercury intrusion porosime-
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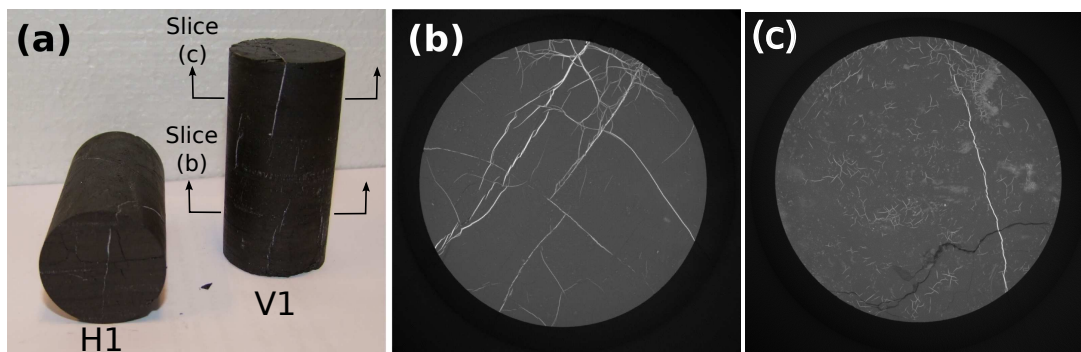


Figure 2: Coal cores 38 mm diameter – slenderness ratio 2:1 used for triaxial tests. (a) Cores picture: coring parallel to the bedding plane (left); coring perpendicular to the bedding plane (right). (b-c) X-ray tomographic reconstruction of cross sections on the bedding plane (image resolution 25 μm - performed at Laboratoire Navier <navier.enpc.fr/Microtomographie>).

178 ter Micromeritics-Autopore IV on a coal specimen of $\sim 1 \text{ cm}^3$ volume with
 179 a crack barely observable without magnification. Maximum mercury pen-
 180 etration at 207 MPa (equivalent to pore size $\sim 6 \text{ nm}$) shows a maximum
 181 cumulative intrusion volume of 0.031 mL/mL (Figure 3-b). The incremental
 182 pore volume curve (derivative of data in Figure 3) shows that most of the
 183 mercury-intruded porosity in this small sample is composed by either pores
 184 smaller than 60 nm or bigger than 10 μm . Elevated mercury pressure may
 185 compress the coal matrix, collapse non-invaded pores, and overestimate mi-
 186 croporosity. Independent low-pressure nitrogen adsorption measurements on
 187 coal crashed from the same seam suggest a peak of pore size distribution
 188 at 40 nm. The Auxiliary Material shows excess sorption measurements on
 189 crushed coal from the same coal seam (performed by BGC-Analytik UG).

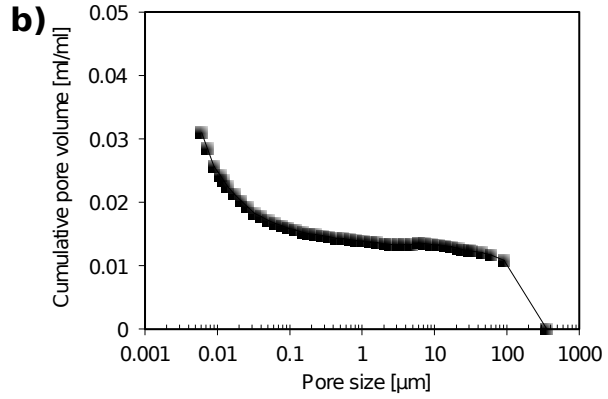
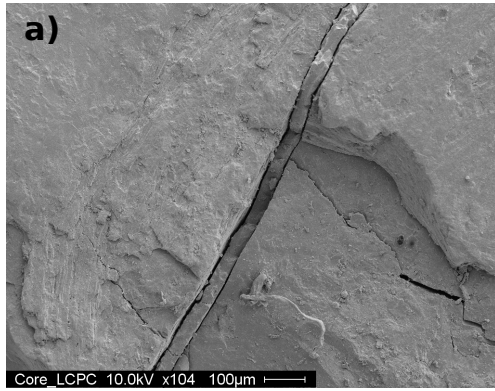


Figure 3: Coal characterization. (a) SEM imaging on a partially calcite-filled fracture. (b) Mercury intrusion porosimetry on a $\sim 1 \text{ cm}^3$ coal sample.

190 3.2. Triaxial cell

191 The experimental apparatus is designed to (1) measure core axial and
 192 radial deformations, (2) control independently axial and radial stresses, (3)
 193 measure CO_2 uptake by the coal specimen, and (4) measure core permeabil-
 194 ity. The following summarizes the triaxial cell characteristics and operation
 195 range. Sanchez Technologies manufactured the triaxial cell (Model: Tr-X
 196 40/60) and provided the fluid pumps. Figure 4 shows a schematic represen-
 197 tation of the triaxial cell.

198 **Specimen.** The triaxial cell fits cylindrical specimens 38 mm (1.5 in) diam-
 199 eter and slenderness 2:1. This specimen size permits accommodating several
 200 submillimeter-sized cleats.

201 **Stresses.** Pressurized silicone oil applies the radial stress (40 MPa maxi-
 202 mum), and a pressure-driven piston applies the axial stress (60 MPa maxi-
 203 mum).

204 **Pore Fluid System.** The CO_2 pore fluid system is managed by upstream
 205 and downstream syringe pumps. The first pump injects and measures CO_2

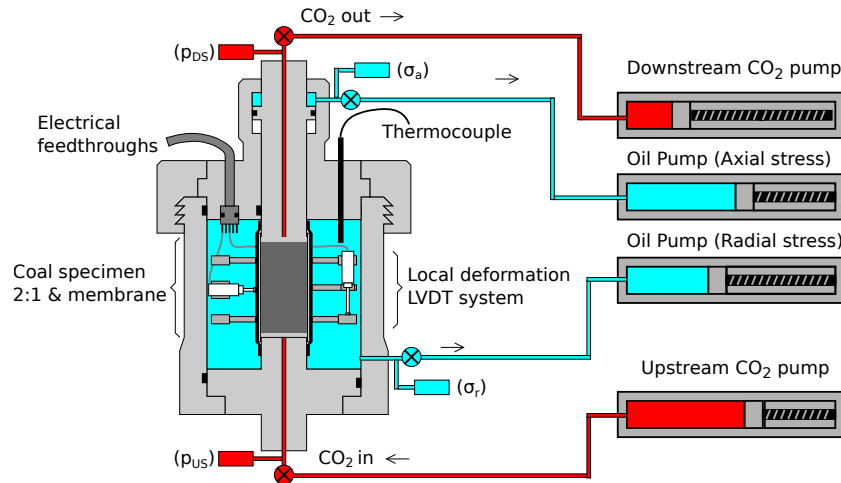


Figure 4: Experimental triaxial device. Main characteristics include: maximum radial stress 40 MPa, maximum axial stress 60 MPa, measurement of local strains, temperature control, and ability to handle supercritical CO₂.

flow rate and injection volume; the second pump controls CO₂ back-pressure. Stainless steel porous plates help distribute evenly the gas on the base and top faces of the specimen. A CO₂-resistant membrane combining layers of teflon and nitrile isolates the CO₂-saturated coal specimen from the surrounding confining fluid.

Strain Measurement. The device includes a specially designed system of displacement transducers (LVDT) to measure local radial and axial strains (three LVDTs for radial measurement and two LVDTs for axial measurements - 1 μm precision manufactured by Solartron - see details in Monfared et al. 2011).

Temperature. Two internal thermocouples measure the temperature of the confining fluid. Electrically-powered heating jackets control specimen and CO₂ pumps temperature. The maximum working temperature is 60°C.

Wiring, Acquisition, and Control. Electrical feedthroughs let measure output electrical signals from transducers placed inside the triaxial cell.

Table 1: List of performed triaxial experiments including their main characteristics. The experimental testing program includes cores drilled perpendicular \perp and parallel \parallel respect to the bedding plane. Coal specimens have a gravimetric water content 2 to 3%. Helium porosity is estimated with the helium skeletal mass density 1510 Kg/m³.

Specimen	Bulk mass density ρ_b [kg/m ³]	Helium porosity [%]	Type of dry loading	Stress/strain condition during CO ₂ injection	Stress/strain condition during permeability test
V1(\perp)	1356	11.2	Anisotropic	Constant stress Constant volume	Changing effective stresses Constrained adsorption
V2(\perp)	1346	12.6	Anisotropic	Constant stress	-
V3(\perp)	1331	13.2	Anisotropic	Constant stress Constant volume	- -
H1(\parallel)	1351	12.3	Anisotropic	Constant stress	-
H2(\parallel)	1318	14.4	Anisotropic	Constant stress	-

Sanchez Technologies provided an electronic acquisition and control system.

Cell calibration. We calibrated the cell with an aluminum specimen to assess end-effects and pressure effects on the cell volume, and measure the compressibility of the nitrile membrane.

4. Experimental results and poromechanical model validation

The objective of the experimental work is to measure the adsorptive, mechanical and transport properties of coal at the core scale to evaluate the developed model as an analytical and predictive tool. Sections 4.1 to 4.3 deal with the measurement of adsorptive-mechanical parameters and poromechanical model validation. Section 4.4 adds steady-state permeability measurements and makes use of the previously developed model to predict

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232 permeability upon adsorption in displacement constrained coal samples (near
10 constant volume condition). All experiments are performed at a temperature
11 39 ± 1°C, and specimens are vacuumed before testing. Table 1 presents a
12 summary of the performed experiments.
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16 Let us begin by considering a conceptual core drilled perpendicular to
17 the bedding plane, such that, the axis of the cylindrical core aligns with
18 the axis of material symmetry (assumed here perpendicular to the bedding
19 plane) and with the axis of the cylindrical triaxial device (axis 3 - Figure
20 5). The principal stresses in directions parallel to the bedding plane are
21 $\sigma_{11} = \sigma_{22} = \sigma_r$ and the principal stress in the direction perpendicular to the
22 bedding plane is $\sigma_{33} = \sigma_a$. These loading conditions impose equal lateral
23 strains $\varepsilon_{11} = \varepsilon_{22} = \varepsilon_r$, to the idealized transverse isotropic coal. The coupled
24 equations (1) reduce to the following set
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$$\begin{cases} \sigma_r = (C_{11} + C_{12})\varepsilon_r + C_{13}\varepsilon_a - b_1 p_c - (1 - b_1)s^a(p_m) \\ \sigma_a = 2C_{13}\varepsilon_r + C_{33}\varepsilon_a - b_3 p_c - (1 - b_3)s^a(p_m) \\ \phi_c - \phi_{c0} = [p_c - s_a(p_m)]/N + 2b_1\varepsilon_r + b_3\varepsilon_a \\ n_T = (1 - \phi_{c0})n_m(p_m, \epsilon_m) + \phi_c \rho_b \end{cases} \quad (3)$$

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245 Strains as a function of stresses, fracture pore pressure, and adsorption stress
46 result in
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$$\begin{bmatrix} \varepsilon_r \\ \varepsilon_a \end{bmatrix} = \begin{bmatrix} \frac{1-\nu}{E} & -\frac{\nu_3}{E_3} \\ -2\frac{\nu_3}{E_3} & \frac{1}{E_3} \end{bmatrix} \begin{bmatrix} \sigma_r + b_1 p_c + (1 - b_1)s^a(p_m) \\ \sigma_a + b_3 p_c + (1 - b_3)s^a(p_m) \end{bmatrix} \quad (4)$$

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54 Analogous equations can be developed from Equation (1) for cores drilled
55 parallel to the bedding plane and loaded in a cylindrical triaxial device.
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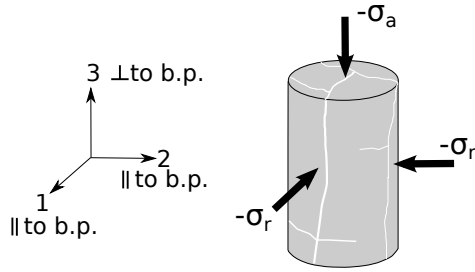


Figure 5: Coal core drilled perpendicular to the bedding plane (b.p.). The triaxial device applies axial stress σ_a and radial stress σ_r which correspond in this case to the seam stresses perpendicular and parallel to the bedding plane respectively.

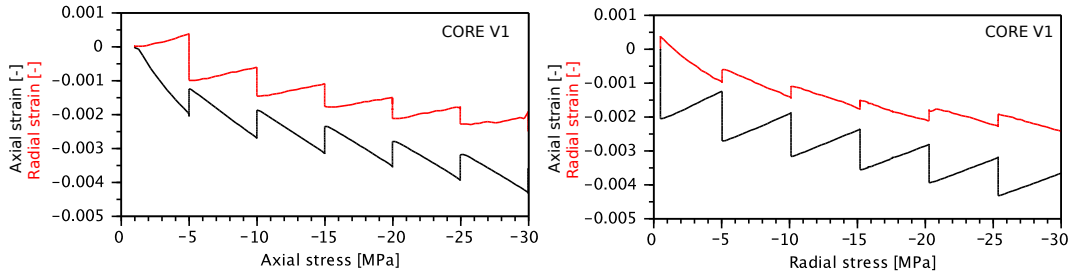


Figure 6: Strain-stress response to drained loading of coal core V1 in dry conditions - no CO_2 in pores. Loading is performed in 5 MPa stress increments (compressive stresses and contraction strains are negative).

249 4.1. Elastic moduli of dry coal specimens

250 Anisotropic loading is applied by steps with increasing axial stress σ_a at
 251 constant radial stress σ_r , and subsequently with increasing radial stress at
 252 constant axial stress. The loading is performed in drained conditions without
 253 CO_2 (no pressure build-up). Figure 6 shows dry loading stress-strain results
 254 from coal core V1.

255 The stress-strain data on cores drilled perpendicular to the bedding plane
 256 lets us recover directly three of the five elastic parameters ($E_3, \nu_3, E/(1-\nu)$)

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257 for linear transverse isotropic elasticity (from Equation 4):

$$\begin{cases} \left. \frac{\partial \varepsilon_r}{\partial \sigma_r} \right|_{p_c, p_m, \sigma_a} = \frac{1-\nu}{E} & ; & \left. \frac{\partial \varepsilon_r}{\partial \sigma_a} \right|_{p_c, p_m, \sigma_r} = -\frac{\nu_3}{E_3} \\ \left. \frac{\partial \varepsilon_a}{\partial \sigma_r} \right|_{p_c, p_m, \sigma_a} = -\frac{2\nu_3}{E_3} & ; & \left. \frac{\partial \varepsilon_a}{\partial \sigma_a} \right|_{p_c, p_m, \sigma_r} = \frac{1}{E_3} \end{cases} \quad (5)$$

258 Stress-strain data from cores drilled parallel to the bedding plane let us
259 recover directly the elastic parameter E . Figure 7 shows the summary of
260 tangent elastic moduli measured for all specimens as a function of axial and
261 radial stresses. Young's elastic moduli values range from ~ 2 to 6 GPa in the
262 range of confining stresses 0.5 to 30 MPa (Figure 7). Core specimens show
263 non-negligible stress-strain nonlinearity, which reduces at confining stresses
264 higher than 15 MPa. Fitting curves represent average stress-dependent elas-
265 tic moduli values that will be used later to estimate secant moduli relevant
266 to the range of testing conditions.

267 4.2. Fracture-induced Biot coefficients

268 Pore-fluid loading is applied by injecting pressurized CO₂. The theoretical
269 strain response to increments of fluid pressure in fractures p_c while keeping
270 constant confining stresses and adsorption stress in the coal matrix (i.e., no
271 variation of the sorbed amount n_r and of the thermodynamic pressure p_m of
272 the fluid in the coal matrix) is derived from Equation (4).

$$\begin{cases} \left. \frac{\partial \varepsilon_r}{\partial p_c} \right|_{\sigma_a, \sigma_r, p_m} = \frac{1-\nu}{E} b_1 - \frac{\nu_3}{E_3} b_3 \\ \left. \frac{\partial \varepsilon_a}{\partial p_c} \right|_{\sigma_a, \sigma_r, p_m} = -\frac{2\nu_3}{E_3} b_1 + \frac{1}{E_3} b_3 \end{cases} \quad (6)$$

273 On the other hand, the stress response of the coal core to CO₂ injection
274 at constant volume and constant adsorption stress conditions is equal to

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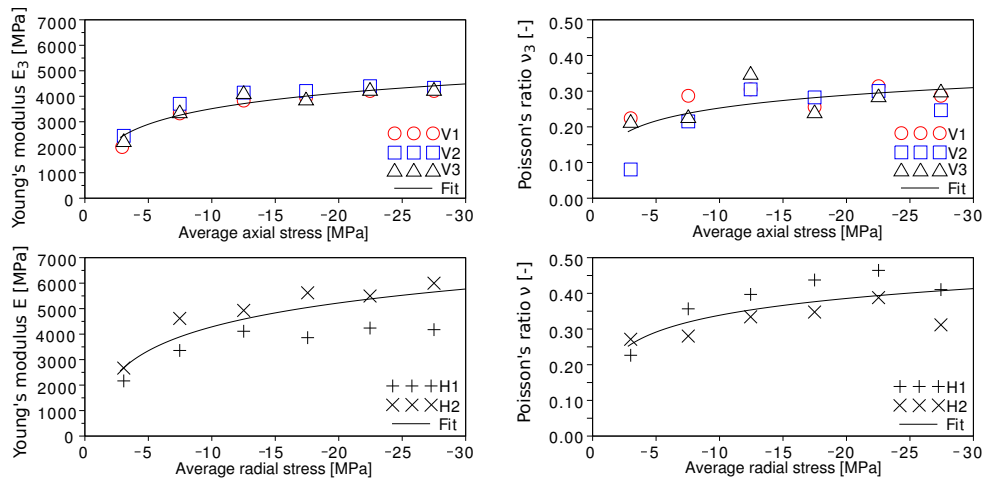


Figure 7: Summary of measured tangent elastic coefficients E , E_3 , ν , ν_3 as a function of axial and radial stress (drained moduli). Fitting functions (Elastic coefficient = $cst_1 + cst_2 \ln(-\text{Stress}/1\text{MPa})$) capture elastic nonlinearity. Coefficients (cst_1 , cst_2) are (1478 MPa, 884 MPa) for E_3 , (1172 MPa, 1352 MPa) for E , (0.133, 0.052) for ν_3 , and (0.182, 0.068) for ν .

$$\begin{cases} \left. \frac{\partial \sigma_r}{\partial p_c} \right|_{\varepsilon_a, \varepsilon_r, p_m} = -b_1 \\ \left. \frac{\partial \sigma_a}{\partial p_c} \right|_{\varepsilon_a, \varepsilon_r, p_m} = -b_3 \end{cases} \quad (7)$$

275 We will note, in particular, that the experimental determination of co-
 276 efficients b_1 and b_3 is difficult because of the overlap of the kinetics of fluid
 277 transfer through the specimen fractures and fluid transfer from/to the coal
 278 matrix. These values will be estimated in the following section from the total
 279 response of coal cores to CO₂ injection including the adsorption response.

280 *4.3. Adsorptive properties*

281 The adsorptive properties of coal cores are tested with CO₂. CO₂ amount
 282 in bulk conditions is computed with Span and Wagner (1996) equation of
 283 state. Constant confining stresses are used to measure adsorptive parameters.
 284 Constant volume experiments will be used to validate the model predictions.
 285 Details follow.

286 *4.3.1. Adsorption under constant confining stresses*

287 In this type of experiments, a pump injects CO₂ at an objective constant
 288 CO₂ pressure (controlled by a pump upstream), while keeping constant to-
 289 tal stress conditions (controlled by axial and radial stress manipulated by
 290 hydraulic servo controllers). Figure 8 shows an example of the time history
 291 response of coal core V1 to CO₂ injection (experiment C.S.V1 - Table 2).
 292 The experiment starts by increasing confining stresses to the level at which
 293 will be kept constant during the entire test ($\sigma_r = \sigma_a = -6$ MPa in this case
 294 – increase not seen in the figure). Then, CO₂ is injected into the specimen at
 295 the objective pressure. CO₂ enters the core from the bottom cap and exits

Table 2: Summary of results of CO₂ injection experiments performed at constant stress conditions.

Test type and specimen	Obj. CO ₂ pressure [MPa]	Total radial stress σ_r [MPa]	Total axial stress σ_a [MPa]	Total CO ₂ uptake n_T [mol/L]	Change of strain $\Delta\varepsilon$ \perp B.P. [-]	Change of strain $\Delta\varepsilon$ \parallel B.P. [-]
C.S.V1	5	-6	-6	1.73	+0.0077	+0.0061
C.S.V2a	10	-11	-12	2.83	+0.0086	-
C.S.V2b	10	-12	-12	2.95	+0.0082	+0.0102
C.S.V3	10	-12	-12	2.98	+0.0100	+0.0082
C.S.H1	5	-7	-7	1.42	-	+0.0058
C.S.H2	10	-11	-12	3.37	-	+0.0072

from the top loading cap. The pipe which exits from the top loading cap is closed with a needle valve, thus, there is a finite dummy volume in the downstream side of the specimen. The fluid pressures at loading caps are continuously measured and are named p_{US} for the pressure upstream and p_{DS} for the pressure downstream (See Figure 8-a). Additional data include axial and radial strains (Figure 8-b) and the amount of injected CO₂ measured from the upstream injection pump (Figure 8-c). The injected amount of CO₂ is corrected to subtract the amount which slowly diffuses through the nitrile membrane. This rate is well identified in experiment signatures and the value ranged from 0.015 to 0.045 mol/day depending on CO₂ pressure and radial effective stress. In adsorption experiments, the specimen is left about 6 days to equilibrate with CO₂. This period of time was sufficient to achieve steady-state conditions according to our strain measurements.

We identify two main regimes during CO₂ injection into the coal core

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310 under constant confining stresses: (1) CO₂ enters fractures and macropores
11 by advection as a function of core fracture permeability, and causes a poroe-
12 lastic expansion due to decrease in effective stress, and (2) CO₂ reaches the
13 coal matrix by advection and diffusion and adsorbs onto the coal matrix,
14 which leads to adsorption-induced swelling (Figure 8-b,c). Both regimes pro-
15 mote core expansion (positive strain) and uptake of CO₂ (positive amount
16 of injected CO₂). Fluid pressure upstream equilibrates with fluid pressure
17 downstream in ~30 min in the experiment shown in Figure 8. The fluid
18 pressure in fractures is expected to be the same everywhere at this point.
19 Afterwards, adsorption-induced swelling prevails. The characteristic time t^*
20 (set at $p_{DS} = 95\%p_{US}$ - based on Figure 8) helps establishing a rough es-
21 timate of the CO₂ uptaken and the swelling strain induced by each type of
22 porosity (macropores and micropores).
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323 Figure 9 shows the data from Figure 8 plotted independently of time.
324 Plotting strains and the injected amount of CO₂ as a function of pressure
325 (Figure 9-a,b) facilitates identifying the overlapping response of fluid filling
326 the cleat pore space and fluid adsorbing on the coal matrix. The reference
327 strains and injected amount of CO₂ measured at the time t^* when there is no
328 more flow through the core are also plotted with point lines in Figure 9-a-d.

329 The data collected from Figures such as 8 and 9 let us measure the to-
330 tal change of strain caused by CO₂ injection and the total CO₂ delivery to
331 the core assembly. The delivery of CO₂ to the piping system (upstream
332 3.9 cm³ and downstream 3.3 cm³) must be subtracted to estimate the net
333 amount of total CO₂ uptake in the coal specimen. In the case of experiment
334 C.S.V1, the coal total uptake is 0.1750 mol - 0.025 mol = 0.150 mol or 1.73

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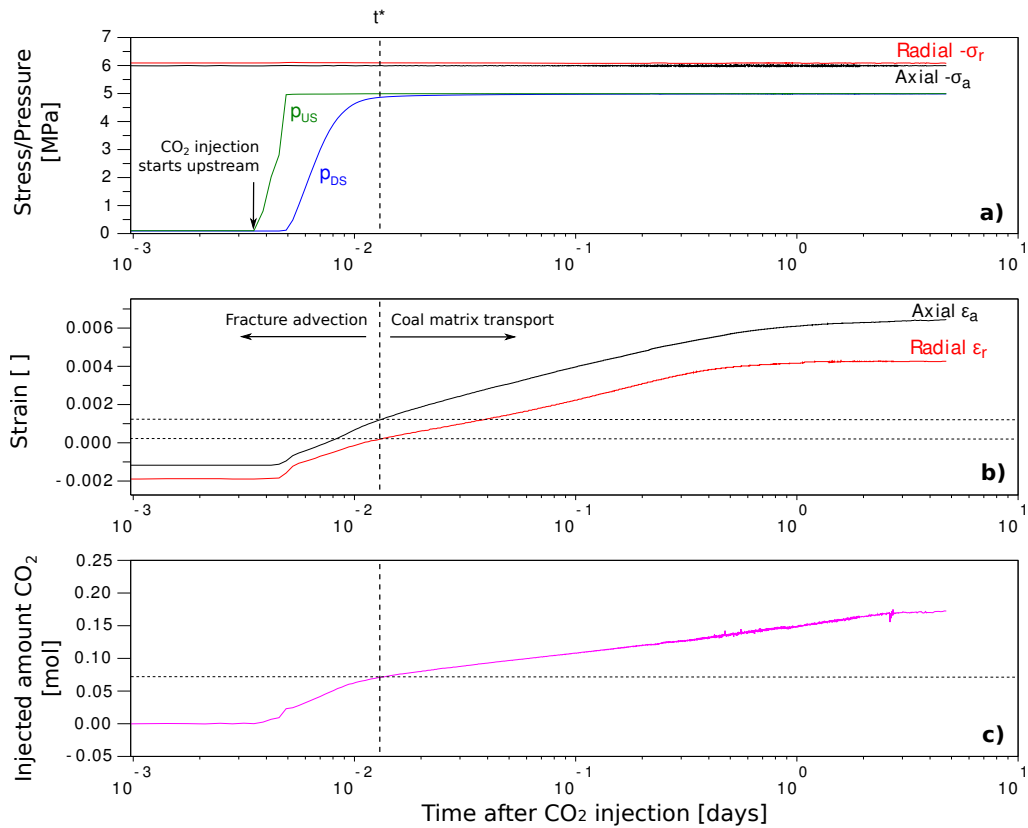


Figure 8: Time evolution of (a) fluid pressure, (b) strains, and (c) injected amount of CO₂ injection experiment C.S.V1 at constant confining stresses $\sigma_r = \sigma_a = -6$ MPa. The objective injection pressure is $p_{US} = p_{DS} = 5$ MPa. Temperature is 39.5°C.

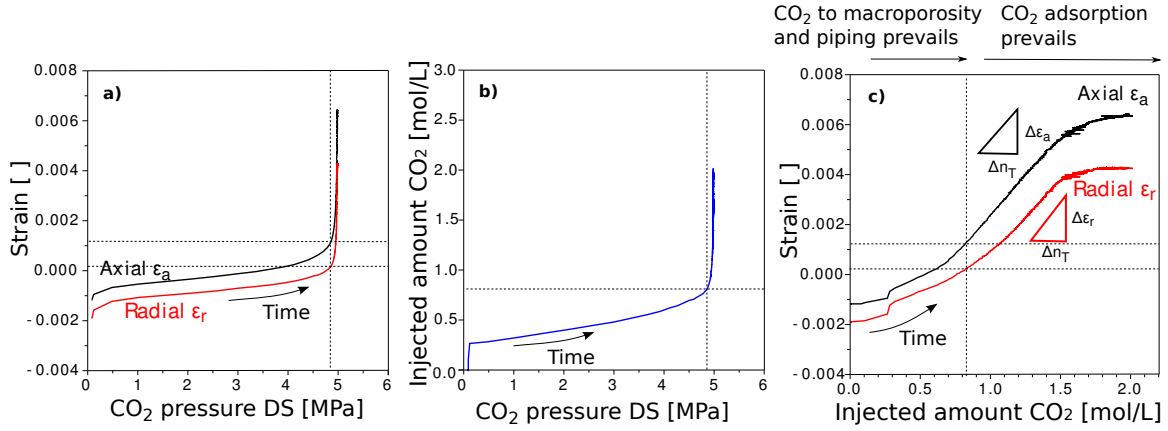


Figure 9: Response of specimen V1 (undeformed volume 86.8 cm³ - experiment C.S.V1) to CO₂ injection at constant confining stresses (same data as in Figure 8). Measurements are plotted independently of time: (a) strain and CO₂ pressure downstream, (b) injected amount per unit volume and CO₂ pressure downstream, and (c) strain and injected amount per unit volume.

mol/L measured per unit volume, and the total change of strain after 5 days is $\epsilon_a \sim +0.0077$ and $\epsilon_r \sim +0.0061$ (includes poroelastic and adsorption-induced swelling). Let us introduce here an additional measurement which is a function only of the coupling between adsorption and strain: the ratio of change of strain to change of uptake amount per unit volume $\Delta\epsilon_a/\Delta n_T$ and $\Delta\epsilon_r/\Delta n_T$ at adsorption-dominated regimes. We compute these values as secant slopes of strain to total uptake after advection is over (after time t^*) and term them “swelling slopes”. For example, these quantities are 0.0043 (mol/L)⁻¹ and 0.0033 (mol/L)⁻¹, respectively for axial and radial strains in experiment C.S.V1 (Figure 9-c).

Table 2 shows a summary of experimental results for all CO₂ injection experiments performed keeping constant confining total stresses. Figure 10

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347 plots the measured total uptake, total change of strains, and swelling slopes
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348 as a function of CO₂ pressure. All experiments show a consistent trend
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349 of declining differential swelling at incremental amount of adsorption (for
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350 example, at injected amounts of CO₂ greater than 1.5 mol/L in Figure 9-c).
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351 The discussion section explores further this phenomenon.

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352 The total strain response upon injection (10-c) predicted by the porome-
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353 chanical model results from the summation of strains caused by poroelastic
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354 effects in macropores (Equation 6) and the strain response to increments of
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355 adsorption stress s^a (a function of the thermodynamic pressure of the fluid
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356 phase in the coal matrix p_m), at constant confining radial σ_r and axial σ_a
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357 stresses and constant cleat fluid pressure p_c . This latter is equal to,

$$\begin{cases} \left. \frac{\partial \varepsilon_r}{\partial p_m} \right|_{\sigma_a, \sigma_r, p_c} &= \left[\frac{1-\nu}{E} (1-b_1) - \frac{\nu_3}{E_3} (1-b_3) \right] \frac{ds^a}{dp_m} \\ \left. \frac{\partial \varepsilon_a}{\partial p_m} \right|_{\sigma_a, \sigma_r, p_c} &= \left[-\frac{2\nu_3}{E_3} (1-b_1) + \frac{1}{E_3} (1-b_3) \right] \frac{ds^a}{dp_m} \end{cases} \quad (8)$$

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358 The total uptake amount per unit volume (10-b) is obtained from Equa-
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359 tion (3). The adsorption swelling slopes (10-d) are calculated from the full
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360 set of equations through numerical simulation.

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361 The following sequence describes the process to determine the adsorptive-
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362 mechanical parameters of the coal core and coal matrix that fit best the
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363 experimental data according to the developed poromechanical model.

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364 1. Estimation of core elastic moduli in dry conditions: We calculate the
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365 secant moduli from Figure (7) for a Terzaghi's effective stress that varies
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366 from 7.5 MPa to 1 MPa (approximate range for the set of experiments
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367 in Table 1). The corresponding values are $E = 2736$ MPa, $E_3 = 2551$
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368 MPa, $\nu = 0.267$, $\nu_3 = 0.198$.

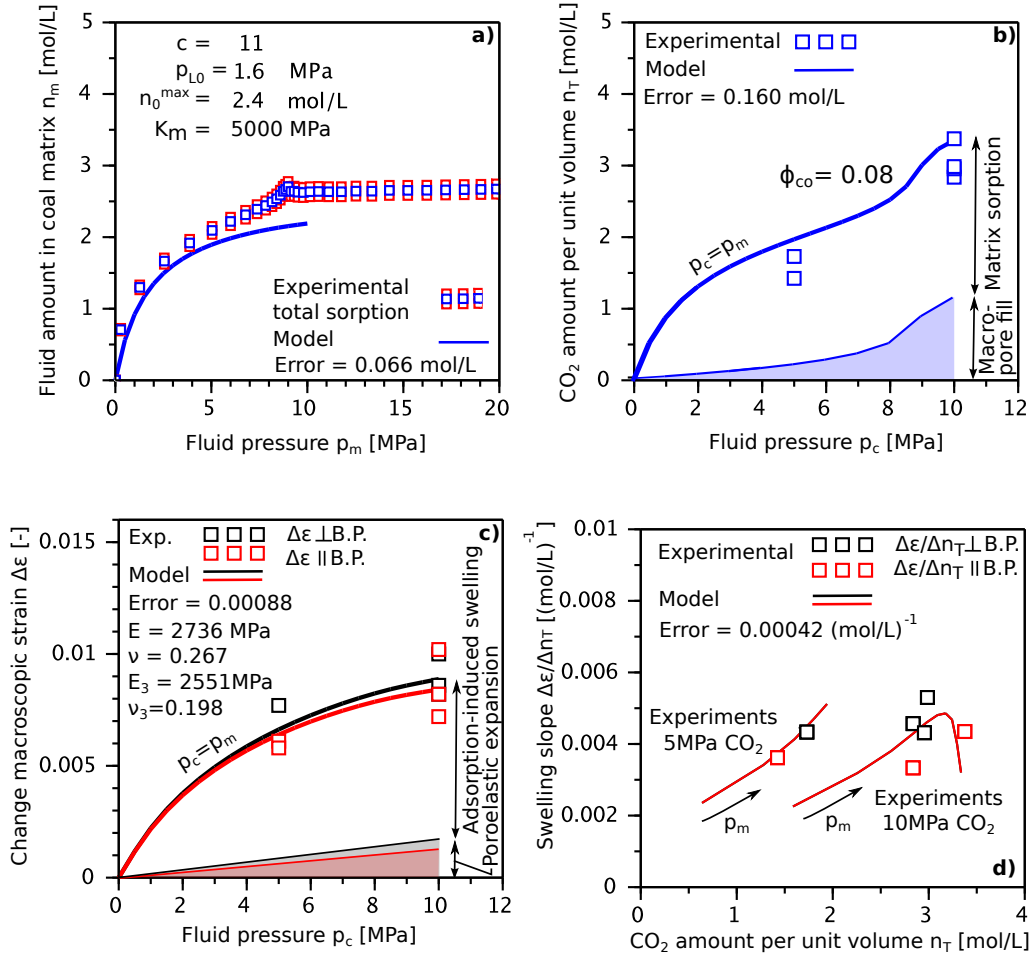


Figure 10: CO₂ uptake and adsorption-induced swelling in coal cores keeping constant confining stresses (See Table 2): experimental values and numerical simulation. Notice that total uptake amounts and change of strains are caused by macropore/fracture poroelastic response and micropore adsorptive response. (a) Coal matrix total sorption amount - from independent manometric tests (red squares indicate uncertainty - Auxiliary Material), (b) core total uptake amount of CO₂ per unit volume, (c) total change of core strains upon CO₂ injection, and (d) ratio of change of swelling strain to change of uptake amount during adsorption at constant pressure in fractures (Δp_c path not shown for model simulation).

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- 369 2. Estimation of coal matrix bulk modulus K_m : We constrain K_m ac-
370 cording to the bulk modulus of the coal core for the highest applied
371 confining stress (~ 30 MPa), which varies between 4000 MPa and 4200
372 MPa. Hence, we adopt an upper bound $K_m = 5000$ MPa. Macro-Biot
373 coefficients and modulus (b_1, b_3, N) are obtained from Equation (A.20).
- 374 3. Estimation of the initial macroporosity ϕ_{c0} : The total initial macroporosity varies from specimen to specimen. Considering that the Helium initial porosity of the set of coal cores varies from 11.2% to 14.4%, image analysis of X-ray slices yields a porosity of fractures (wider than 50 μm) larger than 0.5 %, MPI yields a porosity about 1.5% of pores larger than 0.1 μm , and the microporosity of coal matrix normally ranges from 3% to 10% (Gan et al., 1972; Lin et al., 1978), we set a mean macroporosity $\phi_{c0} = 8\%$ to invert a unique set of coal adsorption parameters. This porosity may seem too high for a coal core, but in this case it comprises pores up to the 0.1 μm scale.
- 384 4. Inversion of the parameters n_0^{max} , p_{L0} , and c ; where n_0^{max} and p_{L0} are parameters of the simplified adsorption isotherm at zero matrix strain $n_{m0}(p_m) = n_0^{max}[p_m/(p_{L0} + p_m)]$ and $c(p_m)$ is fixed to be pressure-independent as suggested by Espinoza et al. (2013). The inversion seeks to fit best (1) the coal matrix total sorption data n_m (these data is collected from an independent sorption experiment on crushed coal from the same coal seam - See Auxiliary Material), (2) the total uptake measured in the triaxial experiments n_T (Table 2), (3) the change of strains upon injection $\Delta\varepsilon$ (parallel and perpendicular to the bedding plane - Table 2), and (4) the measured swelling slopes upon predominant ad-

Table 3: Best fitting measured and inverted model parameters. (Notes: *Indicates uncertainty. The shear modulus perpendicular to bedding C_{44} is not determined in this experimental study.

Core scale	Value		Matrix scale	Value	
E	2736 MPa	Measured	K_m	5000 MPa	Lower Bound
E_3	2551 MPa	Measured	n_0^{max}	2.4 mol/L	Inverted
ν	0.267	Measured	p_{L0}	1.6 MPa	Inverted
ν_3	0.198	Measured	c	11	Inverted
ϕ_0	0.08±0.02	Measured*			

sorption regime $\Delta\varepsilon/\Delta n_T$ (parallel and perpendicular to the bedding plane - Figure 10). The error is calculated with a two-norm criterion as a sum of the relative errors on the prediction of each amount.

The best fitting modeling results are shown in Figure 10. The best fitting parameters are summarized in Table 3. Calculated absolute errors are shown in Figure 10 as well.

Figure 10-b and 10-c show simulation results of total uptake n_T and total strain change $\Delta\varepsilon$ at equilibrium ($p_c = p_m$ at all times). Figures 10-b-c highlights the response expected from each porosity system. Figure 10-d shows the ratio of change of strain to change of total uptake after poroelastic expansion, i.e., change in fracture pressure occurs first (Δp_c) and adsorption-induced swelling follows (Δp_m). This type of simulation (increase of p_c first and p_m later) permits isolating the individual responses of the two porosity systems.

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408 4.3.2. Adsorption under near constant volume conditions and model valida-
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410 The second testing procedure consists on constraining displacements upon
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411 CO₂ injection by imposing a near constant-volume condition. This stress
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412 path is obtained by periodic cancelation of swelling strains upon application
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413 of external total stresses. Figure 11 shows a time history example for exper-
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414 iment C.V.V1b performed at an average objective CO₂ injection pressure of
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415 2 MPa (strictly, this experiment is performed with a pressure gradient for
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416 the sake of measuring permeability: $p_{US} = 2.5$ MPa and $p_{DS} = 1.5$ MPa).
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417 Results show that from an initial condition of total radial stress -3 MPa,
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418 axial total stress -10 MPa and no CO₂, CO₂ injection at near constant-
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419 volume conditions induced a change of radial and axial total stresses of -22
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420 MPa in seven days. Figure 11 shows that the final change in total radial
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421 strain is -0.0001 and the change of axial strain is about $+0.0010$. This flow-
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422 through experiment does not allow a precise measurement of the specimen
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423 CO₂ uptake, hence, it is not reported.

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424 Table 4 summarizes the result of two other similar experiments. The
425 measured change of Terzaghi's effective stress ranges from 19 to 29 MPa.
426 Experiment C.V.V1b was performed to confirm the results from C.V.V1a.
427 Experiment C.V.V3 approached the upper stress limit of the triaxial cell, and
428 hence, constant-volume condition could not be held after 5 hours of starting
429 CO₂ injection. The specimen continued swelling $\Delta\varepsilon_a \sim +0.0007$ and $\Delta\varepsilon_r \sim$
430 $+0.0014$ in 6 days at 36 MPa of hydrostatic confining stress. The fact that
431 the specimen continued swelling indicates that higher confining stresses (>36
432 MPa) should have been applied to keep constant-volume condition. This was

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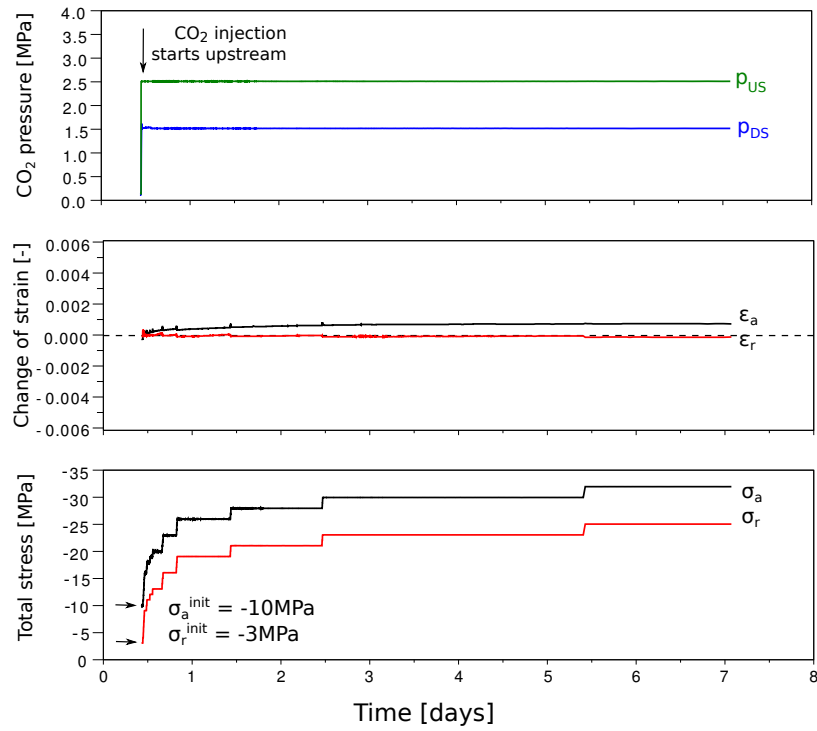


Figure 11: Time history of strains and total stresses upon CO₂ injection in core V1 keeping near constant volume conditions (Experiment C.V.V1b - Table 4).

Table 4: Summary of experimental results of CO₂ injection in cores performed at near constant volume conditions. (*Average pressure)

Test type and specimen	CO ₂ objective pressure [MPa]	Initial radial stress [MPa]	Initial axial stress [MPa]	Total CO ₂ uptake [mol/L]	Final radial stress [MPa]	Final axial stress [MPa]	Change in radial strain [-]	Change in axial strain [-]
C.V.V1a	2*	-3	-10	NA	-23	-30	-0.0005	-0.0002
C.V.V1b	2*	-3	-10	NA	-25	-32	-0.0001	+0.0010
C.V.V3	5	-2	-2	1.67	-36	-36	+0.0007	+0.0014

not a flow-through experiment, therefore it was possible to estimate the total uptake equal to 1.67 mol/L. It includes adsorption during swelling at 36 MPa. Less uptake amount would be expected in perfect isochoric conditions.

Let us now analyze the previous results in light of the double porosity poromechanical model and validate the parameters measured previously. Equation (7) shows that an injection of pore fluid under constant-volume condition ($\Delta\varepsilon_a = \Delta\varepsilon_r = 0$) results in an increase of compressive stress proportional to the macro Biot coefficients b_1 and b_3 . Since b_1 and b_3 are less than one, the increase in compressive stress should be at most equal to the change in fluid pressure. Yet, adsorption-induced stresses must be taken into account in the case of coal. The increase in total stress due to solely changes in adsorption stress (ds^a) can be obtained from Equation (3) and can be termed as a “macroscopic adsorption-induced stress” (macroscopic because weighed by $(1 - b_i)$). The rate of stress change respect to p_m is equal to

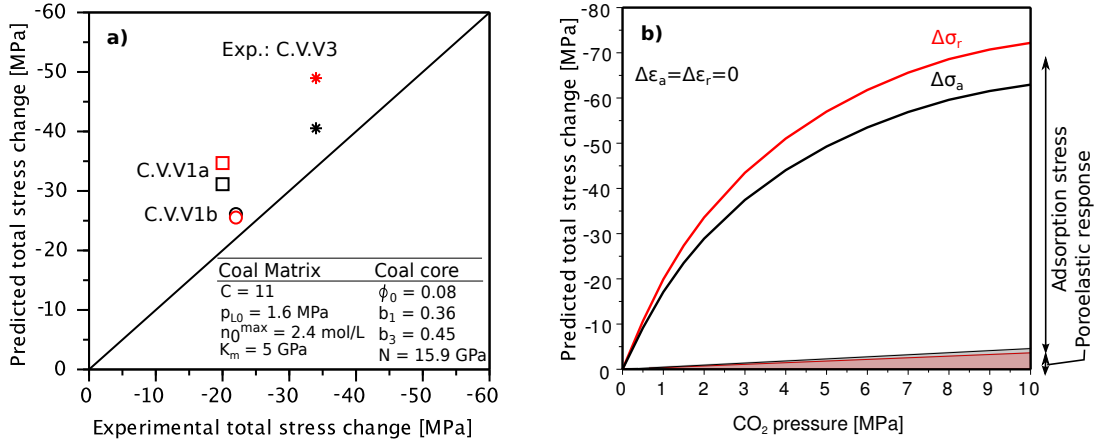


Figure 12: Change of total stress in coal cores induced by CO₂ injection at near constant-volume condition: (a) comparison between experimentally measured values and model predictions and (b) model-predicted change of stress at perfect constant-volume condition.

$$\begin{cases} \left. \frac{\partial \sigma_r}{\partial p_m} \right|_{\varepsilon_a, \varepsilon_r, p_c} = -(1 - b_1) \frac{ds^a}{dp_m} \\ \left. \frac{\partial \sigma_a}{\partial p_m} \right|_{\varepsilon_a, \varepsilon_r, p_c} = -(1 - b_3) \frac{ds^a}{dp_m} \end{cases} \quad (9)$$

Figure 12-a displays the experimentally measured changes in total stress upon CO₂ injection (mostly adsorption stresses) and the model predictions (Equation 3) based on the measurements of drained loading of dry coal specimens (secant moduli $E = 3998 \text{ MPa}$, $E_3 = 3396 \text{ MPa}$, $\nu = 0.333$, $\nu_3 = 0.248$ for a range of effective stress ~ 1 to 25 MPa), the estimated average macroporosity $\phi_{c0} = 0.08$, the lower bound for $K_m = 5000 \text{ MPa}$, and the inverted parameters ($n_{max}^0 = 2.4 \text{ mol/L}$, $p_{L0} = 1.6 \text{ MPa}$, $c = 11$) from swelling experiments at constant confining stress. The simulation results take into account that the sample was not strictly kept in isochoric conditions (i.e., includes experimental strains summarized in Table 4). All experimental values are between 3 to 16 MPa less compressive than the values predicted by the

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10 458 model. The discussion section provides further insight into the mismatch of
11 459 these results. Figure 12-b shows the change of stress predicted under per-
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13 460 fect isochoric condition ($\varepsilon_a = \varepsilon_r = 0$) as a function of CO₂ pressure. The
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15 461 adsorption-induced stress is about 60 MPa at 10 MPa of CO₂ pressure.
16

17 18 462 *4.4. Permeability measurements*

19
20 463 We measured cleat permeability at constant flow rate regimes assuming
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22 464 Darcy compressible fluid flow (viscosity obtained from webbook.nist.gov).
23
24 465 Fluid flow through cleats occurs at pressures higher than 1 MPa and mainly
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26 466 through micron-sized fractures. Therefore, Klinkenberg effect is disregarded.
27

28 29 467 *4.4.1. Permeability of a CO₂-equilibrated core – Effect of effective stresses*

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31 468 We explored the permeability of specimen V1 to CO₂ in axial direction
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33 469 (on the plane of vertical fractures in the seam) varying radial and axial
34
35 470 stresses at mean fracture fluid pressure of ~ 5.25 MPa, and pressure gradient
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37 471 of ~ 1.5 MPa over the length of the core. In order to cancel out the effect of
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39 472 adsorption stresses, the specimen is equilibrated with CO₂ for ~ 6 days at 5
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41 473 MPa prior to permeability measurements, letting the coal matrix swell under
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43 474 constant confining stresses. Thereafter, a constant CO₂ gradient is imposed
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45 475 (upstream 6 MPa and downstream 4.5 MPa). The gradient is kept constant
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47 476 and relatively small to avoid changes in adsorption-induced stresses in time
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49 477 and in space throughout the specimen length. The effect of change of flow
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51 478 rate because of transient mechanically-induced desorption or sorption was
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53 479 neglected (this phenomenon is explained elsewhere (Hol et al., 2012)).

54
55 480 Figure 13 shows measured permeability as a function of radial and axial
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57 481 Terzaghi's effective stresses. The precision of permeability measurements
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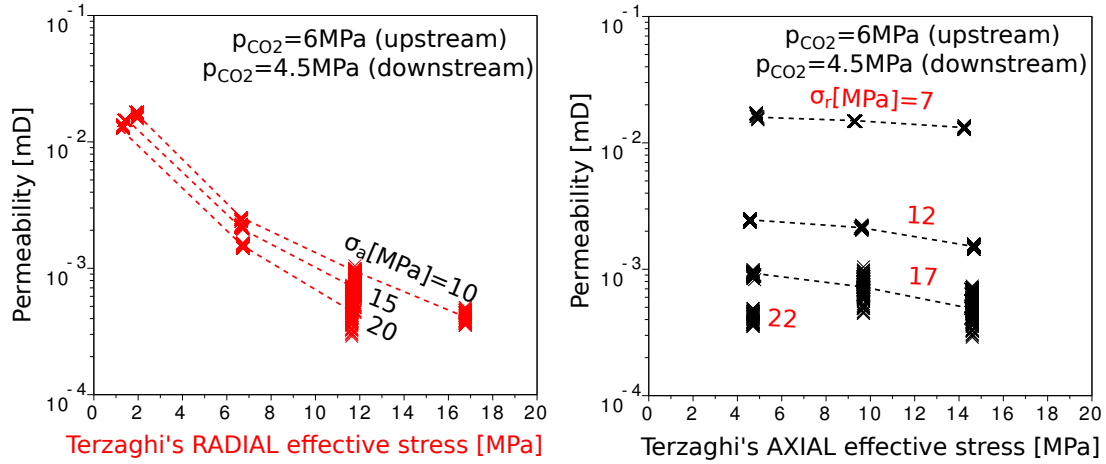


Figure 13: CO₂ permeability of specimen V1 subjected to various levels of effective stress in radial and axial directions. The dashed lines join data points at the same axial (left) or radial stress (right).

decreased with increasing effective stress, because the gradient of pressure was constant at all times. The value of fracture compressibility parameters which best fit Eq. (2) are $\alpha_r = -0.269 \text{ MPa}^{-1}$, $\alpha_a = -0.067 \text{ MPa}^{-1}$, and $k_0 = 0.033 \text{ mD}$ at zero Terzaghi's effective stress in radial and axial directions. The fitting is far from perfect but captures the main trend within the fitted interval (up to $\sigma'_r \sim 17 \text{ MPa}$ and $\sigma'_a \sim 15 \text{ MPa}$). The axial permeability of vertically cored specimens is about 4 times more sensitive to radial than to axial effective stresses. The measured stress sensitivity of permeability is within the range of values measured in the literature -0.23 to -0.46 MPa^{-1} (Somerton et al., 1975).

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492 4.4.2. *Permeability evolution of a core during CO₂ adsorption at near con-*
10 *stant volume condition*
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13 494 In order to resemble the conditions found in coal seams in-situ, we mea-
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15 495 sured coal core permeability during the process of adsorption while keeping
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17 496 near constant-volume condition. Constant-volume condition was chosen over
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19 497 zero-lateral strain condition to avoid shearing the specimen. Figure 14 shows
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21 498 the permeability measured during test C.V.V1b, for which the CO₂ pressure
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23 499 was established upstream at $p_{US} = 2.5$ MPa and downstream at $p_{DS} = 1.5$
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25 500 MPa (Figure 11-a). Permeability decreases more than two orders of magni-
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27 501 tude from 0.02 mD to 0.0001mD as a result of increased Terzaghi's effective
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29 502 stresses as a result of adsorption.

30 503 Let us now predict the variation of permeability for this experiment, based
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32 504 on the previously measured stress-permeability relationship (parameters from
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34 505 Figure 13) and the predicted induced adsorption stresses using the porome-
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36 506 chanical model (Equation 3). Total stresses σ_a and σ_r can be calculated for
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38 507 any equilibrium pressure or combination of p_c and p_m using Equation (3).
39
40 508 Thus, simply replacing the Terzaghi's effective stress $\sigma'_{ij} = -(\sigma_{ij} + p_c)$ in the
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42 509 permeability Equation (2) allows us to calculate the permeability evolution
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44 510 of the coal specimen upon adsorption. The results are plotted in Figure 14.
45
46 511 The predicted final effective radial and axial stress are about 27MPa and the
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48 512 predicted permeability about 3 nD. In Figure (14), the solid line indicates
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50 513 the predicted permeability in the range of stress-permeability fitted from ex-
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52 514 perimental measurements (up to ~ 17 MPa radial effective stress) and the
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54 515 dashed line shows the extrapolated permeability out of this fitting range.
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56 516 The discussion section revisits the overestimation of permeability reduction.

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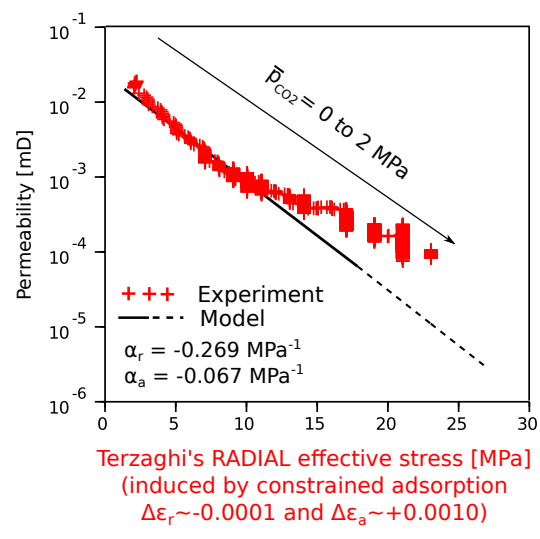


Figure 14: CO₂ permeability evolution of core V1b subjected to constrained adsorption under near constant-volume condition. The specimen develops stresses as a result of CO₂ injection and constrained swelling. Ensuing stresses lower measured permeability up to 100 times. Predicted permeability is calculated based on the change of effective stresses calculated with Equation (3) and permeability parameters from best fit of independent data in Figure (13). The dashed line indicates values of permeability extrapolated out from the experimental fitting range.

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9 517 **5. Discussion**

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11 518 *5.1. Experimental testing*

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14 519 *5.1.1. Specimen representativity*

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16 520 Measurements of rock properties from laboratory tests are usually biased
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18 521 by the length scale of specimens compared to the spatial variability and
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20 522 effective physical properties at reservoir scale. Major fractures such as master
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22 523 cleats cannot be sampled in specimen cores for triaxial testing. Hence, lower
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24 524 elastic moduli and higher permeability are expected at the seam scale than
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26 525 the values measured in core laboratory specimens.

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28 526 An additional non-trivial feature affected by length scale is mechanical
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30 527 anisotropy. Coal laboratory tests in this article and in the literature (Day
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32 528 et al., 2010; Morcote et al., 2010; Espinoza et al., 2013) and shale (Wang,
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34 529 2002) indicate that Young’s modulus in direction perpendicular to the bed-
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36 530 ding plane is lower than in direction parallel to the bedding plane $E_{\perp} < E_{\parallel}$, or
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38 531 equivalently that the Thomsen parameter $\epsilon^* = (C_{\parallel} - C_{\perp})/(2C_{\perp})$ is greater
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40 532 than zero (Thomsen, 1986). However, most master cleats are perpendicu-
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42 533 lar to the bedding plane (Laubach et al., 1998). This fact suggests that
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44 534 there could be a mechanical anisotropy reversal such that at the seam scale
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46 535 $E_{\perp} > E_{\parallel}$.

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48 536 Permeability measured in direction perpendicular to the bedding plane is
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50 537 expected to differ from the permeability in direction parallel to the bedding
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52 538 plane because of the tortuosity (spatial organization) of the cleat network
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54 539 and of the presence of interbedded low-permeability clay-rich layers. Scale
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56 540 issues are still present. In-situ measured seam permeability ranges from
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58 541 0.1 to 100 mD (Scott et al., 2012), which is significantly higher than the

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9 542 permeabilities we measured in the laboratory, which range from 0.0001 to 0.01
10 543 mD. Statistical analyses can help upscale laboratory measured permeability
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12 544 (Lake and Srinivasan, 2004).
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16 545 *5.1.2. Porous structure: a well defined double porosity system?*
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18 546 Various experimental methods suggest the presence of a complex network
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20 547 of pores that is far from being easily divided into two well defined porosity
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22 548 systems. Naked-eye visual observation permits identifying fractures as large
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24 549 as $\sim 500 \mu\text{m}$. X-ray tomography and SEM microscopy permit observing
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26 550 smaller fractures in the range of $200 \mu\text{m}$ to $5 \mu\text{m}$. MIP and N_2 adsorp-
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28 551 tion confirm the presence of small fractures, mesopores and micropores that
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30 552 extend from $5 \mu\text{m}$ down to the nm scale (Figure 3).
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32 553 For the sake of modeling and conceptualization, we have traced a line
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34 554 dividing microporosity and macroporosity based on the predominant flow
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36 555 regimes and poromechanical response. The simplified macroporosity system
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38 556 is the one which is most conductive and has a negligible adsorption-induced
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40 557 mechanical response. The simplified microporosity system is constituted by
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42 558 pores that can uptake fluid at constant pressure in the macropore system and
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44 559 whose solid part can develop a strong adsorption-induced response (strains
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46 560 or stresses). Thus, the definition of macro and microporosity is based on the
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48 561 conceptual ideas presented in Section 3, but may also depend on the length
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50 562 scale of analysis.
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9 563 *5.1.3. Experimental results: uncertainties, unexpected phenomena, and main*
10 *findings*
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13 565 The measurement with the highest uncertainty is the total amount of up-
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15 566 take because of diffusion of CO₂ through the confining membrane. During the
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17 567 time length of experiments, the diffused amount could add up to an amount
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19 568 as large as the effective total uptake amount. Fortunately, these “leaks” were
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21 569 quite steady and measurable. Yet, the uncertainty is estimated to be as large
22
23 570 as 0.5 mol/L. A not-perfectly sealing but deformable membrane performed
24
25 571 better to measure mechanical properties than a tight but rigid membrane,
26
27 572 e.g., lead membrane. The uncertainty in determining total amount of uptake
28
29 573 impacts directly on the determination of the adsorption-strain coupling co-
30
31 574 efficient c , n_{max}^0 , and p_{L0} . We were able to reduce the uncertainty of these
32
33 575 inverted parameters by using additional sorption data on crushed coal from
34
35 576 the same coal seam (Figure 10-a).

36 577 We observed high elastic nonlinearity at low effective stress, as a result of
37
38 578 existing fractures. Nonlinearity lessened at high effective stress as fractures
39
40 579 closed. Using N₂ or He would have facilitated the measurement of porome-
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42 580 chanical properties and macro Biot parameters (b_1, b_3), with less influence of
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44 581 adsorption-induced response. Yet each fluid would yield its own “porome-
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46 582 chanical signature” because each fluid has different affinity with coal surfaces
47
48 583 and can even access different pore sizes. Therefore, we preferred to test only
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50 584 with CO₂.

51 585 The time required for CO₂ uptake and coal equilibration was based on the
52
53 586 measured specimen response through either strains or stresses. The allowed
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55 587 equilibration time was enough to reach asymptotic trends in induced strains
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588 or stresses suggesting that most of the sorption process had already hap-
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11 589 pened. These measurements were adequate to measure the required model
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13 590 parameters. Natural fractures facilitated relatively short equilibration times
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15 591 from a few days to 10 days. Having a non-fractured core would have made
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17 592 measurements much lengthier if not impossible in a practical time frame.

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19 593 The change in swelling slope (Figure 9) in tests performed during adsorp-
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21 594 tion at constant confining stress was a non-expected result. This reduction,
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23 595 observed in other experiments as well, may be due to coal softening (reduc-
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25 596 tion of elastic moduli), as observed previously in the literature (Czaplinski
26
27 597 and Holda, 1982; Masoudian et al., 2014) or CO₂-enhanced coal creep as
28
29 598 documented by Hagin and Zoback (2010) while adsorption-induced swelling
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31 599 is still taking place.

32
33 600 Measured poroelastic and adsorption stresses have an uncertainty of ± 1
34
35 601 MPa. Controlling pressure pumps react with step changes of 1 MPa to
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37 602 changes in strain of 10 μm , i.e., equivalent radial strain $\varepsilon \sim 5 \times 10^{-4}$. The
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39 603 compressibility of the membrane is included in computing radial strains. The
40
41 604 corresponding uncertainty is less than 0.7 $\mu\text{m}/\text{MPa}$.

42 43 605 *5.2. Modeling*

44 45 606 *5.2.1. Main assumptions*

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47 607 A sharp division between micro and macroporosity is not truly realistic (as
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49 608 discussed in Section 6.1.2) but is useful for the sake of isolating predominant
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51 609 phenomena at different length scales. Figures 10 and 12 help put in evidence
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53 610 the response of each porosity system. The double porosity systems is well
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55 611 suited for fluid flow simulation in media with heterogeneous permeability.

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612 Both the coal matrix and the fractured coal may exhibit anisotropic prop-
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613 erties (Pan and Connell, 2011). Non-fractured coal specimens and small
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614 cores generally develop anisotropy controlled by lamination planes $E_{\parallel} > E_{\perp}$
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615 (Espinoza et al., 2013). Although it is possible to develop a model with
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616 anisotropy at both the coal matrix and fractured coal scales, in practical
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617 terms, it is convenient to group anisotropy at one of the two scales. Herein,
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618 we group anisotropy at the macro scale in views of analyzing mechanical
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619 anisotropy induced by master cleats at the fractured coal scale.
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620 5.2.2. Parameter inversion

621 The most important parameters in defining the coupled response are the
622 fractured coal elastic moduli C_{ij} , and coal matrix parameters n_0^{max} and $c(p_m)$,
623 both of which define adsorption stress. Previous model validation studies for
624 coal matrix specimens showed average coupling coefficient $c(p_m)$ for various
625 coal specimens subjected to CH₄ or CO₂ swelling ranging from 5 to 10,
626 with the highest values corresponding to CO₂ and the lowest ones to CH₄
627 (Brochard et al., 2012; Nikoosokhan et al., 2012; Espinoza et al., 2013). The
628 adsorption-strain coupling coefficient is fixed pressure-independent $c(p_m) = c$
629 in this article as suggested by Espinoza et al. (2013). In fact rather than
630 only c , the ratio c/K_m is found to be the one that permits fitting observed
631 adsorption strains and stresses. The coal matrix bulk modulus K_m was fixed
632 to a constant value in this article. Cleat porosity (macroporosity) varies for
633 different samples and would vary also depending on the scale of analysis.
634 Here we chose a constant value $\phi_c = 0.08$ in order to invert a unique set of
635 parameters. Cleat porosity has a second-order importance in fitting induced
636 strains (Figure 10-c) and does not play a role in fitting swelling slopes (Figure
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11 638 The determination of 10 independent parameters is challenging even in
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13 639 well controlled laboratory experiments, which limits the applicability of the
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15 640 full model to industry practice. However, the model remains as a powerful
16
17 641 tool for investigating the effect of anisotropy in the permeability evolution of
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19 642 coal seams.

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22 643 *5.2.3. Model predictions*

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24 644 Implementing the fitted parameters in the model captures well various
25
26 645 measurements including total uptake amount, induced strains/stresses and
27
28 646 swelling slopes. The same parameters (except for elastic moduli) overesti-
29
30 647 mate by 3 to 16 MPa the induced adsorption-induced stress when displace-
31
32 648 ments are constrained upon adsorption. This overestimation may be due to
33
34 649 heterogeneity of coal from sample to sample and measured elastic moduli,
35
36 650 an inaccurate estimation of secant elastic moduli, or also to the fact that
37
38 651 coal may soften with significant amounts of sorbed CO₂ with concomitant
39
40 652 reduction of induced stresses. Creep may also play a role in canceling the
41
42 653 swelling strains as discussed before.

43 654 The overestimation in reduction of permeability (Figure 14) stems from
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45 655 adopting a linear log(permeability)–stress law. Predictions are good in the
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47 656 interval at which the permeability is fitted from experimental data, but ex-
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49 657 trapolations deviate beyond the fitting range. More complex models can
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51 658 capture a “hard permeability” which makes permeability plateau at high ef-
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53 659 fective stress, and thus capture the nonlinear dependence of the logarithm
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55 660 of permeability as a function of effective stress (Liu and Rutqvist, 2010). A
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57 661 simple logarithmic correlation based on Terzaghi’s effective stress seems to

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662 be appropriate over a defined stress range. Yet, coal seams subjected to CH₄
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663 depletion and/or CO₂ injection under in-situ conditions may experience sig-
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664 nificant shear stresses which promotes fracture slip and dilation and coal fines
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665 production with a significant impact on seam permeability as well (Palmer,
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16
666 2008; Okotie and Moore, 2010).

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667 *5.3. Implications on coal bed methane production and CO₂ geological storage*

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668 Our stress-based coupled model offers new insights into the prediction of
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669 natural gas recovery and injectivity during CO₂ injection in unmineable coal
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670 seams. The model can predict order of magnitude changes in permeability
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27
671 applying the appropriate boundary conditions.

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672 We have measured significant changes of stress associated with sorption
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673 under constrained condition, termed as adsorption-induced stresses. Con-
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33
674 versely, desorption is expected to cause stress relaxation. If boundary condi-
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675 tions are close to oedometric (constant vertical stress and zero lateral strain),
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676 then a significant decrease in horizontal stress at constant vertical stress
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677 would be expected upon desorption with ensuing increase of stress anisotropy
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678 that could lead to shear failure and fines production (Okotie and Moore,
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43
679 2010).

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680 **6. Conclusions**

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681 Meaningful coal testing is challenging because of the difficulties in ob-
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682 taining representative fractured coal cores, separating the competing frac-
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683 ture advection and matrix adsorption phenomena, and reproducing reservoir
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684 in-situ conditions during testing in the laboratory.
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685 We performed tests on moderately fractured sub-bituminous/bituminous
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686 coal cores and used CO₂ as a proxy for CH₄. Experimental results from
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687 triaxial testing show that (1) fractured coal has a significant non-linear elastic
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688 behavior, (2) fractured coal cores show mechanical anisotropy, (3) swelling
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689 strains, swelling stresses and total uptake are affected by the response of the
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690 macroporous and microporous systems, and (4) permeability is four times
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691 more sensitive to radial (lateral) stress than to axial (vertical) stress. Overall,
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692 experimental results highlight the difference between the two porous systems
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693 through their poromechanical response. Experiments under near constant
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694 volume condition show that effective stresses increase significantly during
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695 adsorption. The increase in effective stress is the main driver for changing
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696 core permeability. The measured changes of effective stress was as high as
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697 +29 MPa for CO₂ injection at 5 MPa of pressure.

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698 We developed a double porosity poromechanical model to capture the
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699 adsorptive-mechanical response of transverse isotropic fractured coal. The
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700 adsorptive-mechanical coupling is modeled through an adsorption stress func-
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701 tion which depends on the coal matrix properties. The model is fully cou-
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702 pled, therefore, it permits quantifying the influence of adsorption on strain
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703 and stresses as well as the influence of stresses and strains on adsorption.
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704 The discrimination of two porosity levels permits isolating poromechanical
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705 responses and calculating fluid amounts from either fractures or the coal
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706 matrix. Permeability is calculated as function Terzaghi's effective stress.

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707 The developed double porosity model let us recognize the key porome-
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708 chanical properties at each scale and measure relevant parameters. Core
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709 scale measurements and CO₂ injection experimental data matching let us

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710 recover the mechanical-adsorptive properties of the coal matrix such as bulk
711 modulus, isochoric adsorption isotherm, and the adsorption-strain coupling
712 coefficient. The model predicts significant adsorption-induced stresses and
713 order-of-magnitude changes in permeability as measured experimentally.

714 **Acknowledgements**

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718 to H. Daigle who provided the N₂ adsorption results, and to the anonymous
719 reviewers who helped improving this article.

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9 **7. Symbols**

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α	Fracture compressibility coefficient for permeability law [MPa ⁻¹]
ϵ_m	Volumetric strain of the coal matrix [-]
ϵ_{ij}	Strain tensor defined at the fractured coal scale [-]
ϕ_c	Cleat porosity or macroporosity [-]
μ	Chemical potential [J/mol]
ν, ν_3	Poisson's ratio parallel and perpendicular to bedding [-]
σ_{ij}	Total stress tensor defined at the fractured coal scale [Pa]
σ'_{ij}	Terzaghi's effective stress tensor defined at the fractured coal scale [Pa]
ρ_b	Bulk molar density of the fluid [mol/L]
b_1, b_3	Biot coefficient parallel and perpendicular to bedding [-]
c	Adsorption-strain coupling coefficient [-]
k	Permeability [darcy]
k_0	Reference permeability at zero effective stress [darcy]
n_m	Fluid amount in the coal matrix per unit volume of coal matrix [mol/L]
n_0^{max}	Asymptotic parameter of constant matrix volume Langmuir isotherm [mol/L]
n_r	Fluid amount in the coal matrix per unit volume of fractured coal [mol/L]
n_T	Total fluid amount per unit volume of fractured coal REV [mol/L]
p_c	Pressure of fluid in cleats [Pa]
p_m	Thermodynamical pressure of fluid in the coal matrix [Pa]
p_{L0}	Langmuir pressure of constant matrix volume Langmuir isotherm [Pa]
s^a	Coal matrix adsorption stress [Pa]
C_{ij}	fractured coal stiffness tensor coefficient [Pa]
E, E_3	Young's modulus parallel and perpendicular to bedding [Pa]
K_m	Bulk modulus of the coal matrix [Pa]
\bar{V}_b	Molar volume of the fluid in bulk conditions [L/mol]

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722 **Appendix A. Model derivation**

723 The model derivation extends the model developed by Nikoosokhan et al.
724 (2012, 2014) to consider transverse isotropy. The model is based on energy
725 conservation and follows from a thermodynamical formulation.

726 *Appendix A.1. Energy balance equation*

727 Consider a representative elementary volume (REV) of fractured coal
728 (Figure 1). Let us denote the Helmholtz free energy of the solid coal matrix
729 (including the energy of fluid in micropores) per unit volume of undeformed
730 fractured coal as f_m [J/L]. Energy can be added to this system by strain
731 work of the stress tensor $\sigma_{ij}d\varepsilon_{ij}$, by strain work of cleat pressure on fracture
732 walls $p_c d\phi_c$, or by adding fluid to the micropores in the coal matrix μdn_r .
733 Energy conservation under isothermal conditions yields

$$df_m = \sum_{i,j} \sigma_{ij} d\varepsilon_{ij} + p_c d\phi_c + \mu dn_r \quad (\text{A.1})$$

734 being σ_{ij} the total stress tensor acting on a REV of fractured coal (i.e., coal
735 matrix and cleats) [Pa], ε_{ij} the strain tensor of a REV of fractured coal [-], p_c
736 the fluid pressure in cleats [Pa], ϕ_c the Lagrangian porosity of cleats within
737 the REV [-] (calculated respect to the original undeformed reference volume
738 of REV), μ the molar chemical potential of adsorbed fluid in the coal matrix
739 [J/mol], and n_r the number of adsorbed moles in the coal matrix per unit
740 volume of REV of undeformed fractured coal (includes fractures volume)
741 [mol/L]. A Legendre transform lets us change variables, so that the following
742 holds:

$$d(f_m - n_r \mu) = \sum_{i,j} \sigma_{ij} d\varepsilon_{ij} + p_c d\phi_c - n_r d\mu \quad (\text{A.2})$$

743 The development of the model consists in finding expressions to the vari-
 744 ables σ_{ij} , p_c , and n_r as a function of the state variables ε_{ij} , ϕ_c , and μ . This
 745 formulation differs from the one by Espinoza et al. (2013) because it includes
 746 explicitly a well defined fracture porosity and the effect of fracture compress-
 747 ibility. Moreover, in this formulation stresses and strains are defined at the
 748 scale of fractured coal rather than at the scale of the coal matrix.

749 *Appendix A.2. Constitutive equations*

750 *Appendix A.2.1. Total amount of fluid in the coal matrix per unit of volume*
 751 *of fractured coal*

752 Rather than defining a microporosity of the coal matrix, let us define
 753 n_m [mol/L] as the total amount of fluid in the coal matrix per unit volume
 754 of undeformed coal matrix (it does not include fracture volume). The un-
 755 deformed coal matrix volume is chosen as a reference volume, so that the
 756 sorption per unit mass [mol/g] can be obtained knowing the reference bulk
 757 mass density. As suggested by Brochard et al. (2012), the amount of fluid
 758 within the coal matrix depends on the fluid chemical potential μ (or pressure)
 759 and coal matrix volumetric strain ϵ_m . Furthermore, Brochard et al. (2012)
 760 show that the total amount of fluid in a piece of isotropic coal matrix can be
 761 well approximated by a first-order expansion of the total amount of fluid in
 762 the coal matrix with respect to the coal matrix volumetric strain ϵ_m , thus

$$n_m(p_m, \epsilon_m) = n_{m0}(p_m) [1 + c(p_m)\epsilon_m] \quad (\text{A.3})$$

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763 where $n_{m0}(p_m)$ is the adsorption isotherm at zero strain, $c(p_m)$ is a coefficient
764 which quantifies the effect of strain on adsorption capacity, and p_m is the
765 pressure of the bulk fluid at the same chemical potential μ as the adsorbed
766 phase in the coal matrix, termed “thermodynamic pressure”. The coupling
767 coefficient $c(p_m)$ is positive for fluids that make coal swell. This simple linear
768 expansion permits accounting for effects such as swelling strain, adsorption
769 stress, and mechanically induced desorption.

770 In order to add up fluid amounts more easily, let us define n_r the amount of
771 fluid within the coal matrix per unit volume of REV of undeformed fractured
772 coal, which in terms of the amount of fluid within the coal matrix n_m per unit
773 volume of undeformed coal matrix and reference cleat porosity ϕ_{c0} results as

$$n_r(p_m, \varepsilon_{ij}, \phi_c) = (1 - \phi_{c0})n_m(p_m, \epsilon_m) \quad (\text{A.4})$$

774 The volumetric strain at the fractured coal REV scale $\epsilon = \varepsilon_{11} + \varepsilon_{22} + \varepsilon_{33}$
775 is related to the coal matrix volumetric strain ϵ_m in an exact manner through
776 the following relation (Coussy, 2010):

$$\epsilon_m = \frac{\epsilon - (\phi_c - \phi_{c0})}{1 - \phi_{c0}} \quad (\text{A.5})$$

777 where ϕ_{c0} is the reference porosity of the undeformed fractured coal.

778 *Appendix A.2.2. Stress-adsorption relationship*

779 By applying Maxwell’s relations on Eq. (A.2) and taking partial cross
780 derivatives respect to ε_{ij} and μ we obtain

$$\left. \frac{\partial^2(f_m - n_r\mu)}{\partial\varepsilon_{ij}\partial\mu} \right|_{\phi_c} = \left. \frac{\partial^2(f_m - n_r\mu)}{\partial\mu\partial\varepsilon_{ij}} \right|_{\phi_c} \quad (\text{A.6})$$

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781 So that integration leads to,

$$\left. \frac{\partial \sigma_{ij}}{\partial \mu} \right|_{\phi_c, \epsilon_{kl} \neq ij} = - \left. \frac{\partial n_r}{\partial \epsilon_{ij}} \right|_{\phi_c, \mu, \epsilon_{kl} \neq ij} \quad (\text{A.7})$$

782 Let us write $\partial \epsilon_{ij}$ in terms of ϵ_m and ϕ_{c0} and ∂n_r in terms of n_m using Eq.
783 (A.4) and (A.5). This replacement leads to the following equation,

$$\begin{cases} - \left. \frac{\partial n_r}{\partial \epsilon_{ij}} \right|_{\phi_c, \mu} = - \frac{(1-\phi_{c0})}{(1-\phi_{c0})} \left. \frac{\partial n_m}{\partial \epsilon_m} \right|_{\mu} = - \left. \frac{\partial n_m}{\partial \epsilon_m} \right|_{\mu} & \text{if } i = j \\ - \left. \frac{\partial n_r}{\partial \epsilon_{ij}} \right|_{\phi_c, \mu} = 0 & \text{if } i \neq j \end{cases} \quad (\text{A.8})$$

784 By integrating Eq. (A.7) on the molar chemical potential μ and replacing
785 the right hand side with Eq. (A.8), we obtain

$$\sigma_{ij} = \Psi_{ij}(\underline{\underline{\epsilon}}, \phi_c) - \int_{-\infty}^{\mu} \left. \frac{\partial n_m}{\partial \epsilon_m} \right|_{\mu} d\mu \delta_{ij} \quad (\text{A.9})$$

786 Gibbs-Duhem equation lets us express the variation in chemical poten-
787 tial as a function of the thermodynamical pressure as $d\mu = \bar{V}_b(p_m) dp_m$ for
788 isothermal conditions, where \bar{V}_b is the molar volume of the fluid in bulk
789 conditions as a function of temperature and pressure, therefore we can write,

$$\sigma_{ij} = \Psi_{ij}(\underline{\underline{\epsilon}}, \phi_c) - \int_0^{p_m} \left. \frac{\partial n_m}{\partial \epsilon_m} \right|_p \bar{V}_b(p_m) dp_m \delta_{ij} \quad (\text{A.10})$$

790 The first term to the right of the equality Ψ_{ij} represents the poroelastic
791 response of the fractured coal to bulk strains and fracture pressure (without
792 accounting for adsorption phenomena) and the integral term represents the
793 response of the coal matrix to fluid adsorption as a function of fluid pressure
794 and coal matrix strain. The integral term quantifies the stress needed to keep
795 the coal matrix at zero volumetric strain during adsorption, and is termed

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796 “adsorption stress” s^a as motivated by analogy with the theory formulated
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11 by Ravikovitch and Neimark (2006), i.e.,
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$$s^a(p_m) = \int_0^{p_m} \frac{\partial n_m}{\partial \epsilon_m} \bigg|_p \bar{V}_b(p_m) dp_m \quad (\text{A.11})$$

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798 Equation A.10 written in terms of $s^a(p_m)$ is
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$$\sigma_{ij} = \Psi_{ij}(\underline{\epsilon}, \phi_c) - s^a(p_m) \delta_{ij} \quad (\text{A.12})$$

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799 In fact, the integrand of $s^a(p_m)$ can be viewed as a pressure-dependent
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26 Biot-like coefficient b^* [-], (in the sense that $b^* = d\sigma/dp|_\epsilon$), which according
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28 to Eq. (A.3) expands to,
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$$b^*(p_m) = \frac{\partial n_m}{\partial \epsilon_m} \bigg|_p \bar{V}_b(p_m) = c(p_m) n_{m0}(p_m) \bar{V}_b(p_m) \quad (\text{A.13})$$

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34 *Appendix A.2.3. Pressure-adsorption relationship*
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803 By applying Maxwell’s relations on Eq. (A.2) and taking cross partial
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39 derivatives with respect to ϕ_c and μ the following holds
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$$\frac{\partial^2(f_m - n_r \mu)}{\partial \phi_c \partial \mu} \bigg|_{\epsilon_{ij}} = \frac{\partial^2(f_m - n_r \mu)}{\partial \mu \partial \phi_c} \bigg|_{\epsilon_{ij}} \quad (\text{A.14})$$

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805 so that,
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$$\frac{\partial p_c}{\partial \mu} \bigg|_{\epsilon_{ij}, \phi_c} = - \frac{\partial n_r}{\partial \phi_c} \bigg|_{\epsilon_{ij}, \mu} \quad (\text{A.15})$$

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806 By using the micromechanical Eq. (A.5), the latter equation is equivalent
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807 to
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$$\frac{\partial p_c}{\partial \mu} \bigg|_{\epsilon_{ij}, \phi_c} = \frac{\partial n_m}{\partial \epsilon_m} \bigg|_\mu \quad (\text{A.16})$$

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808 Integration of this latter equation on the molar chemical potential μ leads
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11 809 to

$$p_c = \Psi'_{ij}(\underline{\underline{\epsilon}}, \phi_c) + \int_{-\infty}^{\mu} \frac{\partial n_m}{\partial \epsilon_m} \Big|_{\mu} d\mu \quad (\text{A.17})$$

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17 810 Using Gibbs-Duhem equation as before and recalling the definition of
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19 811 adsorption stress we obtain,

$$p_c = \Psi'_{ij}(\underline{\underline{\epsilon}}, \phi_c) + s^a(p_m) \quad (\text{A.18})$$

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25 812 The function Ψ'_{ij} represents the poroelastic response of the fractured coal
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27 813 to bulk strains and changes in porosity (without accounting for adsorption
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29 814 phenomena).

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33 815 *Appendix A.3. Transverse isotropic poroelasticity - Final equations*

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35 816 Let us approximate the behavior of fractured coal assuming transverse
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37 817 isotropy around the axis perpendicular to the bedding plane, from now on
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39 818 assigned to axis number 3. The functions Ψ_{ij} and Ψ'_{ij} for linear elastic trans-
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41 819 verse isotropic poroelastic solids with principal directions of the Biot tensor
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43 820 coinciding with the axes of symmetry of the fractured coal are summarized
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45 821 in the following set of equations Coussy (2004),

$$\begin{cases}
\sigma_{11}(\underline{\underline{\varepsilon}}, \phi_c - \phi_{c0}) = C_{11}\varepsilon_{11} + C_{12}\varepsilon_{22} + C_{13}\varepsilon_{33} - b_1N[(\phi_c - \phi_{c0}) - b_1(\varepsilon_{11} + \varepsilon_{22}) - b_3\varepsilon_{33}] \\
\sigma_{22}(\underline{\underline{\varepsilon}}, \phi_c - \phi_{c0}) = C_{12}\varepsilon_{11} + C_{11}\varepsilon_{22} + C_{13}\varepsilon_{33} - b_1N[(\phi_c - \phi_{c0}) - b_1(\varepsilon_{11} + \varepsilon_{22}) - b_3\varepsilon_{33}] \\
\sigma_{33}(\underline{\underline{\varepsilon}}, \phi_c - \phi_{c0}) = C_{13}\varepsilon_{11} + C_{13}\varepsilon_{22} + C_{33}\varepsilon_{33} - b_3N[(\phi_c - \phi_{c0}) - b_1(\varepsilon_{11} + \varepsilon_{22}) - b_3\varepsilon_{33}] \\
\sigma_{23}(\underline{\underline{\varepsilon}}, \phi_c - \phi_{c0}) = 2C_{44}\varepsilon_{23} \\
\sigma_{31}(\underline{\underline{\varepsilon}}, \phi_c - \phi_{c0}) = 2C_{44}\varepsilon_{31} \\
\sigma_{12}(\underline{\underline{\varepsilon}}, \phi_c - \phi_{c0}) = 2\frac{C_{11}-C_{12}}{2}\varepsilon_{12} \\
p_c(\underline{\underline{\varepsilon}}, \phi_c - \phi_{c0}) = N[(\phi_c - \phi_{c0}) - b_1(\varepsilon_{11} + \varepsilon_{22}) - b_3\varepsilon_{33}]
\end{cases} \tag{A.19}$$

where C_{ij} are elastic coefficients of the stiffness tensor. The macro-poromechanical parameters (b_1, b_3, N) relate to the coal matrix bulk modulus K_m as follows,

$$\begin{cases}
b_1 = 1 - \frac{(C_{11}+C_{12}+C_{13})/3}{K_m} \\
b_3 = 1 - \frac{(2C_{13}+C_{33})/3}{K_m} \\
\frac{1}{N} = \frac{(2b_1+b_3)/3-\phi_{c0}}{K_m}
\end{cases} \tag{A.20}$$

The poromechanical parameters capture the effect of pore/fracture compressibility on the bulk fractured coal elastic moduli. These equations are valid for any transverse isotropic porous solid with a well defined porosity ϕ_c . The last step consists in plugging the macro-poromechanical equations of the fractured coal (Eq. A.19) in the general equations of the stress tensor σ_{ij} (Eq. A.12) and pressure of fluids in cleats p_c (Eq. A.18) developed to consider matrix adsorption and swelling. Hence, the final set of equations results in Equation 1 in Section 2.

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