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# Measurement and modeling of adsorptive-poromechanical properties of bituminous coal cores exposed to CO<sub>2</sub>: 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 CO<sub>2</sub>. 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 CO<sub>2</sub> 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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53 2 Currently, natural gas accounts for roughly 21% of the World's energy  
54 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 CO<sub>2</sub>, conceivable up to  
13 20 GtCO<sub>2</sub> (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<sup>-9</sup> m compose the coal microporosity. The coal  
31 matrix can also host mesopores sized in the order of 10<sup>-8</sup> m. The coal matrix  
32 is capable of adsorbing various gases including CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>, 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 CO<sub>2</sub> 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 CO<sub>2</sub> 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) CO<sub>2</sub> 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 CO<sub>2</sub> 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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55 77 The objective of this study is to measure the coupled poromechanical  
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57 78 response and permeability of coal cores injected with an adsorbate under

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10 different loading and strain boundary conditions, interpret the results in  
11 the context of a double porosity model for transverse isotropic fractured  
12 microporous solids (fully coupled regarding fluid mass, fluid pressure, and  
13 solid stress), and give insight into the evolution of permeability of the coal  
14 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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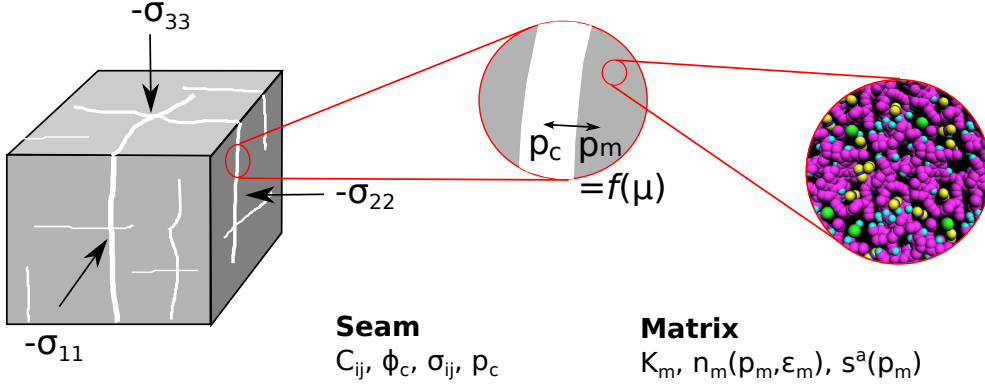


Figure 1: Representative elementary volume of fractured coal and relevant modeling parameters. Fracture zoom sketches the interplay between pressure  $p_c$  of bulk fluid in the cleats, and thermodynamic pressure  $p_m$  of fluid in the coal matrix. Coal matrix zoom shows a molecular representation of 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  $\phi_c$  as a function of the initial porosity  $\phi_{c0}$ , the poroelastic coeffi-  
 118 cient  $N$  (Eq. A.20), the strains of fractured coal  $\varepsilon_{ij}$ , 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  $\varepsilon_m$  of the coal matrix – first term) and  
 123 the amount of fluid in the cleats (which depends on cleat porosity  $\phi_c$  and  
 124 bulk fluid molar density  $\rho_b$  –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  $\alpha_{\parallel}$  and  $\alpha_{\perp}$   
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  $\sim 500$  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  $\text{kg}/\text{m}^3$  (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  $\sim 11.3$  to 13.9%. SEM imaging performed on 1  
46 cm diameter coal cores shows the presence of microfractures ranging from 50  
47  $\mu\text{m}$  to less than 1  $\mu\text{m}$ . Figure 3-a shows a calcite-filled fracture with total  
48 aperture 50  $\mu\text{m}$  being intercepted at  $\sim 90^\circ$  by a smaller 10  $\mu\text{m}$  empty frac-  
49 ture. We measured pore size distribution with a mercury intrusion porosime-  
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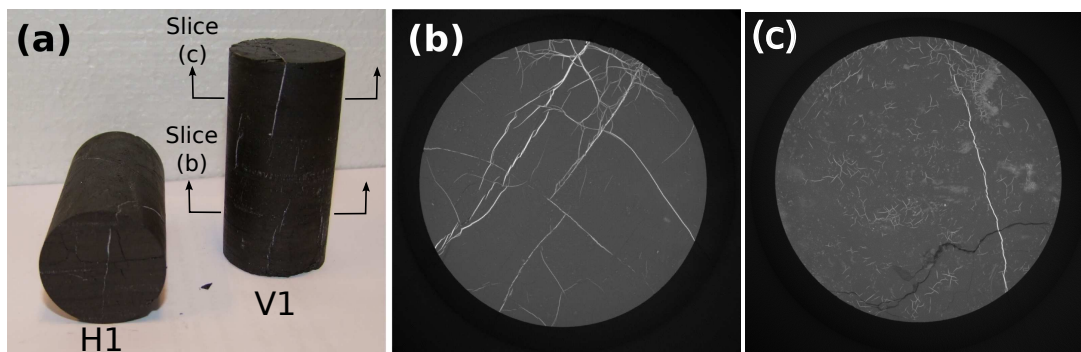


Figure 2: Coal cores 38 mm diameter – slenderness ratio 2:1 used for triaxial tests. (a) Cores picture: coring parallel to the bedding plane (left); coring perpendicular to the bedding plane (right). (b-c) X-ray tomographic reconstruction of cross sections on the bedding plane (image resolution 25  $\mu\text{m}$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

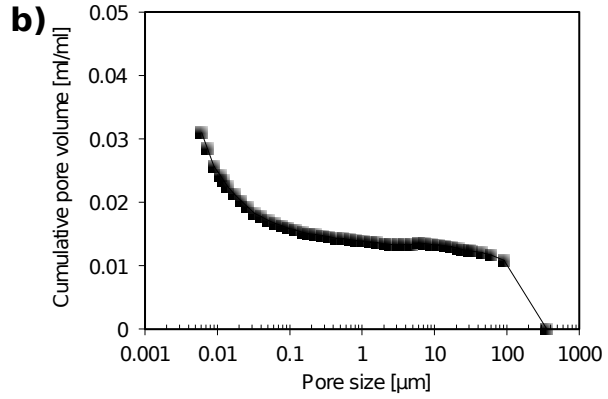
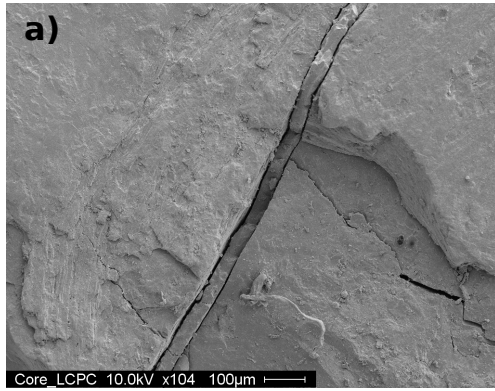


Figure 3: Coal characterization. (a) SEM imaging on a partially calcite-filled fracture. (b) Mercury intrusion porosimetry on a  $\sim 1 \text{ cm}^3$  coal sample.

### 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  $\text{CO}_2$  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  $\text{CO}_2$  pore fluid system is managed by upstream  
 205 and downstream syringe pumps. The first pump injects and measures  $\text{CO}_2$

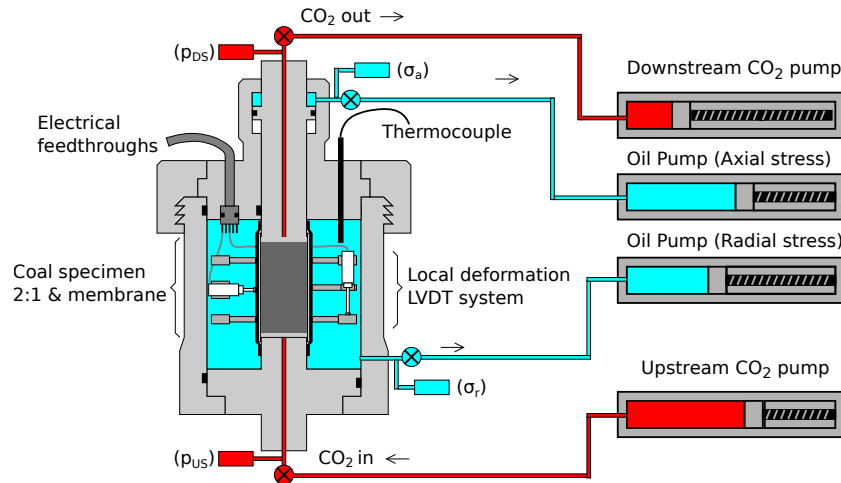


Figure 4: Experimental triaxial device. Main characteristics include: maximum radial stress 40 MPa, maximum axial stress 60 MPa, measurement of local strains, temperature control, and ability to handle supercritical CO<sub>2</sub>.

flow rate and injection volume; the second pump controls CO<sub>2</sub> back-pressure. Stainless steel porous plates help distribute evenly the gas on the base and top faces of the specimen. A CO<sub>2</sub>-resistant membrane combining layers of teflon and nitrile isolates the CO<sub>2</sub>-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 CO<sub>2</sub> 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/m<sup>3</sup>.

Specimen	Bulk mass density $\rho_b$ [kg/m <sup>3</sup> ]	Helium porosity [%]	Type of dry loading	Stress/strain condition during CO <sub>2</sub> 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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247 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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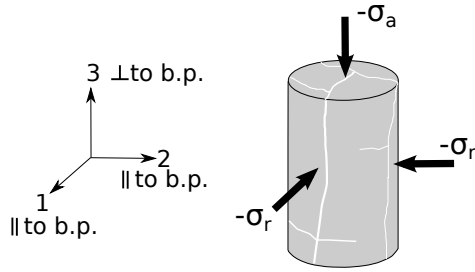


Figure 5: Coal core drilled perpendicular to the bedding plane (b.p.). The triaxial device applies axial stress  $\sigma_a$  and radial stress  $\sigma_r$  which correspond in this case to the seam stresses perpendicular and parallel to the bedding plane respectively.

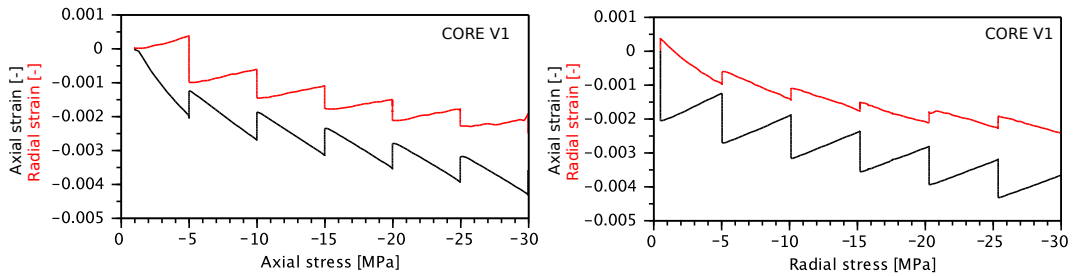


Figure 6: Strain-stress response to drained loading of coal core V1 in dry conditions - no  $\text{CO}_2$  in pores. Loading is performed in 5 MPa stress increments (compressive stresses and contraction strains are negative).

#### 249 4.1. Elastic moduli of dry coal specimens

250 Anisotropic loading is applied by steps with increasing axial stress  $\sigma_a$  at  
 251 constant radial stress  $\sigma_r$ , and subsequently with increasing radial stress at  
 252 constant axial stress. The loading is performed in drained conditions without  
 253  $\text{CO}_2$  (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):  
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$$\begin{cases} \left. \frac{\partial \varepsilon_r}{\partial \sigma_r} \right|_{p_c, p_m, \sigma_a} = \frac{1-\nu}{E} & ; & \left. \frac{\partial \varepsilon_r}{\partial \sigma_a} \right|_{p_c, p_m, \sigma_r} = -\frac{\nu_3}{E_3} \\ \left. \frac{\partial \varepsilon_a}{\partial \sigma_r} \right|_{p_c, p_m, \sigma_a} = -\frac{2\nu_3}{E_3} & ; & \left. \frac{\partial \varepsilon_a}{\partial \sigma_a} \right|_{p_c, p_m, \sigma_r} = \frac{1}{E_3} \end{cases} \quad (5)$$

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17 Stress-strain data from cores drilled parallel to the bedding plane let us  
18 recover directly the elastic parameter  $E$ . Figure 7 shows the summary of  
19 tangent elastic moduli measured for all specimens as a function of axial and  
20 radial stresses. Young's elastic moduli values range from  $\sim 2$  to 6 GPa in the  
21 range of confining stresses 0.5 to 30 MPa (Figure 7). Core specimens show  
22 non-negligible stress-strain nonlinearity, which reduces at confining stresses  
23 higher than 15 MPa. Fitting curves represent average stress-dependent elas-  
24 tic moduli values that will be used later to estimate secant moduli relevant  
25 to the range of testing conditions.  
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#### 267 4.2. Fracture-induced Biot coefficients

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37 Pore-fluid loading is applied by injecting pressurized CO<sub>2</sub>. The theoretical  
38 strain response to increments of fluid pressure in fractures  $p_c$  while keeping  
39 constant confining stresses and adsorption stress in the coal matrix (i.e., no  
40 variation of the sorbed amount  $n_r$  and of the thermodynamic pressure  $p_m$  of  
41 the fluid in the coal matrix) is derived from Equation (4).  
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$$\begin{cases} \left. \frac{\partial \varepsilon_r}{\partial p_c} \right|_{\sigma_a, \sigma_r, p_m} = \frac{1-\nu}{E} b_1 - \frac{\nu_3}{E_3} b_3 \\ \left. \frac{\partial \varepsilon_a}{\partial p_c} \right|_{\sigma_a, \sigma_r, p_m} = -\frac{2\nu_3}{E_3} b_1 + \frac{1}{E_3} b_3 \end{cases} \quad (6)$$

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53 On the other hand, the stress response of the coal core to CO<sub>2</sub> injection  
54 at constant volume and constant adsorption stress conditions is equal to  
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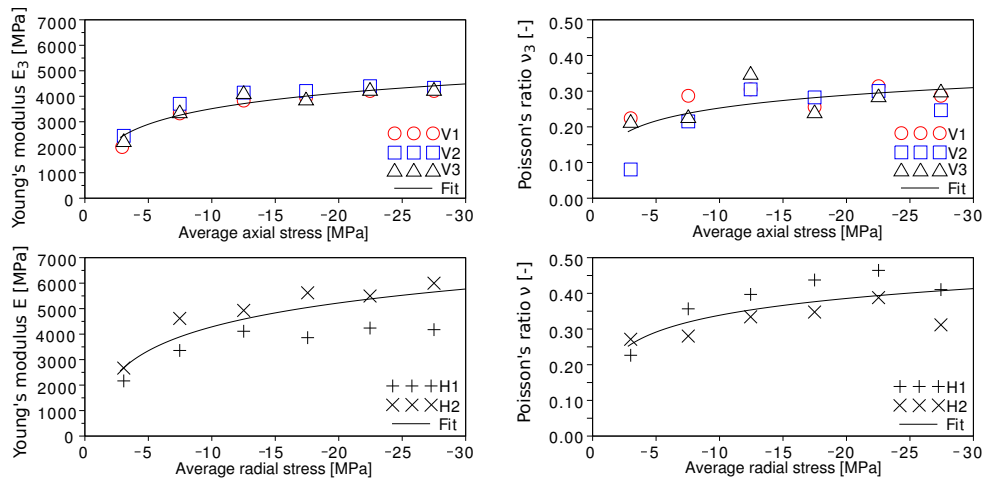


Figure 7: Summary of measured tangent elastic coefficients  $E$ ,  $E_3$ ,  $\nu$ ,  $\nu_3$  as a function of axial and radial stress (drained moduli). Fitting functions (Elastic coefficient =  $cst_1 + cst_2 \ln(-\text{Stress}/1\text{MPa})$ ) capture elastic nonlinearity. Coefficients ( $cst_1$ ,  $cst_2$ ) are (1478 MPa, 884 MPa) for  $E_3$ , (1172 MPa, 1352 MPa) for  $E$ , (0.133, 0.052) for  $\nu_3$ , and (0.182, 0.068) for  $\nu$ .

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

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 CO<sub>2</sub> injection including the adsorption response.

#### 280 *4.3. Adsorptive properties*

281 The adsorptive properties of coal cores are tested with CO<sub>2</sub>. CO<sub>2</sub> 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 CO<sub>2</sub> at an objective constant  
 288 CO<sub>2</sub> 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 CO<sub>2</sub> 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, CO<sub>2</sub> is injected into the specimen at  
 295 the objective pressure. CO<sub>2</sub> enters the core from the bottom cap and exits

Table 2: Summary of results of CO<sub>2</sub> injection experiments performed at constant stress conditions.

Test type and specimen	Obj. CO <sub>2</sub> pressure [MPa]	Total radial stress $\sigma_r$ [MPa]	Total axial stress $\sigma_a$ [MPa]	Total CO <sub>2</sub> 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 CO<sub>2</sub> measured from the upstream injection pump (Figure 8-c). The injected amount of CO<sub>2</sub> 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 CO<sub>2</sub> pressure and radial effective stress. In adsorption experiments, the specimen is left about 6 days to equilibrate with CO<sub>2</sub>. This period of time was sufficient to achieve steady-state conditions according to our strain measurements.

We identify two main regimes during CO<sub>2</sub> injection into the coal core

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310 under constant confining stresses: (1) CO<sub>2</sub> 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) CO<sub>2</sub> 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 CO<sub>2</sub> (positive amount  
16 of injected CO<sub>2</sub>). 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 CO<sub>2</sub> 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 CO<sub>2</sub> 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 CO<sub>2</sub> 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 CO<sub>2</sub> injection and the total CO<sub>2</sub> delivery to  
331 the core assembly. The delivery of CO<sub>2</sub> to the piping system (upstream  
332 3.9 cm<sup>3</sup> and downstream 3.3 cm<sup>3</sup>) must be subtracted to estimate the net  
333 amount of total CO<sub>2</sub> 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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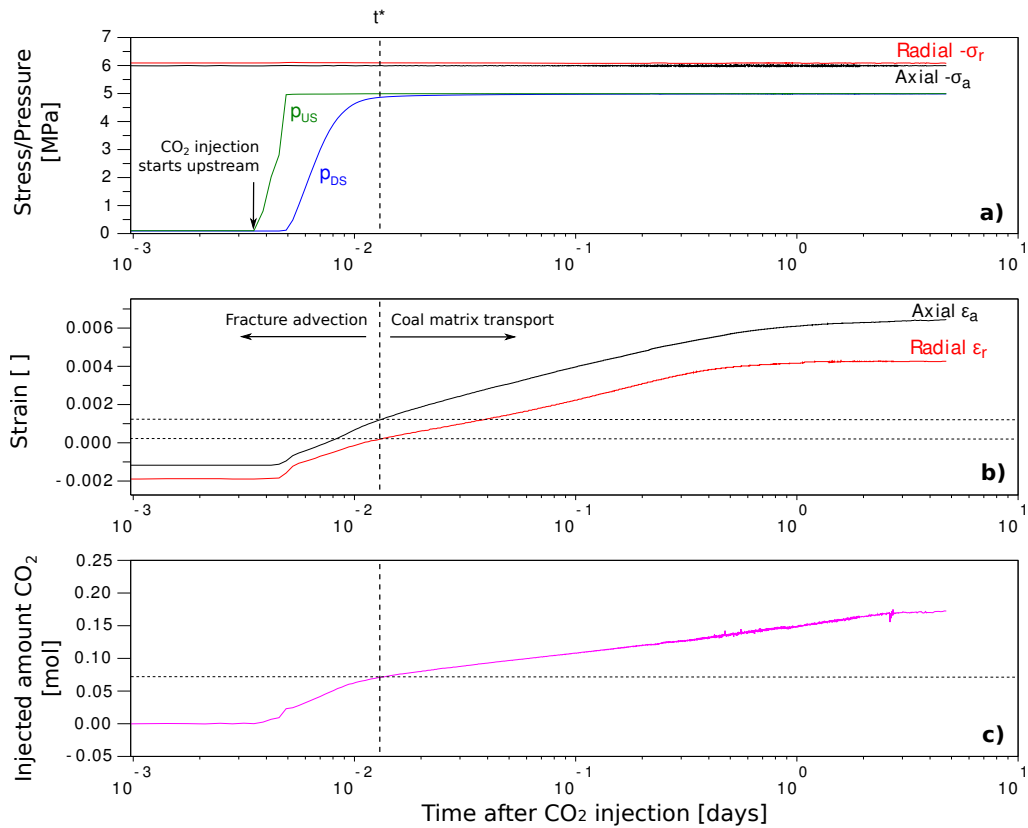


Figure 8: Time evolution of (a) fluid pressure, (b) strains, and (c) injected amount of CO<sub>2</sub> injection experiment C.S.V1 at constant confining stresses  $\sigma_r = \sigma_a = -6$  MPa. The objective injection pressure is  $p_{US} = p_{DS} = 5$  MPa. Temperature is 39.5°C.

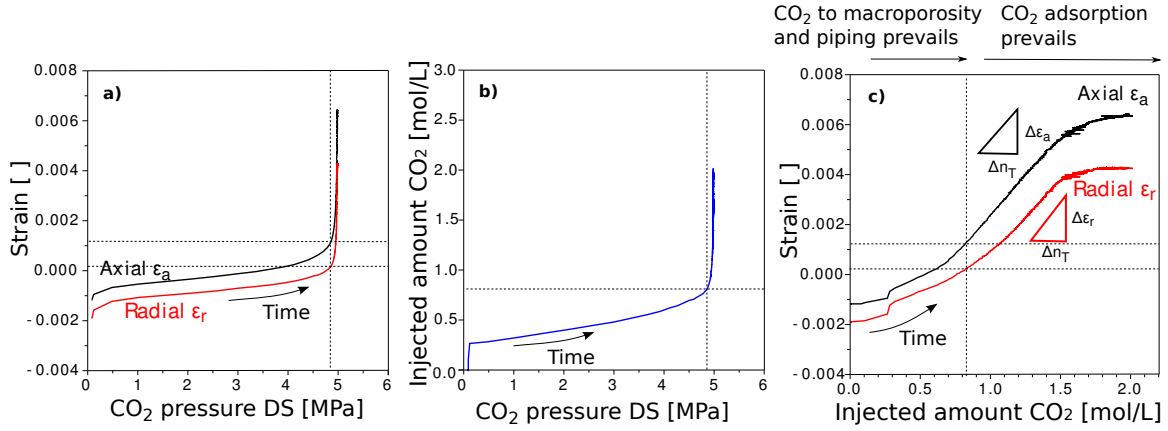


Figure 9: Response of specimen V1 (undeformed volume 86.8 cm<sup>3</sup> - experiment C.S.V1) to CO<sub>2</sub> injection at constant confining stresses (same data as in Figure 8). Measurements are plotted independently of time: (a) strain and CO<sub>2</sub> pressure downstream, (b) injected amount per unit volume and CO<sub>2</sub> 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)<sup>-1</sup> and 0.0033 (mol/L)<sup>-1</sup>, respectively for axial and radial strains in experiment C.S.V1 (Figure 9-c).

Table 2 shows a summary of experimental results for all CO<sub>2</sub> 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 CO<sub>2</sub> pressure. All experiments show a consistent trend  
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13 349 of declining differential swelling at incremental amount of adsorption (for  
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15 350 example, at injected amounts of CO<sub>2</sub> greater than 1.5 mol/L in Figure 9-c).  
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17 351 The discussion section explores further this phenomenon.

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19 352 The total strain response upon injection (10-c) predicted by the porome-  
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21 353 chanical model results from the summation of strains caused by poroelastic  
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23 354 effects in macropores (Equation 6) and the strain response to increments of  
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25 355 adsorption stress  $s^a$  (a function of the thermodynamic pressure of the fluid  
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27 356 phase in the coal matrix  $p_m$ ), at constant confining radial  $\sigma_r$  and axial  $\sigma_a$   
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29 357 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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36 358 The total uptake amount per unit volume (10-b) is obtained from Equa-  
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38 359 tion (3). The adsorption swelling slopes (10-d) are calculated from the full  
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40 360 set of equations through numerical simulation.

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42 361 The following sequence describes the process to determine the adsorptive-  
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44 362 mechanical parameters of the coal core and coal matrix that fit best the  
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46 363 experimental data according to the developed poromechanical model.

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49 364 1. Estimation of core elastic moduli in dry conditions: We calculate the  
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51 365 secant moduli from Figure (7) for a Terzaghi's effective stress that varies  
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53 366 from 7.5 MPa to 1 MPa (approximate range for the set of experiments  
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55 367 in Table 1). The corresponding values are  $E = 2736$  MPa,  $E_3 = 2551$   
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57 368 MPa,  $\nu = 0.267$ ,  $\nu_3 = 0.198$ .

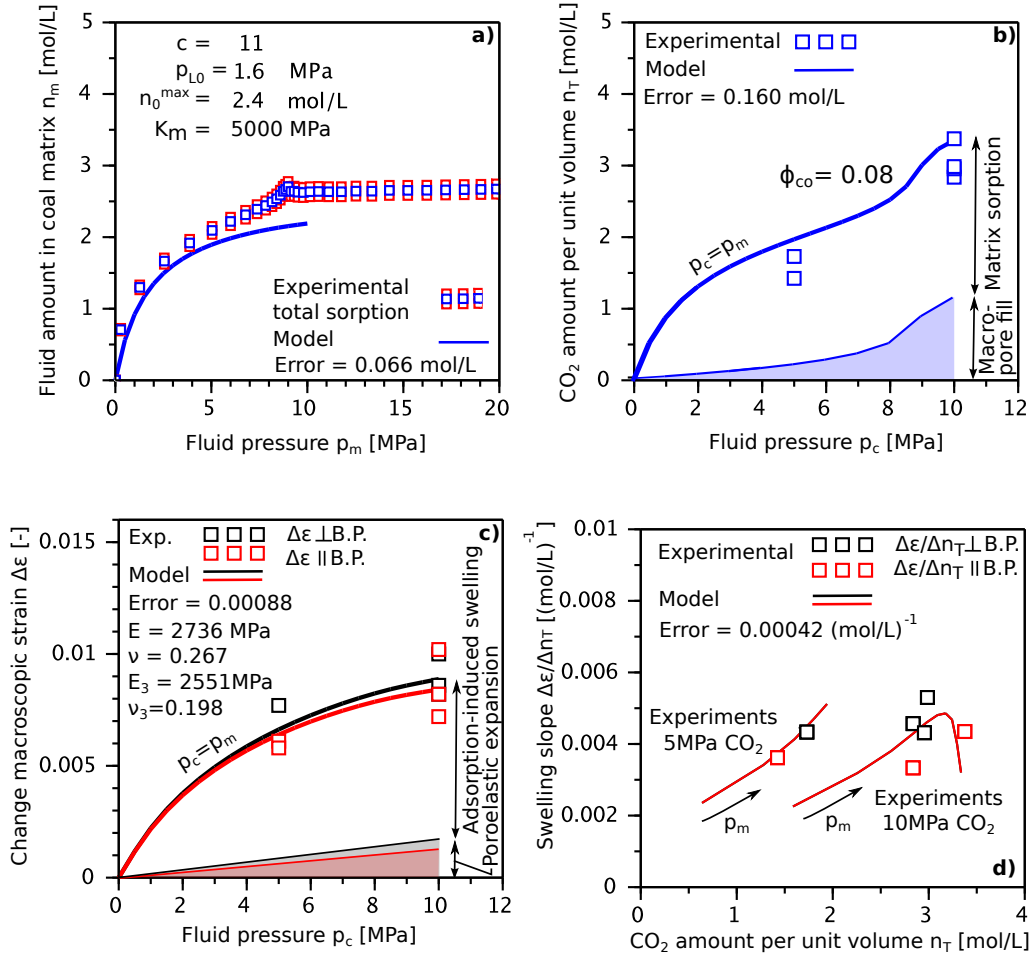


Figure 10: CO<sub>2</sub> uptake and adsorption-induced swelling in coal cores keeping constant confining stresses (See Table 2): experimental values and numerical simulation. Notice that total uptake amounts and change of strains are caused by macropore/fracture poroelastic response and micropore adsorptive response. (a) Coal matrix total sorption amount - from independent manometric tests (red squares indicate uncertainty - Auxiliary Material), (b) core total uptake amount of CO<sub>2</sub> per unit volume, (c) total change of core strains upon CO<sub>2</sub> injection, and (d) ratio of change of swelling strain to change of uptake amount during adsorption at constant pressure in fractures ( $\Delta p_c$  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 ( $\sim 30$  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  $\phi_{c0}$ : 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  $\mu\text{m}$ ) larger than 0.5 %, MPI yields a porosity about 1.5% of pores larger than 0.1  $\mu\text{m}$ , 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  $\mu\text{m}$  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
$\nu$	0.267	Measured	$p_{L0}$	1.6 MPa	Inverted
$\nu_3$	0.198	Measured	$c$	11	Inverted
$\phi_0$	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 ( $\Delta p_c$ ) and adsorption-induced swelling follows ( $\Delta p_m$ ). 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 CO<sub>2</sub> 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  
15  
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 CO<sub>2</sub> injection pressure of  
17  
415 2 MPa (strictly, this experiment is performed with a pressure gradient for  
18  
416 the sake of measuring permeability:  $p_{US} = 2.5$  MPa and  $p_{DS} = 1.5$  MPa).  
19  
417 Results show that from an initial condition of total radial stress  $-3$  MPa,  
20  
418 axial total stress  $-10$  MPa and no CO<sub>2</sub>, CO<sub>2</sub> 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  
23  
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 CO<sub>2</sub> 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 CO<sub>2</sub> 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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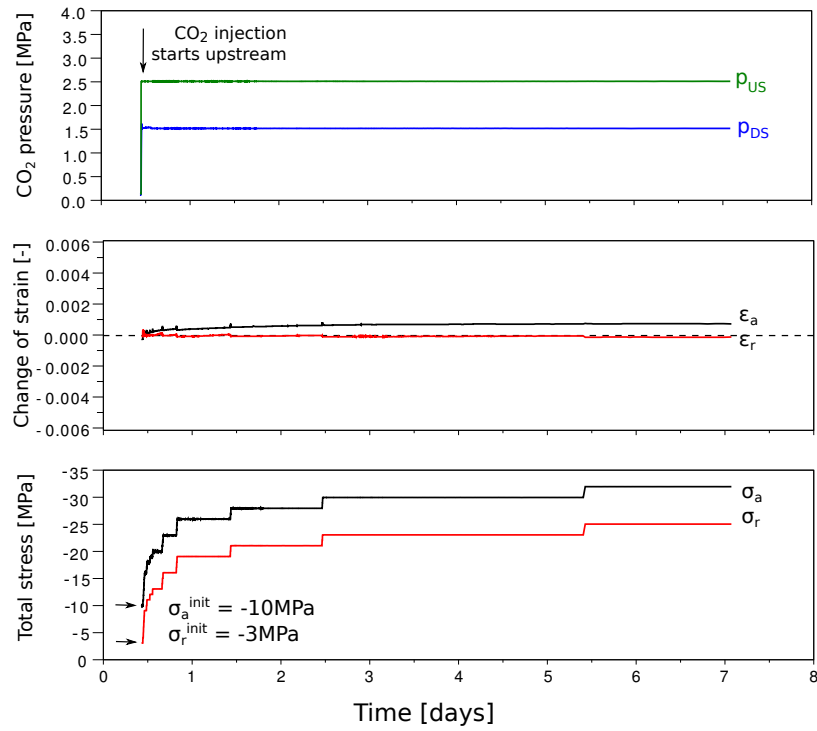


Figure 11: Time history of strains and total stresses upon CO<sub>2</sub> injection in core V1 keeping near constant volume conditions (Experiment C.V.V1b - Table 4).

Table 4: Summary of experimental results of CO<sub>2</sub> injection in cores performed at near constant volume conditions. (\*Average pressure)

Test type and specimen	CO <sub>2</sub> objective pressure [MPa]	Initial radial stress [MPa]	Initial axial stress [MPa]	Total CO <sub>2</sub> 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

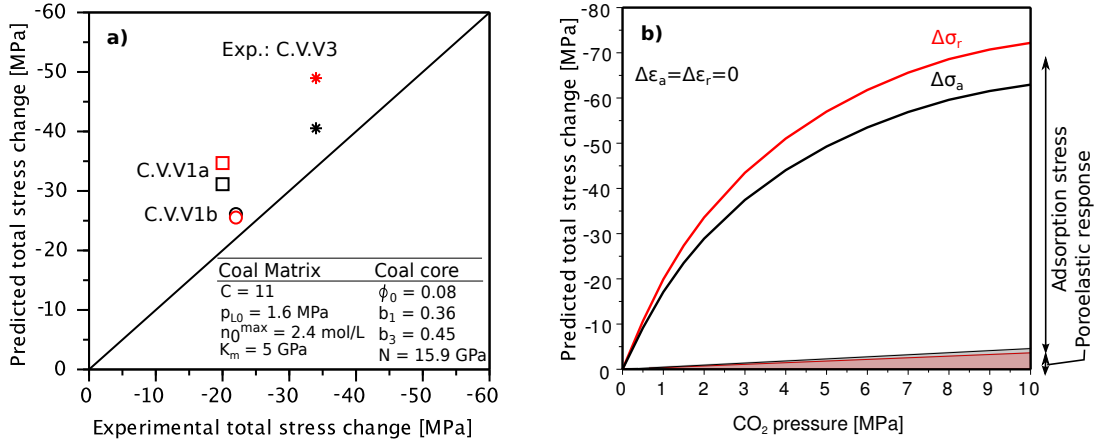


Figure 12: Change of total stress in coal cores induced by CO<sub>2</sub> injection at near constant-volume condition: (a) comparison between experimentally measured values and model predictions and (b) model-predicted change of stress at perfect constant-volume condition.

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

Figure 12-a displays the experimentally measured changes in total stress upon CO<sub>2</sub> 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  $\sim 1$  to  $25 \text{ MPa}$ ), 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 CO<sub>2</sub> pressure. The  
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15 461 adsorption-induced stress is about 60 MPa at 10 MPa of CO<sub>2</sub> pressure.  
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#### 17 18 462 *4.4. Permeability measurements*

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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).  
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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.  
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##### 28 29 467 *4.4.1. Permeability of a CO<sub>2</sub>-equilibrated core – Effect of effective stresses*

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31 468 We explored the permeability of specimen V1 to CO<sub>2</sub> in axial direction  
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33 469 (on the plane of vertical fractures in the seam) varying radial and axial  
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35 470 stresses at mean fracture fluid pressure of  $\sim 5.25$  MPa, and pressure gradient  
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37 471 of  $\sim 1.5$  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 CO<sub>2</sub> for  $\sim 6$  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 CO<sub>2</sub> 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)).  
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55  
56 480 Figure 13 shows measured permeability as a function of radial and axial  
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58 481 Terzaghi's effective stresses. The precision of permeability measurements  
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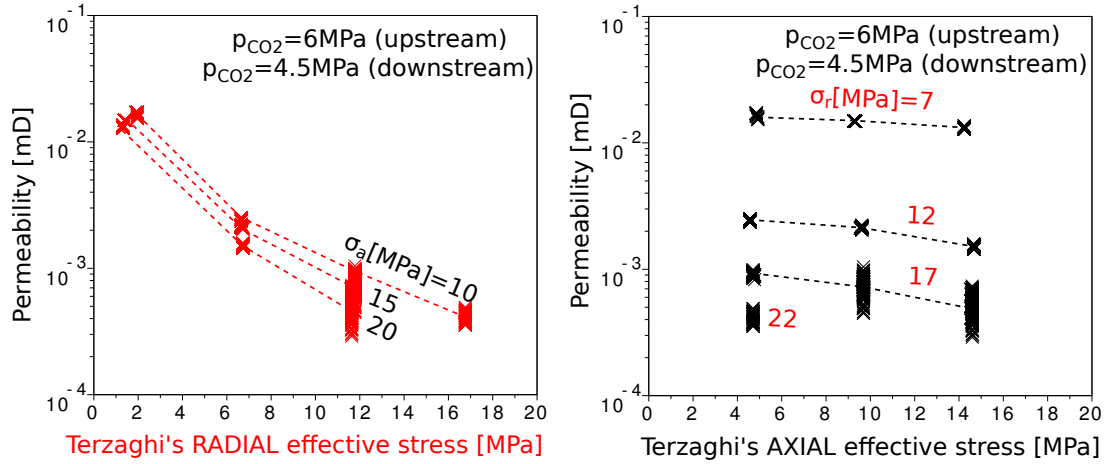


Figure 13: CO<sub>2</sub> permeability of specimen V1 subjected to various levels of effective stress in radial and axial directions. The dashed lines join data points at the same axial (left) or radial stress (right).

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  $-0.46 \text{ MPa}^{-1}$  (Somerton et al., 1975).

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492 4.4.2. *Permeability evolution of a core during CO<sub>2</sub> 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 CO<sub>2</sub> 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  $\sigma_a$  and  $\sigma_r$  can be calculated for  
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38 507 any equilibrium pressure or combination of  $p_c$  and  $p_m$  using Equation (3).  
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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  $\sim 17$  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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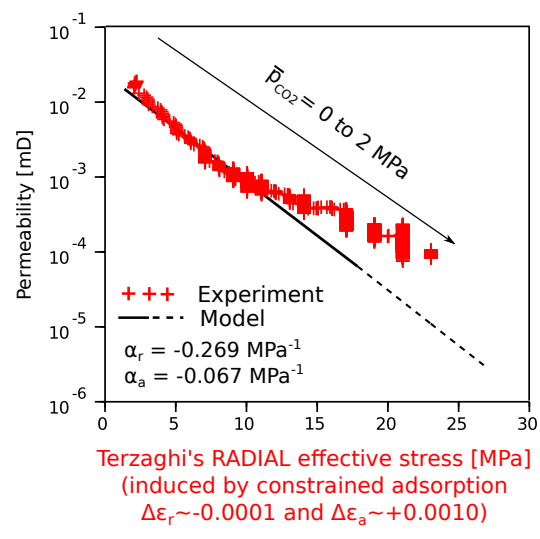


Figure 14: CO<sub>2</sub> permeability evolution of core V1b subjected to constrained adsorption under near constant-volume condition. The specimen develops stresses as a result of CO<sub>2</sub> injection and constrained swelling. Ensuing stresses lower measured permeability up to 100 times. Predicted 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  
37  
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  $\text{N}_2$  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  
41  
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  
45  
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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563 5.1.3. *Experimental results: uncertainties, unexpected phenomena, and main*  
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11 *findings*  
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13 The measurement with the highest uncertainty is the total amount of up-  
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15 take because of diffusion of CO<sub>2</sub> through the confining membrane. During the  
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17 time length of experiments, the diffused amount could add up to an amount  
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19 as large as the effective total uptake amount. Fortunately, these “leaks” were  
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21 quite steady and measurable. Yet, the uncertainty is estimated to be as large  
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23 as 0.5 mol/L. A not-perfectly sealing but deformable membrane performed  
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25 better to measure mechanical properties than a tight but rigid membrane,  
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27 e.g., lead membrane. The uncertainty in determining total amount of uptake  
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29 impacts directly on the determination of the adsorption-strain coupling co-  
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31 efficient  $c$ ,  $n_{max}^0$ , and  $p_{L0}$ . We were able to reduce the uncertainty of these  
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33 inverted parameters by using additional sorption data on crushed coal from  
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35 the same coal seam (Figure 10-a).

36 We observed high elastic nonlinearity at low effective stress, as a result of  
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38 existing fractures. Nonlinearity lessened at high effective stress as fractures  
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40 closed. Using N<sub>2</sub> or He would have facilitated the measurement of porome-  
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42 chanical properties and macro Biot parameters ( $b_1, b_3$ ), with less influence of  
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44 adsorption-induced response. Yet each fluid would yield its own “porome-  
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46 chanical signature” because each fluid has different affinity with coal surfaces  
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48 and can even access different pore sizes. Therefore, we preferred to test only  
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50 with CO<sub>2</sub>.

51 The time required for CO<sub>2</sub> uptake and coal equilibration was based on the  
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53 measured specimen response through either strains or stresses. The allowed  
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55 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  
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27 597 and Holda, 1982; Masoudian et al., 2014) or CO<sub>2</sub>-enhanced coal creep as  
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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  $\pm 1$   
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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  $\mu\text{m}$ , 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  
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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 CH<sub>4</sub> or CO<sub>2</sub> swelling ranging from 5 to 10,  
626 with the highest values corresponding to CO<sub>2</sub> and the lowest ones to CH<sub>4</sub>  
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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9 637 10-d).

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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  
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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  
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26 645 measurements including total uptake amount, induced strains/stresses and  
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28 646 swelling slopes. The same parameters (except for elastic moduli) overesti-  
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30 647 mate by 3 to 16 MPa the induced adsorption-induced stress when displace-  
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32 648 ments are constrained upon adsorption. This overestimation may be due to  
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34 649 heterogeneity of coal from sample to sample and measured elastic moduli,  
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36 650 an inaccurate estimation of secant elastic moduli, or also to the fact that  
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38 651 coal may soften with significant amounts of sorbed CO<sub>2</sub> with concomitant  
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40 652 reduction of induced stresses. Creep may also play a role in canceling the  
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42 653 swelling strains as discussed before.

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44 654 The overestimation in reduction of permeability (Figure 14) stems from  
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46 655 adopting a linear log(permeability)–stress law. Predictions are good in the  
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48 656 interval at which the permeability is fitted from experimental data, but ex-  
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50 657 trapolations deviate beyond the fitting range. More complex models can  
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52 658 capture a “hard permeability” which makes permeability plateau at high ef-  
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54 659 fective stress, and thus capture the nonlinear dependence of the logarithm  
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56 660 of permeability as a function of effective stress (Liu and Rutqvist, 2010). A  
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58 661 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 CH<sub>4</sub>  
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663 depletion and/or CO<sub>2</sub> 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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666 2008; Okotie and Moore, 2010).

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667 *5.3. Implications on coal bed methane production and CO<sub>2</sub> 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 CO<sub>2</sub> 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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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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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 CO<sub>2</sub> as a proxy for CH<sub>4</sub>. 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 CO<sub>2</sub> 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 CO<sub>2</sub> 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**

715 This work was supported by Total S.A. The technical support provided  
716 by E. De Laure and his team at Laboratoire Navier-CERMES is greatly  
717 appreciated. We are thankful to N. Lenoir who performed the X-ray imaging,  
718 to H. Daigle who provided the N<sub>2</sub> adsorption results, and to the anonymous  
719 reviewers who helped improving this article.

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9 **7. Symbols**

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$\alpha$	Fracture compressibility coefficient for permeability law [MPa <sup>-1</sup> ]
$\epsilon_m$	Volumetric strain of the coal matrix [-]
$\epsilon_{ij}$	Strain tensor defined at the fractured coal scale [-]
$\phi_c$	Cleat porosity or macroporosity [-]
$\mu$	Chemical potential [J/mol]
$\nu, \nu_3$	Poisson's ratio parallel and perpendicular to bedding [-]
$\sigma_{ij}$	Total stress tensor defined at the fractured coal scale [Pa]
$\sigma'_{ij}$	Terzaghi's effective stress tensor defined at the fractured coal scale [Pa]
$\rho_b$	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  $\mu dn_r$ .  
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  $\sigma_{ij}$  the total stress tensor acting on a REV of fractured coal (i.e., coal  
735 matrix and cleats) [Pa],  $\varepsilon_{ij}$  the strain tensor of a REV of fractured coal [-],  $p_c$   
736 the fluid pressure in cleats [Pa],  $\phi_c$  the Lagrangian porosity of cleats within  
737 the REV [-] (calculated respect to the original undeformed reference volume  
738 of REV),  $\mu$  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  $\sigma_{ij}$ ,  $p_c$ , and  $n_r$  as a function of the state variables  $\varepsilon_{ij}$ ,  $\phi_c$ , and  $\mu$ . 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  $\mu$  (or pressure)  
 759 and coal matrix volumetric strain  $\epsilon_m$ . 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  $\epsilon_m$ , 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  $\mu$  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  $\phi_{c0}$  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  $\epsilon_m$  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  $\phi_{c0}$  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  $\varepsilon_{ij}$  and  $\mu$  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  $\epsilon_m$  and  $\phi_{c0}$  and  $\partial n_r$  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  $\mu$  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  $\Psi_{ij}$  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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$$14 \quad s^a(p_m) = \int_0^{p_m} \frac{\partial n_m}{\partial \epsilon_m} \Big|_p \bar{V}_b(p_m) dp_m \quad (A.11)$$

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798 Equation A.10 written in terms of  $s^a(p_m)$  is

$$21 \quad \sigma_{ij} = \Psi_{ij}(\underline{\epsilon}, \phi_c) - s^a(p_m) \delta_{ij} \quad (A.12)$$

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799 In fact, the integrand of  $s^a(p_m)$  can be viewed as a pressure-dependent  
800 Biot-like coefficient  $b^*$  [-], (in the sense that  $b^* = d\sigma/dp|_\epsilon$ ), which according  
801 to Eq. (A.3) expands to,

$$30 \quad b^*(p_m) = \frac{\partial n_m}{\partial \epsilon_m} \Big|_p \bar{V}_b(p_m) = c(p_m) n_{m0}(p_m) \bar{V}_b(p_m) \quad (A.13)$$

802 *Appendix A.2.3. Pressure-adsorption relationship*

803 By applying Maxwell’s relations on Eq. (A.2) and taking cross partial  
804 derivatives with respect to  $\phi_c$  and  $\mu$  the following holds

$$40 \quad \frac{\partial^2(f_m - n_r \mu)}{\partial \phi_c \partial \mu} \Big|_{\epsilon_{ij}} = \frac{\partial^2(f_m - n_r \mu)}{\partial \mu \partial \phi_c} \Big|_{\epsilon_{ij}} \quad (A.14)$$

805 so that,

$$47 \quad \frac{\partial p_c}{\partial \mu} \Big|_{\epsilon_{ij}, \phi_c} = - \frac{\partial n_r}{\partial \phi_c} \Big|_{\epsilon_{ij}, \mu} \quad (A.15)$$

806 By using the micromechanical Eq. (A.5), the latter equation is equivalent  
807 to

$$55 \quad \frac{\partial p_c}{\partial \mu} \Big|_{\epsilon_{ij}, \phi_c} = \frac{\partial n_m}{\partial \epsilon_m} \Big|_\mu \quad (A.16)$$

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808 Integration of this latter equation on the molar chemical potential  $\mu$  leads  
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11 809 to

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

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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  $\Psi'_{ij}$  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  $\Psi_{ij}$  and  $\Psi'_{ij}$  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  
44  
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  $\phi_c$ . 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  $\sigma_{ij}$  (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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