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**<sub>1</sub> A transverse isotropic model for microporous solids -  
<sub>2</sub> Application to coal matrix adsorption and swelling**

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3 **Abstract.** Understanding the adsorption-induced swelling in coal is crit-  
4 ical for predictable and enhanced coal bed methane production. The coal ma-  
5 trix is a natural anisotropic disordered microporous solid. We develop an elas-  
6 tic transverse isotropic poromechanical model for microporous solids which  
7 couples adsorption and strain through adsorption stress functions, and ex-  
8 presses the adsorption isotherm as a multivariate function depending on fluid  
9 pressure and solid strains. Experimental data from the literature help invert-  
10 ing the anisotropic adsorptive-mechanical properties of Brzeszcze coal sam-  
11 ples exposed to CO<sub>2</sub>. Main findings include: (1) adsorption-induced swelling  
12 can be modeled by including fluid-specific and pressure-dependent adsorp-  
13 tion stress functions into equilibrium equations, (2) modeling results suggest  
14 that swelling anisotropy is mostly caused by anisotropy of the solid mechan-  
15 ical properties, and (3) the total amount of adsorbed gas measured by im-  
16 mersing coal in the adsorbate overestimates adsorption amount compared  
17 to in-situ conditions up to ~20%. The developed fully coupled model can  
18 be upscaled to determine the coal seam permeability through permeability-  
19 stress relationships.

## 1. Introduction

20 The understanding of adsorption of gases in mesoporous and microporous solids is im-  
21 portant for applications in gas capture, gas separation, and gas desorption/adsorption in  
22 sedimentary rocks. Micropores are defined by the IUPAC as pores with width not exceed-  
23 ing 2 nm. We consider here a broader definition of microporous solids including porous  
24 solids with pores sized in the order of a few nanometers. Natural microporous solids in-  
25 clude coal, kerogen, zeolites, and gas hydrates; while artificial microporous solids include  
26 microcarbon and metal organic frameworks among others [*Ravikovitch and Neimark, 2006;*  
27 *Mazumder et al., 2006; Vandembroucke and Largeau, 2007; Sloan and Koh, 2008; Kowal-*  
28 *czyk et al., 2008; Neimark et al., 2011*]. From another point of view, microporous solids  
29 can be classified as regards their structure in crystalline (ordered structure: metal organic  
30 frameworks, zeolites, hydrates) or disordered (coal, kerogen, microporous carbon). Nano-  
31 sized micropores can host just a few fluid molecules. The adsorption of fluids in these  
32 pores can lead to their expansion or contraction depending on the pore size, geometry,  
33 and fluid-solid interaction [*Kowalczyk et al., 2008; Pijaudier-Cabot et al., 2011; Brochard*  
34 *et al., 2012*]. Macroscopically, adsorption-driven changes of pore size or pressure upscale  
35 as strains (or stresses if strains are prescribed).

36 This coupling between strain and adsorption is particularly important in the context of  
37 underground adsorptive geomaterials, such as coal and organic shales [*Busch et al., 2008;*  
38 *Weniger et al., 2010*]. Hydrocarbon production from these reservoirs imposes significant  
39 changes in stresses coupled with adsorption or desorption leading to strains which affect  
40 the aperture and permeability of natural and generated fractures. The adsorption-strain

41 coupling is critical to defining production rates of natural gas from highly-adsorptive coal  
42 bed seams [*Palmer and Mansoori, 1998; Pan and Connell, 2012*].

43 The adsorption and swelling of various gases including carbon dioxide  $\text{CO}_2$ , methane  
44  $\text{CH}_4$ , and nitrogen  $\text{N}_2$  in coal has been largely studied in the past decades [*Reucroft and*  
45 *Sethuraman, 1987; Ceglarska-Stefanska and Czaplinski, 1993; Levine, 1996; Ceglarska-*  
46 *Stefanska and Zarebska, 2002; Mazumder et al., 2006; Pini et al., 2009*]. Adsorbed amounts  
47 are usually measured in the laboratory in terms of excess adsorbed amount, that is, the  
48 amount of fluid in excess of the amount of fluid which would occupy the pore volume  
49 in bulk conditions. In general for low-medium gas pressure, up to  $\sim 10$  MPa, the rule  
50 “the greater the pressure, the greater the total adsorbed amount” is valid. In this range,  
51 coal swelling is usually proportional to the amount of adsorbed gas. For higher pressures,  
52 the bulk fluid pressure compresses the solid skeleton and may prevail over adsorption-  
53 induced swelling [*Moffat and Weale, 1954; Pan and Connell, 2007; Hol and Spiers, 2012*].  
54 In fact, recent studies show that the application of compressive confining stresses (and  
55 ensuing strains) to coal swollen and equilibrated with  $\text{CO}_2$  at a certain pressure induce the  
56 expulsion of adsorbed gas [*Hol et al., 2011, 2012*]. This desorption induced by mechanical  
57 stresses is not due to the loss of pore accessibility caused by permeability reduction upon  
58 the application of compressive external stresses, phenomenon which is also observed in  
59 coal [*Pone et al., 2009; Wang et al., 2011*].

60 Such a particular behavior of coal motivated the extension of the theory of porome-  
61 chanics to include adsorption phenomena in microporous solids. *Brochard et al.* [2012]  
62 proposed a set of poromechanical equations valid for linear elastic isotropic microporous  
63 solids which considers as main variable the total amount of adsorbed fluid as a function

64 of fluid pressure and solid strain, without a need for introducing the notion of pore vol-  
65 ume. The model is based on first principles, shows good swelling prediction capability,  
66 and eventually leads to the classical theory of poromechanics if adsorption is negligible.

67 Although *Brochard et al.* [2012] developed a model applicable to isotropic coal matrix,  
68 most sedimentary geomaterials are not isotropic. Vertical deposition and overburden at  
69 zero-lateral strain favor transverse isotropic symmetry [*Thomsen*, 1986; *Johnston and*  
70 *Christensen*, 1995; *Vernik and Nur*, 1992]. Anisotropic poromechanical responses can be  
71 quantified by the Biot effective stress coefficient tensor or equivalently by the Skempton  
72 tensor [*Cheng*, 1997; *Cowin*, 2004]. The mechanical and swelling properties of coal are  
73 notably anisotropic, with a major influence of the orientation respect to the bedding plane  
74 and the proximity to cleats [*Cody et al.*, 1988; *Levine*, 1996; *Hol and Spiers*, 2012; *Day*  
75 *et al.*, 2010; *Morcote et al.*, 2010].

76 This article presents an extension of the poromechanical model for adsorptive microp-  
77 orous solids developed by *Brochard et al.* [2012] to take into account material properties  
78 anisotropy. The anisotropy is integrated for both adsorption-induced phenomena and  
79 mechanical properties, and it is implemented by means of a transverse isotropic model  
80 pertinent to sedimentary rocks. The model is used to invert the anisotropic adsorptive-  
81 mechanical properties of a sample of Brzeszcze coal tested by *Hol and Spiers* [2012]. The  
82 model offers further insight into the relationship between swelling and adsorption.

## 2. Model formulation

83 The model is based on energy conservation and follows from a thermodynamical formu-  
84 lation. A more detailed formulation developed for isotropic microporous solids is available  
85 elsewhere [*Brochard et al.*, 2012]. We aim at developing the poromechanical equations for

86 adsorbing microporous solids which exhibit transverse isotropic properties. The interested  
 87 reader will find similar theoretical developments applied to various poromechanical prob-  
 88 lems in [Coussy, 2004, 2010]. The theory of poroelasticity for anisotropic macroporous  
 89 solids has been revisited by several authors [Cheng, 1997; Coussy, 2004; Cowin, 2004].

## 2.1. Energy conservation

90 Consider a representative elementary volume of a microporous solid with free energy per  
 91 unit volume  $f$  [J/L], subjected to stresses  $\sigma_{ij}$  [Pa], and whose adsorbed fluid has a molar  
 92 chemical potential  $\mu$  [J/mol]. Energy conservation under isothermal conditions dictates

$$df = \sum_{i,j} \sigma_{ij} d\varepsilon_{ij} + \mu dn \quad (1)$$

93 being  $\varepsilon_{ij}$  the strain tensor [-], and  $n$  the number of moles of fluid per unit volume of  
 94 undeformed microporous solid [mol/L]. Strains are calculated respect to the original con-  
 95 figuration (Lagrangian formulation). By changing variables, the following holds

$$d(f - n\mu) = \sum_{i,j} \sigma_{ij} d\varepsilon_{ij} - nd\mu \quad (2)$$

96 The first term to the right of the equality represents the incremental work done by  
 97 stresses and the second represents the change of energy associated to changes in the  
 98 chemical potential of the fluid phase. The development of the model consists in finding  
 99 expressions to the variables  $\sigma_{ij}$ , and  $n$  as a function of the other variables. Volume  
 100 average considerations, Maxwell's relations, and the Gibbs-Duhem equation are used to  
 101 this purpose.

## 2.2. Derivation of equations of state

Let us apply Maxwell's relation on the previous energy conservation equality (Eq. 2),

$$\frac{\partial^2(f - n\mu)}{\partial\varepsilon_{ij}\partial\mu} = \frac{\partial^2(f - n\mu)}{\partial\mu\partial\varepsilon_{ij}} \quad (3)$$

Derivation leads to

$$\left. \frac{\partial\sigma_{ij}}{\partial\mu} \right|_{\varepsilon_{kl\neq ij}} = - \left. \frac{\partial n}{\partial\varepsilon_{ij}} \right|_{\mu, \varepsilon_{kl\neq ij}} \quad (4)$$

The equality holds for strains  $\varepsilon_{kl\neq ij}$  constant. By integrating this equation on the molar chemical potential  $\mu$  we obtain the following equation

$$\sigma_{ij} = \Psi_{ij}(\underline{\varepsilon}) - \int_{-\infty}^{\mu} \left. \frac{\partial n}{\partial\varepsilon_{ij}} \right|_{\mu, \varepsilon_{kl\neq ij}} d\mu \quad (5)$$

where  $\Psi_{ij}$  results from this integration and, thus, is a function independent from the fluid chemical potential  $\mu$ . The molar chemical potential  $\mu$  can be re-written using Gibbs-Duhem relation for isothermal conditions  $d\mu = \bar{V}_b(p)dp$ , where  $\bar{V}_b$  is the molar volume of the fluid in bulk conditions [L/mol] as a function of temperature and pressure, and  $p$  is the thermodynamic pressure of the fluid [Pa] defined as the pressure of the bulk fluid at the same chemical potential as the adsorbed phase. Hence, Eq. (5) can be written as follows

$$\sigma_{ij} = \Psi_{ij}(\underline{\varepsilon}) - \int_0^p \left. \frac{\partial n}{\partial\varepsilon_{ij}} \right|_{p, \varepsilon_{kl\neq ij}} \bar{V}_b(p) dp \quad (6)$$

The first term to the right of the equality  $\Psi_{ij}$  represents the “dry” response of the specimen to strains (without the fluid phase) and the integral term represents the response



115 to fluid loading as a function of fluid pressure and solid strain. For example, assuming  
 116 linear elasticity, isotropic material properties, and that the fluid is in bulk conditions,  
 117 *Brochard et al.* [2012] show that Eq. (6) permits recovering the classical equations of  
 118 poroelasticity  $\sigma = K\epsilon - bp$ , where  $K$  is the drained bulk modulus,  $\epsilon$  is the volumetric  
 119 strain, and  $b$  is the Biot coefficient).

### 2.3. Simplification for transverse isotropic coal

120 **Linear elastic transverse isotropy.** The functions  $\Psi_{ij}$  depend on strains  $\underline{\underline{\epsilon}}$  only, hence,  
 121 any linear or non-linear elastic constitutive law may be used. We use transverse isotropic  
 122 linear elasticity for functions  $\Psi_{ij}$  in this formulation. Transverse isotropic materials are  
 123 defined by an axis of symmetry in their elastic properties (say axis 3 in this development).  
 124 The reduced stiffness matrix in Voigt notation  $C_{ij}$  has 5 independent constants. Thus,  
 125 the “dry” response of a generic transversely isotropic porous solid  $\sigma_{ij} = \Psi_{ij}(\underline{\underline{\epsilon}})$  is

$$\begin{cases} \sigma_{11} = C_{11}\epsilon_{11} + C_{12}\epsilon_{22} + C_{13}\epsilon_{33} \\ \sigma_{22} = C_{12}\epsilon_{11} + C_{11}\epsilon_{22} + C_{13}\epsilon_{33} \\ \sigma_{33} = C_{13}\epsilon_{11} + C_{13}\epsilon_{22} + C_{33}\epsilon_{33} \\ \sigma_{23} = 2C_{44}\epsilon_{23} \\ \sigma_{31} = 2C_{44}\epsilon_{31} \\ \sigma_{12} = (C_{11} - C_{12})\epsilon_{12} \end{cases} \quad (7)$$

126 Deformation can be expressed in terms of engineering elastic constants  $E$  (Young mod-  
 127 ulus in direction 1 and 2),  $E_3$  (Young modulus in direction 3),  $\nu$  (Poisson ratio in plane  
 128 1-2),  $\nu_3$  (Poisson ratio in planes 1-3 and 2-3), and  $G_3$  (shear modulus in plane 1-3 and  
 129 2-3) as  $\epsilon_{kl} = \Psi'_{kl}(\underline{\underline{\sigma}})$ ,

$$\left\{ \begin{array}{l} \varepsilon_{11} = (1/E)\sigma_{11} - (\nu/E)\sigma_{22} - (\nu_3/E_3)\sigma_{33} \\ \varepsilon_{22} = -(\nu/E)\sigma_{11} + (1/E)\sigma_{22} - (\nu_3/E_3)\sigma_{33} \\ \varepsilon_{33} = -(\nu_3/E_3)\sigma_{11} - (\nu_3/E_3)\sigma_{22} + (1/E_3)\sigma_{33} \\ 2\varepsilon_{23} = (1/G_3)\sigma_{23} \\ 2\varepsilon_{31} = (1/G_3)\sigma_{31} \\ 2\varepsilon_{12} = [2(1 + \nu)/E]\sigma_{12} \end{array} \right. \quad (8)$$

130 **Multivariate adsorption isotherm.** The derivative  $\partial n / \partial \varepsilon_{ij} \big|_{p, \varepsilon_{kl} \neq ij}$  in the right hand  
131 side of Eq. (6) can be further developed if the total amount of adsorbed fluid  $n(p, \varepsilon_{ij})$  is  
132 known as a function of the thermodynamic pressure of the fluid  $p$  and the strains of the  
133 microporous solid  $\varepsilon_{ij}$ . Considering small strains, this multivariate adsorption isotherm  
134 can be expressed as a first-order expansion with respect to strains, such that,

$$n(p, \varepsilon_{ij}) = n_0(p)[1 + c_1(p)\varepsilon_{11} + c_2(p)\varepsilon_{22} + c_3(p)\varepsilon_{33}] \quad (9)$$

135 where  $n_0(p) = n(p, \underline{0})$  is the total amount of adsorbed fluid at zero solid strain, and  $c_1(p)$ ,  
136  $c_2(p)$ ,  $c_3(p)$  are the adsorption-strain coupling functions for each direction [-]. Coefficients  
137  $c_i(p)$  capture the coupling between adsorption and strain, and depend on fluid pressure,  
138 type of gas, pore shape and size distribution. *Brochard et al.* [2012] performed molecular  
139 simulations of CH<sub>4</sub> adsorption in disordered organic microporous solids and validated this  
140 first-order expansion (Eq. 9) in the isotropic case, for volumetric strains up to  $\sim 10\%$ . Such  
141 strains are sufficiently large to consider that this first-order expansion is valid for coal in  
142 practical conditions. We will consider that this validity remains in the case of transverse  
143 isotropy. Also, *Brochard et al.* [2012] show that adsorption-strain coupling coefficients can  
144 be negative when adsorption induces pore shrinkage or positive if pore expansion prevails.  
145 Transverse isotropy in the adsorption-strain coupling imposes  $c_1(p) = c_2(p)$ .

146 Various adsorbing materials, including coal exposed at moderate gas pressure, are well  
 147 described by Langmuir adsorption isotherms as a function of pressure or more appropri-  
 148 ately as a function of the fluid fugacity [*Fowler, 1935; Vandamme et. al., 2010; Hol et al.,*  
 149 *2011*]. Hence, assuming a Langmuir-type of adsorption as a function of pressure for the  
 150 undeformed microporous coal leads to

$$n_0(p) = n_0^{max} \frac{p}{\beta + p} \quad (10)$$

151 with  $n_0^{max}$  being the maximum total amount of adsorbed fluid at zero solid strain [mol/L],  
 152 and  $\beta$  the Langmuir pressure [Pa] at which one half of the adsorption capacity is reached  
 153 while keeping zero solid strain.

154 From Eq. (9) and (10), the derivatives of  $n(p, \varepsilon_{ij})$  with respect to strains  $\varepsilon_{ij}$  result:

$$\left. \frac{\partial n}{\partial \varepsilon_{ij}} \right|_{p, \varepsilon_{kl} \neq ij} = c_i(p) n_0(p) \delta_{ij} \quad (11)$$

155 Eq. (9) implicitly assumes that the principal directions of the tensor  $\partial n / \partial \varepsilon_{ij} \big|_{p, \varepsilon_{kl} \neq ij}$  co-  
 156 incide with the geometrical axes of symmetry. Hence, these derivatives are non-zero only  
 157 with respect to strain components  $\varepsilon_{ii}$  and vanish for shear strains components  $\varepsilon_{ij}$  with  
 158  $i \neq j$ . Back to Eq. (6), adsorption-induced strains and stresses on the anisotropic micro-  
 159 porous solid can be quantified with terms named  $s_i^a(p) = \int_0^p \partial n / \partial \varepsilon_{ij} \big|_{p, \varepsilon_{kl} \neq ij} \bar{V}_b dp$ , which,  
 160 with the help of Eq. (11), become

$$s_i^a(p) = \int_0^p c_i(p) n_0(p) \bar{V}_b(p) dp \quad (12)$$

161 We call these terms “adsorption stresses” as motivated by an analogy with the theory  
 162 formulated by *Ravikovitch and Neimark* [2006]. Physically, these terms quantify the  
 163 stresses needed to keep the microporous solid at zero volumetric strain during adsorption.

164 **Final equations.** Combining the linear elastic constitutive equations (Eq. 7) with  
 165 adsorption stresses (Eq. 12), the set of equations (Eq. 6) becomes

$$\left\{ \begin{array}{l} \sigma_{11} = C_{11}\varepsilon_{11} + C_{12}\varepsilon_{22} + C_{13}\varepsilon_{33} - s_1^a(p) \\ \sigma_{22} = C_{12}\varepsilon_{11} + C_{11}\varepsilon_{22} + C_{13}\varepsilon_{33} - s_1^a(p) \\ \sigma_{33} = C_{13}\varepsilon_{11} + C_{13}\varepsilon_{22} + C_{33}\varepsilon_{33} - s_3^a(p) \\ \sigma_{23} = 2C_{44}\varepsilon_{23} \\ \sigma_{31} = 2C_{44}\varepsilon_{31} \\ \sigma_{12} = (C_{11} - C_{12})\varepsilon_{12} \end{array} \right. \quad (13)$$

166 The model relies on five mechanical parameters  $C_{11}$ ,  $C_{33}$ ,  $C_{12}$ ,  $C_{13}$ ,  $C_{44}$  (stiffness co-  
 167 efficients which can be expressed as a function of  $E$ ,  $\nu$ ,  $E_3$ ,  $\nu_3$ ,  $G_3$ ), one fluid equation  
 168 of state  $\bar{V}_b(p)$  at a given temperature, two characteristic total adsorption parameters  
 169  $n_0^{max}$  and  $\beta$  for the microporous solid at zero strain, and two adsorption-strain coupling  
 170 pressure-dependent functions  $c_1(p)$  and  $c_3(p)$ .

### 3. Application example - Unconstrained coal swelling

171 Let us consider a piece of coal matrix immersed in  $\text{CO}_2$  under unconstrained dis-  
 172 placement boundary conditions (unjacketed: no membrane or wall, the solid is free  
 173 to swell), such that  $\text{CO}_2$  pressure applies isotropic loading and no shear on the solid  
 174 ( $\sigma_{11} = \sigma_{22} = \sigma_{33} = -p$ ;  $\sigma_{ij} = 0$  for  $i \neq j$ ). The coal is in thermodynamical equilibrium,  
 175 thus, the chemical potential of the bulk fluid at pressure  $p$  is the same as the chemical  
 176 potential of the adsorbed fluid. Isotropic loading in a transverse isotropic medium imposes  
 177  $\varepsilon_{11} = \varepsilon_{22}$  in the general case  $C_{11} \neq C_{12}$  (from Eq. 13),  $\varepsilon_{11} = \varepsilon_{22} = \varepsilon_1$  the principal strain

178 on the bedding plane, and  $\varepsilon_{33} = \varepsilon_3$  the principal strain perpendicular to the bedding  
 179 plane, hence, Eq. (13) reduces to

$$\begin{cases} \sigma_{11} = \sigma_{22} = -p = (C_{11} + C_{12})\varepsilon_1 + C_{13}\varepsilon_3 - s_1^a(p) \\ \sigma_{33} = -p = 2C_{13}\varepsilon_1 + C_{33}\varepsilon_3 - s_3^a(p) \end{cases} \quad (14)$$

180 Let us write the principal strains  $\varepsilon_1$  and  $\varepsilon_3$  as function of stresses using the engineering  
 181 elastic constants. Swelling strains result

$$\begin{bmatrix} \varepsilon_1 \\ \varepsilon_3 \end{bmatrix} = \begin{bmatrix} \frac{1-\nu}{E} & -\frac{\nu_3}{E_3} \\ -2\frac{\nu_3}{E_3} & \frac{1}{E_3} \end{bmatrix} \begin{bmatrix} s_1^a(p) - p \\ s_3^a(p) - p \end{bmatrix} \quad (15)$$

182 Recent experimental results show the swelling of cylindrical Brzeszcze coal specimens  
 183 immersed in CO<sub>2</sub> up to 100 MPa of fluid pressure (specimen size 4mm length and 4mm  
 184 diameter [*Hol and Spiers, 2012*]). Strains are measured in two perpendicular directions in  
 185 the bedding plane (noted  $\varepsilon_x$  and  $\varepsilon_y$ ), at 45° between the  $x$  and  $y$  axes, and perpendicular to  
 186 the bedding plane (this latter noted  $\varepsilon_z$ ). Results show a clear orthotropic swelling of coal  
 187 specimens, being the strain perpendicular to the bedding plane the highest  $\varepsilon_z > \varepsilon_y \sim \varepsilon_x$ .  
 188 We consider the major principal strain  $\varepsilon_3 = \varepsilon_z$ . We calculate the other two principal  
 189 strains from the original data set based on the three strains measured in the bedding plane.  
 190 Figures 1-a and 1-d show the principal strains for two coal specimens 364-1 and 364-2,  
 191 as named in the source reference. Although the experimental results show an orthotropic  
 192 response, we fit a transverse isotropic model in views of upscaling to coal seams with  
 193 transverse isotropic properties and to reduce the number of inverted parameters. The  
 194 experimental principal strain  $\varepsilon_3$  and the mean of the experimental principal strains on  
 195 the bedding plane  $(\varepsilon_1 + \varepsilon_2)/2$  are used to validate the transverse isotropic model.

196 Additional data help us invert the coal adsorptive-mechanical properties; these data  
197 include: (1) total amount of adsorption in Brzeszcze coal immersed in CO<sub>2</sub> up to 15 MPa  
198 (estimated from excess adsorbed amount measurements at 45°C by *Gensterblum et al.*  
199 [2010] with best fitting adsorbed phase volume per unit mass of coal  $V_{ads}/m_{coal} = 0.055$   
200 cm<sup>3</sup>/g - see Auxiliary Material); (2) the dry coal mass density  $\rho \sim 1343$  kg/m<sup>3</sup> [*Hol et al.*,  
201 2011]; (3) the molar volume of CO<sub>2</sub> at 40°C as a function of pressure (from equation of  
202 state by *Span and Wagner* [1996]). The coal is expected to adsorb slightly less CO<sub>2</sub> at  
203 45°C than at 40°C, temperature at which swelling strains are measured. In the calculation  
204 performed here to estimate total adsorbed amounts from excess adsorbed amounts, we  
205 adopt the procedure proposed by *Ottiger et al.* [2006]. We thus neglect the effect of  
206 mesopores and adsorption-induced deformations on the adsorption process, which is a  
207 very simplistic approach. However, correctly interpreting gravimetric adsorption data  
208 by taking into account the full coupling between adsorption and strain still remains a  
209 challenging issue. While this issue is not fully solved, in addition to the interpretation  
210 proposed here, as alternatives, one can aim at using models based directly on excess  
211 adsorption data (e.g., *Vermorel et al.* [2013a, b]) or instead at measuring total adsorbed  
212 amounts directly (e.g., as proposed by *Hol et al.* [2011]).

213 In order to bound the inverted parameters, we follow two paths for the inversion pro-  
214 cedure. First, we explore the anisotropy of adsorption-induced strains, and conclude  
215 that assuming isotropic and pressure-independent adsorption stresses is reasonable for  
216 the analyzed coal and permits recovering a plausible estimate of the coal mechanical  
217 anisotropy. Second, mechanical isotropy is assumed and the adsorption-strain coupling  
218 coefficients are recovered (still considered pressure-independent), so that, the anisotropy

of adsorption-strain coupling coefficients is determined. Last, we show that introducing a pressure-dependent coupling coefficients  $c_1(p) \neq c_3(p)$  (based on the experimentally measured swelling strains and anisotropic mechanical elastic constants) enables a better the fitting of the experimental data.

### 3.1. Mechanical anisotropy and isotropic adsorption stresses

We first investigate whether adsorption-induced stresses can be assumed isotropic or not. According to our poromechanical model (Eq. 15), having equal coupling coefficients  $c_1(p) = c_3(p)$  or equivalently  $s_1^a(p) = s_3^a(p) = s^a(p)$  leads to,

$$\begin{cases} \varepsilon_1 = [s^a(p) - p] \left( \frac{1-\nu}{E} - \frac{\nu_3}{E_3} \right) \\ \varepsilon_3 = [s^a(p) - p] \left( \frac{1-2\nu_3}{E_3} \right) \end{cases} \quad (16)$$

which means that under such an assumption the ratio  $\varepsilon_1/\varepsilon_3 = f(E, E_3, \nu, \nu_3)$  should be constant and independent of gas pressure and adsorption stresses. Figures 1-b and 1-e show the experimental ratios between the principal strains for both specimens. It is observed that  $\varepsilon_1/\varepsilon_3$  and  $\varepsilon_2/\varepsilon_3$  decrease slightly with pressure by about  $\sim 10\%$  for fluid pressure  $p < 10$  MPa, which indicates slight anisotropy of adsorption stresses only in this range of pressure. Hence, as a first approximation we set adsorption stresses equal in all directions  $s_1^a = s_3^a = s^a$  with constant adsorption coupling functions so that  $c_1(p) = c_3(p) = c$ . Assuming isotropic adsorption stresses leads to recovering an upper bound for mechanical anisotropy, which can be assessed by the parameter  $(C_{11} - C_{33})/(2C_{33})$ . Additional assumptions help reduce the number of inverted parameters: non-diagonal stiffness coefficients are approximately equal  $C_{12} \sim C_{13}$  (as observed in many shales with weak anisotropy from an extensive data compilation by Wang [2002]), and one of the

238 Poisson ratios is assumed  $\nu_3 = 0.28$  according to typical coal Poisson ratios determined  
 239 indirectly from elastic waves velocity [Morcote et al., 2010].

240 By assuming  $C_{12} \sim C_{13}$ , the following moduli ratio holds,

$$\frac{E}{E_3} = \frac{\nu_3 + \nu_3\nu - \nu}{\nu_3^2} \quad (17)$$

241 hence, given  $\nu_3$ ,  $\nu = f(E/E_3)$ . Given the total adsorbed amount and swelling principal  
 242 strains of coal immersed in CO<sub>2</sub> as a function of pressure ( $\varepsilon_1$ ,  $\varepsilon_2$ ,  $\varepsilon_3$ ), and a coupling  
 243 coefficient  $c$ , the parameters  $n_0^{max}$  and  $\beta$  can be determined to best fit Eq. (9). Therefore,  
 244 the free fitting parameters are  $E$ ,  $E_3$ , and  $c$ . The shear modulus in planes which contain  
 245 the symmetry axis  $G_3$  is not needed for this particular application.

246 We fit our model simultaneously to the swelling strain and total adsorption amount data  
 247 by forward simulation and 2-norm error estimation. Modeled strains  $\varepsilon_1$  and  $\varepsilon_3$  are set to  
 248 fit experimentally measured  $(\varepsilon_1 + \varepsilon_2)/2$  and  $\varepsilon_3$  respectively. The calculated error surfaces  
 249 are available as Auxiliary Material. Figures 1-a,c,d,f show the best-fitting modeling results  
 250 along with the experimental data and best fitting parameters. Notice that  $E$  and  $E_3$  are  
 251 the specimen “dry” or “drained” moduli. The unjacketed response of the same specimens  
 252 to Helium (specimen 364-1:  $d\varepsilon_3/dp = 0.0050$  %/MPa,  $d\varepsilon_1/dp \sim 0.0040$  %/MPa; specimen  
 253 364-2:  $d\varepsilon_3/dp = 0.0049$  %/MPa,  $d\varepsilon_1/dp \sim 0.0040$  %/MPa - original data set from *Hol and*  
 254 *Spiers* [2012]) suggests stiffer moduli in the direction parallel to the bedding plane. From  
 255 our inversion results, we indeed find that  $E > E_3$ . The inverted mechanical anisotropy in  
 256 terms of the Thomsen parameter  $\varepsilon^* = (C_{11} - C_{33})/(2C_{33})$  is about 0.137 (specimen 364-1)  
 257 and 0.082 (specimen 364-2), in agreement with Thomsen parameters measured in other  
 258 coals  $\varepsilon^* \sim 0.05 - 0.10$  [Morcote et al., 2010] and shales  $\varepsilon^* = 0.17 \pm 0.08$  [Wang, 2002].



259 Inverted bulk moduli  $K=4.21$  GPa for specimen 364-1 and  $K=3.58$  GPa for specimen 364-  
 260 2 are in the range of values for bituminous coal: 0.79 to 5.3 GPa (spanning several scales  
 261 and loading rates [*Mazumder, 2007; Morcote et al., 2010; Pan et al., 2010; Masoudian*  
 262 *et al., 2013*]).

### 3.2. Mechanical isotropy and anisotropic adsorption stresses

263 Let us now assume isotropic mechanical properties and find the pressure-independent  
 264 adsorption-strain coupling coefficients  $c_1$  and  $c_3$  which best fit the experimental data.  
 265 Having  $E = E_3$  and  $\nu = \nu_3$ , Eq. (15) leads to

$$\begin{cases} \varepsilon_1 = \frac{1-\nu}{E} [s_1^a(p) - p] - \frac{\nu}{E} [s_3^a(p) - p] \\ \varepsilon_3 = -\frac{2\nu}{E} [s_1^a(p) - p] + \frac{1}{E} [s_3^a(p) - p] \end{cases} \quad (18)$$

266 We simplify the problem similarly as done above by assuming one known Poisson ratio  
 267  $\nu = 0.28$ . Forward simulation and error estimation let us recover the best fitting parameters  
 268  $E$ ,  $c_1$ , and  $c_3$ . Figure 2 shows the simulation results for the best fitting parameters.  
 269 Auxiliary Material contains the error surfaces for best fitting parameter determination.  
 270 The fact that  $c_3 > c_1$  implies that adsorption stresses would be higher in the direction  
 271 perpendicular to the bedding plane. We find that the upper bound for adsorption stress  
 272 anisotropy is  $(c_3 - c_1)/(2c_1) \sim 0.048$  for specimen 364-1 and 0.030 for specimen 364-2. The  
 273 fitting is overall poorer than the one observed by concentrating anisotropy on mechanical  
 274 elastic moduli.

### 3.3. Pressure-dependent adsorption-strain coupling coefficients

275 Let us use now the values of  $E_3$ ,  $E$ ,  $\nu_3$ , and  $\nu$  found in Section 3.1 and relax the previous  
 276 assumption of pressure-independent coupling coefficients. By taking the derivatives of Eq.

277 (15) with respect to pressure and multiplying by the inverse of the compliance matrix,  
 278 the following holds,

$$\begin{bmatrix} \frac{ds_1^a(p)}{dp} - 1 \\ \frac{ds_3^a(p)}{dp} - 1 \end{bmatrix} = \begin{bmatrix} \frac{1-\nu}{E} & -\frac{\nu_3}{E_3} \\ -2\frac{\nu_3}{E_3} & \frac{1}{E_3} \end{bmatrix}^{-1} \begin{bmatrix} \frac{d\varepsilon_1}{dp} \\ \frac{d\varepsilon_3}{dp} \end{bmatrix} \quad (19)$$

279 Hence, calculating the value of  $ds_i^a(p)/dp$ , the coupling coefficients follow from Eq.  
 280 (12) and are equal to  $c_i(p) \sim 8$  to 11 (Figure 3-a,c - Note:  $d\varepsilon_1/dp$  is calculated from  
 281 experimental  $(\varepsilon_1 + \varepsilon_2)/2$  as a function of pressure). The difference between  $c_3(p)$  and  
 282  $c_1(p)$  is maximum at pressure  $p < 10$  MPa as suggested by Figure 1. The lower bound for  
 283 anisotropy of adsorption stresses is estimated with the average difference  $\overline{|c_3 - c_1|}/(2\overline{c_1})$   
 284 equal to 0.013 for specimen 364-1 and 0.011 for specimen 364-2. We find that assuming  
 285 a constant coupling coefficient with pressure does not capture perfectly the coal matrix  
 286 linear compressibility observed experimentally at high pressures  $p > 10$  MPa (See Figures  
 287 1-a-d). In contrast, a better curve matching is observed by considering pressure-dependent  
 288 coupling coefficients  $c_1(p) \neq c_3(p)$ . The recalculated swelling strains are plotted with the  
 289 refitted adsorption variables  $n_0^{max}$  and  $\beta$  in Figure 3-b,d for both specimens.

## 4. Discussion

### 4.1. Comparison with empirical approaches and non-empirical theories

290 Early empirical developments simplify the effect of coal swelling in reservoir geome-  
 291 chanics to be analogous to that of thermal dilation [*Palmer and Mansoori, 1998; Shi and*  
 292 *Durucan, 2004*], such that, coal swelling is expressed empirically by fitting a Langmuir  
 293 isotherm directly to swelling strains, or assuming a linear relationship between swelling  
 294 and Langmuir-fitted adsorption amount (See a comprehensive review in *Pan and Connell*

295 [2012]). Such empirical developments are sometimes too simplistic, as here for instance,  
 296 monotonic Langmuir functions would not be able to fit well the non-monotonic swelling  
 297 strains shown in Figure 1.

298 Other non-empirical models have been recently proposed to explain and predict the  
 299 adsorption-induced swelling of coal. First, *Hol and Spiers* [2012] proposed a thermody-  
 300 namical model to explain the Brzeszcze coal swelling data we use in this article. They base  
 301 their formulation on the separation of adsorption-induced swelling and bulk fluid-driven  
 302 compression (similar to Eq. 15), with the additional hypothesis of a linear relationship  
 303 between total adsorption amount and adsorption strain. Our model shows fair agreement  
 304 with this hypothesis for samples immersed in the adsorbate at low gas pressure  $p < 10$   
 305 MPa, although deviations may occur at higher pressures when the bulk fluid pressure  
 306 compression of the coal matrix prevails decreasing the total amount of adsorption (Figure  
 307 1-c,f). Anisotropy modeling is not included by *Hol and Spiers* [2012], although it has  
 308 been considered by the same authors elsewhere [*Hol et al.*, 2012]. Second, *Vermorel et al.*  
 309 [2013a] developed a poromechanical formulation based on properties defined at the molec-  
 310 ular scale, such as microporosity or microporous skeleton stiffness  $K_s$ . This formulation  
 311 proposes an adsorption-mechanical coupling based on the definition of an apparent micro-  
 312 porosity which depends on excess  $n_{ex}$  and total  $n_t$  adsorbed amounts. We find the isotropic  
 313 version of our formulation compatible with theirs if  $\partial n / \partial \epsilon|_p = (1 - K/K_s)n_t / (n_t - n_{ex})$   
 314 where  $\epsilon$  is the volumetric deformation (see Eq. 11). Third, *Pan and Connell* [2011] de-  
 315 veloped a swelling model for mesoporous solids which includes orthotropy. They base  
 316 their model on a crossed-tube analogous structure. In contrast, our model does not as-  
 317 sume a particular geometry and mechanical properties ensues naturally from continuum

318 mechanics. Last, *Coudert et al.* [2011] present a general thermodynamical formulation  
 319 of adsorption-strain couplings in soft microporous zeolites and metal organic frameworks.  
 320 Their formulation shares many common points with our model.

321 All these non-empirical models have shown an advance in adsorption-strain understand-  
 322 ing and good fitting results. We believe our model is thermodynamically and mechan-  
 323 ically rigorous, based on well-defined quantities, valid for any strain or stress boundary  
 324 conditions, and easily expandable to more complex constitutive laws (orthotropy, non-  
 325 linearity). Our theoretical model has the key advantage over empirical developments of  
 326 having fully coupled equations which relate adsorption not only to strain but also to stress.  
 327 Furthermore, our model enables the prediction of the influence of adsorption on strains  
 328 and stresses as well as the influence of stresses and strains on the amount of adsorbed  
 329 fluid. The prediction of stresses is advantageous to use permeability-stress laws instead  
 330 of permeability-porosity laws.

## 4.2. Adsorption isotherms for samples immersed in the adsorbate

331 We emphasize that adsorption isotherms of free-swelling coal samples immersed in the  
 332 adsorbate represent just one path on the multivariate adsorption isotherm  $n(p, \varepsilon_{ij})$  which  
 333 describes how total adsorbed amount depends on fluid pressure and solid strains. Thus,  
 334 the measurements from this kind of experiments are only relevant to this pressure-strain  
 335 path and are not representative of the adsorption isotherm with the real boundary in-situ  
 336 conditions. As shown in Figures 1-c,f and 2-b,d, the difference in adsorbed amount for a  
 337 coal sample immersed in CO<sub>2</sub> (displacement-unconstrained, free swelling) and coal sam-  
 338 ple kept at zero strain is  $\sim 20\%$ . External confining stresses, estimated at about  
 339  $\sim 90$  MPa with our model, must be applied in order to prevent any deformation of the

340 Brzeszcze coal sample (CO<sub>2</sub> pressure  $p = 10$  MPa - from  $\varepsilon_{ij} = 0$  in Eq. 13). In-situ  
341 conditions lie in between these two scenarios. In general, the adsorption isotherm is a  
342 multivariate function which depends on pressure and strains. Hence, there may be ad-  
343 sorption/desorption at constant fluid pressure driven by strain changes such as mechanical  
344 stretching and compression (Eq. 11 - See experimental evidence in [*Hol et al.*, 2012; *Hol*  
345 *and Spiers*, 2012]).

### 4.3. Relation between volumetric strain and total adsorption for samples immersed in the adsorbate

346 Our model predicts an approximately linear relationship between total adsorption  
347 amount and strain when adsorption-induced swelling prevails for adsorption experiments  
348 with coal samples immersed in the adsorbate, i.e., at low-medium fluid pressure (Figure  
349 4-a,b,  $p < 10$  MPa). However, as predicted by our model (no adsorption amount experi-  
350 mental data in this pressure range), total adsorption amount may decrease as bulk fluid  
351 pressure takes more relevance and compresses the coal matrix. Improved certainty in the  
352 determination of total adsorption amount  $n$  would help corroborate this last statement.  
353 Recall the fact that we follow a Lagrangian mechanical description, therefore, adsorption  
354 per unit volume of undeformed coal matrix [mol/L] may be converted to adsorption per  
355 unit mass [mol/kg] through the initial mass density [kg/L]. Recently published experi-  
356 mental data of simultaneous measurement of adsorption and swelling up to 15 MPa show  
357 a similar shape of the swelling-adsorption signature as the one predicted by our model  
358 before the onset of coal matrix contraction (see Fig. 8 from *Day et al.* [2008]). Other  
359 phenomena that may affect the shape of the adsorption-strain curve include creep at con-

stant Terzaghi's effective stress and coal matrix adsorption-induced softening [Czaplinski  
and Holda, 1982; Hagin and Zoback, 2010; Masoudian et al., 2013].

#### 4.4. Adsorption-strain coupling expressed with Biot-like coefficients

The Biot coefficient  $\alpha$  was first defined as the ratio of change in fluid content  $\zeta$  to  
dilation  $\epsilon$  in a jacketed (drained) test, such that for a conventional poroelastic material  
 $\zeta = \alpha\epsilon$  [Biot and Willis, 1957]. Biot noted that  $\alpha$  can also be identified to  $1 - K/K_{sk}$   
(where  $K$  and  $K_{sk}$  are the bulk moduli measured in drained and unjacketed conditions)  
which provides a further interpretation of  $\alpha$  different from the first definition. The fact  
that they yield equal values results from the symmetry of the coefficients in the poroelastic  
matrix and in turn from the existence of a thermodynamic potential with pressure  $p$  and  
change in fluid content  $\zeta$  as conjugate variables. In conventional poroelasticity the density  
of the fluid into the porosity of the material corresponds to its bulk density, therefore, the  
Biot coefficient  $\alpha$  is less than 1 as the volume of fluid  $\zeta$  entering the specimen during a  
drained test cannot be greater than its dilation  $\epsilon$ .

However, this is not the case in adsorptive materials such as coal where the density  
of the adsorbed fluid can be much greater than the density of the bulk fluid. In the  
framework of the theory developed here for transverse isotropic microporous solids, the  
incremental amount of fluid content  $d\zeta = d(n\bar{V}_b)$  during a jacketed test is (from Eq. 9)

$$d[n(p, \varepsilon_{ij})\bar{V}_b(p)] = b_1(p)d\varepsilon_1 + b_2(p)d\varepsilon_2 + b_3(p)d\varepsilon_3 \quad (20)$$

where  $b_i(p) = c_i(p)n_0(p)\bar{V}_b(p)$  can be coined as a (tangent) Biot-like coefficient, consis-  
tently with the first definition given by Biot. The fluid content  $n\bar{V}_b$  is the volume (per

unit volume of coal) that the amount of adsorbed fluid would occupy in it were in bulk  
 conditions. Therefore the volume change  $d(n\bar{V}_b)$  can be much larger than the volumetric  
 dilation increment  $d\epsilon$  leading to coefficients  $b_i(p)$  exceeding 1. This does not contradict  
 thermodynamics. Due to the symmetry already discussed, this coefficient  $b_i(p)$  can still be  
 identified to  $1 - K/K_{unj}$  which is equal to  $ds_i^a/dp$  here (Eq. 12), but theunjacketed bulk  
 modulus  $K_{unj}(p)$  does not reveal the solid stiffness alone. It is a macroscopic property  
 which also accounts for the adsorptive properties of the fluid molecules onto the solid pore  
 walls. *Vermorel et al.* [2013b] defines an analogous Biot-like pressure-dependent coefficient  
 as  $b(p) = b^\infty n_T(p)/[n_T(p) - n_{excess}(p)]$  where  $b^\infty < 1$  is the asymptotic Biot coefficient  
 obtained at high fluid pressure.

Given the coal adsorption properties  $(n_0^{max}, \beta, c_i)$  and the CO<sub>2</sub> bulk molar volume  $\bar{V}_b$ ,  
 the Biot-like coefficient ranges from  $b^t \sim 40$  to 1 for low CO<sub>2</sub> pressure  $p = 0$  to  $\sim 10$  MPa  
 (adsorption prevails) and decreases asymptotically to  $\sim 0.74$  as CO<sub>2</sub> pressure increases  
 (plotted in Figure 4-c,d as solid line for pressure-independent  $c$  and as symbols for pressure-  
 dependent  $c_i(p)$ ). The  $b(p)$  curve exhibits an inflexion point which roughly coincides with  
 the limit between adsorption-dominated swelling  $b > 1$  and bulk fluid-driven compression  
 $b < 1$  (Figure 4-c,d).

Although they remain nontrivial, variations of the introduced Biot-like coefficients  $b_i$   
 with the fluid pressure are simpler than variations of the coupling coefficients  $c_i$ , in the  
 sense that at least the former are monotonic. The complexity of the variations of the  
 coefficients  $c_i(p)$  stems from the fact that those coefficients involve characteristics of the  
 solid, of the fluid, and of their interaction. In contrast, since  $b_i(p) = c_i(p)n_0(p)\bar{V}_b(p)$ , the  
 Biot-like coefficients are corrected for variations of density of the bulk fluid. Therefore,

402 when looking for a physical interpretation of the inverted parameters of the model, one  
 403 might find more useful focusing on the Biot-like coefficients rather than on the more  
 404 convoluted coupling coefficients  $c_i$ . For instance, in the isotropic case, *Vermorel et al.*  
 405 [2013b] propose the physical rule of thumb that the Biot-like coefficient  $b(p)$  is proportional  
 406 to the ratio of the density  $\rho_{sorbed}$  of the adsorbed phase to the density  $\rho_{free}$  of the bulk  
 407 phase (i.e.,  $b(p) \propto \rho_{sorbed}(p)/\rho_{free}(p)$ ).

#### 4.5. Implications for coal bed methane recovery

408 The determination of strains and effective stress on the direction of the bedding plane  
 409 in coal seams subjected to primary or secondary methane recovery is critical to predict  
 410 changes in fracture permeability. Various permeability-stress relationships have been pro-  
 411 posed to predict permeability, generally in the form:  $\log(k/k_0) = a(\sigma_h + p - \sigma_{h0} - p_0)$   
 412 (being  $k_0$  the reference permeability at total horizontal stress  $\sigma_{h0}$  and fluid pressure  $p_0$   
 413 and  $a \sim 0.1$  to  $0.2 \text{ MPa}^{-1}$  [*Somerton et al.*, 1975; *Pan and Connell*, 2012]). Thus, a  
 414 general model that predicts stresses in the anisotropic coal seam as a function of fluid  
 415 pressure and boundary conditions is required for realistic predictions and simulations of  
 416 coal bed methane recovery. The coal seam can be modeled as a double-porosity reservoir  
 417 rock, where cleats account for macroporosity and coal matrix micropores account for the  
 418 microporosity. In order to become practical at the scale of the seam, the constitutive  
 419 laws derived here at the scale of the coal matrix must be upscaled to the scale of the  
 420 coal seam by incorporating the presence of the cleats, in the spirit of what was done by  
 421 *Nikoosokhan et al.* [2012]. In this upscaling, the presence of the cleats only induces regular  
 422 poromechanical effects, in the sense that cleats are macropores in which adsorption-effects  
 423 are negligible, and the coal matrix is responsible for adsorption-induced phenomena. This



424 extension of our model will allow to incorporate, in a thermodynamically sound manner,  
425 both anisotropy and adsorption-induced phenomena into the constitutive laws of coal  
426 seams.

## 5. Conclusions

427 Coal matrix is a natural disordered microporous solid. Material properties are often  
428 anisotropic due to sedimentation and diagenetic processes. Although coal adsorptive  
429 properties have been widely studied, the physical link between adsorption and induced-  
430 strain has been relegated to a second plane. The knowledge of adsorptive and mechanical  
431 properties is critical for calculating coal bed gas reserves and storage capacity, and enabling  
432 predictable exploitability.

433 We extended a poromechanical model developed for microporous solids to take into  
434 account adsorptive-mechanical anisotropy through adsorption stresses and a stiffness ma-  
435 trix. Our model ensues from first principles and continuum mechanics, and applies to any  
436 displacement or stress boundary conditions.

437 Experimental data from the literature help validate our model and invert adsorptive-  
438 mechanical model parameters. The model is able to simulate the competition between  
439 adsorption-induced adsorption and bulk fluid-driven compression in adsorption experi-  
440 ments of coal samples immersed in CO<sub>2</sub>.

441 Remarkable lessons from this study and the Brzeszcze coal include: (1) swelling  
442 anisotropy is likely to be caused principally by mechanical anisotropy ( $E > E_3$ ), in  
443 fact, concentrating anisotropy on elastic moduli yields better fitting than concentrat-  
444 ing anisotropy on adsorption stresses, (2) assuming a coupling coefficient  $c$  independent of  
445 fluid pressure is reasonable and yields acceptable predictions, (3) adsorption measurements

446 in coal samples immersed in the adsorbate represent just one path on the multivariate  
447 adsorption isotherm.

448 The developed fully coupled model permits inverting transverse isotropic mechanical-  
449 adsorptive properties of microporous coal matrix. Upscaling to a coal seam model which  
450 includes both macroporous cleats and microporous adsorptive coal matrix would yield the  
451 coupled constitutive equations of the coal seam. This set of fully coupled equations enable  
452 the prediction of strains and effective stress in different directions respect to the bedding  
453 plane under in-situ anisotropic state of stresses. The calculated effective stresses can be  
454 used through permeability-stress relationships for determining seam permeability.

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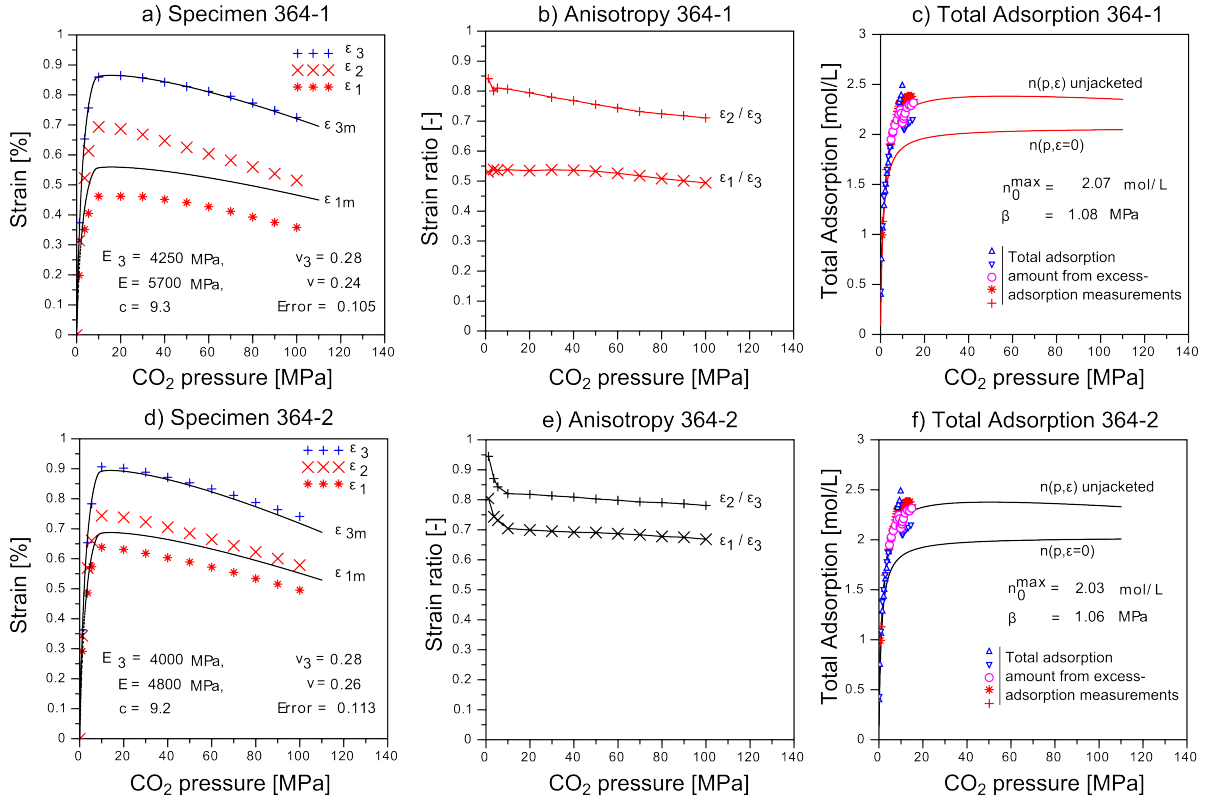
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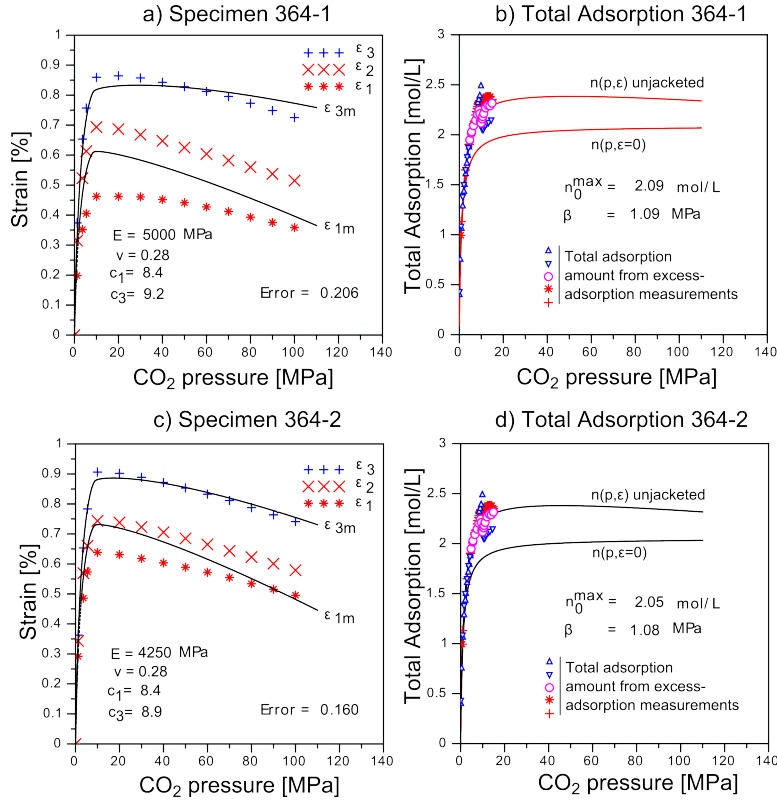
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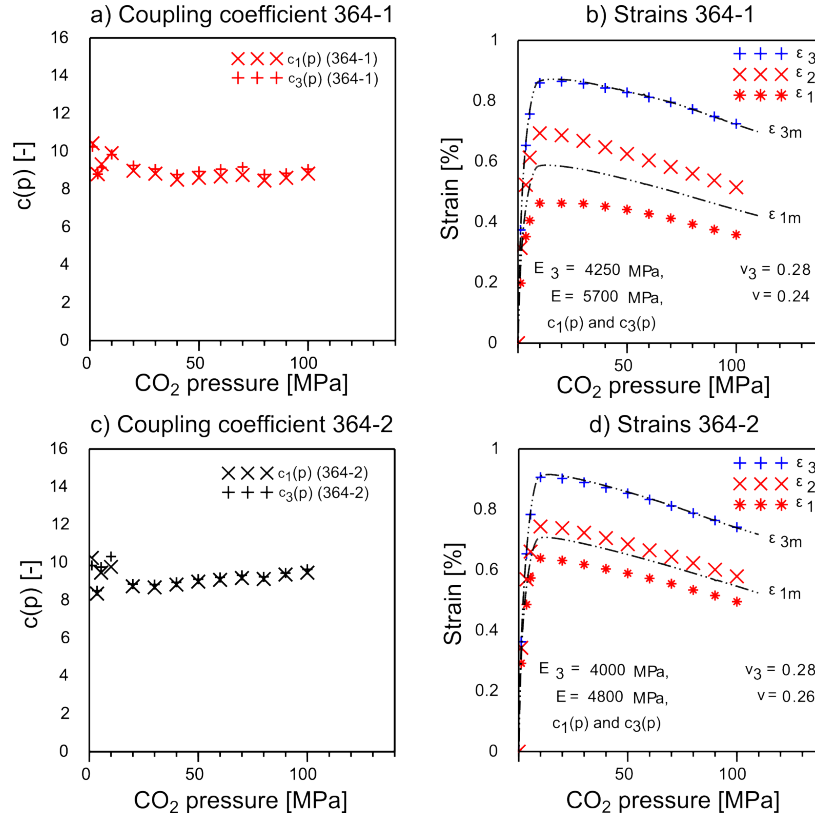
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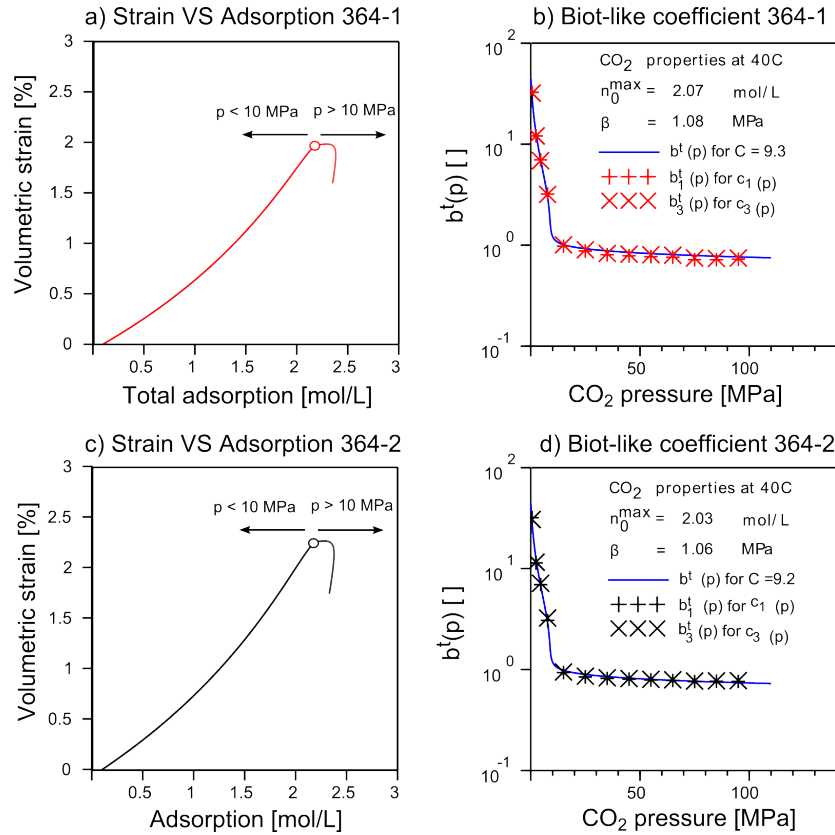
**Figure 1.** Adsorption-induced swelling of coal immersed in CO<sub>2</sub>. (a) Experimental measurements (symbols: principal strains  $\epsilon_1, \epsilon_2, \epsilon_3$  - original data set from *Hol and Spiers* [2012]) and transverse isotropic microporous model predictions considering mechanical anisotropy only  $c_1(p) = c_3(p) = c$  (lines:  $\epsilon_{1m}, \epsilon_{3m}$  fit  $(\epsilon_1 + \epsilon_2)/2$  and  $\epsilon_3$  respectively). (b) Swelling anisotropy: strain ratios  $\epsilon_1$  and  $\epsilon_2$  respect to the major principal strain  $\epsilon_3$  perpendicular to the bedding plane. (c) Total adsorption amount estimated from excess sorption measurements (symbols - see Auxiliary Material) and model fitted prediction (Eq. 9). (a,b,c) Specimen 364-1. (d,e,f) Specimen 364-2.



**Figure 2.** Adsorption-induced swelling of coal immersed in CO<sub>2</sub>. (a) Experimental measurements (symbols: principal strains  $\epsilon_1, \epsilon_2, \epsilon_3$  - original data set from *Hol and Spiers* [2012]) and transverse isotropic microporous model predictions considering anisotropy on adsorption stresses only  $E = E_3$  and  $\nu = \nu_3$  (lines:  $\epsilon_{1m}, \epsilon_{3m}$  fit  $(\epsilon_1 + \epsilon_2)/2$  and  $\epsilon_3$  respectively). (b) Total adsorption amount estimated from excess sorption measurements (symbols - Auxiliary Material) and model fitted prediction (Eq. 9). (a,b) Specimen 364-1. (c,d) Specimen 364-2.



**Figure 3.** Determination of adsorption-strain coupling coefficients as a function of pressure for finding minimum adsorption stress anisotropy. (a) Calculated coupling coefficient  $c_i(p)$  in the bedding plane and perpendicular to the bedding plane (Eq. 19). (b) Transverse isotropic microporous model predictions (lines:  $\epsilon_{1m}, \epsilon_{3m}$ ) considering  $c_i(p)$  previously calculated and refitted  $n_0^{max}$  and  $\beta$  compared to experimental data - original data set from *Hol and Spiers* [2012]. (a,b) Specimen 364-1 - refitted  $n_0^{max} = 2.04$  mol/L and  $\beta = 1.08$ . (d,e) Specimen 364-2 - refitted  $n_0^{max} = 2.00$  mol/L and  $\beta = 1.06$ .



**Figure 4.** Insight into the poromechanics of coal matrix. (a) Model-predicted volumetric strain as a function of total adsorption amount for coal immersed in CO<sub>2</sub> (model parameters from Figure 1). (b) Biot-like pressure dependent functions  $b_i^t(p)$ . (a,b) Specimen 364-1. (c,d) Specimen 364-2.