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1 Modeling transient variations of permeability in coal seams at the reservoir scale

2 by N. Abouloifa, M. Vandamme¹, and P. Dangla

3 Navier, Ecole des Ponts, Univ Gustave Eiffel, CNRS, Marne-la-Vallée, France

4

5 Abstract

6 Production of fluid from or injection of fluid into a coal seam leads to variations of
7 permeability of the seam, resulting from adsorption of fluid in the coal matrix in particular.
8 One subtle effect is that, if the fluid pressure in the cleat increases, after an immediate
9 opening of the cleat, one expects the cleat aperture (and hence the permeability) to decrease
10 for a transient time, as a consequence of the fluid diffusion from the cleat to the coal matrix.
11 In this work, we aim to model such transient variations of permeability by proposing
12 constitutive equations at the fractured coal scale. Permeability depends on the complete
13 history of pressures over time. The constitutive equations rely on Boltzmann's superposition
14 principle, which requires kernels as inputs. One can identify the kernels with finite-element
15 simulations of the response of an individual cleat subjected to a history of fluid pressure. We
16 also propose approximate versions of those kernels, which only depend on a few parameters
17 with a physical meaning. Examples of fluid injection simulations into a coal seam making use
18 of the constitutive equations here derived are presented.

19

20 1 Introduction

21 Unminable coal seams could contribute to the storage of CO₂ (i.e., carbon dioxide) up to
22 20 Gt (Gale, 2004). Given that anthropogenic emissions of CO₂ are estimated at 10 Gt per year
23 (Knorr, 2009), coal seams could contribute to store about two years of those emissions, which
24 is small but non-negligible. Unminable coal seams contain CH₄ (i.e., methane) naturally:
25 proven resources are 2 billion standard cubic feet, and contingent resources are estimated at
26 300 billion standard cubic feet (Moore, 2012). Natural gas presently contributes to about 21%
27 of the world's energy supply (International Energy Agency, 2019). Production of CH₄ can be
28 enhanced by injecting CO₂ into the seam –a process known as CO₂-enhanced coal bed
29 methane recovery (or CO₂-ECBM). During both injection of CO₂ (Oudinot et al., 2011; Pekot
30 and Reeves, 2002) and production of CH₄ (Palmer and Mansoori, 1998; Moore et al., 2011;
31 Scott et al., 2012), significant variations of permeability are observed. The reason beyond
32 those variations is well known: they are the consequence of adsorption-induced volume
33 variations of the coal (Gray, 1987).

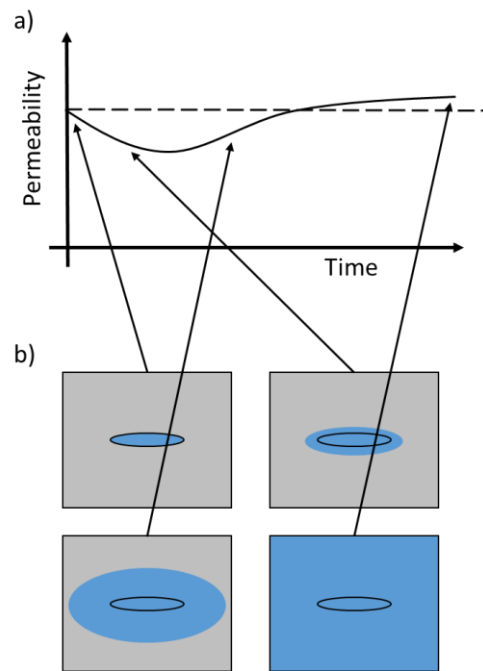
34 Naturally, coal is a fractured material (Laubach et al., 1998): those natural fractures are
35 called cleats, vary in size, are mostly vertical, and are spaced by about a centimeter. Cleats
36 govern the seam's permeability (Mazumder et al., 2006; Pan and Connell, 2007). Between
37 cleats, one finds the coal matrix (Harpalani and Schraufnagel, 1990), porous, with pores down
38 to a sub-nanometric size. The variations of permeability observed during production or
39 injection are due to the cleats' opening or closure, which is consecutive to shrinkage or
40 swelling of the coal matrix, respectively. Dimensional variations of the coal matrix observed
41 upon variations of the pressure of the fluid it contains are known to be due to adsorption

¹ Corresponding author

42 effects (Levine, 1996). In the smallest pores of the material, most of the fluid it contains is
43 indeed adsorbed, i.e., in intermolecular interaction with the solid skeleton's atoms. Because
44 of those intermolecular interactions, an unconstrained piece of coal matrix tends to swell
45 upon an increase of fluid pressure. The magnitude of those pressure-induced dimensional
46 variations depends on the fluid. Upon CO₂ injection, the injection induces increased CO₂
47 pressure in the bed and coal matrix swelling. In the confined conditions that prevail in the
48 underground, this swelling translates into cleats closure, and finally into permeability
49 decrease.

50 Subtle adsorption-induced variations of permeability can take place. Let us consider a
51 sample of fractured coal submitted to constant confining stresses, in which we inject fluid.
52 Because the cleat system's permeability is larger than that of the coal matrix, fluid will first
53 penetrate the cleats (such flow of fluid or transfer of mass through the cleats is also referred
54 to as seepage (Barenblatt et al., 1960)). It will decrease the effective stress, hence leading to
55 an aperture of the cleats and increasing the sample's permeability. Immediately after the
56 injection, fluid will start penetrating the coal matrix through Fickian diffusion (Moore, 2012).
57 At early times, the concentration of fluid in the coal matrix must be larger in the vicinity of the
58 cleat than far from it (see Figure 1-b). Even if the sample is under constant confining stresses,
59 the coal matrix's swelling will be localized around the cleat, which will tend to close the cleat.
60 At large times, the concentration of fluid in the coal matrix should be homogeneous. In this
61 case, if the sample is under constant confining stresses and the coal matrix is homogeneous,
62 swelling of the coal matrix should translate into a homothetic swelling of the whole sample
63 and hence of the cleats as well, leading to a long-term increase of permeability (see Figure 1-
64 a at large times). As a result, even if fluid pressure in the cleats is constant over time, one can
65 expect a non-monotonic and complex variation of permeability, as displayed in Figure 1-a. This
66 complex variation results from the transient diffusion of fluid through the coal matrix and the
67 induced heterogeneity of fluid pressure in the matrix. We will refer to those variations as
68 "transient variations of permeability."

69



70

71 *Figure 1: Schematics of (a) transient variations of permeability and (b) distribution of fluid in*
 72 *the coal. The black ellipse represents the cleat, and we display the injected fluid in blue. The*
 73 *dashed line on subfigure (a) indicates the initial permeability.*

74

75 The study of those transient variations of permeability has gained significant interest
 76 recently. With finite-element simulations of coal samples (Peng et al., 2014b; Qu et al., 2014;
 77 X. Liu et al., 2018) or through review of laboratory data (Liu et al., 2011a), several studies
 78 showed that the diffusion of fluid through the coal matrix could yield non-monotonic
 79 variations of permeability of significant magnitude. Assuming local thermodynamic
 80 equilibrium (i.e., assuming that, inside a representative elementary volume of fractured coal,
 81 the fluid's thermodynamic pressure is homogeneous, i.e., the same at any location in the coal
 82 matrix as in the cleats) can yield significant errors.

83 On laboratory samples submitted to constant confining stresses, several groups
 84 (Robertson, 2005; Pini et al., 2009; Wang et al., 2011) observed non-monotonic variations of
 85 permeability with the pore fluid's pressure—a phenomenon known as permeability rebound.
 86 Liu et al. (2011b), Peng et al. (2014a), and Qu et al. (2014) proposed to explain this
 87 permeability rebound by the fact that experimental data would have been acquired before
 88 reaching an equilibrium or a steady-state. Therefore, this permeability rebound would be a
 89 consequence of transient variations of permeability. Experimentally, Wei et al. (2019b)
 90 observed non-monotonic evolutions of permeability with time for a coal sample submitted to
 91 constant confining stresses and fluid pressures, which, according to them, suggests a
 92 transition from a swelling nearby the cleat to further away. Note, however, that other theories
 93 can explain the permeability rebound without invoking transient effects. For instance, by
 94 assuming that a fraction of the bulk coal swelling or of the matrix coal swelling translates into
 95 the closure of the cleats, models can capture permeability rebound. This ability holds
 96 independent of whether the fraction is considered constant (Liu and Rutqvist, 2010; Connell
 97 et al., 2010; Chen et al., 2012; Guo et al., 2014; Lu et al., 2016; Z. Liu et al., 2018) or pressure-

98 dependent (Peng et al., 2017). Heterogeneity of coal (Izadi et al., 2011; Chen et al., 2013), as
99 well as gas slippage (Niu et al., 2018) or a transition between different flow regimes (Wang et
100 al., 2019) could also explain complex variations of the permeability with pore pressure for a
101 piece of coal submitted to constant confining stresses.

102 Our aim in this work is to model transient variations of permeability in reservoir
103 simulations. As Peng et al. (2014b), Qu et al. (2014), and X. Liu et al. (2018) showed, those
104 transient variations of permeability can be modeled numerically as soon as the coal matrix
105 around the cleat is meshed and the diffusion of fluid through the coal matrix is explicitly
106 modeled. However, such an approach cannot be employed to perform reservoir simulations,
107 as the number of elements required to perform such simulation would be much too large.
108 Consequently, we aim at deriving a model at a scale such that the representative elementary
109 volume is a piece of fractured coal.

110 At our scale of interest, i.e., at the scale where a piece of fractured coal is our
111 representative elementary volume (seen as a smeared porous medium), many have modeled
112 adsorption-induced variations of permeability (for a review, see Pan and Connell (2012)). Most
113 models (e.g., Palmer and Mansoori (1998), Shi and Durucan (2004), Cui and Bustin (2005),
114 Vandamme et al. (2010), Pijaudier-Cabot et al. (2011), Wu et al. (2011), Brochard et al. (2012),
115 Espinoza et al. (2014)) assume local thermodynamic equilibrium. This assumption means that,
116 by nature, those models are unable to capture the non-monotonic variations of permeability
117 displayed in Figure 1-a, as those variations are the direct consequence of the heterogeneity of
118 the concentration of fluid in the coal matrix within the representative elementary volume. In
119 contrast, others aimed at relaxing this assumption of local thermodynamic equilibrium (Wu et
120 al., 2010; Liu et al., 2011b; Wu et al., 2011; Peng et al., 2014a; Vandamme et al., 2014; Zang
121 and Wang, 2017; Wei et al., 2019a).

122 Barenblatt et al. (1960) modeled fluid flow in fractured rocks by introducing variables
123 averaged over a scale much larger than the spacing between fractures. They considered two
124 pressures of fluid at each point in space, i.e., one in the fractures and one in the pore space in
125 the rock between the fractures. They considered the transfer of fluid between fractures and
126 rock porosity, but, by construction, in their model, the fluid pressure is homogeneous in the
127 whole rock porosity at each point in space. In the same spirit, but specifically for coal, Wu et
128 al. (2010, 2011) and Vandamme et al. (2014) introduced kinetics of transfer between cleats
129 and coal matrix. But they considered that the thermodynamic pressure of fluid in the coal
130 matrix is homogeneous: such simplification makes it impossible to capture the non-monotonic
131 variations of permeability displayed in Figure 1-a. To solve this issue, the model of Liu et al.
132 (2011b) and its extension to double-porosity media (Peng et al., 2014a) introduce the notion
133 of local and global swelling. In fact, in those models, in a representative elementary volume
134 of fractured coal, one pressure of the fluid in the cleats and one pressure of the fluid in the
135 coal matrix is defined, which can differ. But fluid in the cleats is considered to make the cleat's
136 vicinity swell so that the cleats aperture (and thus the permeability) decreases with increased
137 fluid pressure in the cleat. Upon fluid injection in an unconfined sample, both the fluid
138 pressure in the cleats and the coal matrix vary. After an immediate opening of the cleats, the
139 cleats' fluid pressure tends to close the cleats in the short term. In unconfined conditions, the
140 fluid pressure in the matrix tends to open the cleats in the long term. Those two kinetics
141 compete. Zang and Wang (2017) also relax the local thermodynamic equilibrium condition by
142 introducing a non-equilibrium swelling and defining the kinetics that governs this non-

143 equilibrium swelling related to fluid diffusion in the coal matrix. Like the model of Liu et al.
144 (2011b), this model can capture non-monotonic evolutions of permeability upon injection into
145 an unconfined piece of coal, depending on the kinetics of increase of fluid pressure in the
146 cleats and the coal matrix. Wei et al. (2019a) consider that the transient variation of the cleats'
147 aperture is related to the gradient of fluid pressure in the coal matrix. Therefore, they
148 postulate that it is associated with the rate of coal matrix swelling. By doing so, they end up
149 with a model able to capture non-monotonic transient variations of permeability.

150 In this work, we aim to propose constitutive equations that can model transient
151 permeability variations in reservoir simulations. Hence, we formulate them at a scale such
152 that the representative elementary volume is a small piece of fractured coal. Our approach is
153 original and alternative to the previous paragraph's models, as it relies on the Boltzmann
154 superposition principle. In such a framework, we can model the transient variations of
155 permeability predicted by simulations of fluid injection in an individual cleat with constitutive
156 equations formulated at a scale above (i.e., at a scale at which the representative elementary
157 volume contains both coal matrix and cleats). The fact that this upscaling is exact represents
158 the main interest of the model we propose.

159 Section 2 starts by performing finite-element simulations of fluid injection in a cleat
160 surrounded by the coal matrix. Those simulations, performed at a scale lower than the one at
161 which we aim at deriving our model, make it possible to understand the process at stake better
162 and serve as a reference case. This section also presents the principle of the modeling
163 proposed, which relies on the Boltzmann superposition principle. In section 3, we derive the
164 constitutive equations able to capture transient variations of permeability at the scale of a
165 representative elementary volume of fractured coal. We also implement those equations and
166 simulate fluid injection into a reservoir by using the results of the finite-element simulations
167 performed in section 2 as input. In section 4, we propose an engineering version of our model,
168 with which we can model transient variations of permeability by introducing just a few
169 parameters with a physical meaning.

170 2 Transient variations of permeability of an individual cleat whose permeability is governed 171 by its aperture

172 This section considers a system made of a single cleat surrounded by an isotropic coal
173 matrix. The cleat is cylindrical and infinitely long. The history of pressure $p_c(t)$ of the fluid in
174 the cleats is imposed and is homogeneous in the cleat. Consequently, the problem is 2-
175 dimensional, and we focus on what happens in a slice perpendicular to the direction of the
176 cleat (i.e., in the plane visible in Figure 1-b). We aim to simulate numerically (see section 2.2)
177 and then model (see section 2.3) how this system reacts to the history of pressure, particularly
178 in terms of permeability and transfer of fluid from the cleat to the coal matrix.

179 We give the various geometries of cleats considered in this study in Figure 2. All cleats
180 are embedded in a square with edges with a length equal to 1 cm. We consider one cleat with
181 a rectangular cross-section (called 'rectangular cleat') of dimensions 0.05 cm by 0.5 cm. We
182 also consider cleats with an elliptical cross-section (called 'elliptical cleats') of a long axis $A =$
183 0.25 cm and of short axis a . Various ratios a/A are considered: 0.03, 0.10, 0.30, 1.00. The cleat
184 with aspect ratio $a/A = 1$ corresponds to a cleat with a circular cross-section.

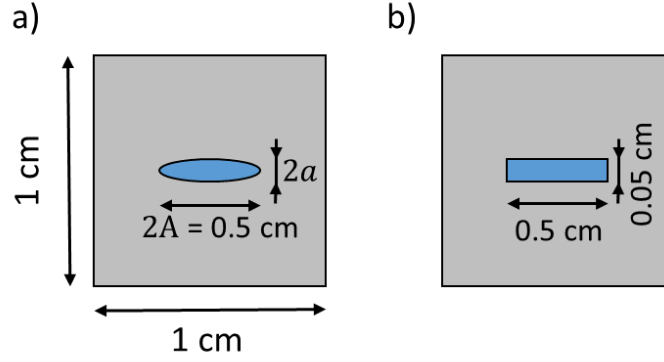


Figure 2: Various geometries of the cross-section of cleats considered in this study: cleats with (a) an elliptical cross-section and (b) a rectangular cross-section.

The intrinsic permeability $\kappa(t)$ of the cleat is assumed to be governed by its aperture and to follow the celebrated Kozeny-Carman relationship (Coussy, 2010):

$$\kappa \propto \frac{(\phi_c)^3}{(1-\phi_c)^2}, \quad (1)$$

where ϕ_c is the porosity associated with the cleat system and is defined as:

$$\phi_c = \frac{V_c}{V_0} \quad (2)$$

where V_c is the actual volume of cleats in the volume of fractured coal of interest. In the reference configuration, the volume of fractured coal of interest is V_0 . Under the assumption that the porosity is much smaller than 1, Eq. (1) reduces to:

$$\kappa = \kappa_0 \left(\frac{V_c}{V_{c,0}} \right)^3 = \kappa_0 \left(1 + \frac{\Delta V_c}{V_{c,0}} \right)^3 \quad (3)$$

where κ_0 is the permeability in the reference configuration, $V_{c,0}$ is the volume of the cleats in the reference configuration, and ΔV_c is the variation of the volume of the cleats.

2.1 Equations governing the problem

We consider that the cleat is subjected to a fluid pressure $p_c(t)$. At a given time t , the thermodynamic pressure $p_m(y, z, t)$ of the fluid in the coal matrix (where y and z are coordinates in the plane of interest) is, in the generic case, heterogeneous.

The Langmuir adsorption isotherm relates the mass concentration $c_m(y, z, t)$ of fluid in the coal matrix (per unit volume of coal matrix in the reference configuration) to the thermodynamic pressure $p_m(y, z, t)$ of the fluid in the coal matrix through:

$$c_m(y, z, t) = c_{m,max} \frac{p_m(y, z, t)/p_{L0}}{1 + p_m(y, z, t)/p_{L0}} \quad (4)$$

where $c_{m,max}$ and p_{L0} are Langmuir parameters.

The pressure $p_c(t)$ of fluid in the cleats imposes the thermodynamic pressure $p_m(y, z, t)$ of fluid on the edge of the cleat and hence the concentration $c_{edge}(t)$ of fluid in the coal matrix on the edge of the cleat, through:

$$c_{edge}(p_c(t)) = c_{m,max} \frac{p_c(t)/p_{L0}}{1 + p_c(t)/p_{L0}} \quad (5)$$

208 The transport of fluid in the coal matrix is assumed to be diffusive and to follow Fick's
209 law (Moore, 2012) so that the mass flow vector \underline{w}_m of fluid through the coal matrix verifies:

$$\underline{w}_m = -D \nabla c_m \quad (6)$$

210 where D is the diffusivity of the fluid through the coal matrix. For engineering applications, it
211 is common to consider that the diffusivity D is constant (which we will assume here), while in
212 practice, it may not be (Zhao et al., 2019). The mass concentration of fluid then verifies:

$$\frac{\partial c_m}{\partial t} = D \Delta c_m \quad (7)$$

213 The coal matrix is assumed to behave in a linear elastic manner. We note its stiffness
214 tensor \underline{C}_m . Its bulk modulus is K_m and its Poisson's ratio ν_m . Adsorption roughly makes a piece
215 of coal swell proportionally to the concentration c_m of fluid in the coal matrix, so that the
216 constitutive equation of the coal matrix is:

$$\underline{\underline{\sigma}} = \underline{C}_m : \left(\underline{\underline{\varepsilon}} - \varepsilon^a(c_m) \underline{\underline{1}} \right) \text{ with } \varepsilon^a(c_m) = \alpha c_m \quad (8)$$

217 where ε^a is an adsorption strain and α a parameter governing the magnitude of adsorption-
218 induced swelling. Likewise, we can express this equation as:

$$\underline{\underline{\sigma}} = \underline{C}_m : \underline{\underline{\varepsilon}} + \sigma^a(c_m) \underline{\underline{1}} \text{ with } \sigma^a(c_m) = -3K_m \varepsilon^a(c_m) = -3K_m \alpha c_m \quad (9)$$

219 where we call $\sigma^a(c_m)$ adsorption stress.

220 2.2 Numerical solution of response to a step loading

221 We perform two-dimensional simulations with Abaqus. We use plane-strain conditions
222 and consider no confining stress, i.e., $\sigma_0 = 0$. Note that, by considering non-zero constant
223 confining stress, we would have calculated identical variations of aperture. We give properties
224 and conditions in Table 1. We obtained material properties in this table from the literature.
225 The maximum adsorbed amount $c_{m,max}$ expressed in kg.m^{-3} is calculated from the maximum
226 adsorbed amount of 2.4 mol.L^{-1} in Espinoza et al. (2014) as: $c_{m,max} = (2.4 \text{ mol.L}^{-1}) * M_{\text{CO}_2}$
227 where $M_{\text{CO}_2} = 44 \text{ g/mol}^{-1}$ is the molar mass of CO_2 . The parameter α expressed in $\text{m}^3.\text{kg}^{-1}$ is
228 calculated from the swelling coefficient $4 * 10^{-3} \text{ L.mol}^{-1}$ in Espinoza et al. (2014) as $\alpha = 4 * 10^{-3} \text{ L.mol}^{-1} / M_{\text{CO}_2}$.
229

230

231 Table 1: Input parameters for computation

Parameter	Value	Reference
Diffusivity D of fluid in coal matrix ($\text{m}^2 \cdot \text{s}^{-1}$)	$1.0 \cdot 10^{-9}$	Order of magnitude from Nazarova et al. (2014)
Bulk modulus K_m of coal matrix (GPa)	5.0	(Espinoza et al., 2014)
Poisson's ratio ν_m of coal matrix (1)	0.2	Order of magnitude from Espinoza et al. (2014)
Maximum adsorbed amount $c_{m,max}$ in Langmuir isotherm ($\text{kg} \cdot \text{m}^{-3}$)	105.6	Calculated from Espinoza et al. (2014)
Characteristic pressure p_{L0} in Langmuir isotherm (MPa)	1.6	Espinoza et al. (2014)
Parameter $\alpha = \Delta \varepsilon^a / \Delta c_m$ governing the magnitude of adsorption-induced swelling ($\text{m}^3 \cdot \text{kg}^{-1}$)	$9.1 \cdot 10^{-5}$	Calculated from Espinoza et al. (2014)
Temperature T ($^{\circ}\text{C}$)	40	
Confining stress σ_0 (MPa)	0	

232

233 We apply a step loading of fluid pressure in the cleat:

$$p_c(t) = \begin{cases} 0 & \text{if } t \leq 0 \\ p_{c,0} & \text{if } t > 0 \end{cases} \quad (10)$$

234 Consequently, the hydric boundary conditions on the edge $\partial\Omega_{in}$ of the cleat are:

$$c_m(y, z, t) = c_{edge}(t) = \begin{cases} 0 & \text{if } t \leq 0 \\ c_{m,max} \frac{p_{c,0}/p_{L0}}{1 + p_{c,0}/p_{L0}} & \text{if } t > 0 \end{cases} \quad (11)$$

235 On all boundaries other than the edge of the cleat, we impose no flux.

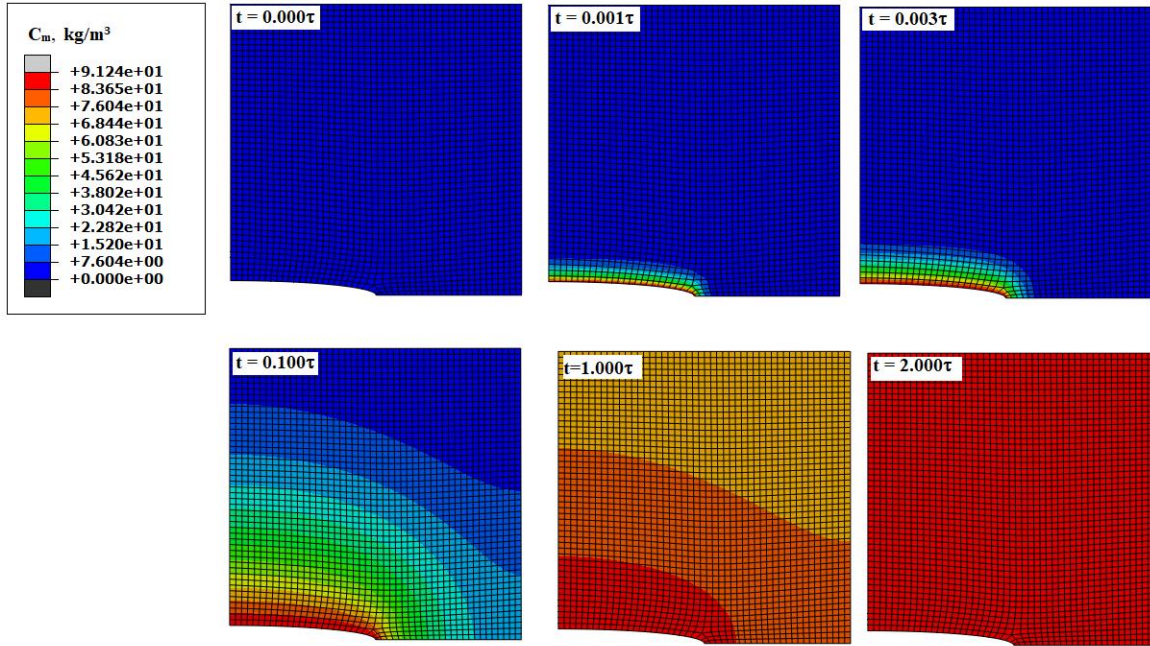
236 The mechanical boundary conditions on the edge $\partial\Omega_{in}$ of the cleat are:

$$\underline{\underline{\sigma}} \cdot \underline{\underline{n}} = \begin{cases} 0 & \text{if } t \leq 0 \\ -p_c \underline{\underline{n}} & \text{if } t > 0 \end{cases} \quad (12)$$

237 As stated in Table 1, on the outer boundaries, we impose no stress. On the boundaries that
238 correspond to planes of symmetry of the modeled system, we impose mechanical boundary
239 conditions that are consistent with the symmetry, namely free sliding parallel to the boundary
240 and no displacement perpendicular to the boundary.

241 We give an example of the distribution of concentration around a cleat with an elliptical
242 cross-section in Figure 3.

243



244

245 *Figure 3: Distribution of fluid concentration c_m in the coal matrix at various times around a*
 246 *cleat with an elliptical cross-section and an aspect ratio $a/A = 0.03$.*

247 The variation ΔV_c of the cleat's volume is calculated from the displacements $\underline{\xi}$ of the
 248 edges of the cleat according to:

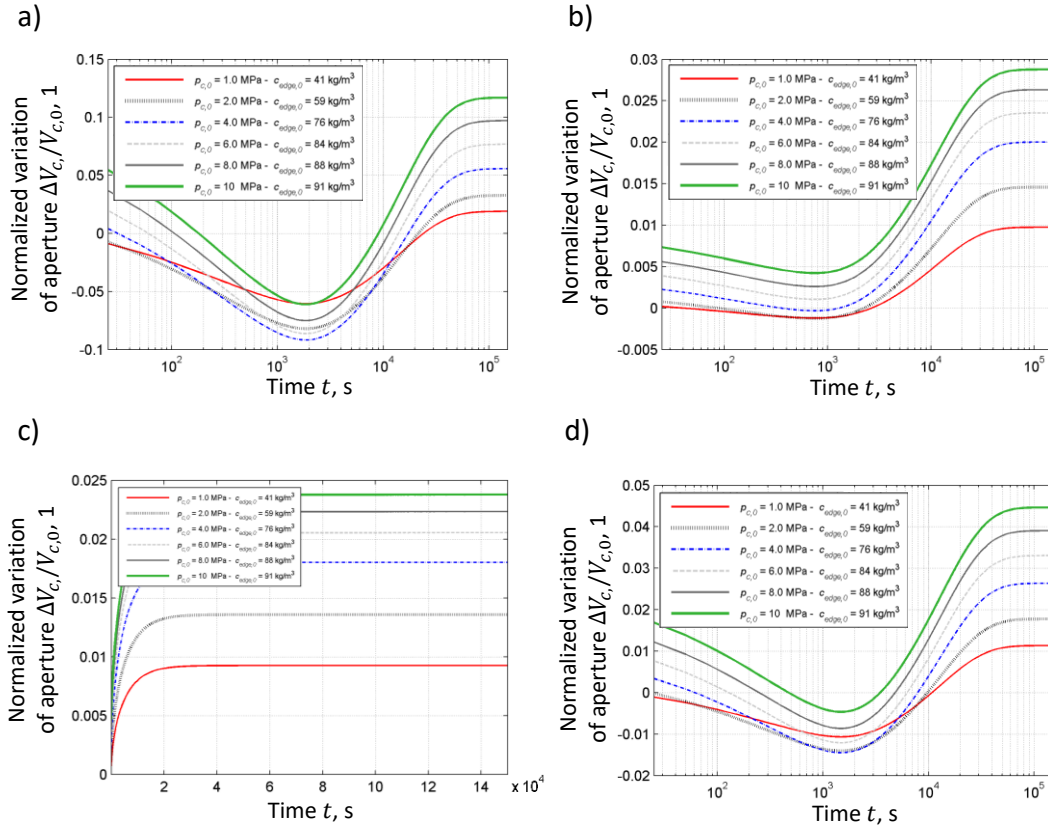
$$\Delta V_c = - \int_{\partial\Omega_{in}} \underline{\xi} \cdot \underline{n} dS \quad (13)$$

249 where \underline{n} is the vector normal to the inner surface $\partial\Omega_{in}$ of the region occupied by the coal
 250 matrix. For some of the geometries considered, normalized variations $\Delta V_c/V_{c,0}$ of the aperture
 251 of the cleat are displayed in Figure 4. From those variations of aperture, variations of
 252 permeability can be readily obtained with the Kozeny-Carman equation (3), as displayed in
 253 Figure 5.

254 On those figures, one observes that, for cleats with a circular cross-section, permeability
 255 increases monotonically with time. For all other cleats with a cross-section with an aspect ratio
 256 different from 1.0, permeability varies in a nonmonotonic manner, first decreasing at early
 257 times before increasing at larger times. In all cases, permeability at equilibrium is larger than
 258 permeability at early times since, in unconfined conditions, the coal matrix's swelling
 259 translates into a homothetic swelling of the sample and hence of the cleat as well.

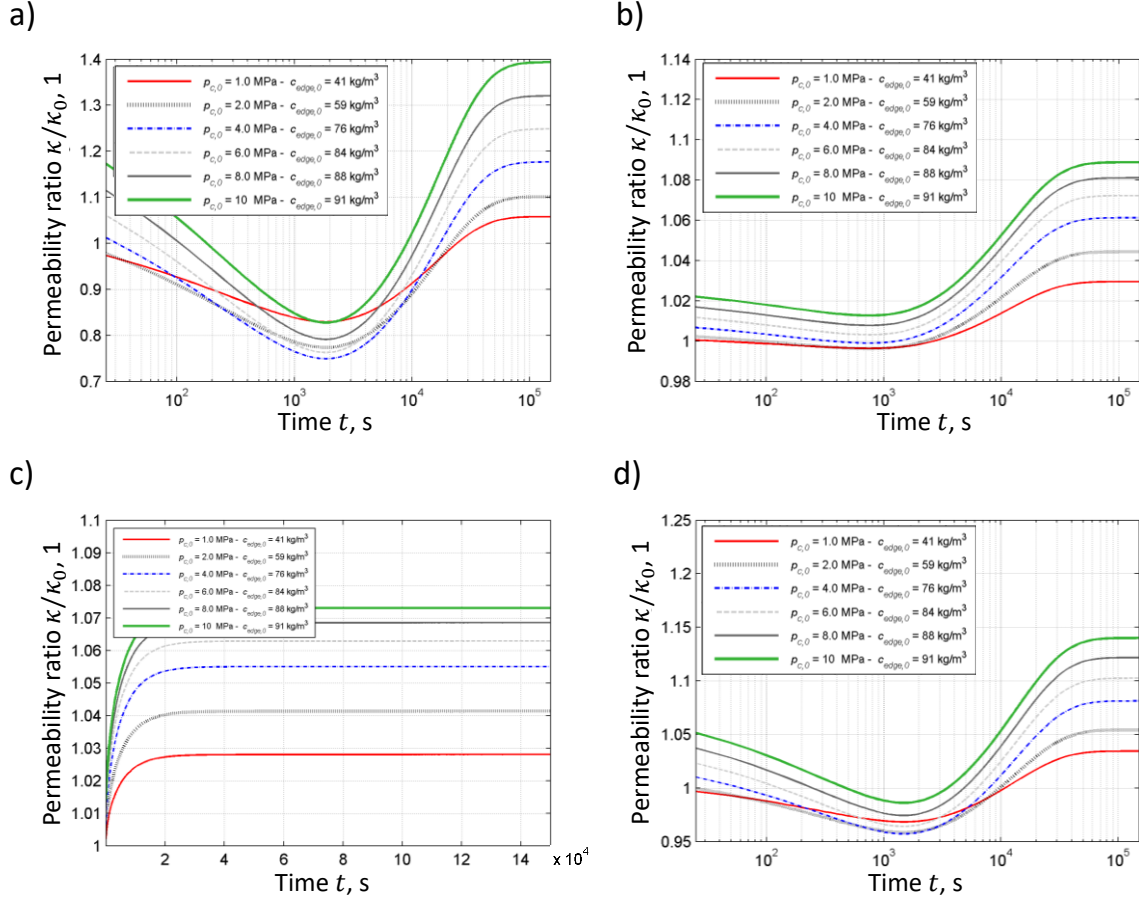
260 For a given aspect ratio, the geometry of the cross-section (namely elliptical (see Figure
 261 4-b and Figure 5-b) or rectangular (see Figure 4-d and Figure 5-d)) impacts the evolutions of
 262 permeability over time, but not very significantly. However, we note that, for given fluid
 263 pressure, the immediate increase of permeability is larger for the cleat with a rectangular
 264 cross-section (see Figure 4-d) than with an elliptical one (see Figure 4-b). For cleats with an
 265 elliptical cross-section, when the aspect ratio decreases (i.e., when the cleat is flatter), both
 266 the transient reduction of permeability and the immediate increase of permeability (i.e., the
 267 poroelastic effect due to the mechanical pressure of the fluid on the pore walls) are more
 268 pronounced. In contrast, the characteristic time to reach equilibrium seems roughly

269 independent of the cleat's cross-section geometry. The time at which permeability is the
 270 lowest is equal to about 1500s for the cleat with a rectangular cross-section whose aspect
 271 ratio is 0.3 and for cleats with an elliptical cross-section whose aspect ratio is 0.1 or 0.03. This
 272 time is equal to about 750s for the cleat with an elliptical cross-section whose aspect ratio is
 273 0.3. For sufficiently flat cleats, the time at which permeability is the lowest is roughly
 274 independent of the cleat's cross-section geometry. We will discuss those trends further in a
 275 quantitative analysis of the time-dependent response performed in Section 4.2.



276
 277 *Figure 4: Evolution over time of cleat aperture for cleats with various geometries subjected to*
 278 *various fluid pressures: (a) elliptical cross-section with aspect ratio $a/A = 0.03$, (b) elliptical*
 279 *cross-section with aspect ratio $a/A = 0.3$, (c) elliptical cross-section with aspect ratio*
 280 *$a/A = 1.0$, (d) rectangular cross-section with aspect ratio $a/A = 0.3$.*

281



282

283 *Figure 5: Evolution over time of permeability for cleats with various geometries subjected to*
 284 *various fluid pressures: (a) elliptical cross-section with aspect ratio $a/A = 0.03$, (b) elliptical*
 285 *cross-section with aspect ratio $a/A = 0.3$, (c) elliptical cross-section with aspect ratio*
 286 *$a/A = 1.0$, (d) rectangular cross-section with aspect ratio $a/A = 0.3$.*

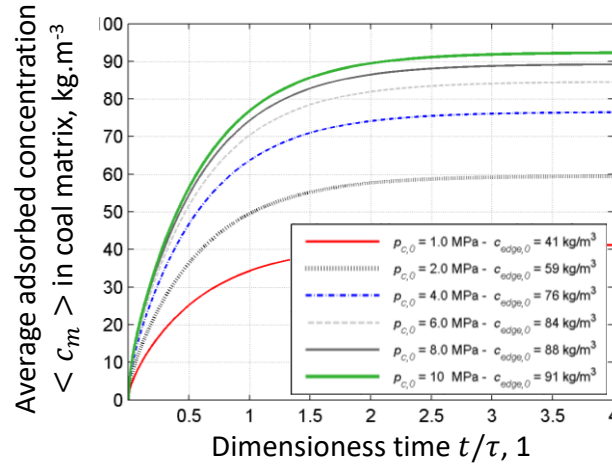
287 From the simulations performed, the flow rate dm_m/dt of fluid from the cleats to the
 288 coal matrix (expressed per unit volume V_0 of fractured coal in the reference configuration) can
 289 be calculated by spatial integration of the flux on the edges of the cleat:

$$\frac{dm_m}{dt} = -\frac{1}{V_0} \int_{\partial\Omega_{in}} \underline{w}_m \cdot \underline{n} dS \quad (14)$$

290 The amount m_m of fluid in the coal matrix (still per unit volume V_0 of fractured coal in the
 291 reference configuration) can be calculated by integration over time of Eq. (14).

292 The average mass $\langle c_m \rangle$ of fluid in the coal matrix per unit volume of coal matrix in
 293 the reference configuration is displayed in Figure 6 for the example of the elliptical cleat with
 294 an aspect ratio $a/A = 0.03$. In this figure, time is displayed in a dimensionless manner by
 295 dividing it by a characteristic time $\tau = 25,000$ s, which corresponds to $\tau = L_{diff}^2/D$, where
 296 $D = 1 \times 10^{-9}$ m²/s is the diffusion coefficient of the coal matrix and $L_{diff} = 0.5$ cm is a
 297 drainage length equal to half of the characteristic spacing between cleats. The value toward
 298 which $\langle c_m(t) \rangle$ should converge at large times is known: it should tend toward $c_{edge,0}$, since
 299 the concentration of fluid in the coal matrix per unit of coal matrix should be homogeneous
 300 and equal to its value $c_{edge,0}$ on the edge of the cleat. We noticed however a slight

301 discrepancy, as $\langle c_m(t) \rangle$ tended toward values that slightly differed from the expected one.
 302 We attributed this discrepancy to numerical errors. In the rest of the manuscript, we rescaled
 303 all values of $c_m(t)$ (and consequently of $m_m(t) = (1 - \phi_{c,0})c_m(t)$ and of \dot{m}_m) based on the
 304 calculated asymptotic value of $c_m(t)$, to ensure that the rescaled $c_m(t)$ properly converged
 305 toward the expected value $c_{edge,0}$. Hence, the rescaled $m_m(t)$ also properly converges toward
 306 its expected value, namely $(1 - \phi_{c,0})c_{edge,0}$.



307

308 *Figure 6: Evolution of average mass $\langle c_m(t) \rangle$ of fluid in coal matrix per unit volume of coal*
 309 *matrix, for the elliptical cleat with an aspect ratio $a/A = 0.03$. The characteristic time τ used*
 310 *to make the time dimensionless is $\tau = 25,000$ s.*

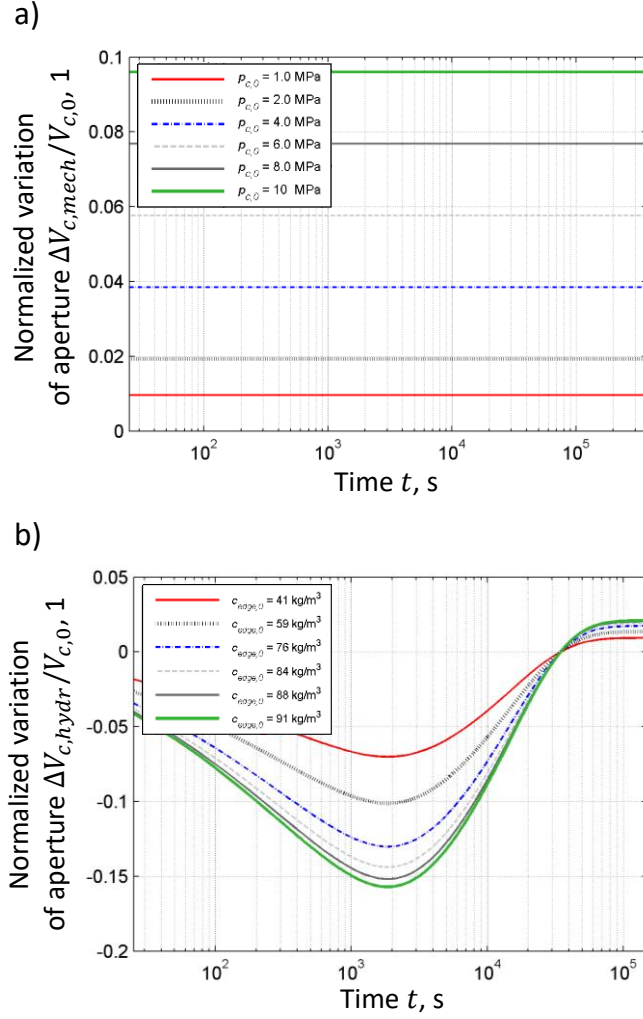
311 2.3 Principle of the solution to a generic loading: analogy with linear viscoelasticity

312 This section is devoted to explaining how, based on the cleat's response to a step loading
 313 of fluid pressure simulated in section 2.2, we can calculate the cleat's response to a generic
 314 fluid pressure evolution. The approach is based on the Boltzmann superposition principle.

315 Part of the variation ΔV_c of the cleat's volume is due to the deformation induced by the
 316 fluid's mechanical pressure on the cleat's walls and by the confining stress potentially applied
 317 to the system, while another part of the variation is due to the diffusion of fluid in the coal
 318 matrix and to the swelling it induces, i.e.:

$$\Delta V_c = \Delta V_{c,mech} + \Delta V_{c,hydr} \quad (15)$$

319 where $\Delta V_{c,mech}$ is the contribution due to the mechanical loading (called 'mechanical
 320 contribution') and $\Delta V_{c,hydr}$ is the contribution due to the diffusion of fluid in the coal matrix
 321 (called 'hydraulic contribution'). For the case of the elliptical cleat with an aspect ratio $a/A =$
 322 0.03 , we display the two contributions in Figure 7.



323

324 *Figure 7: For an elliptical cleat with aspect ratio $a/A = 0.03$, mechanical contribution*
 325 *$\Delta V_{c,mech}$ and hydraulic contribution $\Delta V_{c,hydr}$ to the variation ΔV_c of the volume of the cleat.*

326 The mechanical contribution $\Delta V_{c,mech}$ to the variation ΔV_c of the cleat's volume depends
 327 linearly on the pressure $p_c(t)$ of the fluid in the cleats and on the confining stress $\sigma_0(t)$:

$$\frac{\Delta V_{c,mech}(t)}{V_{c,0}} = \frac{p_c(t)}{K_{c,p}} + \frac{\sigma_0(t)}{K_{c,\sigma}} \quad (16)$$

328 where $K_{c,p}$ and $K_{c,\sigma}$ characterize the stiffness of the cleat. Since we performed the calculations
 329 in section 2.2 in the absence of any confining stress (i.e., $\sigma_0 = 0$), we restrict ourselves to the
 330 case where the variation $\Delta V_{c,mech}/V_{c,0}$ is proportional to the fluid pressure $p_c(t)$:

$$\frac{\Delta V_{c,mech}(t)}{V_{c,0}} = \frac{p_c(t)}{K_{c,p}} \quad (17)$$

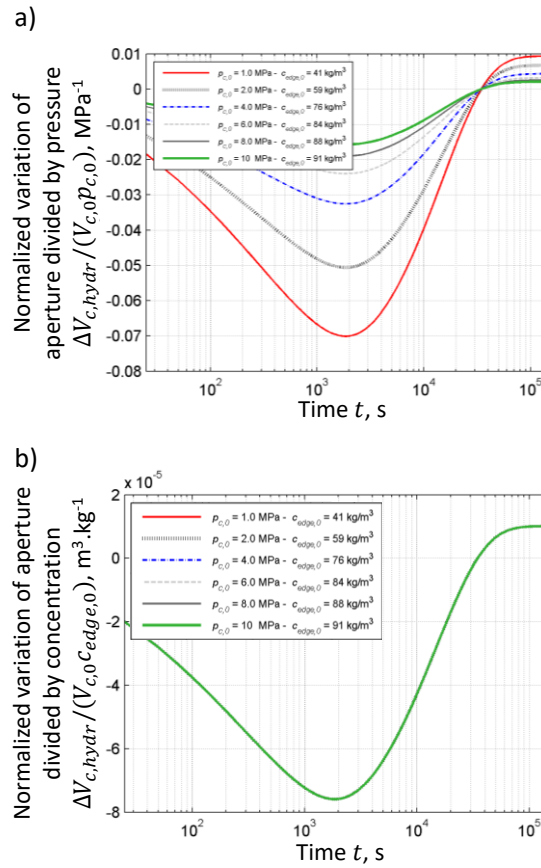
331 In contrast, even in the case of a step of fluid pressure (i.e., $p_c(t) = p_{c,0}$), the hydraulic
 332 contribution $\Delta V_{c,hydr}(t)$ at any time t is not proportional to the pressure $p_{c,0}$ of fluid in the
 333 cleats, as can be observed in Figure 8-a in the case of the elliptical cleat with aspect ratio
 334 $a/A = 0.03$. But transport of fluid is a linear process (see Eq. (7)), and swelling depends
 335 linearly on the concentration $c_{edge,0}$ on the edge of the cleat as well (see Eqs. (8) and (9)), so
 336 that the hydraulic contribution $\Delta V_{c,hydr}(t)$ at any time t is proportional to this concentration,

337 as can be observed in Figure 8-b. If we consider this concentration $c_{edge,0}$ on the edge of the
 338 cleat as the variable with which to formulate the problem, we can hence consider coal as a
 339 Boltzmann-type material, for which the superposition of the action implies the superposition
 340 of the responses, and for which one can express the evolution of the aperture of the cleat as
 341 follows:

$$\frac{\Delta V_{c,hydr}}{V_{c,0}} = \begin{cases} J_{\delta_c}(t) \cdot c_{edge,0} & \text{if } t > 0 \\ 0 & \text{if } t \leq 0 \end{cases} \quad (18)$$

342 Here we call $J_{\delta_c}(t)$ the kernel of the variation of aperture due to the ingress of fluid in the coal
 343 matrix. It is the response to a unit step of concentration $c_{edge,0} = 1 \text{ kg/m}^3$.

344



345

346 *Figure 8: For elliptical cleat with aspect ratio $a/A = 0.03$, contribution of the fluid in the*
 347 *matrix to the variation of aperture (a) normalized by the pressure of the fluid in the cleat or*
 348 *(b) normalized by the concentration on the edge of the cleat.*

349 Let us now consider a generic injection of fluid following a history of pressure $p_c(t)$ in
 350 the cleat. This injection causes a history of concentration $c_{edge}(t)$ on the edge of the cleat
 351 (see Eq. (5)). We consider that this function $c_{edge}(t)$ is piecewise continuous and
 352 differentiable. We note τ_i the instances where c_{edge} is discontinuous and $[c_{edge}]_i$ the
 353 corresponding jumps of the concentration on the edge of the cleat. Therefore, the
 354 concentration function $c_{edge}(t)$ can be rewritten as:

$$c_{edge}(t) = \int_{-\infty}^t Y(t-\tau)dc_{edge} + \sum_{\tau_i < t} [c_{edge}]_i Y(t-\tau_i) \quad (19)$$

355 where $Y(t)$ is the Heaviside function. The response $\Delta V_{c,hydr}(t)$ of the cleat aperture to the
 356 variations of concentration is derived directly from the application of Boltzmann superposition
 357 principle to Eq. (19):

$$\frac{\Delta V_{c,hydr}}{V_{c,0}} = \int_{-\infty}^t J_{\delta_c}(t-\tau)dc_{edge} + \sum_{\tau_i < t} [c_{edge}]_i J_{\delta_c}(t-\tau_i) \quad (20)$$

358 We can write Eq. (20) by using Stieltjes integrals as follows:

$$\frac{\Delta V_{c,hydr}}{V_{c,0}} = J_{\delta_c}(t) \otimes c_{edge}(t) = \int_{\tau=-\infty}^t J_{\delta_c}(t-\tau) \frac{\partial c_{edge}}{\partial \tau}(\tau) d\tau \quad (21)$$

359 with:

$$\frac{\partial c_{edge}}{\partial t} = \left\{ \frac{\partial c_{edge}}{\partial t} \right\} + \sum_{\tau_i < t} [c_{edge}]_i \delta(t-\tau_i) \quad (22)$$

360 where $\delta(t)$ is the Dirac function, $\{ \}$ is the derivative of the continuous part of the
 361 concentration $c_{edge}(t)$, and where \otimes denotes the Stieltjes convolution product.

362 Combining Eqs. (3), (15), (16), and (21), we can finally express the variation of
 363 permeability for a generic loading:

$$\begin{aligned} \kappa(t) &= \kappa_0 \left(1 + \frac{\Delta V_{c,mech}}{V_{c_0}} + \frac{\Delta V_{c,hydr}}{V_{c_0}} \right)^3 \\ &= \kappa_0 \left(1 + \frac{p_c(t)}{K_{c,p}} + \frac{\sigma_0(t)}{K_{c,\sigma}} + J_{\delta_c}(t) \otimes c_{edge}(t) \right)^3 \end{aligned} \quad (23)$$

364 where $c_{edge}(t) = c_{edge}(p_c(t))$, as given by Eq. (5). Note that, although the variations of
 365 volume ΔV_c satisfy Boltzmann superposition principle when expressed versus the amount of
 366 adsorbed fluid on the edge of the cleat, the variations of permeability do not. Likewise, when
 367 expressed versus the pressure p_c of the fluid in the cleats, the variations of volume ΔV_c do not
 368 satisfy the Boltzmann superposition principle.

369 Note that the amount $m_m(t)$ of fluid in the coal matrix (per unit volume V_0 of fractured
 370 coal in the reference configuration), as well as the flux $\dot{m}_m(t)$ of fluid from the cleats to the
 371 coal matrix (still per unit volume V_0 of fractured coal in the reference configuration), also
 372 satisfy the Boltzmann superposition principle, so that one can write:

$$m_m(t) = J_{m_m}(t) \otimes c_{edge}(t) = \int_{\tau=-\infty}^t J_{m_m}(t-\tau) \frac{\partial c_{edge}}{\partial \tau}(\tau) d\tau \quad (24)$$

373 and:

$$\dot{m}_m(t) = J_{\dot{m}_m}(t) \otimes c_{edge}(t) = \int_{\tau=-\infty}^t J_{\dot{m}_m}(t-\tau) \frac{\partial c_{edge}}{\partial \tau}(\tau) d\tau \quad (25)$$

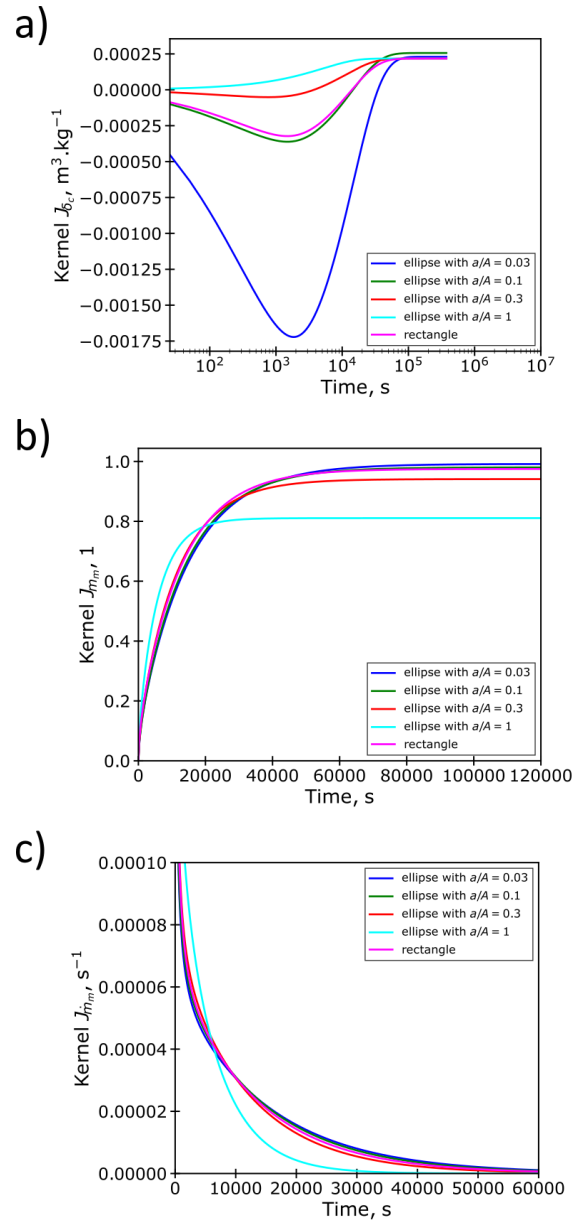
374 where $J_{m_m}(t)$ and $J_{\dot{m}_m}(t)$ are two kernels which verify:

$$J_{\dot{m}_m}(t) = dJ_{m_m}/dt \quad (26)$$

375 The three kernels $J_{\delta_c}(t)$, $J_{m_m}(t)$, and $J_{\dot{m}_m}(t)$ are displayed in Figure 9 for the various
 376 geometries of cleats. For what concerns the variation of aperture $\Delta V_{c,mech}(t)/V_{c,0}$ due to the

377 mechanical pressure of the fluid in the cleat (see Eq. (16)), the stiffness $K_{c,p}$ can be directly
 378 calculated from the numerical simulations of individual cleats performed in section 2.2. This
 379 stiffness characterizes how much the cleat's volume increases instantaneously when the fluid
 380 pressure in the cleat increases. From those simulations, we find that the stiffness $K_{c,p}$ is equal
 381 to 104.2 MPa, 381.1 MPa, 1.121 GPa, 2.558 GPa, for the cleats with an elliptical cross-section
 382 with aspect ratio $a/A = 0.03, 0.1, 0.3, 1$, respectively, and to 161.3 MPa for the cleat with
 383 rectangular cross-section. We observe that the shape of the cleat seems to have little effect
 384 on the time at which the aperture of the cleat is minimal, but has a significant effect on the
 385 magnitude of the transient variation of this aperture: the smaller the aspect ratio a/A , the
 386 more pronounced those variations are.

387



388

389 *Figure 9: Kernels (a) $J_{\delta_c}(t)$, (b) $J_{m_m}(t)$, and (c) $J_{\dot{m}_m}(t)$ for all geometries of cleats, which*
 390 *define the response of the cleat in terms of relative volume variation $\Delta V_c(t)/V_0$, average*
 391 *amount $m_m(t)$ of fluid in the coal matrix per unit volume of fractured coal, and rate $\dot{m}_m(t)$*

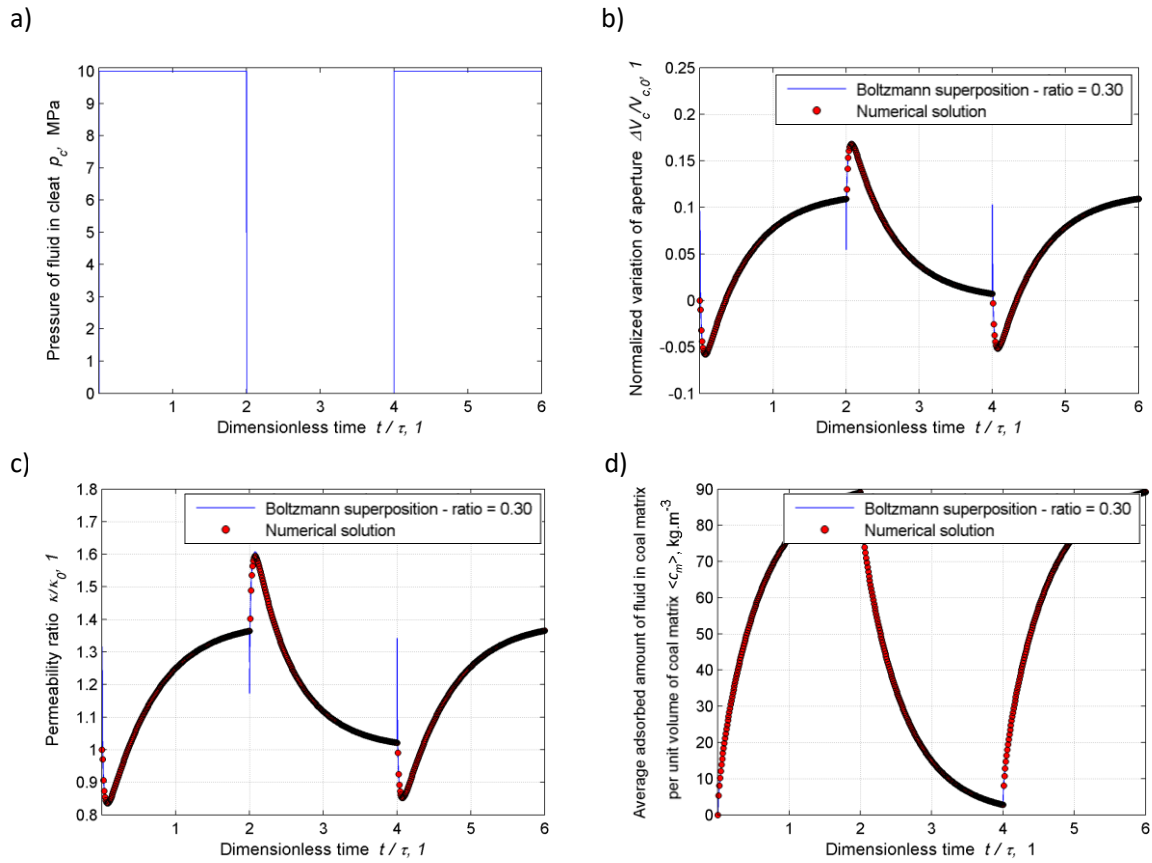
392 *of variation of this same amount, respectively. The characteristic time τ used to make the*
393 *time dimensionless is $\tau = 25,000$ s.*

394

395 2.4 A numerical verification of response of individual cleat to a generic loading

396 This section is dedicated to validating the approach proposed in section 2.3, based on
397 the Boltzmann superposition principle, to calculate the response of an individual cleat to a
398 generic history $p_c(t)$ of the pressure of fluid in the cleat. We perform the validation on the
399 elliptical cleat with an aspect ratio $a/A = 0.03$ submitted to no confining stress (i.e., $\sigma_0 = 0$).
400 The cleat is submitted to various histories $p_c(t)$ of the pressure of fluid. We calculate the
401 cleat's response in two manners: 1) plane-strain finite-element calculations identical to the
402 ones performed in section 2.2 but for the history $p_c(t)$ of the pressure of fluid in the cleat,
403 and 2) calculations based on the method proposed in section 2.3 that uses the Boltzmann
404 superposition principle. From both approaches, we obtain: the relative volume variation
405 $\Delta V_c(t)/V_0$ of the cleat, the permeability $\kappa(t)$ over time (calculated from the relative volume
406 variation $\Delta V_c(t)/V_0$ with Kozeny-Carman equation (3)), and the average amount $\langle c_m(t) \rangle$
407 of fluid in the coal matrix per unit volume of coal matrix (with $\langle c_m(t) \rangle = m_m(t)/(1 -$
408 $\phi_{c,0})$, where $m_m(t)$ is the average amount of fluid in the coal matrix per unit volume of
409 fractured coal). When aiming at using the Boltzmann superposition principle (section 2.3), the
410 relative volume variation $\Delta V_c(t)/V_0$ and the average amount $m_m(t)$ of fluid in the coal matrix
411 (per unit volume of fractured coal) are calculated with Eq. (15) (which uses Eqs. (16) and (21))
412 and Eq. (24), respectively, by using the kernels displayed in Figure 9.

413 We display the results of the comparison in Figure 10, Figure 11, and Figure 12, for what
414 concerns the relative volume variation $\Delta V_c(t)/V_{c,0}$ of the cleat, the permeability $\kappa(t)$, and the
415 average amount $\langle c_m(t) \rangle$ of fluid in the coal matrix per unit volume of coal matrix,
416 respectively. All calculations performed with Boltzmann superposition principle are in perfect
417 agreement with the finite-element calculations, which validates the applicability of this former
418 method introduced in section 2.3.



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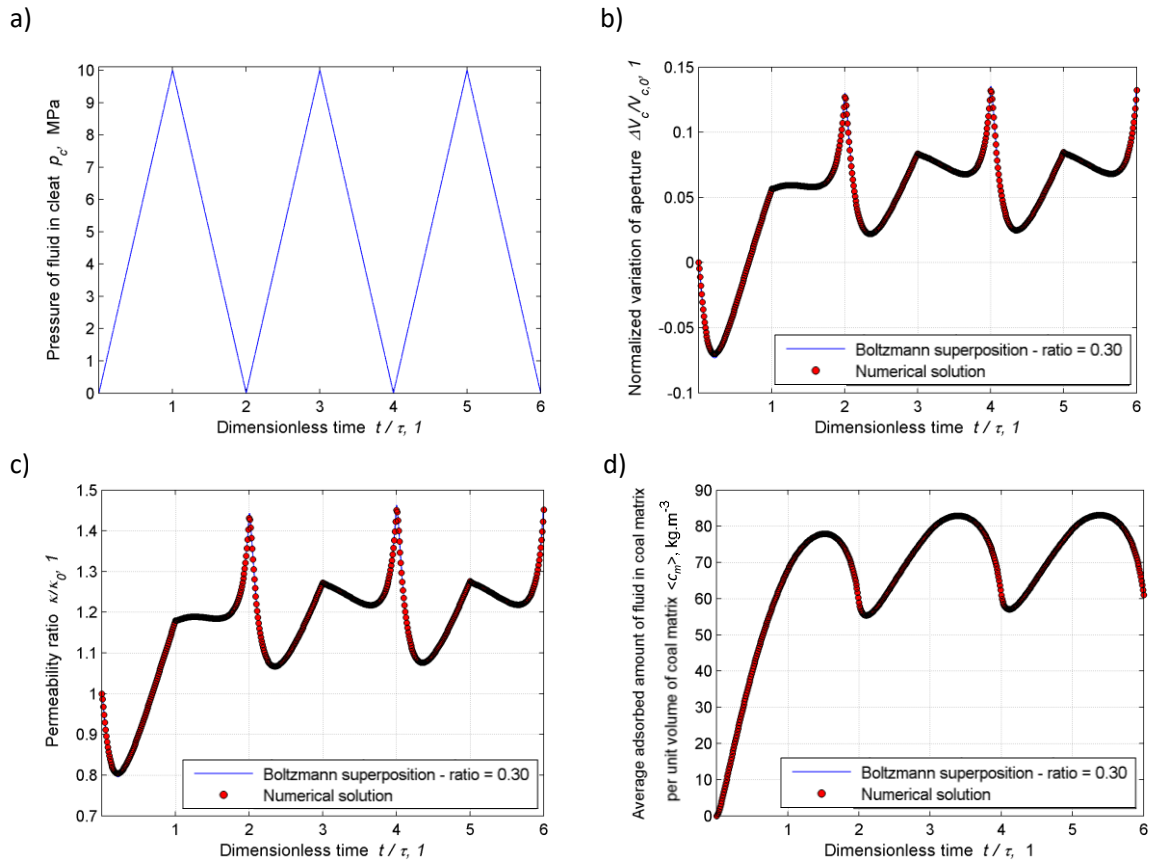
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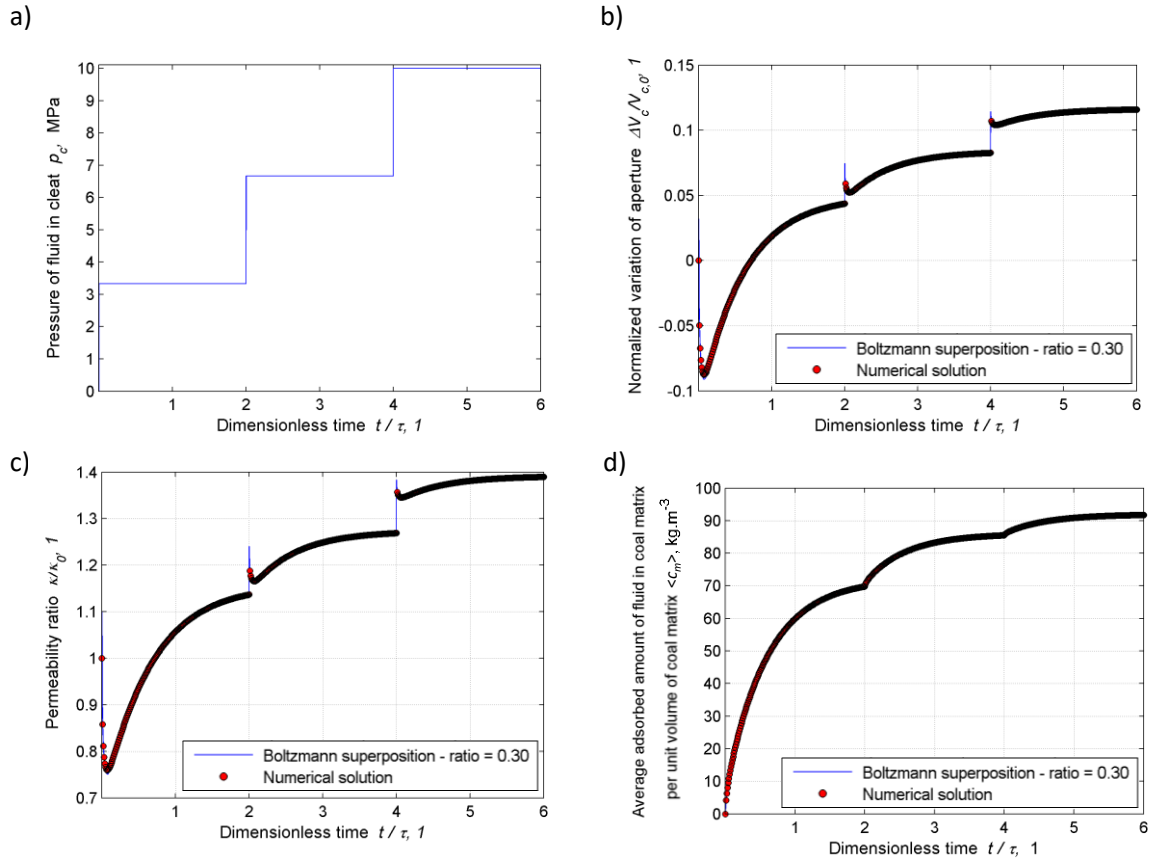
Figure 10: Response of an individual elliptical cleat with an aspect ratio $a/A = 0.03$ to a periodic rectangular loading: (a) history $p_c(t)$ of pressure of fluid considered, (b) variation $\Delta V_c(t)/V_0$ of the relative volume of the cleat, (c) permeability $\kappa(t)$, and (d) average amount $\langle c_m(t) \rangle$ of fluid in the coal matrix per unit volume of coal matrix. The characteristic time τ used to make the time dimensionless is $\tau = 25,000$ s.



426

427 *Figure 11: Response of an individual elliptical cleat with an aspect ratio $a/A = 0.03$ to a*
 428 *triangular loading: (a) history $p_c(t)$ of pressure of fluid considered, (b) variation $\Delta V_c(t)/V_0$ of*
 429 *the relative volume of the cleat, (c) permeability $\kappa(t)$, and (d) average amount $\langle c_m(t) \rangle$ of*
 430 *fluid in the coal matrix per unit volume of coal matrix. The characteristic time τ used to make*
 431 *the time dimensionless is $\tau = 25,000$ s.*

432



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Figure 12: Response of an individual elliptical cleat with an aspect ratio $a/A = 0.03$ to a staircase loading: (a) history $p_c(t)$ of pressure of fluid considered, (b) variation $\Delta V_c(t)/V_0$ of the relative volume of the cleat, (c) permeability $\kappa(t)$, and (d) average amount $\langle c_m(t) \rangle$ of fluid in the coal matrix per unit volume of coal matrix. The characteristic time τ used to make the time dimensionless is $\tau = 25,000$ s.

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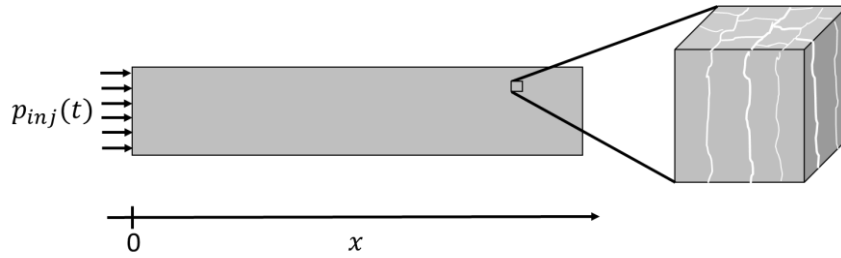
451

3 Modeling at the scale of the coal seam integrating transient variations of permeability obtained by FEM calculations at the scale of an individual cleat

We now consider the problem of an injection into a cylindrical coal structure of direction \underline{e}_x , of length $L = 200$ m, with an arbitrary cross-section whose characteristic dimension is much larger than the centimeter, so that there are many cleats over the cross-section (see Figure 13). The cleat with an elliptic cross-section of aspect ratio $a/A = 0.03$ considered in section 2 is assumed to be representative of the cleats in the sample. The sample initially contains CO₂ at $p_{init} = 1$ MPa. At time $t = 0$ s, CO₂ is injected at one end of the cylinder at a pressure $p_{inj}(t) = 10$ MPa while the other end and the lateral surface remain sealed. The process is assumed isothermal, remaining at 40°C. At this temperature, we consider an equation of state for CO₂ of the form:

$$\rho(p) = M_{\text{CO}_2} \left(\frac{p}{RT} + \frac{c_1}{1 + \exp\left(-\left(\frac{p - c_2}{c_3}\right)\right)} \right) \quad (27)$$

452 where p is the pressure of the fluid, ρ is its mass density, $R = 8.314 \text{ J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1}$ is the ideal gas
 453 constant, and c_1, c_2, c_3 are parameters. A fit of this equation to the actual equation of state
 454 of CO_2 at 40°C (Lemmon et al., 2020) yields $c_1 = 2.19 \cdot 10^4 \text{ mol}\cdot\text{m}^{-3}$, $c_2 = 9.92 \cdot 10^6 \text{ Pa}$, and
 455 $c_3 = 1.21 \cdot 10^6 \text{ Pa}$, with an error smaller than 12% over the whole range of pressures
 456 considered, even in supercritical conditions (i.e., above approximately 7.5 MPa). We consider
 457 that the properties of the material are those given in Table 1. We aim at solving the evolutions
 458 of the system over time.



459

460 *Figure 13: 1-dimensional problem solved at the scale of a structure. The inset is a zoom on a*
 461 *representative elementary volume. In the cleats, the fluid pressure is $p(x, t)$.*

462

463 The initial hydraulic conditions read:

$$p_c(x, 0) = p_{init} \quad (28)$$

464 and the hydraulic boundary conditions read:

$$\begin{cases} p_c(x = 0, t) = p_{inj}(t) = 10 \text{ MPa} \\ \left. \frac{\partial p_c}{\partial x} \right|_{x=L} = 0 \end{cases} \quad (29)$$

465 We assume that the hydro-mechanical response of any representative elementary
 466 volume of material to a history of pressure $p_c(x, t)$ of fluid in the cleats is the same as the one
 467 identified with the Abaqus calculations performed in section 2 for a system containing an
 468 individual cleat. We will study the hydro-mechanical response of a representative elementary
 469 volume subjected to generic mechanical boundary conditions in section 4.

470 The problem considered is one-dimensional: at any time, all variables depend on the
 471 coordinate x only. Consequently, solving the problem means, in particular, finding out the
 472 evolutions in time of the pressure $p_c(x, t)$ of fluid in the cleat, of the average amount $m_m(x, t)$
 473 of fluid in the coal matrix (per unit volume of fractured coal), and the permeability $\kappa(x, t)$.
 474 Since the fluid is injected from one end of the sample, in the generic case, the pressure $p_c(x, t)$
 475 is expected to be heterogeneous, and hence the parameters $m_m(x, t)$ or $\kappa(x, t)$ as well. We
 476 expect the degree of heterogeneity and how heterogeneity remains to depend on the
 477 permeability κ_0 of the fractured coal: we will vary this parameter from $\kappa_0 = 10^{-13} \text{ m}^2$ to $\kappa_0 =$
 478 10^{-11} m^2 (which corresponds to the low permeability domain of Moore (2012)), while keeping
 479 other properties (see Table 1) constant.

480 Solving the problem requires first formulating the fractured coal's constitutive
 481 equations, the next section's focus.

482 3.1 Governing equations at the scale of the coal seam

483 We can write the mass balance equation for a representative elementary volume of
 484 fractured coal as:

$$\frac{\partial m}{\partial t} = -\frac{\partial w}{\partial x'} \quad (30)$$

485 where w is the macroscopic fluid mass flow through the cleat and m is the total fluid mass
 486 content (per unit volume V_0 of fractured coal in the reference configuration) which is
 487 composed of the fluid mass content m_c in the cleat and the fluid mass content m_m in the coal
 488 matrix (still per unit volume V_0 of fractured coal in the reference configuration), i.e., $m =$
 489 $m_c + m_m$.

490 The macroscopic flow (i.e., transfer of mass) of fluid through the cleats (also called
 491 seepage) is a viscous flow generally considered (Harpalani and Chen, 1997) to obey Darcy's
 492 law (Darcy, 1856):

$$w(x, t) = -\frac{\rho_F \kappa}{\eta_F} \frac{\partial p_c}{\partial x} = -k \frac{\partial p_c}{\partial x} \quad (31)$$

493 where $k = \rho_F \kappa / \eta_F$, $p_c = p_c(x, t)$, $\kappa = \kappa(x, t)$, and where ρ_F and η_F are the bulk mass density
 494 and the viscosity of the fluid (here CO₂), respectively. Under isothermal conditions, $\rho_F =$
 495 $\rho_F(p_c)$. For CO₂, we consider the equation of state given in Eq. (27). We assume that the
 496 viscosity $\eta_F = 47.83 \cdot 10^{-3}$ Pa.s (which corresponds to the viscosity of CO₂ at 313.15K and 10
 497 MPa (Lemmon et al., 2020)) does not depend on the pressure of the fluid.

498 The mass m_c of fluid in the cleat is related to the cleat porosity ϕ_c through $m_c = \rho_F \phi_c$.
 499 Assuming that the variations of the mass of fluid in the cleat due to the deformation of the
 500 cleat are negligible when compared to variations of mass due to variations of density, the mass
 501 m_c of fluid in the cleats can be written as follows:

$$m_c = \rho_F \phi_{c,0} \quad (32)$$

502 We neglect microscopic flow through the coal matrix in the direction (Ox) (i.e., $\underline{w}_m \cdot \underline{e}_x =$
 503 0). Therefore, one can adapt Eq. (24) as follows:

$$m_m(x, t) = J_{m_m}(t) \otimes c_{edge}(x, t) \quad (33)$$

504 Likewise, for what concerns the evolutions of permeability, by taking into account that $\sigma_0 =$
 505 0, we can adapt Eq. (23) as:

$$\kappa(x, t) = \kappa_0 \left(1 + \frac{p_c(x, t)}{K_c} + J_{\delta_c}(t) \otimes c_{edge}(x, t) \right)^3 \quad (34)$$

506 where $c_{edge}(x, t) = c_{edge}(p_c(x, t))$, as given by Eq. (5).

507 The kernels $J_{\delta_c}(t)$ and $J_{m_m}(t)$ required for the computation are those displayed in
 508 Figure 9 for the elliptical cleat with an aspect ratio $a/A = 0.03$.

509

510 3.2 Resolution of the problem with finite volume method

511 We divide space into $N_{nodes} + 1$ equispaced nodes, with node 0 located in $x_0 = 0$ and
 512 node N_{nodes} located in $x_{N_{nodes}} = L$. We note $d_{ij} = L/N_{nodes}$ the distance between
 513 neighboring nodes i and j . We note all variables at node i with a subscript i . We note the time
 514 step Δt .

515 We obtain the finite volume scheme by a discrete balance in the elements of the mesh.
 516 Integrating Eq. (30) over any element i of length V_i and discretizing it in time with a backward
 517 Euler scheme yields, for all elements i :

$$V_i(m_i(t + \Delta t) - m_i(t)) + \Delta t \sum_j w_{ij}(t + \Delta t) = 0 \quad (35)$$

518 where w_{ij} expresses an approximation of the outflow from element i to element j for any i
 519 and any neighbor j of i :

$$w_{ij}(t + \Delta t) = -k_{ij}(t) \frac{p_j(t + \Delta t) - p_i(t + \Delta t)}{d_{ij}} \quad (36)$$

520 where d_{ij} is the distance between the center of element i and element j . In this equation k_{ij}
 521 is an explicit evaluation of the permeability coefficient $k = \rho_F \kappa / \eta_F$ at the interface between
 522 element i and element j , here $k_{ij} = (k_i + k_j)/2$.

523 At node 0, the pressure is imposed:

$$p_0(t + \Delta t) - p_{inj} = 0 \quad (37)$$

524 Equation (35) is solved through an iterative process. Since m_m is a functional of $c(t)$, we
 525 have to evaluate the change δm_m for any change δc between t and $t + \Delta t$:

$$\delta m_m = \int_t^{t+\Delta t} J_{m_m}(t + \Delta t - u) \frac{\partial}{\partial u}(\delta c) du \quad (38)$$

526 Approximating δc by $\delta[c(t + \Delta t)](u - t)/\Delta t$ between t and $t + \Delta t$, we finally get:

$$\delta m_m = \left(\frac{1}{\Delta t} \int_0^{\Delta t} J_{m_m}(\Delta t - u) du \right) \delta c(t + \Delta t) \approx J_{m_m}(\Delta t) \delta c(t + \Delta t) / 2 \quad (39)$$

527 So, the set of equations (35), formally $E_i = 0$ for all $i \geq 1$, can be solved iteratively through a
 528 Newton method, $A_{ij} \delta p_j = -E_i$, and using the jacobian matrix:

$$A_{ij} = V_i \left(\phi_{c,0} \frac{d\rho_F}{dp_i} + \frac{J_{m_m}(\Delta t)}{2} \frac{dc_{edge}}{dp_i} \right) \delta_{ij} + \Delta t \left(\left(\sum_{r \in \text{neighbors}} \frac{k_{ir}(t)}{d_{ir}} \right) \delta_{ij} - \frac{k_{ij}(t)}{d_{ij}} \right) \quad (40)$$

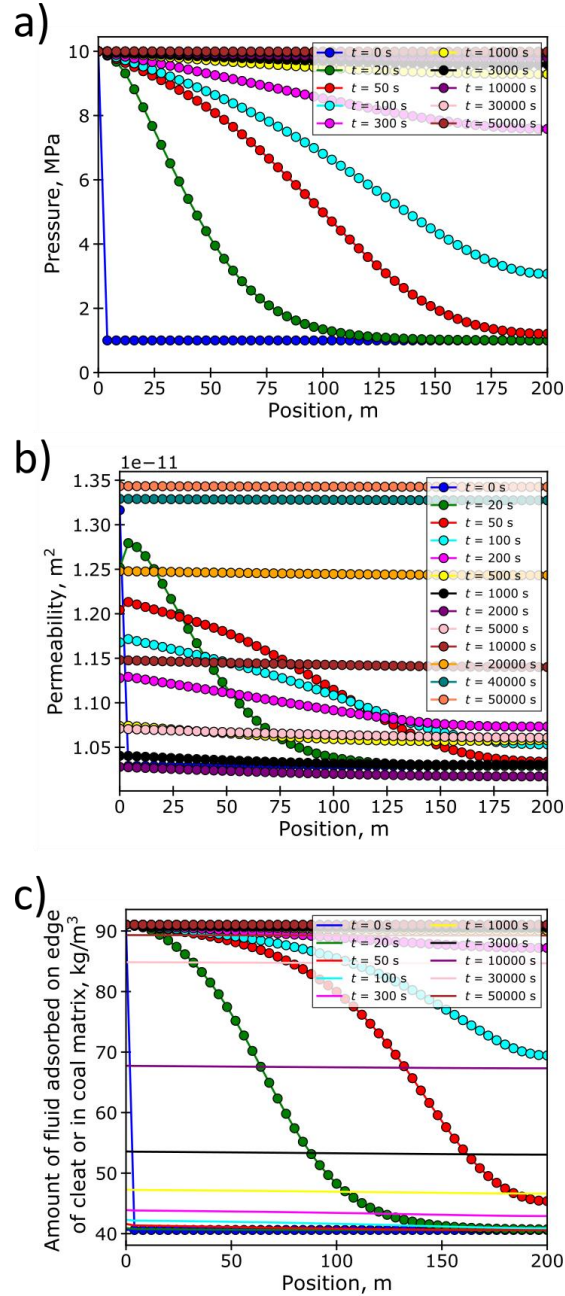
529 The Python code to solve this problem is available on the GitHub repository of the
 530 corresponding author².

531

² If you base future research on this code, please acknowledge it appropriately in your publications and cite the present article.

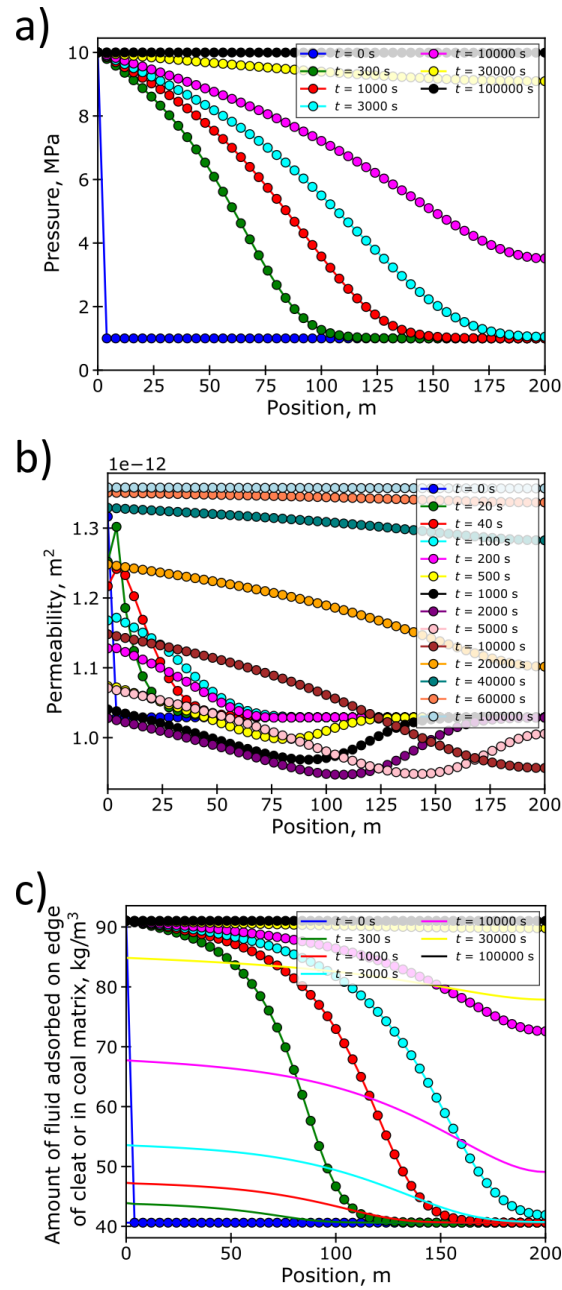
532 **3.3 Results and discussion of the various kinetics involved**

533 We display the results in Figure 14, Figure 15, and Figure 16 for the samples with a
 534 reference permeability κ_0 equal to 10^{-11} m^2 , 10^{-12} m^2 and 10^{-13} m^2 , respectively. Note that the
 535 times at which we display the results vary from figure to figure since the pressure in the cleats
 536 needs more time to reach its asymptotic value when the permeability κ_0 is lower.



537
 538 *Figure 14: Distribution of (a) pressure p_c in cleats, (b) permeability κ , and (c) concentrations*
 539 *of adsorbed fluid during injection into a coal sample with a reference permeability $\kappa_0 =$*
 540 *10^{-11} m^2 . In sub-figure c are displayed the concentration c_{edge} of fluid adsorbed on the edge*
 541 *of the cleat (with symbols) and the average concentration $\langle c_m \rangle$ of fluid adsorbed in the*
 542 *coal matrix (with solid lines). Both concentrations are expressed per unit volume of the coal*
 543 *matrix.*

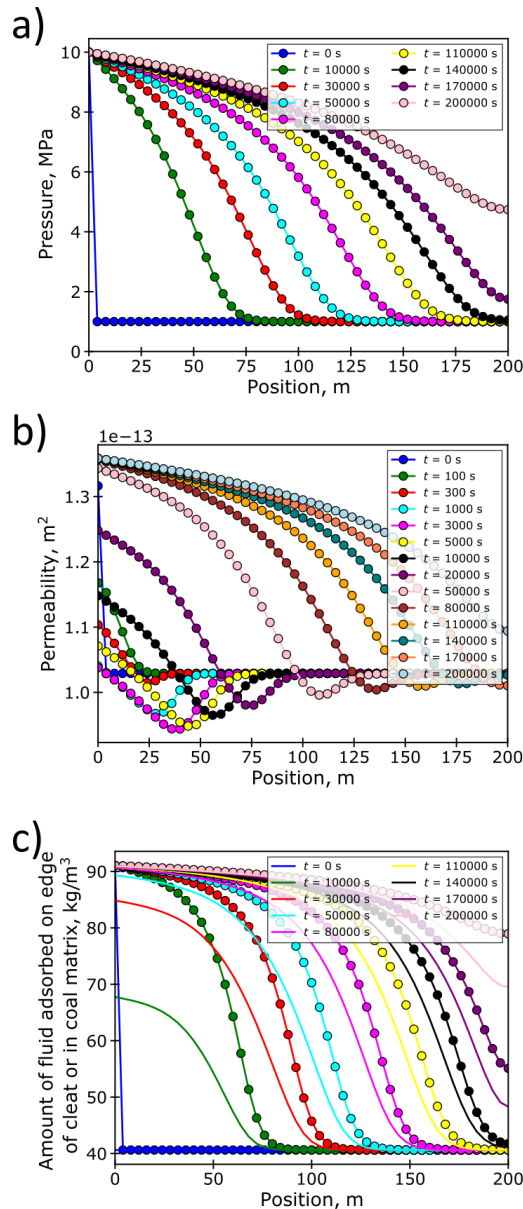
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545

546 *Figure 15: Distribution of (a) pressure p_c in cleats, (b) permeability κ , and (c) concentrations*
 547 *of adsorbed fluid during injection into a coal sample with a reference permeability $\kappa_0 =$*
 548 *$10^{-12} m^2$. In sub-figure c are displayed the concentration c_{edge} of fluid adsorbed on the edge*
 549 *of the cleat (with symbols) and the average concentration $\langle c_m \rangle$ of fluid adsorbed in the*
 550 *coal matrix (with solid lines). Both concentrations are expressed per unit volume of the coal*
 551 *matrix.*

552



553

554 *Figure 16: Distribution of (a) pressure p_c in cleats, (b) permeability κ , and (c) concentrations*
 555 *of adsorbed fluid during injection into a coal sample with a reference permeability $\kappa_0 =$*
 556 *10^{-13} m^2 . In sub-figure c are displayed the concentration c_{edge} of fluid adsorbed on the edge*
 557 *of the cleat (with symbols) and the average concentration $\langle c_m \rangle$ of fluid adsorbed in the*
 558 *coal matrix (with solid lines). Both concentrations are expressed per unit volume of the coal*
 559 *matrix.*

560

561 One can observe different behaviors depending on the permeability κ_0 . For the system
 562 with the highest permeability $\kappa_0 = 10^{-11} \text{ m}^2$ (see Figure 14), the pressure p_c in the cleats
 563 becomes homogeneous before a significant amount of fluid has diffused into the coal matrix
 564 (since at time $t = 1000 \text{ s}$, the average concentration $\langle c_m \rangle$ of fluid in the coal matrix is still
 565 low almost everywhere in the sample (see Figure 14-c)). Consequently, diffusion in the coal
 566 matrix mostly happens after the pressure in the cleats has become homogeneous. The
 567 average amounts of fluid adsorbed in the coal matrix and the transient adsorption-induced

568 variations of permeability are relatively homogeneous compared to the concentration on the
569 edge of the cleats. Also, the evolution of the permeability over time is rather complex, as it is
570 nonmonotonic and shaped like the letter N (i.e., it increases, decreases, and then increases
571 back). In contrast, for the system with the lowest permeability $\kappa_0 = 10^{-13} \text{ m}^2$ (see Figure 16),
572 transfer of fluid from/to the coal matrix happens faster than transport of fluid through the
573 cleat system: when the pressure in the cleats becomes homogeneous, the system has reached
574 equilibrium. One can check that, during the injection, at any position $x > 100 \text{ m}$ and at any
575 time t , the average concentration of fluid in the coal matrix is relatively close to the one given
576 by Eq. (5), i.e., relatively close to the concentration of fluid adsorbed on the cleat's edge.
577 Locally, for all those positions, we always remain relatively close to local thermodynamic
578 equilibrium. Far from the injection point, the permeability almost increases monotonically
579 with time while displaying the same pattern of heterogeneity as the pressure. Finally, the
580 system with the intermediate permeability $\kappa_0 = 10^{-12} \text{ m}^2$ (see Figure 15) manifests a
581 behavior that is intermediate between the one observed for the system with permeability
582 $\kappa_0 = 10^{-11} \text{ m}^2$ (see Figure 14) and $\kappa_0 = 10^{-13} \text{ m}^2$ (see Figure 16).

583 The results show a competition between two kinetics: 1) advective transfer through the
584 cleat network and 2) diffusive exchange between cleats and coal matrix. A characteristic time
585 τ_{diff} of diffusion of fluid from the cleats to the coal matrix is:

$$\tau_{diff} = (L_{diff})^2 / D, \quad (41)$$

586 where L_{diff} is a characteristic drainage length around each cleat (and is therefore equal to
587 half a characteristic distance between cleats). In the simulations at the scale of an individual
588 cleat performed in section 2, for which $L_{diff} \sim 0.5 \times 10^{-2} \text{ m}$, we find $\tau_{diff} \sim 2.5 \times 10^4 \text{ s}$.

589 We define a characteristic time τ_{advec} of the advective transfer through the cleats network as
590 the characteristic time required for the pressure on the edge of the reservoir to be disturbed
591 by the injection. We can obtain this characteristic time readily from an observation of Figure
592 14, Figure 15, and Figure 16: we find that $\tau_{advec} \sim 1 \cdot 10^2 \text{ s}$, $\tau_{advec} \sim 1 \cdot 10^4 \text{ s}$, and $\tau_{advec} \sim 1 \cdot$
593 10^5 s , for the systems with $\kappa_0 = 1 \cdot 10^{-11} \text{ m}^2$, $\kappa_0 = 1 \cdot 10^{-12} \text{ m}^2$, and $\kappa_0 = 1 \cdot 10^{-13} \text{ m}^2$,
594 respectively.

595 From the orders of magnitude just calculated, we infer that the assumption of local
596 thermodynamic equilibrium is reasonably valid if:

$$\tau_{advec} \gg \tau_{diff}. \quad (42)$$

597 In such a case, transient variations of permeability occur on the time scale τ_{diff} , which is too
598 short to impact the transfer process at the structural scale (which occurs on the time scale
599 τ_{advec}) significantly. At all positions that verify this criterion, at any time t , the average
600 concentration $\langle c_m \rangle$ of fluid in the coal matrix is relatively close to the one given by Eq. (5).
601 In contrast, if τ_{advec} is not much larger than τ_{diff} , the assumption of local thermodynamic
602 equilibrium is not valid anymore, and transient variations of permeability can impact the
603 dynamics of transfer at the structural scale (if the magnitude of those variations is significant).
604 The results displayed in Figure 14 to Figure 16 support this discussion regarding the various
605 kinetics involved in the process: only in the system with reference permeability $\kappa_0 = 1 \cdot 10^{-13}$
606 m^2 and further from about 100 m from the point of injection is the condition (42) respected
607 and the assumption of local thermodynamic equilibrium appears to be reasonably valid.

608 In contrast, in the case where $\tau_{advec} \ll \tau_{diff}$, the fluid has time to flow through the
 609 cleats before diffusing significantly through the coal matrix. The transient variations of
 610 permeability occur once the pressure of fluid in the cleats has become homogeneous.
 611 Consequently, the transient variations of permeability occur homogeneously as well.

612 4 Modeling at the scale of the coal seam integrating transient 613 variations of permeability: an engineering approach

614 In section 3, we showed that one could perform calculations at the structural scale as
 615 soon as two kernels are known: the kernel $J_{\delta_c}(t)$ that governs how the volume or aperture of
 616 the cleats varies over time and the kernel $J_{\dot{m}_m}(t)$ that governs the rate at which fluid is being
 617 transferred from the cleats to the coal matrix. We obtained those kernels through finite-
 618 elements simulations in section 2.3. However, the need to resort to finite-element simulations
 619 of a representative cleat is impractical for the engineer, who first of all does not know the
 620 cleats' geometry. To respond to this issue, in this section, we propose a generic form for those
 621 kernels.

622 In section 2, also, we performed all calculations in plane-strain. In this section, we derive
 623 constitutive equations of a representative elementary volume of fractured coal submitted to
 624 a generic state of strains and stresses to give more flexibility to use the proposed model.

625 Finally, up to now, we considered exclusively that variations of permeability are due to
 626 variations of aperture or volume of the cleat through the Kozeny-Carman relationship (3).
 627 However, instead of using Kozeny-Carman-type laws to model permeability in coal, many use
 628 stress-based permeability laws. This section also explains how to model transient variations
 629 of permeability when using those latter permeability laws.

630 4.1 Constitutive equations of a representative elementary volume of fractured coal 631 including transient variations of permeability under a generic state of strains or 632 stresses

633 We consider a representative elementary volume of isotropic fractured coal under a
 634 generic state of strains or stresses. Suppose the coal matrix fluid is homogeneously distributed
 635 (without needing necessarily to be in equilibrium with the fluid in the cleats). Under the
 636 assumption that deformations have a negligible impact on the amounts of fluid in the cleats
 637 and the coal matrix, the poromechanical behavior of this representative elementary volume
 638 is given by (Espinoza et al., 2016):

$$\left\{ \begin{array}{l} \sigma = K\varepsilon - bp_c - (1 - b)\sigma^a(c_m) \\ s_{ij} = 2Ge_{ij} \\ \phi_c - \phi_{c,0} = b\varepsilon + \frac{p_c}{N} - \frac{\sigma^a(c_m)}{N} \\ m_T = \phi_{c,0}\rho_F(p_c) + (1 - \phi_{c,0})c_m \end{array} \right. , \quad (43)$$

639 where c_m is the mass of fluid in the coal matrix per unit volume of coal matrix (such that $m_m =$
 640 $(1 - \phi_{c,0})c_m$, since m_m is the mass of fluid in the coal matrix per unit volume of fractured
 641 coal); m_T is the total mass of fluid in the fractured coal per unit volume of fractured coal; σ ,
 642 s_{ij} , ε , and e_{ij} are the volume stress, shear stresses, volume strain, and shear strains,

643 respectively; K , G , b , and N are the bulk modulus, shear modulus, Biot coefficient, and Biot
 644 modulus, respectively; σ^a is the adsorption stress that intervened in Eq. (9).

645 As explained in section 2, transient variations of permeability are due to the fluid
 646 distribution's heterogeneity in the coal matrix. To capture those transient effects, we,
 647 therefore, propose to extend the set of Eqs. (43) into:

$$\left\{ \begin{array}{l} \sigma = K\varepsilon - bp_c - (1 - b)\sigma^a(\langle c_m \rangle) \\ s_{ij} = 2Ge_{ij} \\ \phi_c - \phi_{c,0} = b\varepsilon + \frac{p_c}{N} - \frac{\sigma^a(\langle c_m \rangle)}{N} + \Delta\phi_{c,t} \\ m_T = \phi_{c,0}\rho_F(p_c) + (1 - \phi_{c,0})\langle c_m \rangle \end{array} \right. \quad (44)$$

648 where $\langle c_m \rangle$ denotes the volume average of c_m over the coal matrix, and where the
 649 function $\Delta\phi_{c,t}$ captures the transient variations of the porosity associated with the cleat
 650 system. Note that, like in section 3.1, these functions neglect the impact of deformation on
 651 the fluid amounts.

652 With the same reasoning as the one performed in section 2.3, we can obtain the average
 653 amount $\langle c_m \rangle$ of fluid in the coal matrix through convolution:

$$\langle c_m(t) \rangle = J_{\langle c_m \rangle}(t) \otimes c_{edge}(t) \quad (45)$$

654 where the function $J_{\langle c_m \rangle}(t)$ is a kernel. Since $m_m(t) = (1 - \phi_{c,0})\langle c_m(t) \rangle$, we have
 655 $J_{\langle c_m \rangle}(t) = J_{m_m}(t)/(1 - \phi_{c,0})$. Given the shape of the kernel $J_{m_m}(t)$ (see Figure 9-b), for the
 656 kernel $J_{\langle c_m \rangle}(t)$ we propose the function:

$$J_{\langle c_m \rangle}(t) = 1 - \exp(-t/\tau_{diff}), \quad (46)$$

657 where τ_{diff} is the characteristic time for the fluid in the representative elementary volume to
 658 diffuse through the coal matrix. This kernel tends toward one at large times and thus verifies
 659 that when a step of concentration $c_{edge,0}$ is applied on the edge of the cleat, at large times,
 660 the average concentration of fluid in the coal matrix tends toward $c_{edge,0}$.

661 Because of the presence of the term $\langle c_m(t) \rangle$, the set of equations (44) includes some
 662 dependency versus time, such that the porous solid behaves apparently like a poroviscoelastic
 663 solid. Consequently, we could rewrite this set of equations in the framework of a functional
 664 approach to poroviscoelasticity (Coussy, 2004) by introducing an apparent relaxation bulk
 665 modulus $K(t)$, a shear modulus $G(t)$, and a Biot coefficient $b(t)$, but such rewriting is out of
 666 the scope of this study, which is focused on the modeling of transient evolutions of
 667 permeability mostly.

668 In Eq. (44), variations $\phi_c - \phi_{c,0}$ of cleat porosity are composed of various terms: 1)
 669 terms that depend on time-independent variables, 2) a term that takes into account the
 670 average swelling of the coal matrix and takes into account the history of concentration, 3) a
 671 transient term $\Delta\phi_{c,t}$. Again, we can assert that this transient variation $\Delta\phi_{c,t}$ of the cleat pore
 672 volume can be obtained through a convolution product:

$$\Delta\phi_{c,t} = J_{\phi_c}(t) \otimes c_{edge}(t) \quad (47)$$

673 where the function $J_{\phi_c}(t)$ is a kernel. For this kernel $J_{\phi_c}(t)$, we propose a function of the
 674 form:

$$J_{\phi_c}(t) = -\frac{\Delta\phi_{c,max}}{c_{m,max}} \exp(-t/\tau_{diff})(1 - \exp(-t/\tau_{closing})), \quad (48)$$

675 where it is reasonable to consider that the characteristic time τ_{diff} is the same as in the
 676 previous equation, where $\tau_{closing}$ is another characteristic time, related to the time for the
 677 cleat to reach its minimum aperture, and where the parameter $\Delta\phi_{c,max}$ controls the
 678 magnitude of the maximal reduction of the cleat's aperture.

679 When permeability is given by the Kozeny-Carman relationship (3), we have all
 680 equations needed to perform the calculations. In comparison with classical poromechanical
 681 formulations that disregard the transient phenomena here considered, to capture those
 682 transient phenomena, we need therefore to introduce two kernels, namely the function
 683 $J_{<c_m>}(t)$ that governs how the average concentration of fluid in the coal matrix varies and the
 684 function $J_{\phi_c}(t)$ that governs how the cleat porosity varies. To define those two kernels, we
 685 need only three parameters: the characteristic time τ_{diff} needed for the fluid to diffuse
 686 through the coal matrix (which is the only parameter required to define $J_{<c_m>}(t)$), a
 687 parameter $\Delta\phi_{c,max}$ that controls the magnitude of the maximal transient reduction of the
 688 cleat's aperture, and a characteristic time $\tau_{closing}$ for the cleat to reach its minimal aperture.

689 In contrast, when the permeability law is stress-based, we propose to adapt it into:

$$\kappa = \kappa_0 \exp(\sigma + p_c - \sigma_t) \quad (49)$$

690 where σ is the total volume stress and σ_t is a transient stress. We assume that we can obtain
 691 this transient stress through the following convolution product:

$$\sigma_t = J_{\sigma}(t) \otimes c_{edge}(t) \quad (50)$$

692 where, for the kernel $J_{\sigma}(t)$, we propose:

$$J_{\sigma}(t) = \frac{\sigma_{t,max}}{c_{m,max}} \exp(-t/\tau_{diff}) (1 - \exp(-t/\tau_{closing})) \quad (51)$$

693 where $\sigma_{t,max}$ is a parameter that controls the magnitude of the maximal transient stress. Note
 694 that, in the absence of sorption, Eq. (49) leads to a permeability of coal, which depends on the
 695 Terzaghi effective stress (as was considered analytically by Connell et al. (2010) or
 696 experimentally by Somerton et al. (1975), for instance). In contrast, by considering a Kozeny-
 697 Carman-type aperture-based permeability relationship (Eq. (1)), with the set of poroelastic
 698 equations (44), one instead finds that the permeability depends on the Biot effective stress.

699 Here again, in comparison with classical poromechanical formulations that disregard the
 700 transient phenomena here considered, to capture those transient phenomena, we need to
 701 use two kernels, namely the function $J_{<c_m>}(t)$ that governs how the average concentration
 702 of fluid in the coal matrix varies and the function $J_{\sigma}(t)$ that governs how the effective stress
 703 acting on the cleat porosity varies. To define those two functions, we need only three
 704 parameters: the characteristic time τ_{diff} for the fluid in the representative elementary
 705 volume to diffuse through the coal matrix, the characteristic time $\tau_{closing}$ for the cleat to reach

706 its minimum aperture and a characteristic maximal transient variation $\sigma_{t,max}$ of the effective
 707 stress acting on the cleat porosity.

708 4.2 Verification of the validity of the proposed kernels

709 As already explained in section 4.1, the kernel $J_{<c_m>}(t)$ that intervenes in Eq. (45) and
 710 was defined in Eq. (46) is related to the kernel $J_{m_m}(t)$ introduced in Eq. (24) through:

$$J_{m_m}(t) = (1 - \phi_{c,0})J_{<c_m>}(t). \quad (52)$$

711 Consequently, given Eq. (26), we have:

$$J_{\dot{m}_m}(t) = (1 - \phi_{c,0})dJ_{<c_m>}/dt. \quad (53)$$

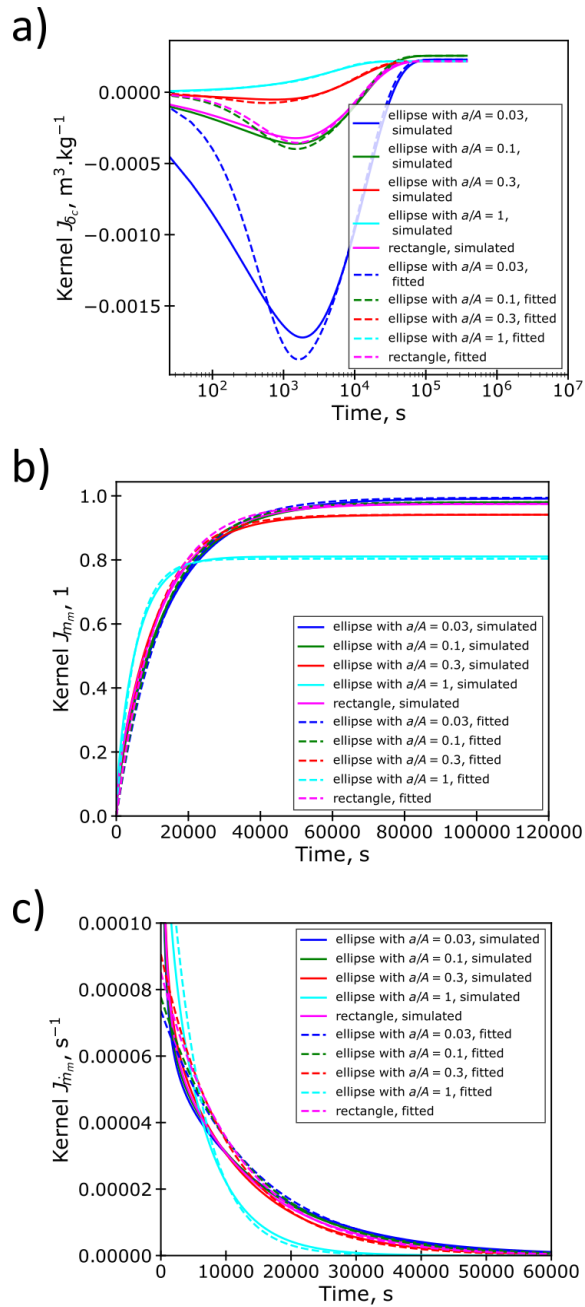
712 The relation between the kernel $J_{\phi_c}(t)$ (that intervened in Eq. (47) and that we defined in Eq.
 713 (48)) and kernels introduced in section 2.3 is more complicated. An examination of the
 714 equation governing $\phi_c - \phi_{c,0}$ in the set of equations (44), together with the fact that the
 715 adsorption stress σ^a depends linearly on the average concentration $<c_m>$ in the coal matrix
 716 (see Eq. (9)), show that the kernel J_{ϕ_c} must depend linearly on the kernels J_{δ_c} and J_{m_m} . Given
 717 that the kernel J_{ϕ_c} captures variations of pore volume relative to the reference volume of
 718 porous solid (see Eq. (47)) while the kernel J_{δ_c} captures variations of pore volume relative to
 719 the reference pore volume (see Eq. (18)), we pose:

$$J_{\delta_c} = \frac{1}{\phi_{c,0}}J_{\phi_c} + \beta J_{m_m} \quad (54)$$

720 where β is a proportionality factor that we can calculate from the simulated kernels, as $\beta =$
 721 $\lim_{t \rightarrow +\infty} J_{\delta_c} / J_{m_m}$. Based on the simulations performed in section 2.2, for the elliptical cleat with
 722 aspect ratio $a/A = 0.03$, we find $\beta = 2.35 \cdot 10^{-4} \text{ m}^3 \cdot \text{kg}^{-1}$. Relation (54) explains that the
 723 kernel J_{ϕ_c} can converge toward 0 in the long term, while Figure 8-b displays a contribution
 724 $\Delta V_{c,hydr}$ to the variation of volume of the cleats that converges toward a strictly positive value:
 725 in Eq. (44), we capture this net increase of cleat volume due to hydric effects in the long term
 726 by terms other than $\Delta\phi_{c,t}$.

727 Based on these relations, we can assess the validity of the generic forms (46) and (48)
 728 proposed for the kernels $J_{<c_m>}(t)$ and $J_{\phi_c}(t)$. To do so, for each cleat geometry, we fit the
 729 parameters τ_{diff} , $\tau_{closing}$ and $\Delta\phi_{t,max}$ which define the kernels $J_{<c_m>}(t)$ and $J_{\phi_c}(t)$ so that
 730 the kernels $J_{m_m}(t)$ and $J_{\dot{m}_m}(t)$, $J_{\delta_c}(t)$, as calculated through relations (52), (53), and (54),
 731 respectively, are as close as possible from their numerical evaluation by finite-element
 732 calculations (see Figure 9). Note that, in this fit, all parameters other than τ_{diff} , $\tau_{closing}$ and
 733 $\Delta\phi_{t,max}$ that are required for the evaluation of the kernels (e.g., $c_{m,max}$ and $\phi_{c,0}$) are already
 734 known and can be found in Table 1.

735 We display the results of this fit for all cleat geometries in Figure 17 and give the fitted
 736 parameters in Table 2. The figure shows that the form chosen for the kernels $J_{<c_m>}(t)$ and
 737 $J_{\phi_c}(t)$ is appropriate, in the sense that those kernels can be successfully fitted to kernels
 738 obtained numerically with finite-element simulations of the explicit diffusion of the fluid
 739 through the coal matrix.



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742 *Figure 17: Kernels (a) $J_{\delta_c}(t)$, (b) $J_{m_m}(t)$, and (c) $J_{\dot{m}_m}(t)$ for all geometries of cleats*
 743 *considered in this study, as evaluated with finite-element simulations (solid lines) and as*
 744 *obtained by fitting the generic functions proposed for the kernels $J_{<c_m>}(t)$ and $J_{\phi_c}(t)$*
 745 *(dashed lines).*

746

747 Table 2: Results of fit of kernels $J_{<c_m>}(t)$ and $J_{\phi_c}(t)$

Geometry of cleat's cross-section	Characteristic time τ_{diff} , s	Characteristic time $\tau_{closing}$, s	Characteristic maximal reduction of cleat porosity $\Delta\phi_{c,max}$, 1
Elliptic with aspect ratio $a/A = 0.03$	13502.2	503.4	$1.390 \cdot 10^{-3}$
Elliptic with aspect ratio $a/A = 0.1$	12615.8	538.7	$1.064 \cdot 10^{-3}$
Elliptic with aspect ratio $a/A = 0.3$	10388.4	200.8	$6.085 \cdot 10^{-4}$
Elliptic with aspect ratio $a/A = 1$	5103.9	204.5	$-6.396 \cdot 10^{-4}$
Rectangular with aspect ratio $a/A = 0.1$	11447.5	600.1	$1.251 \cdot 10^{-3}$

748

749 The values provided in Table 2 make it possible to confirm an observation already given
750 in section 2.2: the transient decrease of permeability (characterized by the parameter
751 $\Delta\phi_{c,max}$) is all the more pronounced that the aspect ratio a/A of the cross section of the cleat
752 differs from 1.0. The characteristic time τ_{diff} of the process of diffusion also decreases with
753 an increasing aspect ratio a/A , but we believe that this effect is mostly due to the fact that, in
754 our study, by construction, with an increasing aspect ratio a/A , the porosity of the cleats
755 increases and hence the volume of coal matrix toward which fluid diffuses is lower. The
756 magnitude of this characteristic time τ_{diff} is consistent with the value $L_{diff}^2/D =$
757 $(0.5 \times 10^{-2} \text{ m}^2)^2/(10^{-9} \text{ m}^2 \cdot \text{s}^{-1}) = 2.5 \cdot 10^4 \text{ s}$ expected from the inter-cleats spacing. Also,
758 as already discussed in section 2.2, the characteristic time $\tau_{closing}$ is mostly independent of
759 the aspect ratio when this aspect ratio differs sufficiently from 1 (i.e., when the cleat is flat
760 enough). Figure 17 shows that this characteristic time $\tau_{closing}$ is about half-an-order-of-
761 magnitude smaller than the actual time for the cleat to reach its minimal aperture (and hence
762 for the permeability to be minimal). At a given aspect ratio, the three parameters are shown
763 in Table 2, which define the various kernels, are roughly independent of the geometry of the
764 cleat's cross-section, as can be inferred from an observation of the parameters for the elliptic
765 and rectangular cleats with aspect ratio $a/A = 0.1$.

766 Parameters (e.g., permeability (Wold et al., 2008)) can vary significantly from the lab
767 scale to the field scale. In the same spirit, the parameters proposed here in Table 2, obtained
768 by calculations performed at the scale of an individual cleat, may differ from parameters
769 relevant for calculations at the scale of the seam. However, Figure 17 shows that the kernels'
770 form that we propose is valid for all geometries assumed for the cleat. Consequently, one
771 could expect that this kernels' form should also be relevant for calculations performed at the
772 seam scale, even though the three parameters that define the kernels quantitatively may
773 differ from those given in Table 2. This difference must stem from the difference of scale and
774 the fact that the actual cleats' geometry is not elliptical.

775

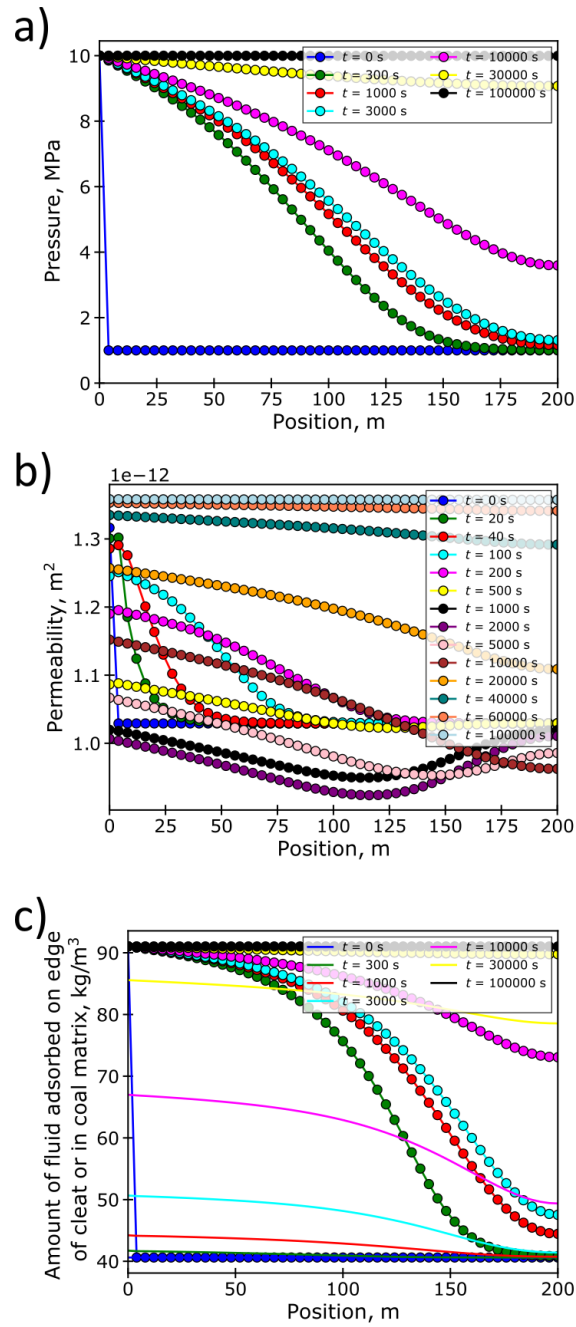
776 4.3 Example of calculation with the proposed kernels

777 In this section, we consider again the problem treated in section 3 of a one-dimensional
778 injection into a cylindrical coal structure of length $L = 200 \text{ m}$ and of arbitrary cross section,

779 in which cleats with an elliptic cross-section of aspect ratio $a/A = 0.03$ is assumed to be
 780 representative. We consider the same initial conditions (i.e., the sample contains CO₂ at 1
 781 MPa) and hydraulic boundary conditions (i.e., CO₂ is injected at 10 MPa at one end of the
 782 cylinder while the other surfaces remain sealed). However, in contrast to what we did in
 783 section 3 in which we calculated the needed kernels with finite-element simulation, here we
 784 use the kernels $J_{<c_m>}(t)$ and $J_{\phi_c}(t)$ given by Eqs. (46) and (48), respectively, together with
 785 the fitted parameters given in Table 2 for the cleat of interest, i.e., $\tau_{diff} = 13502.2$ s,
 786 $\tau_{closing} = 503.4$ s, and $\Delta\phi_{c,max} = 1.390 \cdot 10^{-3}$. The same code as in section 3.2 is used, but
 787 now the kernels $J_{m_m}(t)$ and $J_{\dot{m}_m}(t)$, $J_{\delta_c}(t)$ are calculated from the kernels $J_{<c_m>}(t)$ and $J_{\phi_c}(t)$
 788 through relations (52), (53), and (54). For such one-dimensional simulations, assuming like in
 789 Section 3 that the hydro-mechanical couplings are all lumped in the proposed kernels, the
 790 knowledge of Biot coefficient of Biot modulus is not needed. In contrast, generic 3-
 791 dimensional simulations require those parameters.

792 The results of the simulation of the injection, displayed in Figure 18, compare well with
 793 the results obtained with the kernels calculated with the finite-element simulations (see
 794 Figure 15). Nevertheless, we observe some differences at the earliest times, when the
 795 proposed kernels differ the most from the simulated ones (see Figure 17-a in particular).
 796 However, the good agreement confirms that the generic form (46) and (48) of the kernels
 797 $J_{<c_m>}(t)$ and $J_{\phi_c}(t)$ is appropriate to capture adsorption-induced transient variations of
 798 permeability.

799



800

801 *Figure 18: Distribution of (a) pressure p_c in cleats, (b) permeability κ , and (c) concentrations*
 802 *of adsorbed fluid during an injection in a coal sample with a reference permeability $\kappa_0 =$*
 803 *$10^{-12} m^2$, based on the generic kernels $J_{<c_m>}(t)$ and $J_{\phi_c}(t)$ given by Eqs. (46) and (48),*
 804 *respectively. For sub-figure c, the same comments apply as for Figure 15-c.*

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806 4.4 Limitations and perspectives

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808 We identify the following limitations and perspectives:

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- In the simulations of an injection of CO₂ into a coal sample or coal reservoir (see section 3), we made some assumptions to use the kernels calculated with finite-element simulations in section 2. Indeed, we assumed that fluid diffuses through the coal matrix only perpendicular to the cleats and neighboring slices do not restrain the swelling of a slice of coal perpendicular to the cleat. By relaxing those assumptions, the actual kernels to be used would differ from the ones calculated. But we do not expect the kernels' form to differ significantly from the generic forms proposed in section 4. Consequently, for the simulations performed in section 4.3, relaxing the assumptions above-mentioned would come back to use input parameters that would differ from the ones given in Table 2.
 - The calculations that we performed in sections 3 and 4 at the scale of a coal sample or coal seam were based on the idea that some representative cleat exists. However, we know that the cleat system of coal seams is complex and somehow fractal, involving various geometries (i.e., shape, aperture, or aspect ratio) of the cleats and their spacing. Consequently, one should extend the approach presented in this work to such a case. We see no a priori impossibility in performing this extension. One could express the permeability of the representative elementary volume of fractured coal as a function of the aperture of the various families of cleats and could introduce kernels specific to each family of cleats.
 - We discussed in section 3.3 the notion of local thermodynamic equilibrium. Depending on the coal's properties and the distance to the injection well, we found out that assuming local thermodynamic equilibrium could be reasonable or not. Even if, for a given coal seam, the assumption of local thermodynamic equilibrium is not valid at some locations in the seam or at some time during the injection process, assessing the quantitative impact of this assumption on the seam's response (in particular on the predicted flow rates) would be interesting.
 - The proposed approach could be applied to other physical processes that induce heterogeneity inside the representative elementary volume of fractured coal. For instance, injection of a hot fluid would cause heterogeneity of temperature and thermal dilation inside the coal matrix and induce transient permeability variations. However, the approach here proposed, relying on the Boltzmann superposition principle, is limited to processes for which the time-dependent phenomena and their consequences can be expressed linearly. If nonlinearities enter the picture, then the method could be extended by introducing nonlinear convolutions in Volterra's formalism (Ogunfunmi, 2007). However, such formulation is significantly more complex than the one used in this study and requires a larger number of kernels, whose identification could prove difficult. As proposed in this study, as much as possible, aiming at formulating the governing equations linearly of the time-dependent physical process seems to be a reasonable approach to try before considering nonlinear developments.

850

851 5 Conclusions

852 In this work, we aimed at finding out how to model transient variations of permeability
853 due to heterogeneous adsorption-induced swelling of the coal matrix by formulating
854 constitutive equations at the scale of a representative elementary volume of fractured coal.
855 The need to develop constitutive equations at this scale rather than at a lower scale stems
856 from the will to perform simulations at the scale of a coal sample or a coal reservoir, without
857 meshing explicitly coal matrix and cleat network and solving for the transport through the coal
858 matrix. To better understand the physical processes involved, we first performed finite-
859 element simulations of an individual cleat surrounded by a coal matrix, solving the transport
860 of fluid through the coal matrix and the coal matrix's swelling. Then we derived the
861 constitutive equations that we were seeking for, using the just mentioned finite-element
862 calculations as inputs. Finally, we solved those constitutive equations with the finite-volume
863 method to simulate CO₂ injection into a coal structure. The conclusions of this work are:

- 864 • We confirm that adsorption-induced transient variations of permeability can be
865 due to the heterogeneity of the concentration of fluid in the coal matrix. The
866 magnitude of those transient variations of permeability depends significantly on
867 the aspect ratio of the cleat's cross-section (see Figure 5). In contrast, a
868 characteristic duration τ_{diff} of those transient variations is relatively
869 independent of the geometry of the cleat's cross-section (see Figure 5 again).
870 This characteristic duration τ_{diff} should rather depend on the diffusivity D of
871 the fluid through the coal matrix and on the characteristic distance L_{diff}
872 between neighboring cleats (see Eq. (41)).
- 873 • Transient variations of permeability can be rigorously captured with constitutive
874 equations formulated at the scale of a representative elementary volume of
875 fractured coal. To do so, we relied on the Boltzmann superposition principle. The
876 resulting constitutive equation (see Eq. (23)) involves a convolution product so
877 that the permeability $\kappa(t)$ at a given time t depends on the full history of
878 pressure $p_c(t')$ of fluid in the cleats until time t , i.e., at all times $t' < t$.
- 879 • Although transient adsorption-induced variations of permeability are involved,
880 we could model them with a not-too-complex use of the Boltzmann
881 superposition principle. The reason why we succeeded in doing so is the
882 following: we can express the swelling as depending linearly on the
883 concentration of fluid in the coal matrix (see Eq. (8)), and the transport of fluid
884 through the coal matrix can be well modeled with a linear diffusion equation (see
885 Eq. (7)) so that all-time dependent processes and their consequences depend
886 linearly on the history $c_{edge}(t)$ of concentration of fluid in the coal matrix in the
887 vicinity of the cleat. In this approach, all nonlinearity is concentrated into the
888 instantaneous relation (5) between this concentration and the pressure $p_c(t)$ of
889 the fluid in the cleats.
- 890 • At the scale of a representative elementary volume of fractured coal, to
891 formulate a complete set of constitutive equations that can capture transient
892 variations of permeability, we need to use convolutions productions that rely on

893 two kernels: one that governs the evolution of the aperture of the cleat
894 (depending on how we write the equations, this kernel is $J_{\delta_c}(t)$ (see Eq. (34) in
895 section 3.1) or $J_{\phi_c}(t)$ (see Eq. (47) in section 4.1) and one that governs the
896 evolution of the amount of fluid in the coal matrix (depending on how we write
897 the equations, this kernel is $J_{m_m}(t) = dJ_{m_m}(t)/dt$ (see Eq. (33) in section 3.1)
898 or $J_{<c_m>}(t)$ (see Eq. (45) in section 4.1)). Those two kernels can be identified with
899 finite-element simulations of the transport of fluid through the coal matrix and
900 the swelling it induces (see section 2.2 and Figure 9).

- 901 • At this same scale of a representative elementary volume of fractured coal, we
902 proposed three-dimensional constitutive equations that can capture transient
903 permeability variations in Eq. (44). Those constitutive equations require the
904 knowledge of two kernels (i.e., $J_{\phi_c}(t)$ and $J_{<c_m>}(t)$), for which we propose
905 generic forms (see Eqs. (46) and (48)) that can be used by the engineer.
906 Altogether, those two kernels depend on only three parameters with an explicit
907 physical meaning: a characteristic time τ_{diff} of diffusion of fluid through the coal
908 matrix, a characteristic time $\tau_{closing}$ related to the closure of the cleats, and a
909 parameter $\Delta\phi_{c,max}$ that controls the magnitude of the maximal transient
910 variation of permeability. For those who prefer to model permeability as
911 depending on stresses rather than on aperture of the cleat, we propose an
912 appropriate adaptation of the permeability law (see Eq. (49)), which requires the
913 knowledge of 1 kernel (see Eq. (50)) for which we also propose a generic form
914 (see Eq. (51)) that also depends on three parameters with the same explicit
915 physical meaning (i.e., the same two characteristic times and a parameter $\sigma_{t,max}$
916 that controls the magnitude of the maximal transient variations of effective
917 stress).
- 918 • In the process of injection of fluid in a coal bed, two kinetics are at stake, namely
919 the kinetics of advective transfer through the cleats (with a characteristic time
920 τ_{advec}) and the kinetics of transfer from the cleats to the coal matrix (with a
921 characteristic time τ_{diff}). If those characteristic times verify Eq. (42), i.e.,
922 $\tau_{advec} \gg \tau_{diff}$, local thermodynamic equilibrium is ensured: in a small
923 representative elementary volume of fractured coal, one can consider that the
924 chemical potential of the fluid is homogeneous (i.e., the same in the cleat as
925 anywhere in the coal matrix). We provide an expression for the characteristic
926 time τ_{diff} in Eq. (41). For a given system, it is possible that the assumption of
927 local thermodynamic equilibrium could be valid far from the injection well but
928 not reasonable in the vicinity of the injection well.

929 We discussed the limitations and perspectives in section 4.4.

930

931

932 [Nomenclature](#)

Symbol	Meaning	Unit
A_{ij}	Jacobian matrix	$\text{kg}\cdot\text{m}^{-2}\cdot\text{Pa}^{-1}$
a/A	Aspect ratio of elliptical cleat	1
α	Parameter governing magnitude of adsorption-induced swelling	$\text{m}^3\cdot\text{kg}^{-1}$
b	Biot coefficient of fractured coal	1
β	Proportionality factor	$\text{m}^3\cdot\text{kg}^{-1}$
c_{edge}	Amount of fluid adsorbed on edge of cleat	$\text{kg}\cdot\text{m}^{-3}$
\mathbf{C}_m	Stiffness tensor of coal matrix	Pa
c_m	Mass concentration of fluid in coal matrix, per unit volume of coal matrix in reference configuration	$\text{kg}\cdot\text{m}^{-3}$
$c_{m,max}$	Maximum adsorbed amount in Langmuir isotherm	$\text{kg}\cdot\text{m}^{-3}$
D	Diffusivity of fluid in coal matrix	$\text{m}^2\cdot\text{s}^{-1}$
$\partial\Omega_{in}$	Inner surface of system	N.A.
$\partial\Omega_{out}$	Outer surface of system	N.A.
$\Delta\phi_{c,t}$	Transient variations of porosity associated to cleat system	1
$\Delta\phi_{c,max}$	Maximal transient variation of porosity associated to cleat system	1
$\underline{\underline{e}}$	Shear strain tensor	1
$\underline{\underline{\varepsilon}}$	Strain tensor	1
ε	Volume strain	1
ε^a	Adsorption strain	1
ϕ_c	Porosity associated to cleats	1
η_F	Dynamic viscosity of fluid	$\text{Pa}\cdot\text{s}$
$J_{\langle c_m \rangle}$	Kernel associated to average mass concentration of fluid in coal matrix per unit volume of coal matrix	1
J_{δ_c}	Kernel associated to variation of aperture of cleats	$\text{m}^3\cdot\text{kg}^{-1}$
J_{m_m}	Kernel associated to average mass concentration of fluid in coal matrix per unit volume of fractured coal	1
$J_{\dot{m}_m}$	Kernel associated to rate of variation of average mass concentration of fluid in coal matrix per unit volume of fractured coal	s^{-1}
J_{ϕ_c}	Kernel associated to variation of cleat porosity	$\text{m}^3\cdot\text{kg}^{-1}$
J_{σ}	Kernel associated to transient stress	$\text{Pa}\cdot\text{m}^3\cdot\text{kg}^{-1}$
K	Bulk modulus of fractured coal	Pa
$K_{c,p}$	Parameter characterizing stiffness of cleat	Pa
$K_{c,\sigma}$	Parameter characterizing stiffness of cleat	Pa
K_m	Bulk modulus of coal matrix	Pa
k	Permeability coefficient	$\text{kg}\cdot\text{m}^{-1}\cdot\text{Pa}^{-1}\cdot\text{s}^{-1}$
κ	Intrinsic permeability of fractured coal	m^2
κ_0	Intrinsic permeability of fractured coal in reference configuration	m^2
$\underline{\underline{\xi}}$	Displacement	m
L	Length of system	m

M_F	Molar mass of fluid	kg.m^{-3}
M_{CO_2}	Molar mass of CO_2	kg.m^{-3}
m	Mass of fluid in fractured coal per unit volume of fractured coal	kg.m^{-3}
m_c	Mass of fluid in cleat per unit volume of fractured coal	kg.m^{-3}
m_m	Mass of fluid in coal matrix per unit volume of fractured coal	kg.m^{-3}
N	Biot modulus	Pa
\underline{n}	Outward normal vector	N.A.
ν_m	Poisson's ratio of coal matrix	1
p_c	Pressure of fluid in cleats	Pa
$p_{c,0}$	Step of pressure of fluid in cleats	Pa
p_{init}	Initial pressure of fluid in coal seam	Pa
p_{inj}	Pressure of injection in coal seam	Pa
p_{L0}	Characteristic pressure in Langmuir isotherm	Pa
p_m	Thermodynamic pressure of fluid in coal matrix	Pa
ρ_F	Mass density of fluid	kg.m^{-3}
R	Ideal gas constant	$\text{J.mol}^{-1}.\text{K}^{-1}$
$\underline{\underline{s}}$	Shear stress tensor	Pa
$\underline{\underline{\sigma}}$	Stress tensor	Pa
σ	Volume stress	Pa
σ^a	Adsorption stress	Pa
σ_0	Imposed confining stress	Pa
σ_t	Transient stress	Pa
$\sigma_{t,max}$	Characteristic maximal transient stress	Pa
t	Time	s
Δt	Time step	s
T	Temperature	K
τ	Characteristic time	s
τ_{advec}	Characteristic time of transfer through cleats network	s
$\tau_{closing}$	Characteristic time for cleats to reach their minimal aperture	s
τ_{diff}	Characteristic time of diffusion of fluid from cleats to coal matrix	s
V_c	Volume of cleats	m^3
$V_{c,0}$	Volume of cleats in reference configuration	m^3
V_0	Volume of fractured coal in reference configuration	m^3
V_i	Volume of element i	m
ΔV_c	Variation of volume of cleats	m^3
$\Delta V_{c,mech}$	Variation of volume of cleats due to mechanical effects	m^3
$\Delta V_{c,hydr}$	Variation of volume of cleats due to ingress of fluid in coal matrix	m^3
w	Mass flow rate of fluid through cleats	$\text{kg.m}^{-2}.\text{s}^{-1}$
w_m	Mass flow rate of fluid through coal matrix	$\text{kg.m}^{-2}.\text{s}^{-1}$
x	Position	m
Δx	Distance between nodes	m
Y	Heaviside function	1

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