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Pore-scale hydro-mechanical modeling of gas transport in coal matrix

Ahmad Mostafa¹, Luc Scholtès², Fabrice Golfier¹

¹Université de Lorraine, CNRS, Laboratoire GeoRessources, Nancy, France

²Université Clermont-Auvergne, CNRS, IRD, OPGC, Laboratoire Magmas et Volcans, Clermont-

Ferrand, France

Corresponding author email: ahmad.mostafa@univ-lorraine.fr

ABSTRACT:

We present a 3D model coupling a discrete element model and a pore network model specifically developed to describe the different diffusion mechanisms at stake in coal matrix as well as the associated adsorption induced deformations. The material is assumed to be saturated with gas and diffusion occurs through the combination of Knudsen diffusion within the pore space, surface diffusion at the solid surface, and adsorption-desorption at the pore-solid interface. The model is hydro-mechanically coupled in the sense that changes in pore pressure produce hydrostatic forces that deform the solid skeleton, while deformation of the pore space induces pore pressure changes that promote inter-pore flow. Sorption induced deformations are taken into account by considering an additional pressure term related to the concentration of gas within the medium (the so-called solvation pressure). The implemented transport models are verified against analytical solutions describing diffusion in porous media with and without sorption-desorption, and a comparison is made with a swelling experiment performed on a coal specimen to illustrate the relevance of the proposed approach for describing adsorption induced deformation. As a result, this new pore-scale model offers a precise way to assess coal matrix sorption induced deformation and contributes to the knowledge of CBM storage and transport processes.

Keywords: coal, gas transport, sorption, DEM, PNM, solvation pressure.

24 **1 Introduction**

25 Coalbed methane (CBM), also known as coal seam gas (CSG), has drawn much attention lately as an alternative
26 energy resource. Production curves of CBM reservoirs are very different, however, from the ones of hydrocarbon
27 conventional reservoirs (Liu et al., 2011). As emphasized by several studies (Li et al., 2017; Mostaghimi et al.,
28 2017, 2016; Privalov et al., 2020; Wang et al., 2018), transport and poromechanical properties of coal are strongly
29 driven by topological and morphological features of its pore space.

30 Coal is fractured by nature; it is a dual porosity/permeability system made up of a porous matrix surrounded by
31 fractures known as cleats (Figure 1, scale II). The orientation of this quasi-orthogonal cleat network including
32 tensile fractures or face cleats, and compressive and strike-slip fractures or butt cleats, depends on the principal
33 stress' directions (Laubach et al., 1998) and provides preferential pathways for fluid flow with fracture apertures
34 up to 100 microns. In contrast, methane gas is stored within the low porosity coal matrix with pore sizes generally
35 varying from a few to several dozens of nanometers (Li et al., 2017; Wang et al., 2018).

36 The cleat-matrix system compartmentalizes the transport and mechanical properties of coal. Knudsen and surface
37 diffusions prevail in the nanometer-sized pores of the matrix, while molecular diffusion and two-phase Darcy
38 flow occur mainly within the cleat network. All these transport mechanisms induce mechanical couplings related
39 to both (i) the pore pressure changes which may alter the effective stress and consequently impact the bulk volume
40 of the coal and, (ii) the sorption processes which contribute to swell or shrink the coal matrix (Pini et al., 2009;
41 Wang et al., 2011). Indeed, coal can sorb various gases including CO₂, CH₄ and N₂, and the adsorption of these
42 gases induces swelling strains (Ceglarska-Stefańska and Czapliński, 1993; Ceglarska-Stefańska and Zarębska,
43 2002; Pan and Connell, 2007). The magnitude of adsorption-induced deformation depends on the pores structure
44 as well as on the nature of the gas adsorbed. It is well known for instance that the swelling is much higher with
45 CO₂ than with CH₄ (Brochard et al., 2012), hence advocating the development of enhanced coal bed methane
46 (ECBM) technology combining CBM recovery and CO₂ sequestration.

47 If many experimental studies have attempted to accurately determine this volumetric deformation rate (Majewska
48 et al., 2010) and have given precious insights into the changes in pore size distribution during
49 adsorption/desorption processes, they did not completely succeed to capture the dynamics of mass transfer within
50 the cleat-matrix system since measurements are generally carried out at equilibrium (Wang et al., 2018). Also,

51 coal measurements are strongly affected by sample preparation, material composition, environment, and
52 methodology (Mostaghimi et al., 2017).

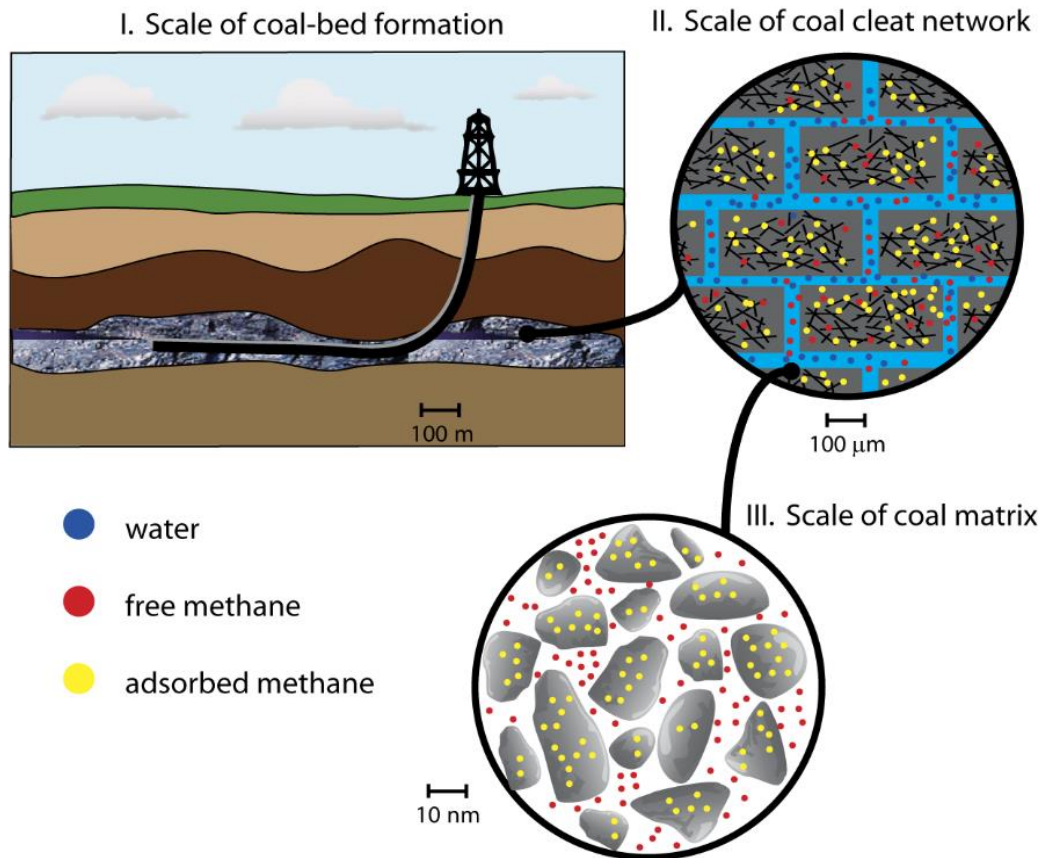
53

54 The inherent couplings between the physical processes at stake and the multiscale features of coal need to be
55 explored further to better assess the macroscopic response of the coal matrix and the sorption induced volumetric
56 deformation. If pioneering works (see for instance the review by (Liu et al., 2011)) have focused on permeability
57 models using continuum (Connell, 2016; Guo et al., 2016) or dual-porosity approaches (Bertrand et al., 2017; Ma
58 et al., 2017; Perrier et al., 2018; Wu et al., 2010), much attention has been paid in the last years to study the hydro-
59 mechanical (HM) behavior of coal matrix at the pore-scale. For instance, Liu et al. (Liu and Mostaghimi, 2017)
60 have derived a model based on the lattice Boltzmann method taking into account fluid-rock interactions with
61 permeability and porosity variations to investigate the reactive transport of CO₂-saturated brine in coal fractures.
62 Youjun and Vafai, (Youjun and Vafai, 2017) have simulated fluid flow in pores using a digital SEM image of a
63 coal rock sample. More recently, Sampath et al. (Sampath et al., 2020) have developed a HM model taking into
64 account CO₂ diffusion and adsorption-induced mechanical deformation in coal matrix. To the best of our
65 knowledge, however, a coupled HM model that incorporates the different transport mechanisms combining the
66 diversity of nanoscale processes with the complexity of pore network topology has never been derived for coal
67 matrix.

68 In this work, we propose a 3D pore-scale model for coal matrix based on a pore network model (PNM) describing
69 gas transport coupled to a discrete element model (DEM) describing the mechanical behavior. The model is based
70 on the framework of the pore scale finite volume (PFV) scheme implemented in the open-source platform YADE
71 DEM (Šmilauer et al., 2015). The DEM-PFV coupled approach was initially designed for upscaling fluid flow in
72 granular materials (Chareyre et al., 2012) and has been afterward used to describe hydro-mechanical processes in
73 both soils (Catalano et al., 2014; Scholtès et al., 2015) and rocks (Papachristos et al., 2017). Lately, Caulk et al.
74 (Caulk et al., 2020) extended its capability by incorporating heat transfer to the scheme and the associated
75 possibility to describe thermo-hydro-mechanical processes. Here, we go further by introducing mass transport and
76 sorption induced deformation processes to its formulation.

77 In summary, the equations governing the transport and HM schemes are first presented and derived accordingly
78 to the geometric and numerical characteristics of the proposed model. Then, a validation exercise is provided

79 where each component of the transport model is challenged against analytical solutions of Fick's law considering
 80 either pore-pore, pore-particle or particle-particle diffusion mechanisms. Finally, the coupled HM model is used
 81 to simulate an experiment from the literature where a coal sample experience swelling due to gas adsorption.



82

83

Figure 1 Different scales of a coalbed reservoir.

84 2 Methodology

85 The porous coal matrix is modeled based on the assumption that the solid phase is made up of densely packed
 86 spherical particles bonded one with another, and the pore space idealized as a network of interconnected pores.

87 The following sections detail the mechanical and transport models, the assumptions made and their respective
 88 implementation in YADE DEM.

89 2.1 Mechanical scheme

90 The DEM is a numerical method that models geomaterials as assemblies of particles interacting one with another
 91 according to predefined contact laws as initially proposed by Cundall and Strack (Cundall and Strack, 1979). Each
 92 particle is defined through its own mass, size and position. The numerical scheme relies on an iterative temporal

93 integration of Newton's second law of motion to describe the movement of each particle depending on the forces
 94 they are subjected to. To model coal, we use spherical particles and define elastic-brittle-force displacement laws
 95 between the particles, as proposed in the bonded particle model (BPM) introduced by (Scholtès and Donzé, 2013)
 96 to simulate rock like materials. The interaction force F between two particles a and b is decomposed into a normal
 97 and a shear component.

98 The normal force is computed as:

$$99 \quad F_n = k_n \Delta D \quad (1)$$

100 where ΔD is the relative displacement between a and b , and k_n is the normal stiffness defined by:

$$101 \quad k_n = E_{eq} \frac{R_a R_b}{R_a + R_b} \quad (2)$$

102 with E_{eq} an elastic modulus, and R_a and R_b the radii of a and b respectively. In compression, F_n can increase
 103 indefinitely. In tension, a maximum acceptable force is defined as a function of the interparticle tensile strength t
 104 such as $F_{n,max} = t A_{int}$, with $A_{int} = \pi[\min(R_a, R_b)]^2$. If $F_n \geq F_{n,max}$, tensile rupture occurs.

105 The shear force is computed incrementally such as:

$$106 \quad F_s = \{F_s\}_{updated} + k_s \Delta u_s \quad (3)$$

107 with k_s the shear stiffness defined as $k_s = P k_n$ with $0 < P < 1$, and Δu_s the relative incremental tangential
 108 displacement. F_s can increase up to a threshold value defined by a Mohr-Coulomb type criterion such as $F_{s,max} =$
 109 $c + F_n \tan(\varphi)$, with c the interparticle cohesion, and φ the interparticle friction angle. If $F_s \geq F_{s,max}$, shear rupture
 110 occurs.

111 In addition, the integration of the equations of motion being done through an explicit finite difference scheme, a
 112 local non viscous damping is used to dissipate kinetic energy and to ease the convergence of the simulated system
 113 toward quasi-static equilibrium (see (Duriez et al., 2016) for details). Basically, the resultant force $\vec{F} = \vec{F}_n +$
 114 \vec{F}_s considered in Newton's second law of motion is damped by a force \vec{F}^d defined as:

$$115 \quad \vec{F}^d = -\alpha \text{sign} \left(\sum \vec{F} \cdot \left(\vec{v} + \frac{\Delta t^m}{2} \vec{a} \right) \right) \sum \vec{F} \quad (4)$$

116 with α a damping coefficient ($0 < \alpha < 1$), \vec{v} and \vec{a} the velocity and acceleration of the particle respectively, and
 117 Δt^m the mechanical time step.

118 2.2 *Mass transport model*

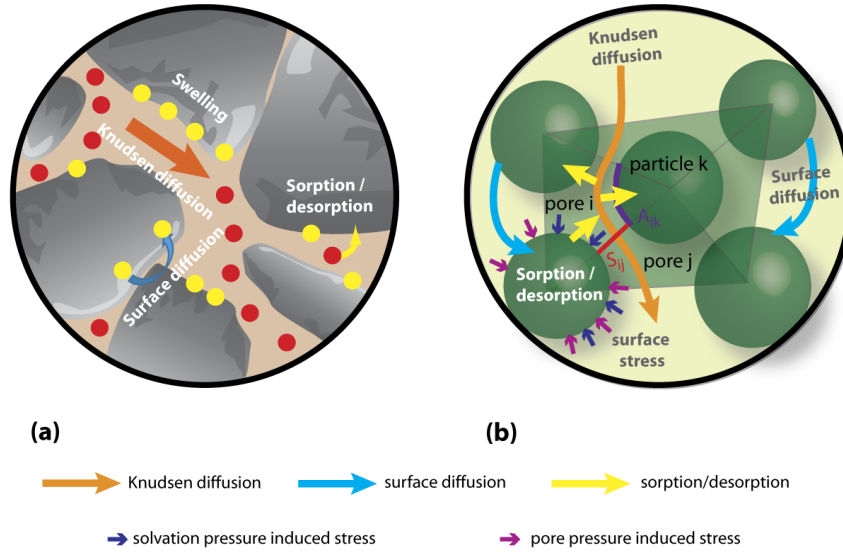
119 The diffusion of gas molecules within coal is rather complex due to the multi-scale architecture of the material,
120 with pore sizes varying over several orders of magnitude (Jing et al., 2017). Theory of gas transport in porous
121 media has a long history from the simple Fick's law for binary systems, to the Stefan-Maxwell equations for
122 multicomponent mixtures and ultimately, the Dusty-Gas Model, based on the Chapman-Enskog kinetic theory
123 and including the couplings between the various mechanisms (Reinecke and Sleep, 2002; Thorstenson and
124 Pollock, 1989). Indeed, different transport modes manifest and drive gas migration in nanoporous media,
125 depending on the pore size and on the flow regime (Do, 1998):

- 126 (i) Viscous flow in which displacement of molecules is induced by the mean velocity and governed by
127 the total pressure gradient which acts as driving force.
- 128 (ii) Molecular or continuum diffusion where molecular-molecular collision prevails. In the gas mixture,
129 each species moves relative to each other, and the driving force is the molar fraction gradient for an
130 isothermal system.
- 131 (iii) Knudsen diffusion in which molecule-wall collisions dominate compared to collisions between
132 molecules. Contrarily to molecular diffusion, each species moves independently from each other.
133 The driving force here is the partial pressure gradient, which reduces to the total pressure gradient
134 for a single species gas flow.
- 135 (iv) Surface diffusion in which adsorbed molecules moves along solid surface (pore walls) from one
136 adsorption site to another (Choi et al., 2001).

137 These different transport mechanisms may compete and combine in a complex way. The relative magnitude of
138 collisions between molecules relative to collisions between molecules and walls (molecular vs Knudsen diffusion)
139 is classically expressed through the dimensionless Knudsen number K_n , defined as the ratio between the molecular
140 mean free path length λ which is a function of pore pressure, and the mean pore diameter d . For nanoporous
141 material such as coal, and at low pore pressures, $K_n \gg 1$ typically, meaning that Knudsen diffusion generally
142 prevails.

143 In the present study, we assume that water was previously expelled from coal matrix (residual water is expected
144 to be trapped in the smallest pores that do not participate to flow) so that single-phase gas flow occurs within the
145 pore network. The gaseous mixture is only composed of methane (an ideal gas assumption is employed) and a

146 part of gas is sorbed on the coal solid phase. Due to the very low permeability of the matrix and considering
 147 single-species gas phase, only Knudsen and surface diffusions are thus considered (Figure 2a).



148

149

Figure 2 Mass transport and associated diffusion mechanisms in a) coal matrix, b) DEM-PFV model.

150 The DEM-PFV approach discretizes the pore space of the particle assembly into tetrahedra built from a weighted
 151 Delaunay triangulation of the particles' centers (Figure 2b). Each tetrahedron contains both a solid fraction
 152 resulting from the intersection between the tetrahedron and the associated vertex spheres, and a fluid fraction (the
 153 pore) resulting from the remainder of the tetrahedron volume. Each pore is thus connected to 4 neighboring pores
 154 and is in contact with 4 particles through surface areas where mass transport occurs by diffusion.

155 2.2.1 Mass balance equation for pores

156 Gas molecules are transported from one pore to another and are adsorbed/desorbed from and to the solid particles.
 157 Let V_f be the volume of a pore i saturated with fluid (gas). Integrating the continuity equation in pore i gives:

$$158 \int_{V_f} \frac{\partial c_i}{\partial t} dV + \int_{V_f} \bar{\nabla} \cdot \bar{J}_{pore,i} dV = 0 \quad (5)$$

159 where c_i is the concentration of gas in pore i , and $\bar{J}_{pore,i}$ represents the gas fluxes coming in and out of pore i .

160 Applying the divergence theorem to Equation (5) gives:

$$161 \int_{V_f} \frac{\partial c_i}{\partial t} \cdot dV + \int_{\partial V_f} \bar{J}_{pore,i} \cdot \bar{n} dS = 0 \quad (6)$$

162 Since each pore i is connected to 4 neighboring pores and in contact with 4 particles, the contour surface of each
 163 pore can be calculated as:

164
$$\partial V_f = \sum_{j=1}^4 \partial V_{pore,i-pore,j} + \sum_{k=1}^4 \partial V_{pore,i-particle,k} = \sum_{j=1}^4 S_{ij} + \sum_{k=1}^4 A_{ik} \quad (7)$$

165 with S_{ij} and A_{ik} the intersection surface areas between pore i and pore j , and between pore i and particle k
 166 respectively.

167 In our modeling approach, Knudsen diffusion describes the pore-pore diffusion fluxes and adsorption/desorption
 168 describes the pore-particle diffusion fluxes (Figure 2). Equation (7) can then be written as follows:

169
$$\int_{V_f} \frac{\partial c_i}{\partial t} dV = -\sum_{j=1}^4 \int_{S_{ij}} \bar{J}_{ij}^K \cdot \bar{n} dS - \sum_{k=1}^4 \int_{A_{ik}} \bar{J}_{ik}^{ad} \cdot \bar{n} dS \quad (8)$$

170 where \bar{J}_{ij}^K is the Knudsen diffusion flux between pore i and pore j , and \bar{J}_{ik}^{ad} is the adsorption/desorption flux
 171 between pore i and particle k .

172 2.2.1.1 Knudsen diffusion

173 For a single-species phase flow along a straight circular capillary tube, the Knudsen diffusion flux can be defined
 174 as (Do, 1998):

175
$$\bar{J}^K = -D^K \bar{\nabla} c \quad (9)$$

176
$$\bar{J}^K = -D^K \frac{1}{RT} \bar{\nabla} p$$

177 with $p = cRT$ the gas pressure, R the gas constant (J/mol/K) and T the absolute temperature (K). The Knudsen
 178 diffusion coefficient D^K (m²/s) is given by:

179
$$D^K = \frac{2r}{3} \sqrt{\frac{8RT}{\pi M_g}} \quad (10)$$

180 with r the pore hydraulic radius, and M_g the molar mass (kg/m³).

181 The Knudsen flux may thus be written in terms of concentration between pore i and pore j such as:

182
$$\bar{J}_{ij}^K = -\frac{2r}{3} \sqrt{\frac{8RT}{\pi M_g}} \bar{\nabla} c_{ij} \quad (11)$$

183 with r computed as proposed by (Chareyre et al., 2012) in the PFV scheme. Then the spatial integration of
 184 Equation (11) between pores i and j gives:

185
$$\int_{S_{ij}} \bar{J}_{ij}^K \cdot \bar{n} dS = - \frac{S_{ij}}{L_{ij}} \frac{2r}{3} \sqrt{\frac{8RT}{\pi M_g}} (c_j^t - c_i^t) \quad (12)$$

186 where L_{ij} is the distance between the centers of pore i and pore j .

187 2.2.1.2 Sorption

188 Sorption generally refers to the transport and attachment of a solute to the surface of a solid phase. Sorption
 189 processes are usually investigated at equilibrium through isotherms that relate the amount of adsorbed solute to
 190 the bulk concentration at constant temperature. In our modeling approach, we define kinetics formulations
 191 balancing the relative rates of adsorption and desorption (Raouf et al., 2012).

192 ➤ **Linear first order sorption**

193 Linear sorption is characterized by an infinite number of sites on the particles' surfaces. As a result, solid particles
 194 can adsorb an infinite number of gas molecules according to the following relation:

195
$$\frac{\partial s}{\partial t} = (K_{att}c - K_{det}s) \quad (13)$$

196 where s (mol/m³) is the amount of solute adsorbed onto the solid phase surface, c (mol/m³) is the concentration
 197 of solute in the mobile phase (gas), K_{att} and K_{det} are respectively the adsorption and desorption coefficients
 198 defining the rate of adsorption/desorption, and t is the time (s). At equilibrium we recover the linear relationship:

199
$$K_d = \frac{K_{att}}{K_{det}}$$

200 where $\frac{\partial s}{\partial t} = 0$ then $s = \frac{K_{att}}{K_{det}}c = K_d \cdot c \quad (14)$

201 ➤ **Langmuir sorption**

202 If adsorption is limited by the number of sites on the solid surface (Liu et al., 2019), the following nonlinear
 203 relation can be used:

204
$$\frac{\partial s}{\partial t} = \frac{s}{s_{max}} K_{det} - \left(1 - \frac{s}{s_{max}}\right) K_{att}c \quad (15)$$

205 where the sorption is proportional to the number of available sites $\left(1 - \frac{s}{s_{max}}\right)$, s_{max} being the maximum amount
 206 of sorbed concentration. At equilibrium, we recover:

207
$$\frac{\partial s}{\partial t} = 0, \text{ then } \frac{s}{s_{max}} = \frac{K_d \cdot c}{1 + K_d c} \quad (16)$$

208 In our model, the diffusion fluxes between pores and particles can thus be described either by the linear relation
 209 of Equation (13) or by the nonlinear relation (Langmuir isotherm) of Equation (15). For instance, the spatial
 210 integration of the pore-particle diffusion flux between pore i and particle k in the case where it is governed by the
 211 Langmuir isotherm is defined as:

212
$$\int_{A_{ik}} \bar{J}_{ik}^{ad} \cdot \bar{n} dS = \frac{A_{ik}}{A_k} \left[K_{det} \frac{s_k^t}{s_{k,max}} - \left(1 - \frac{s_k^t}{s_{k,max}} \right) K_{att} c_i \right] \times V_p \quad (17)$$

213 where s_k (mol/m³) is the solute concentration adsorbed on the surface of particle k , A_k is the overall surface of
 214 particle k , and V_p is the volume of particle k .

215 Finally, considering both sorption Equation (17) and Knudsen diffusion Equation (12), the evolution of each pore
 216 concentration is obtained by an explicit integration over time such as:

217
$$c_i^{t+\Delta t} = \sum_{i=1}^4 \frac{V_p A_{ik}}{V_f A_k} \left[\left(1 - \frac{s_k^t}{s_{k,max}} \right) K_{att} c_i^t - K_{det} \frac{s_k^t}{s_{k,max}} \right] \Delta t^{\frac{ad}{des}} + \sum_{i=1}^4 \frac{S_{ij}}{V_f L_{ij}} \frac{2r}{3} \sqrt{\frac{8RT}{\pi M_g}} (c_j^t - c_i^t) \Delta t^K + c_i^t \quad (18)$$

218 with $\Delta t^{\frac{ad}{des}}$ and Δt^K the associated time steps.

219 2.2.2 Mass balance equation for particles

220 The amount of gas molecules adsorbed on the solid particles results from the transport of gas molecules both from
 221 neighboring solid particles and from pores (Figure 2b). Particle-particle flux occurs by surface diffusion, while
 222 pore-particle flux results from adsorption/desorption process. Let V_p be the volume of a particle k . Integrating the
 223 mass balance equation on particle k gives:

224
$$\int_{V_p} \frac{\partial s_k}{\partial t} dV + \int_{V_p} \bar{\nabla} \cdot \bar{J}_{particle,k} dV = 0 \quad (19)$$

225 where s_k is the adsorbed concentration on particle k , and $\bar{J}_{particle,k}$ represents the gas fluxes coming in and out of
 226 particle k .

227 Applying the divergence theorem to Equation (19) gives:

228
$$\int_{V_p} \frac{\partial s_k}{\partial t} dV + \int_{\partial V_p} \bar{J}_{particle,k} \cdot \bar{n} dS = 0 \quad (20)$$

229 Since each particle is connected to N particles (N depends on the spatial arrangement of the particles within the
230 medium), and to 4 pores, the contour surface of each particle can be calculated as:

231
$$\partial V_p = \sum_{l=1}^N \partial V_{particle,k-particle,l} + \sum_{i=1}^4 \partial V_{particle,k-pore,i} = \sum_{l=1}^N S_{kl} + \sum_{i=1}^4 A_{ik} \quad (21)$$

232 where S_{kl} is the intersection surface area between particle k and particle l , and A_{ki} is the intersection surface area
233 between particle k and pore i (Figure 2b).

234 Equation (21) can then be written as follows:

235
$$\int_{V_p} \frac{\partial s_k}{\partial t} dV = - \sum_{l=1}^N \int_{S_{kl}} \bar{J}_{kl}^S \cdot \bar{n} dS - \sum_{i=1}^4 \int_{A_{ki}} \bar{J}_{ki}^{ad} \cdot \bar{n} \cdot dS \quad (22)$$

236 where \bar{J}_{kl}^S is the surface diffusion flux between particle k and particle l , and \bar{J}_{ki}^{ad} is the adsorption/desorption flux
237 between particle k and pore i .

238 2.2.2.1 Surface diffusion

239 Particle-particle flux is governed by surface diffusion in which gas molecules are transported from the surface of
240 a grain to the surface of a neighboring grain. In essence, molecules jump from an adsorption site to another
241 adsorption site. The diffusive flux is thus driven by the gradient of sorbed concentration between and within
242 particles and the surface diffusion flux can be expressed as:

243
$$\bar{J}^S = -D^S \bar{\nabla} s \quad (23)$$

244 The surface diffusion coefficient D^S is known to be a function of temperature following the Arrhenius equation
245 and strongly depends on the surface loading (Do, 1998). For sake of simplicity, however, it will be kept constant
246 hereafter.

247 The spatial integration of the diffusive flux between particles k and l is defined as:

248
$$\int_{S_{kl}} \bar{J}_{kl}^S \cdot \bar{n} dS = - \theta \frac{D^S S_{kl}}{L_{kl}} (s_k^t - s_l^t) \quad (24)$$

249 where S_{kl} is the intersection surface area between particle k and particle l computed here as $S_{kl} = 4r_l r_k$, L_{kl} is the
250 distance between the centers of particle k and particle l , r_l and r_k are the radiuses of particle k and particle l ,

251 and θ is a scaling parameter enabling to constrain the transport properties of the simulated medium depending on
 252 the structural characteristics of the particle assembly (θ is related to the particle connectivity which is a function
 253 of the particle size distribution of the packing as well of its porosity).

254 2.2.2.2 Sorption

255 Particle-pore diffusion processes are governed by the same mechanisms than pore-particle diffusion processes
 256 (see Equation (17)).

257 Finally, combining both sorption Equation (17) and surface diffusion Equation (24), the evolution of each particle
 258 concentration is obtained by an explicit integration over time such as:

$$259 \quad s_k^{t+\Delta t} = \sum_{i=1}^4 \frac{A_{ik}}{A_k} \left[\left(1 - \frac{s_k^t}{s_{k,max}} \right) K_{att} c_i^t - K_{det} \frac{s_k^t}{s_{k,max}} \right] \Delta t^{\frac{ad}{des}} + \sum_{l=1}^m \theta \frac{D^S S_{kl}}{V_p L_{kl}} (s_k^t - s_l^t) \Delta t^{sd} + s_k^t \quad (25)$$

260 with $\Delta t^{\frac{ad}{des}}$ and Δt^{sd} the associated time steps.

261 2.3 Hydro-mechanical coupling

262 The hydraulic forces acting on a solid particle immersed in a fluid result from both the pressure and viscous stress
 263 acting on its surface (Chareyre et al., 2012). In coal matrix with nanometer pores, pore velocities are very low
 264 (they are actually neglected in the mass transport model) and hence are the viscous stress forces. We thus only
 265 consider the contribution of the normal pressure forces which result from pressure losses within the pore space as
 266 follows:

$$267 \quad \vec{F}_{ij}^k = A_{ij}^k (p_i - p_j) \vec{n}_{ij} \quad (26)$$

268 where $p_i - p_j$ represents the pressure difference between pore i and pore j , A_{ij}^k is the intersection surface area
 269 between particle k and the pore throat between pore i and pore j , \vec{n}_{ij} is the unit vector pointing from pore i to pore
 270 j . As shown in (Catalano et al., 2014) and in (Scholtès et al., 2015), the HM coupling resulting from Equation (26)
 271 enables the DEM-PFV model to simulate conventional poromechanical behaviors as described by Biot's theory
 272 (Biot, 1941).

273 The classical Biot's theory states that the mechanical behavior of a porous medium depends on the bulk fluid
 274 pressure only (p_f) without considering its detailed composition. This means that two pore fluids with different
 275 compositions having the same bulk pressure should produce the same volumetric deformation. This is not verified

276 for swelling materials like coal which shows different deformation amplitudes depending on the nature of the pore
 277 fluid (Ottiger et al., 2008). It is now well known that the deformation of coal is not solely governed by the pore
 278 bulk pressure: as for all swelling materials, sorption processes induce additional deformation related to the nature
 279 of the sorbed molecules (Brochard et al., 2012). Gas molecules get adsorbed at the surface of the pores and induces
 280 a surface excess free energy. An additional interfacial force is thus exerted and resulting stress develops within
 281 the porous medium. Different approaches were proposed to introduce these nanoscale features into an extended
 282 poromechanical equation to explain the sorption-induced deformations from a continuum description. First works
 283 have introduced the so-called solvation pressure to describe swelling since in confined pores, pressure is not
 284 anymore a scalar (Gor and Neimark, 2010; Kowalczyk et al., 2008; Ustinov and Do, 2006; Yang et al., 2010). A
 285 second class of models, based on an energy balance approach, relates the changes in surface potential energy
 286 due to gas adsorption to the elastic energy (“Dilatation of Porous Glass - SCHERER - 1986 - Journal of the
 287 American Ceramic Society - Wiley Online Library,” n.d.; Dolino et al., 1996; Grosman and Ortega, 2008; Pan
 288 and Connell, 2007). Other researchers also have reformulated the poroelastic constitutive equations by introducing
 289 an apparent porosity and an interaction free energy that are both related to the Gibbs adsorption isotherm (Mushrif
 290 and Rey, 2009; Perrier et al., 2018; Pijaudier-Cabot et al., 2011; Sampath et al., 2020; Vermorel and Pijaudier-
 291 Cabot, 2014). In contrast, Vandamme and co-workers (Brochard et al., 2012; Espinoza et al., 2014, 2013;
 292 Nikoosokhan et al., 2014) have made use of the bulk pressure and of the swelling strain to express the sorbed
 293 amount or the adsorption-induced pressure.

294 In the present work, the sorption-induced deformations are interpreted in terms of the solvation pressure or
 295 adsorption pressure p_s , that is related to the amount of gas molecules attached to the solid skeleton. Specifically,
 296 the total pore pressure in each pore i is equal to the sum of the bulk fluid pressure p_f and of the solvation pressure
 297 p_s such as:

$$298 \quad p_i = p_{i,f} + p_{i,s} \quad (27)$$

299 with $p_{i,f}$ and $p_{i,s}$ defined as follows:

300
$$p_{i,s} = \sum_{k=1}^4 \frac{A_{i,k} \times p_s^k}{A_{i,k}} \quad (28)$$

301
$$p_{i,f} = c_i RT \quad (29)$$

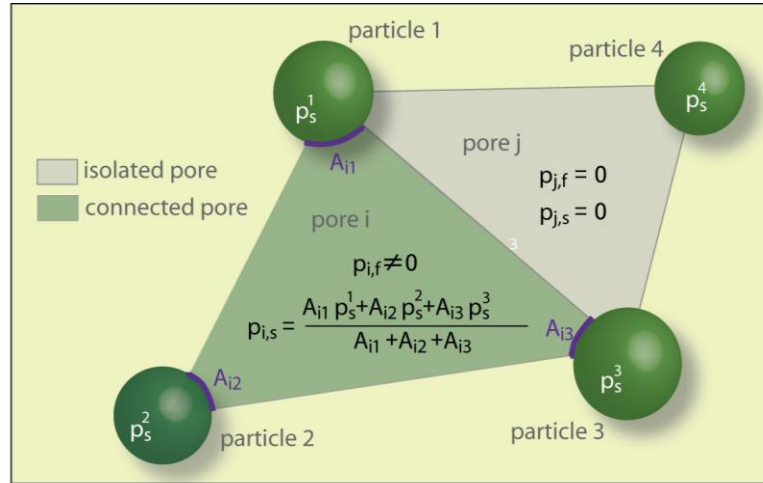
302 where the solvation pressure related to the amount of gas molecules attached to the particle k is denoted p_s^k and
 303 computed as

304
$$p_s^k = \alpha s_k RT \quad (30)$$

305 A_{ik} is the intersection surface area between particle k and pore i , and α is a coupling coefficient. Hereafter, α is
 306 fixed constant but it is not necessarily the case (Brochard et al., 2012).

307 Although the solvation pressure is dependent of the adsorbed amount on a grain and hence should be specific to
 308 a given particle, it is also a pressure which is summed up with the bulk pressure specific to a pore. To further
 309 explain how this additional pressure term is calculated within the pore space, we illustrate in Figure 3 the different
 310 situations that can be encountered:

- 311 • Case 1: for connected pores (belonging to the connected porosity which will be further discussed in
 312 Section 3), we compute a contact surface weighted average solvation pressure for each particle in contact
 313 with pore i , and the fluid pressure value depends on the gas pressure gradient within the percolating pore
 314 space.
- 315 • Case 2: for isolated pores (belonging to the unconnected porosity), the fluid pressure and the solvation
 316 pressure are fixed to zero resulting in a zero-total pore pressure. These pores also have no contribution
 317 on the fluxes related to the surrounding pores and particles.



318

319

Figure 3 2D representation of pore pressure distribution within the pores of the DEM-PFV

320

model: distinction between connected pores and unconnected pores

321 3 Model implementation

322 3.1 Scaling strategy

323 If the interparticle properties presented in Section 2.1 can be calibrated so that the emergent mechanical properties
 324 of the DEM model match those of the material to model (see (Scholtès and Donzé, 2013) for details of the
 325 procedure), the diffusion mechanisms at stake require some adjustments of the transport model. For instance, coal
 326 matrix has a porosity that is significantly smaller than the porosity of an assembly of spherical particles. Basically,
 327 the pore space of the model is scaled so that the effective volume of fluid is $V_{f,eff}$ equal to:

$$328 \quad V_{f,eff} = V_f \frac{n_m}{n_a} \quad (31)$$

329 with n_a the porosity of the spherical particle assembly, and n_m the porosity of the actual material.

330 Scaling the diffusive fluxes is actually sufficient to match the required porosity without changing the volume of
 331 both the pore space and the solid phase in the discretized equations, resulting in a total mass balance that accurately
 332 represents the material to model as follows:

333 For particles:
$$\bar{J}_{eff} = \bar{J} \frac{1-n_a}{1-n_m} \quad (32)$$

334 For cells:
$$\bar{J}_{eff} = \bar{J} \frac{n_a}{n_m} \quad (33)$$

335 with \bar{J} the non-scaled flux calculated according to the previous sections and \bar{J}_{eff} the effective (“scaled”) diffusive
336 flux.

337 In addition, we also introduced a procedure to adjust the poromechanical response of our DEM-PFV model since
338 the Biot coefficient of a DEM assembly is intrinsically equal to 1 given the rigid particle assumption of the DEM
339 formulation. As a matter of fact, according to the poroelasticity theory (Detournay and Cheng, 1993), the
340 contribution of the pore pressure p to the overall deformation of porous media is defined through the effective
341 stress such as

342
$$\sigma' = \sigma - bp \quad (34)$$

343 where σ' is the effective stress, σ is the total stress, and b is the Biot coefficient equal to $1 - \frac{K}{K_s}$ with K the bulk
344 modulus of the medium, and K_s the bulk modulus of the solid phase. This effective stress produces volumetric
345 strain ε_v which, for a swelling material, presents an additional term of adsorption induced strain ε_s as follows:

346
$$\varepsilon_v = -\frac{1}{K}(\sigma - bp_f) + \varepsilon_s \quad (35)$$

347 where $\sigma = \frac{\sigma_{kk}}{3}$ is the mean stress and ε_s is defined such as:

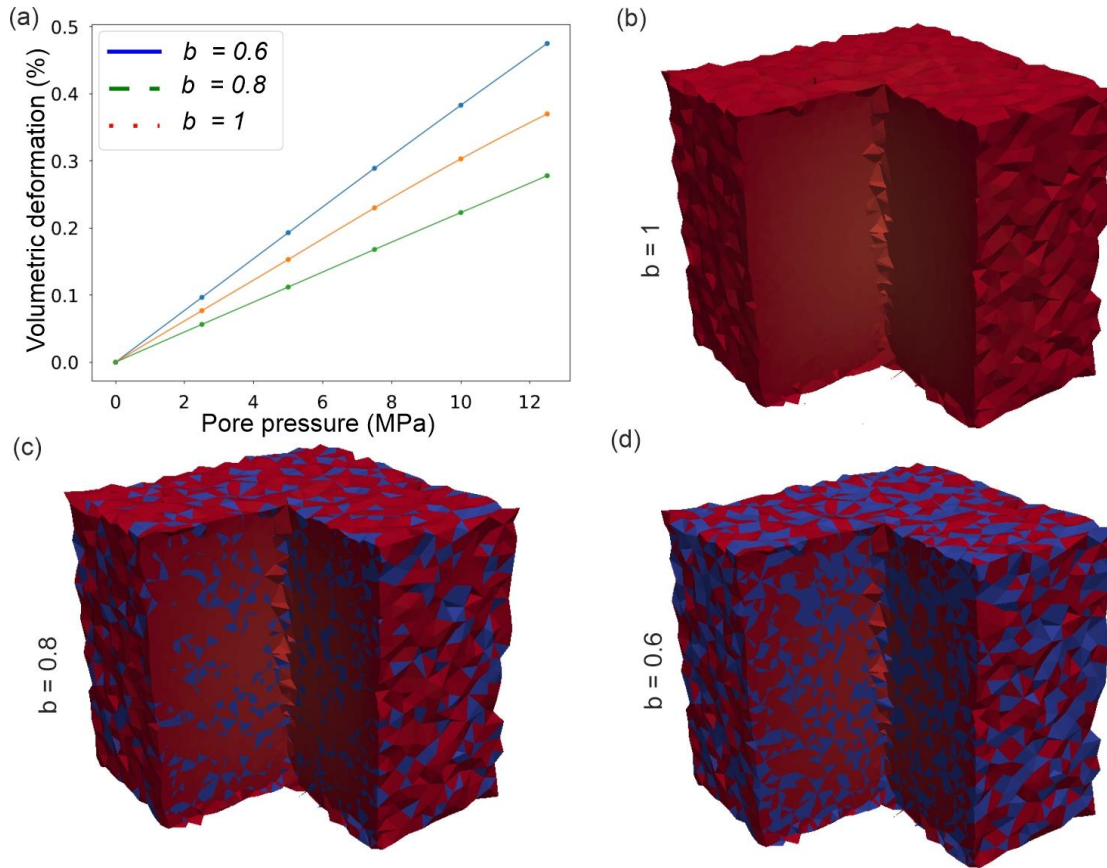
348
$$\varepsilon_s = b \frac{ps}{K} \quad (36)$$

349 Instead of applying a global scaling factor to Equation (27), we introduced the possibility to neutralize a certain
350 amount of pores within the pore network so that the connected porosity of the particle assembly $n_{a,c}$ is equal to:

351
$$n_{a,c} = bn_a \quad (37)$$

352 with b directly equal to the Biot coefficient of the material. The neutralized pores (unconnected porosity)
353 correspond to tetrahedra randomly chosen within the triangulated mesh where gas transport calculations are
354 simply not performed. These isolated pores thus participate to the overall deformability of the particle assembly

355 due to their compliance without contributing to its poromechanical response as illustrated in Figure 4.



356

357 Figure 4 Illustration of the connected porosity n_{ac} and of its effect on the Biot coefficient, Equation (37): a) Volumetric deformation
 358 as a function of pore pressure increase and b,c,d) pressure distribution at equilibrium ($p_{c\infty}=12.5$ MPa) in numerical samples with
 359 different pre-defined Biot coefficients (the connected pores are in red and the isolated pores in blue).

360 3.2 Numerical stability

361 Numerical stability needs to be considered in every explicit numerical scheme as any effect can only move by a
 362 maximum of one spatial grid block in one time step. For instance, in the general case of a 2D diffusion problem,
 363 the explicit discretization reads:

364
$$\frac{u_i^{t+\Delta t} - u_i^t}{\Delta t} = D \frac{u_{i+1}^t - 2u_i^t + u_{i-1}^t}{\Delta x^2} \quad (38)$$

365 where D is the diffusion coefficient. Solving Equation (38) for $u_i^{t+\Delta t}$ leads to:

366
$$u_i^{t+\Delta t} = ru_{i+1}^t + (1 - 2r)u_i^t + ru_{i-1}^t \quad (39)$$

367 with $r = D \frac{\Delta t}{\Delta x^2}$ the Fourier Number. Applying the Von Neumann stability analysis, the FTCS (Forward Time
 368 Centered Space) method is satisfied only if the following condition is fulfilled:

369
$$r \leq \frac{1}{2} \text{ then } \Delta t \leq \frac{\Delta x^2}{2D} \quad (40)$$

370 In our model, in addition to the mechanical time step that governs the solid particles motion, three-time steps exist
 371 due to the three different types of transport mechanisms occurring respectively between pore-pore, pore-particle
 372 and particle-particle. To ensure the stability of the numerical scheme, the simulation time step Δt is set up equal
 373 to a fraction of the minimum time step $\Delta t_{min} = \min\left(\Delta t^m, \Delta t^K, \Delta t^{des}, \Delta t^{sd}\right)$ required to ensure the stability of the
 374 numerical scheme (in all our simulations, the time step was defined equal to $0.8 \Delta t_{min}$).

375 **4 Verification of the transport models**

376 To verify the correct implementation of the transport equations in our DEM-PFV model, we compared its
 377 predictions to analytical solutions of Fick's law considering different 1D problems. For that purpose, we built up
 378 a pseudo 1D numerical assembly with dimensions equal to $L_0 \times 0.2L_0 \times 0.2L_0$ m, made up of 1,000 particles with
 379 radii varying between 7.1 and 13 mm, and 5,500 cells. In all the verification examples presented below, HM
 380 couplings were not considered, and the simulated medium was non deformable (the mechanical scheme was
 381 turned off and the particles fixed in space and time). In the present section and for the sake of comparison, all
 382 results and variables will be presented dimensionless. The x -coordinate, the Knudsen diffusion coefficient D^K and
 383 the concentration c are normalized respectively by the sample length L_0 , the gas diffusion coefficient D_0 and the
 384 inlet concentration c_0 . The dimensionless time is defined as $t = Dt_0 / L^2$ where t_0 is the physical time. Similar
 385 normalization holds for the sorbed concentration s and the surface diffusion coefficient D^s . The different test cases
 386 with the corresponding parameter values used in the simulations and analytical solutions are gathered in Table 1.

387

388

389 Table 1.

390 Numerical parameters of the different verification test cases.

Parameter	Test case 1	Test case 2	Test case 3
n	0.45	0.47	0.45
Dimensionless time step Δt	9e-7	9e-6	9e-7
D^K/D_0	1	-	1
D^S/D_0	-	1	-
θ	-	1.12	-
$K_d = k_{att}/k_{det}$	-	-	2
k_{att}	-	-	0.116
k_{det}	-	-	0.058

391 4.1 Test case 1: Knudsen diffusion

392 First, we consider diffusion of non-sorbing species within a nanoporous material. As a consequence, only Knudsen
393 diffusion occurs, and both sorption and surface diffusion may be neglected. The boundary value problem is thus
394 defined by the following initial and boundary conditions:

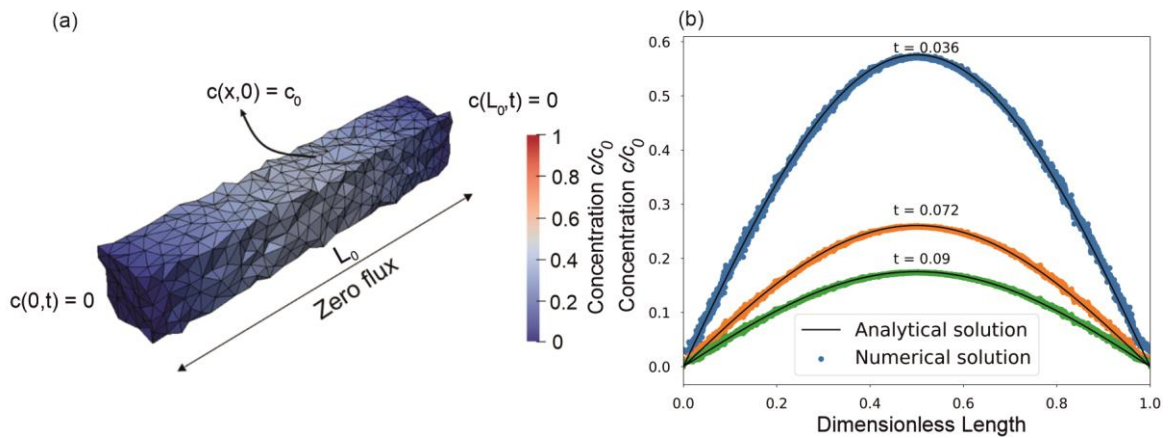
$$395 \quad \frac{\partial c}{\partial t} = D^K \frac{\delta^2 c}{\delta x^2}, \quad \forall x \in [0, L_0] \quad (41)$$

$$396 \quad \begin{aligned} c(x, 0) &= c_0, \quad \forall x \in [0, L_0] \\ c(0, t) &= c(L_0, t) = 0 \end{aligned}$$

397 Using Fourier series, the unsteady solution of this 1D diffusion problem can be written as follows:

$$398 \quad c(x, t) = \sum_{m=1}^{\infty} \frac{2}{\pi} \left(\frac{1 - \cos(m\pi)}{m} \right) \sin \left(m\pi \frac{x}{L_0} \right) e^{-\frac{D^K}{n_a L_0^2} (m\pi)^2 t_0} \quad (42)$$

399 The numerical assembly, illustrated in Figure 5a, was subjected to the initial and boundary conditions defined in
400 Equation (41) and the simulation was run with the parameters presented in Table 1.



401
402 Figure 5 a) Concentration distribution in pores at $t=0.04$. b) Numerical and analytical concentration profiles in pores at different times.

403 As shown in Figure 5b, the concentration distribution in the numerical model matches perfectly the analytical
 404 solution in space and time, confirming the accuracy of the numerical scheme for simulating Knudsen diffusion.

405 4.2 Test case 2: surface diffusion

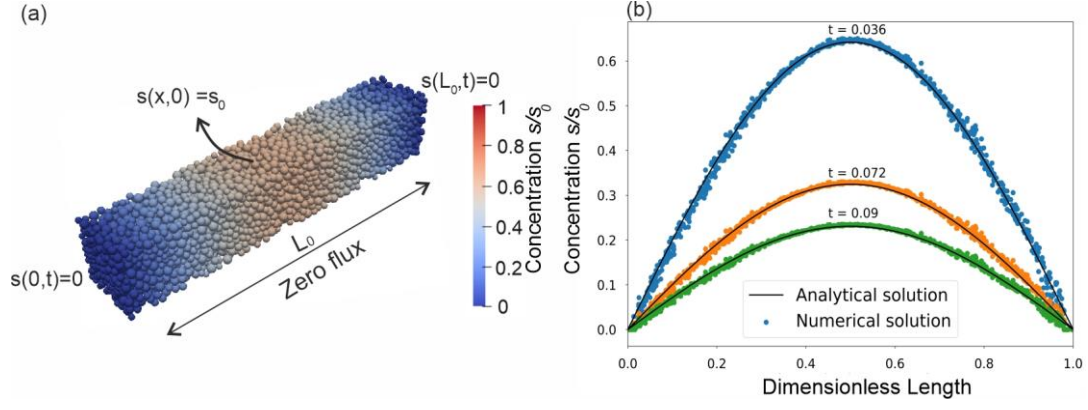
406 The second test case assumes that the gaseous species is now irreversibly adsorbed and may only diffuse through
 407 the solid grains. Surface diffusion is thus considered as the only effective transport mechanism and the problem
 408 is defined by the following initial and boundary conditions:

$$\begin{aligned}
 409 \quad \frac{\partial s}{\partial t} &= D^s \frac{\delta^2 s}{\delta x^2}, \quad \forall x \in [0, L_0] \\
 410 \quad s(x, 0) &= s_0, \quad \forall x \in [0, L_0] \\
 411 \quad s(0, t) &= s(L_0, t) = 0
 \end{aligned}
 \tag{43}$$

412 This 1D diffusion problem is very similar to the first one and the analytical solution in terms of Fourier series can
 413 be written as follows:

$$414 \quad s(x, t) = \sum_{m=1}^{\infty} \frac{2}{\pi} \left(\frac{1 - \cos(m\pi)}{m} \right) \sin \left(m\pi \frac{x}{L_0} \right) e^{-\left(\frac{D^s}{(1-n_a)L_0^2} \right) (m\pi)^2 t_0}
 \tag{44}$$

415 The numerical values used in the simulation are presented in Table 1. The parameter θ was initially calibrated
 416 since the number of contacts between particles depends on the packing characteristics (samples are generated
 417 using a random packing procedure, considering a pre-defined size distribution). Figure 6a displays an example of
 418 the concentration field in the solid grains at a given time of the simulation, while a comparison of the numerical
 419 solutions with averaged concentration profiles at different times is provided in Figure 6b. As for test case 1, the
 420 concentration distribution in the numerical model matches perfectly the analytical solution in space and time,
 421 showing the correctness and reliability of our surface diffusion model and of its implementation.



422

423 Figure 6 Concentration distribution in particles at $t = 0.04$. b) Comparison of numerical and analytical concentration profiles in particles at
424 different times.

425 4.3 Test case 3: Knudsen diffusion with adsorption/desorption

426 For this test case, we consider diffusion of a sorbing species within a non-swelling porous material. Here, we used
427 a linear isotherm to describe the solute adsorption onto the solid grains (see Section 2.2.1.2) and surface diffusion
428 is neglected (no transport within the solid phase). The system is initially at thermodynamic equilibrium so that
429 partitioning between gas and solid phases is respected. The adsorption and desorption kinetics coefficients
430 K_{att} and K_{det} are chosen high enough so that the equilibrium state is verified at each time step. Based on these
431 assumptions, the 1D boundary value problem is defined as follows:

$$432 \quad \frac{\partial c}{\partial t} = D^K \frac{\delta^2 c}{\delta x^2}, \quad \forall x \in [0, L_0]$$

$$433 \quad s(x, t) = K_d c(x, t), \quad \forall x \in [0, L_0] \quad (45)$$

$$434 \quad c(x, 0) = c_0; s(x, 0) = K_d c_0, \quad \forall x \in [0, L_0]$$

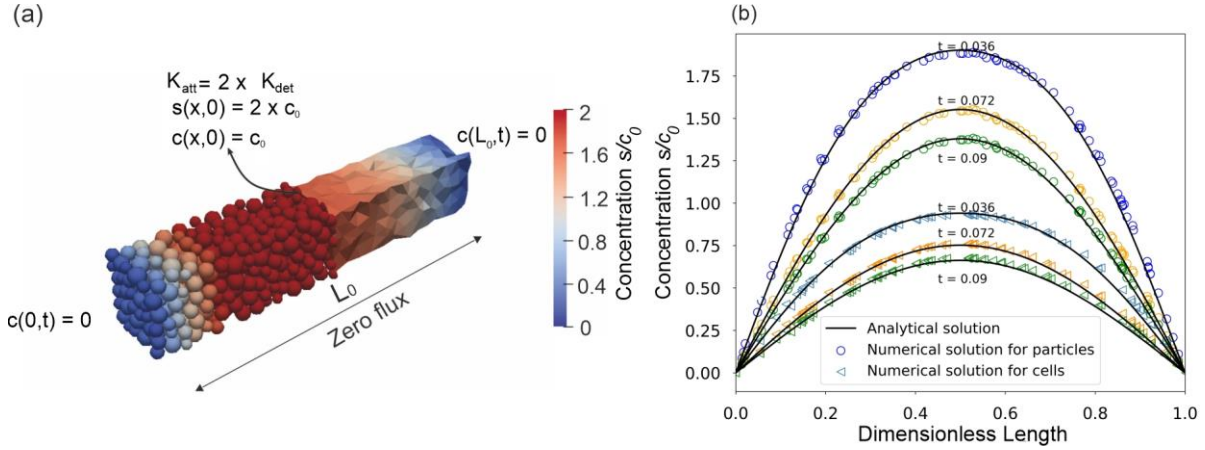
$$435 \quad c(0, t) = c(L_0, t) = 0$$

436 and the transient analytical solution is given by:

$$437 \quad c(x, t) = \sum_{m=1}^{\infty} \frac{2}{\pi} \left(\frac{1 - \cos(m\pi)}{m} \right) \sin \left(m\pi \frac{x}{L} \right) e^{-\left(\frac{D^K}{n_a(1+K_d)L_0^2} \right) (m\pi)^2 t_0} \quad (46)$$

438 The numerical parameters used for this test case are presented in Table 1. Figure 7a displays an example of the
439 concentration fields in both pores and grains obtained at a given time while a comparison of the numerical
440 solutions with a averaged concentration profiles at different times is provided in Figure 7b. Here a gain, a very good

441 agreement between the numerical and analytical solutions is obtained. We may also observe that our kinetics
 442 formulation converges well to the equilibrium since the partitioning between gas and solid grains, driven by $K_d =$
 443 2, is always respected. This comparison validates the coupling method between diffusion and sorption processes.



444

445 Figure 7. a) Concentration distribution in particles and pores simulated numerically at $t = 0.04$. b) Comparison of numerical and analytical
 446 concentration profiles in pores and particles at different times.

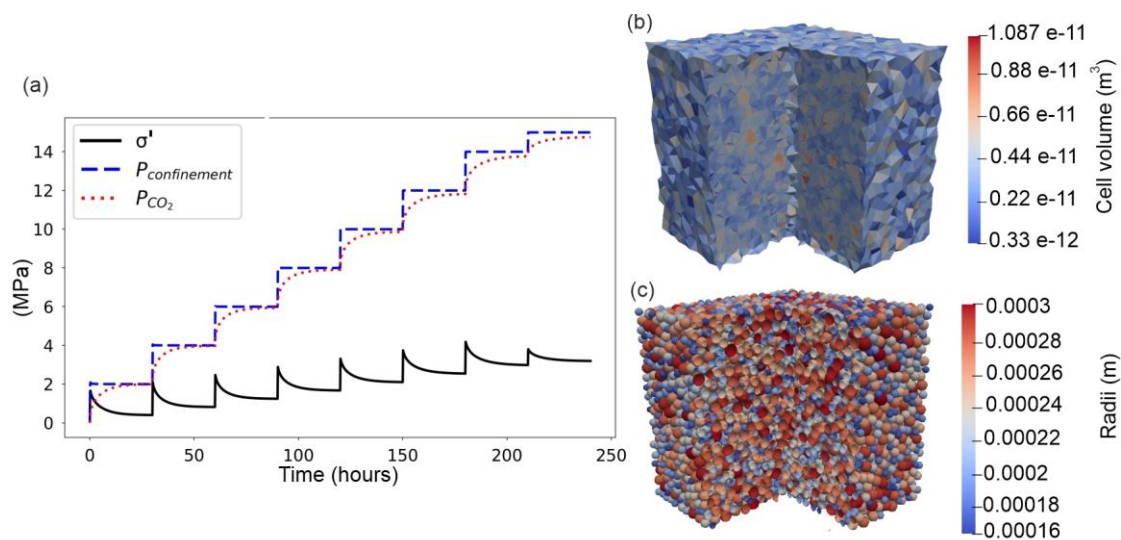
447 5 Hydro-mechanical behavior: comparison with a swelling experiment

448 To verify the implementation and validity of our HM scheme (Section 2.3), we compared the predictions of our
 449 model to the experimental results of Day et al. (Day et al., 2008a) who performed a swelling experiment on a 30
 450 x 10 x 10 mm block of coal under confined conditions. The CO₂-induced coal swelling was measured for different
 451 values of CO₂ pressure by optical method. The experiment performed at 55°C on the Australian bituminous coal
 452 from the Bowen Basin (referred to as sample 3 in Day et al. (Day et al., 2008a) and as Qld8 in Day et al. (Day et
 453 al., 2008b)) was used as reference. Note that the same data set (but at 40°C) was also used by (Sampath et al.,
 454 2020) for model validation purpose but some discrepancies exist between our two investigations. Indeed, due to
 455 the lack of experimental data available, a part of their numerical properties were obtained from another coal
 456 experiment and they discarded the highest pressures of swelling experiment (higher than 12MPa) for comparison.
 457 In the present analysis, we did our best to carry out a fair comparison by using the full range of experimental data
 458 available and a complete set of consistent physical properties for this coal. All the parameter values are gathered
 459 in Table 2. Because the elastic properties and Biot coefficient were not provided either in (Day et al., 2008b,
 460 2008a), we set the value of b based on the value used by Sampath et al. (Sampath et al., 2020). The Young modulus
 461 E and the Poisson ratio ν were chosen in order to reproduce the behavior observed by Day et al. (Day et al., 2008a)
 462 with a non-sorbing gas (a volumetric contraction of 0.06% was reached at 15MPa of helium pressure). The s_{max}

463 value was inferred from the maximum sorption capacity of the coal W_0 in the isotherm model calculated by Day
 464 et al. (Day et al., 2008b) (68.4 kg of CO₂ per ton of rock for the sample Qld 8). Finally, values of D^K and D^S were
 465 taken from the literature (Dong et al., 2017) whereas K_{att} , K_{det} were fixed arbitrarily. D^S is fixed constant but D^K
 466 varies as a function of the pore radius, as expressed in Equation (10), around an average value found in the
 467 literature (see Table 2). Note that the values chosen for these transport and kinetic parameters do not affect the
 468 steady-state behavior and hence, the comparison we made which is based on equilibrium states.

469 5.1 Sample preparation

470 A 10 x 10 x 10 mm particle assembly made up of 10,000 particles was generated and the interparticle properties
 471 calibrated following the approach proposed by Scholtès and Donzé (Scholtès and Donzé, 2013). The coal sample
 472 is considered isotropic although, even for the matrix, this assumption may be questioned (Day et al., 2008a). The
 473 calibration was done by performing uniaxial compression tests on the numerical sample following a trial and
 474 errors approach to determine the adequate interparticle properties to match both Young modulus and Poisson ratio
 475 of the coal. In addition, both the porosity and Biot coefficient of the numerical sample were defined to match
 476 those of the tested coal by following the procedures presented in Section 3.1.



477

478 Figure 8 Numerical set up for simulating the swelling experiment of Day et al. (2008): a) boundary pressures and resulting mean effective
 479 stress, b) pore size distribution, c) particles size distribution.

480 Table 2

481 Numerical parameters used to simulate the coal matrix experiment by Day et al. [57]. The star symbol denotes
 482 the calibration parameters.

Parameters	DEM-PFV model	Sources
Young modulus E (GPa) *	4.550	<i>computed from Day et al., 2008 (Day et al., 2008a) (sample 3)</i>
Poisson ratio ν (-)*	0.225	<i>computed from Day et al., 2008 (Day et al., 2008a) (sample 3)</i>
Temperature T (K)	328	<i>Day et al., 2008 (Day et al., 2008a) (sample 3)</i>
Gas constant R (J.mol ⁻¹ .K ⁻¹)	8.314	N/A
Molar mass M_g (g.mol ⁻¹)	44.01	N/A
Bulk density (kg.m ⁻³)	1220	<i>Day et al., 2008 (Day et al., 2008b) (sample Qld 8, Table 3)</i>
Skeletal density (kg.m ⁻³)	1303	<i>Day et al., 2008 (Day et al., 2008b) (sample Qld 8, Table 3)</i>
s_{max} (mol.m ⁻³)	2025	<i>Day et al., 2008 (Day et al., 2008b) (sample Qld 8, Table 2)</i>
K_d (mol.m ⁻³) *	0.0017	-
K_{att} (s ⁻¹)	17e-11	-
K_{det} (mol.m ⁻³ .s ⁻¹)	1e-7	-
D^K (×10 ⁻¹² m ² .s ⁻¹)	3	<i>Dong et al., 2017 (Dong et al., 2017) (Table 4)</i>
D^S (×10 ⁻¹² m ² .s ⁻¹)	30	<i>Dong et al., 2017 (Dong et al., 2017) (Table 4)</i>
Biot coefficient b (-)	0.8	<i>Sampath et al., 2020 (Sampath et al., 2020) (Table 2)</i>
Coupling coefficient α (-) *	20.2	-
Porosity of assembly n_a (%)	32	N/A
Porosity of coal sample n_m (%)	7.1	<i>Day et al., 2008 (Day et al., 2008b) (sample Qld 8, Table 3)</i>

483 5.2 Experimental procedure

484 Day et al. (Day et al., 2008a) measured the dimensional changes of a sample contained within a pressure cell
 485 where CO₂ pressure was increased up to 15 MPa. In our experiment, the numerical sample is confined in between
 486 6 frictionless boundary walls to control the confining pressure $p_{confinement}$. To reproduce the swelling experiment
 487 of Day et al. (Day et al., 2008a), we imposed CO₂ pressure both as boundary stress ($p_{confinement}$) and as boundary
 488 pressure (p_{CO_2}) on all the sample boundaries. $p_{confinement}$ is adjusted by displacing the boundary walls. During
 489 the simulation, p_{CO_2} and $p_{confinement}$ were increased stepwise from equilibrium states and the volumetric
 490 deformation ϵ_v , solvation pressure (p_s), adsorbed amount of gas (s) and fluid pressure (p_f) were recorded at each
 491 step (Figure 8a).

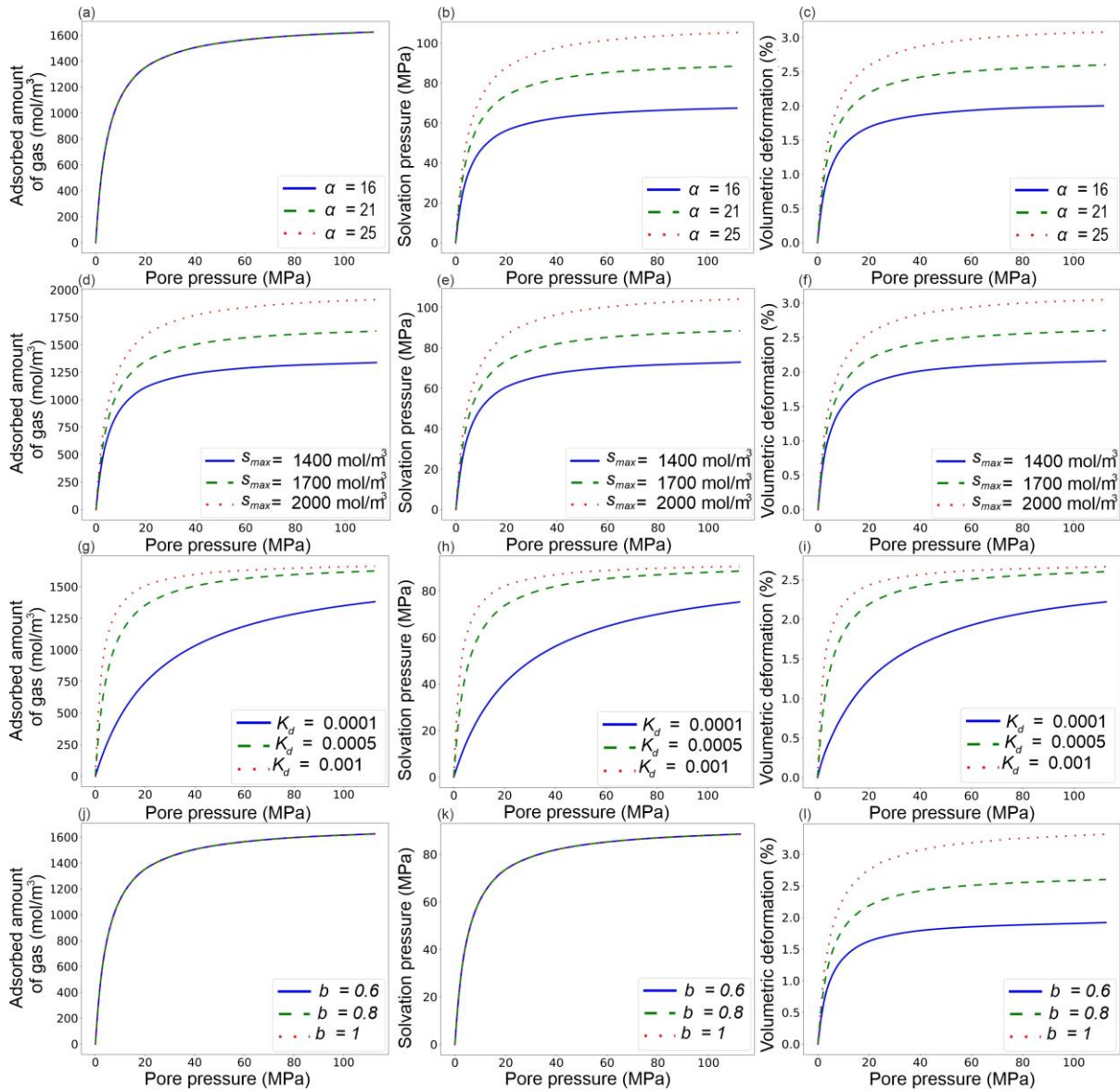
492 Since the Biot coefficient is lower than one, the effective stress varies at each CO₂ injection pressure step. Day et
 493 al. (Day et al., 2008a) however suggest that the mechanical compression caused by the pore pressure increase can

494 be neglected (0.06% of contraction for non-sorbing gas against 2% of swelling deformation with CO₂ at 15Mpa).
495 In other words, Day et al. investigated throughout this experiment the influence of sorption-induced swelling on
496 the poromechanical behavior of the material without taking into consideration the effect of the fluid bulk pressure.

497 5.3 Parametric study

498 Before directly comparing the model results with the experimental observations, a parametric study is presented
499 to highlight the influence of the model parameters on the emergent behavior. The results are summarized in Figure
500 9 and the parameters roles assessed as follows:

- 501 • The coupling term α controls the intensity of the solvation pressure p_s (Equation (30), Figure 9b), hence
502 the amplitude of the associated deformation (Equation (35), Figure 9c). Thus, α needs to be adjusted as
503 a function of the swelling potential of the material with respect to the nature of the adsorbed fluid as, for
504 example, CO₂ induces larger swelling than CH₄ in coal (Duruca et al., 2009). p_s is directly proportional
505 to α (Figure 9b), giving the opportunity to set its value so as to match experimental evidence of swelling
506 for each combination of material and adsorbed fluid. One has to note that α does not influence the amount
507 of molecules that can be adsorbed on the solid phase (Figure 9a).
- 508 • According to Equations (15) and (16), s_{max} determines the maximum amount of gas molecules that can
509 be adsorbed on the surfaces of the solid phase. Depending on the nature of the fluid-solid interaction at
510 play, s_{max} must be adjusted because it also affects the solvation pressure and thus the volumetric
511 deformation (Figure 9d-e-f). In contrast, K_d controls the adsorption/desorption kinetics (Figure 9g-h-i)
512 without influencing the maximum amount of adsorbed gas molecules, hence leading to the same plateau
513 value for the adsorbed amount of gas molecules, solvation pressure and volumetric deformation.
- 514 • Finally, the Biot coefficient b does not influence neither the amount of adsorbed gas molecules nor the
515 solvation pressure (Figure 9j-k) while it directly influences the volumetric deformation (Figure 9l), as
516 described by the poromechanical equation (Equation (35)).



517

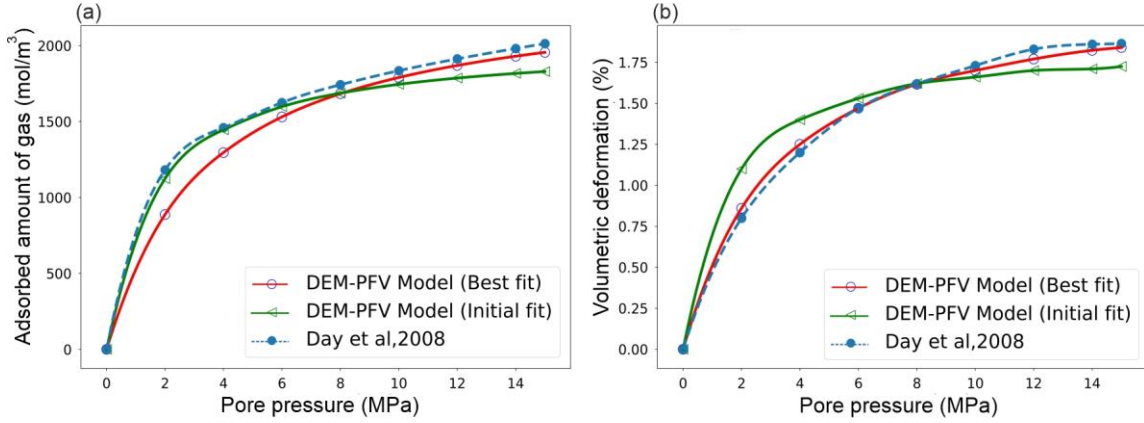
518

Figure 9 Evolution of adsorbed amount of gas s (left), solvation pressure p_s (middle) and volumetric deformation ε_v (right) versus pore pressure p_f for: (a,b,c) different α values and fixed parameters ($s_{max}=1700, K_d=0.0005, b=0.8$), (d,e,f) different s_{max} values and fixed parameters ($\alpha=21, K_d=0.0005, b=0.8$), (g,h,i) different K_d values and fixed parameters ($\alpha=21, s_{max}=1700, b=0.8$), (j,k,l) different b values

520

and fixed parameters ($\alpha=21, K_d=0.0005, s_{max}=1700$).

521



523

524 Figure 10 Swelling experiment: comparison of the DEM-PFV model predictions to the experiment of Day et al. (Day et al., 2008a):

525 evolutions of a) adsorbed amount of gas and, b) volumetric deformation as functions of the pore pressure.

526 The calibration of the model was done through the following procedure:

- 527 1. Determine the interparticle elastic properties (E_{eq} and P) through uniaxial or triaxial compression testing
- 528 (trial and error adjustments) to match both Young modulus E and Poisson ratio ν of the material.
- 529 2. Set b equal to the measured value and scale the numerical sample connected porosity $n_{a,c}$.
- 530 3. Determine the experimental solvation pressure p_s by using the extended poromechanical equations,
- 531 (Equations (35) and (36)) such that

$$532 \quad p_s = \frac{K \varepsilon_v + (\sigma - b p_f)}{b} \quad (47)$$

533 where $K = \frac{E}{3(1-2\nu)}$ is the bulk modulus and $\sigma = \frac{\sigma_{kk}}{3}$ is the mean stress and, from the value of s_{max} ,

534 estimate α using the plateau value of p_s .

- 535 4. Finally, calibrate K_d (trial and error adjustments) so that the numerical results fit with the experiments in
- 536 terms of solvation pressure or swelling deformation.

537 The numerical results presented in Figure 10 (referred to as the initial fit) match the experimental observations

538 for the following set of parameters: $\alpha = 20.02$, $K_d = 0.0017 \text{ mol.m}^{-3}$ with the s_{max} value inferred from the

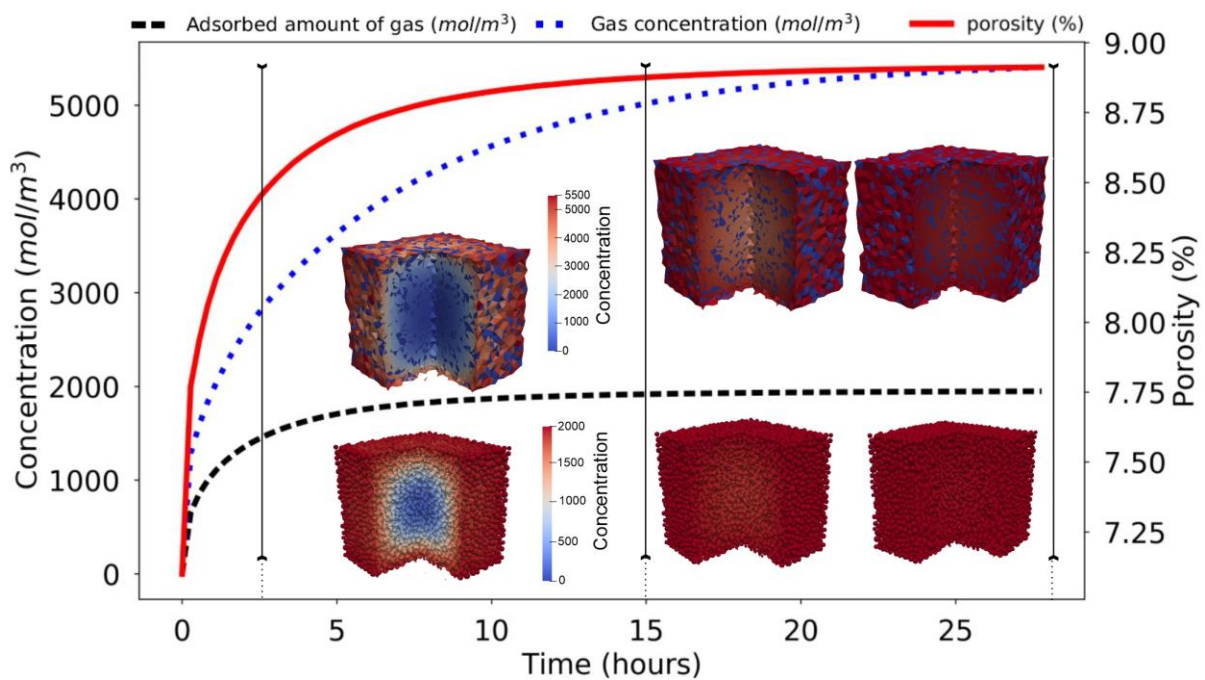
539 experiment ($s_{max} = 2025 \text{ mol.m}^{-3}$). The fit to experimental data point is not ideal yet. In their study, Day et al.

540 (Day et al., 2008b) have explored a pore filling isotherm model (Dubinin–Radushkevich model) which has been

541 found to provide a better fit to experimental adsorption data than the classical Langmuir model, especially at high

542 pressures, above 6 MPa. A comparison between this isotherm model and ours is shown in Figure 10a and confirms
 543 this observation. This discrepancy, inherent to the use of Langmuir isotherm model, is also observed when
 544 comparing volumetric deformations in Figure 10b (an average error of 19% is found). Note that a better fit can be
 545 achieved when fitting also the s_{max} value. A second simulation, referred to as the best fit, where $s_{max} = 2400$
 546 $\text{mol}\cdot\text{m}^{-3}$, $K_d = 0.0008 \text{ mol}\cdot\text{m}^{-3}$ and $\alpha = 20.02$ are shown in Figure 10a and Figure 10b. The error is thus reduced
 547 to 2%.

548 Additional insights can be gained from numerical simulations into the dynamics of mass transfer processes within
 549 the coal material. Figure 11 shows the changes in averaged gas and sorbed concentrations with time in the porous
 550 sample between two successive constant pressure steps. The corresponding porosity variation is also exhibited.
 551 As an illustrative example, we focused on the transient behavior corresponding to an increase from 0 to 15 MPa
 552 of CO_2 pressure. Results indicate that the gas diffuses first through the solid phase. As expected from diffusion
 553 values of Table 2 and accordingly to the literature (Dong et al., 2017; Mathias et al., 2020), surface diffusion is
 554 the predominant transport mechanism within coal matrix. This increase in sorbed concentration within the coal
 555 sample results in swelling and hence, volumetric strain and porosity increase. Owing to the values used for K_{att}
 556 and K_{det} , the desorption rate is not large enough and equilibrium is reached faster in the solid phase than in the gas
 557 phase (Figure 11). As a consequence, swelling occurs before the pore pressure reaches its equilibrium state in
 558 agreement with Sampath et al. results (Sampath et al., 2020).



559

560

Figure 11. Evolution of porosity, fluid concentration and adsorbed amount of gas with time.

561 **6 Conclusion**

562 A new 3D pore-scale model for porous swelling materials is proposed to investigate coal bed methane production.
563 In our model, a DEM model where the material is represented as an assembly of particles interacting one with
564 another according to predefined contact laws is coupled to a PNM approach for describing transport mechanisms
565 between and within pores and solid grains. The material is assumed to be saturated with gas and various
566 mechanisms for gas transport across the coal matrix have been considered including Knudsen diffusion, surface
567 diffusion and sorption. Mechanical couplings inherent to such swelling materials are also taken into account. The
568 model simulates both (i) the interplay between pore pressure and external stress with variations in the effective
569 stress which may impact the mechanical behavior of the medium and (ii) the sorption processes which contribute
570 to swell or shrink the material. This latter mechanism is considered through the addition of an additional pressure
571 term, classically called the solvation pressure, related to the sorbed concentration.

572 The implementation of the model was tested against analytical solutions and compared to a swelling experiment.
573 The model was found to be robust and a suitable tool for describing adsorption induced deformation. This
574 comprehensive investigation reveals the complex physics at stake during methane adsorption and the numerical
575 results give precious insights onto the internal dynamics of gas within the coal matrix. In particular, simulations
576 reveal that coal matrix swelling occurs before the pore pressure reaches its equilibrium state since surface diffusion
577 prevails. The present model is thus capable to describe the strong coupling between transport, chemical (sorption)
578 and mechanical processes and can be used to predict methane recovery. Future work will focus on introducing
579 cleat network to assess the effect of cleat intensity and facilitate more comprehensive analysis on the permeability
580 changes at the coal rock mass scale.

581

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