

# 1                    **On modelling of consolidation processes in geological materials**

2

## 3    **Abstract**

4    Low-permeability materials may be seen as natural geological barriers for radioactive waste  
5    repositories. However, to ensure their safe performance, a good understanding of their  
6    mechanical properties is required. Although the standard Biot's poroelastic model is widely  
7    used to estimate the key properties of these materials, experimental observations differ from  
8    this mathematical formulation and suggest that a more complex rock deformation behaviour to  
9    include a creep effect is needed. In this study, the Biot's differential equations are modified to  
10   include a rheological skeleton. In comparison with other existing models, here we propose a  
11   formulation with a minimal parametric uncertainty: we show that with just one additional  
12   physically-based parameter, the experimental creep behaviour is properly described. This  
13   enhanced model is implemented within a finite element framework and employed in a fitting  
14   algorithm to extract the hydro-mechanical properties from experimental data. To illustrate its  
15   generality, we analyse laboratory tests performed on three different types of materials: (a) an  
16   unlithified lower Oligocene clay from Belgium (Boom Clay), (b) an indurated Jurassic  
17   mudrock (Callovo-Oxfordian mudstone) and (c) a Triassic siltstone (Mercia Mudstone  
18   Formation). Numerical fits to the data support the validity of this approach and demonstrate its  
19   applicability to a range of low-permeability materials regardless of mineralogy or burial  
20   history.

21

22    *Keywords: consolidation tests, Biot's model, elasticity, viscosity, creep, porous medium*

## 23 **1. Introduction**

24 Diagenetic processes occurring during burial will have a profound effect on the hydro-  
25 mechanical (HM) behaviour of mudrocks (Horseman and Harrington, 1996). However, the  
26 properties of a mudrock are not solely governed by diagenesis alone and a number of processes  
27 occurring before, during and after can play an important role in defining the structural  
28 characteristics of such materials. Most important of these is the role of stress history, which  
29 can be affected as a direct result of both tectonic and erosional forces combining to produce  
30 deformation, uplift and exhumation. The importance of these processes and their impact on the  
31 HM behaviour of mudrocks can be profound (Bjerrum, 1967; Skempton, 1970 and Novello,  
32 1987). In a geological repository for radioactive waste, the ability to predict long-term changes  
33 in rock properties over protracted periods of time is a central requirement in the development  
34 of any safety case. In many geological disposal concepts, clay-based formations are considered  
35 favourable options for the hosting of such underground repositories. Thus, understanding  
36 changes in HM behaviour as a repository undergoes either burial or exhumation is fundamental  
37 to the long-term prediction of both natural and engineered barriers. Central to this  
38 understanding is an ability to quantitatively model these processes in order to test material  
39 sensitivities, validate repository concepts and allow scenario analyses to be undertaken. With  
40 this in mind, laboratory experiments measuring the consolidation (loading) and rebound  
41 (unloading) of rock samples are undertaken to provide essential data with which to test and  
42 validate HM models.

43 Experiments on sediments and sedimentary rocks have shown that additional volume strain can  
44 accumulate, even after the sediment is fully consolidated to the applied stress (Atkinson and  
45 Bransby, 1978). Bishop and Lovebury (1969) demonstrated that remoulded London clay still  
46 showed creep three years after primary consolidation was complete. The mechanisms of  
47 secondary consolidation possibly include: (i) grain surface diffusion, (ii) time-dependent crack

48 generation associated with a redistribution of stored strain energy and (iii) diffusion in  
49 microfractures, with stress corrosion weakening the fracture tips. Thus, this creep behaviour  
50 should be considered in the mathematical formulation designed to extract the hydro-mechanical  
51 properties from experimental data.

52 The analysis of the consolidation of soil media was first addressed in a one-dimensional setting  
53 by Terzaghi (1925) and was later generalized by Biot (1941). Since these first contributions,  
54 where soil was described as an ideal linear elastic material, significant progress has been made  
55 to account for more realistic deformation behaviours. In these enhanced models, the standard  
56 Biot's consolidation theory is usually modified to account for, among others, viscoelastic,  
57 elasto-plastic, elasto-viscoplastic or damage soil skeletons. As contributions in this direction,  
58 and without attempting to be complete, we refer to the models proposed by Oka et al. (1986),  
59 Bardet (1992), Manoharan and Dasgupta (1995), Fowler and Noon (1999), Hamiel et al. (2004)  
60 and references therein.

61 These extended mathematical models led to a more appropriate characterisation of the  
62 consolidation of porous media. Nevertheless, their main disadvantage arises from the  
63 requirement of additional parameters for both the solid skeleton and the fluid. Determination  
64 from experimental data in low and ultra-low permeability materials can be challenging or even  
65 unfeasible and hence, simple models such as the standard Biot's consolidation theory are still  
66 preferred when characterising materials for real-life applications.

67 In this paper, a viscoelastic model with a minimal parametric uncertainty is proposed. In this  
68 contribution, the standard Biot's poroelastic model (Section 3) is modified to include the creep  
69 effect observed in experimental tests (Section 4). In contrast to some other techniques, only  
70 one additional parameter with respect to the classical Biot's model is needed. The simplicity  
71 of this approach and the clear physical meaning of the three parameters involved is used here  
72 to derive an algorithm for parameter identification, which successfully performs with

73 experimental data obtained from consolidation experiments conducted on different kinds of  
74 clay-based materials (Section 5).

75

## 76 **2. Experimental set-up and test methodology**

77 Testing was undertaken using a BGS custom-designed isotropic permeameter consisting of five  
78 main components: (1) a specimen assembly, (2) a 70 MPa rated pressure vessel and associated  
79 confining pressure system, (3) a fluid injection system, (4) a backpressure system, and (5) a  
80 National Instruments data acquisition system. Each specimen was sandwiched between two  
81 stainless steel end-caps and jacketed in heat-shrink Teflon to exclude confining fluid and  
82 provide a flexible pressure seal. A unique ‘lock-ring’ arrangement (Figure 1) was then placed  
83 over the jacketed specimen, so as to provide a leak-tight seal. The inlet and outlet zones for  
84 permeant flow through the specimen were provided by porous filter discs mounted between  
85 the sample and the load bearing surface of the end-caps. Once complete the sample assembly  
86 was then inserted into the pressure vessel and an isotropic stress applied using water.

87 Volumetric flow rates were controlled or monitored using a pair of syringe pumps operated  
88 from a single digital control unit. Each pump can operate in either a constant pressure or  
89 constant flow mode. A programme written in LabVIEW™ elicited data from each pump at  
90 pre-set time intervals. Testing was performed in an air-conditioned laboratory at a nominal  
91 temperature of 20 °C. All pressure sensors were calibrated against laboratory standards.

92 Analysis in this paper relates to data from experiments conducted on three different clay-based  
93 natural geological barrier materials: (i) Boom Clay, (ii) Callovo-Oxfordian claystone and (iii)  
94 Mercia Mudstone/microsparstone. Further details relating to these samples are given in the  
95 following sections, but a summary of their geotechnical properties is also given in Table 1. To  
96 minimise possible osmotic swelling of samples, a synthetic porewater solution was prepared  
97 for use as the backpressure fluid and as the permeant during all hydraulic testing. In the absence

98 of pore-fluid composition data for the Mercia mudstone, a salt-saturated solution was made  
 99 using crushed halite from close in the succession to the sampling location (Harrington et al.,  
 100 2018). Each test consisted of a hydration phase, an initial hydraulic test and a consolidation  
 101 phase.

102



Figure 1. A sample of Mercia Mudstone after preparation (left), arranged within the isotropic test assembly (centre) and as a 2D x-ray image (right).

103

104 Table 1. Geotechnical properties of test samples used to provide experimental data for model  
 105 validation. In the absence of test data, values marked # are based on average values quoted by  
 106 Harrington et al. (2017).

Sample	Length [mm]	Diameter [mm]	Bulk density [Mg.m <sup>-3</sup> ]	Dry density [Mg.m <sup>-3</sup> ]	Void ratio [-]
Boom Clay	42.67	49.92	2.05	1.68	0.60
Callovo-Oxfordian	48.38	50.18	2.45 <sup>#</sup>	2.32 <sup>#</sup>	0.17 <sup>#</sup>
Mercia Mudstone	48.76	54.42	2.32	2.10	0.30

107

108

### 109 3. Numerical model: linear elastic skeletal deformation

#### 110 3.1 Governing equations

111 The fluid flow through a compressible porous medium may be described by Biot's model, see  
112 Biot (1941). In this model, the governing equation for flow is obtained by combining Darcy's  
113 law with the mass conservation equation, thus leading to

$$114 \quad \nabla \cdot \left( \frac{k}{\mu} \nabla p \right) = \phi \beta \frac{\partial p}{\partial t} + \frac{\partial}{\partial t} (\nabla \cdot \mathbf{u}) \quad (1)$$

115 where  $\mathbf{u}$  is the solid displacement [m],  $p$  is the fluid pressure [Pa],  $k$  is the intrinsic permeability  
116 [m<sup>2</sup>],  $\mu$  is the dynamic viscosity of the fluid [Pa·s],  $\phi$  is the porosity [-] and  $\beta$  is the  
117 compressibility of the fluid [Pa<sup>-1</sup>]. Note that an isotropic permeability, represented by the scalar  
118  $k$ , is here assumed.

119 The classical Biot's model assumes an elastic deformation of the matrix. Thus, Equation (1) is  
120 coupled to the mechanical equilibrium equation

$$121 \quad \nabla \cdot \boldsymbol{\tau} + \mathbf{f} = \mathbf{0} \quad (2)$$

122 where  $\mathbf{f}$  is the body force per unit volume of the medium [N/m<sup>2</sup>] and  $\boldsymbol{\tau}$  is the total stress on the  
123 medium [Pa], which can be expressed as

$$124 \quad \boldsymbol{\tau} = \boldsymbol{\sigma} - \alpha p \mathbf{I} \quad (3)$$

125 where  $\boldsymbol{\sigma}$  is the effective stress tensor [Pa],  $\alpha$  is the Biot's coefficient [-] and  $\mathbf{I}$  is the identity  
126 tensor. Under the assumption of small strains and assuming an isotropic linear elastic material,  
127 the effective stress tensor takes the form

$$128 \quad \boldsymbol{\sigma} = \lambda \text{tr}(\boldsymbol{\varepsilon}) \mathbf{I} + 2G \boldsymbol{\varepsilon} \quad (4)$$

129 where  $\lambda$  is the first Lamé's constant [Pa],  $G$  is the shear modulus [Pa] and  $\text{tr}$  stands for the  
130 trace operator. For the sake of simplicity,  $\alpha = 1$  is here assumed. This is a reasonable  
131 assumption for a saturated porous medium and leads to the equation

$$132 \quad \frac{E}{2(1+\nu)(1-2\nu)} \nabla(\nabla \cdot \mathbf{u}) + \frac{E}{2(1+\nu)} \nabla^2 \mathbf{u} - \nabla p = -\mathbf{f} \quad (5)$$

133 where  $E$  is the Young's modulus [Pa] and  $\nu$  is the Poisson's coefficient [-]. Note that the  
 134 relationships  $\boldsymbol{\varepsilon} = \nabla^s \mathbf{u}$ ,  $\lambda = \frac{Ev}{(1+\nu)(1-2\nu)}$  and  $G = \frac{E}{2(1+\nu)}$  have here been used.

135

### 136 3.2 Finite element formulation

137 The numerical solution of Biot's model is usually approached using the Galerkin finite element  
 138 method. Thus, Equations (1) and (5) are first cast in a weak form to be subsequently linearised.

139 Following standard procedures, the solid displacements and the fluid pressure at time  $t$  can be  
 140 expressed as

$$141 \quad \mathbf{u}(\mathbf{x}) \cong \mathbf{u}^h(\mathbf{x}) = \mathbf{N}(\mathbf{x})\mathbf{u} \quad (6.1)$$

$$142 \quad p(\mathbf{x}) \cong \mathbf{p}^h(\mathbf{x}) = \mathbf{N}(\mathbf{x})\mathbf{p} \quad (6.2)$$

143 where  $\mathbf{N}$  is the matrix of standard finite element shape functions,  $\mathbf{u}$  is the standard nodal  
 144 displacement vector and  $\mathbf{p}$  is the standard fluid pressure vector. These approximations lead to  
 145 the coupled-system of discretized equations

$$146 \quad \mathbf{L}^T \frac{d\mathbf{u}}{dt} + \mathbf{H}\mathbf{p} + \mathbf{S} \frac{d\mathbf{p}}{dt} = -\mathbf{F}_p \quad (7.1)$$

$$147 \quad \mathbf{K}\mathbf{u}^{n+1} - \mathbf{L}\mathbf{p}^{n+1} = \mathbf{F}_u \quad (7.2)$$

148 which upon application of the backward Euler finite difference time scheme becomes

$$149 \quad \begin{bmatrix} \mathbf{K} & -\mathbf{L} \\ \mathbf{L}^T & \Delta t\mathbf{H}+\mathbf{S} \end{bmatrix} \cdot \begin{bmatrix} \mathbf{u}^{n+1} \\ \mathbf{p}^{n+1} \end{bmatrix} = \begin{bmatrix} \mathbf{F}_u \\ \mathbf{L}^T\mathbf{u}^n + \mathbf{S}\mathbf{p}^n - \Delta t\mathbf{F}_p \end{bmatrix} \quad (8)$$

150 In order to obtain a symmetric system, here the sign convention normally adopted for the  
 151 variable  $\mathbf{p}$  is reversed. Hence, for the coupled problem of flow through a deformable medium,  
 152  $\mathbf{p}$  is negative for compressive pressure whereas it is positive for tensile pressure thus leading  
 153 to the system

$$154 \quad \begin{bmatrix} \mathbf{K} & \mathbf{L} \\ \mathbf{L}^T & -\Delta t\mathbf{H}-\mathbf{S} \end{bmatrix} \cdot \begin{bmatrix} \mathbf{u}^{n+1} \\ \mathbf{p}^{n+1} \end{bmatrix} = \begin{bmatrix} \mathbf{F}_u \\ \mathbf{L}^T\mathbf{u}^n - \mathbf{S}\mathbf{p}^n + \Delta t\mathbf{F}_p \end{bmatrix} \quad (9)$$

155 with the matrices defined in Table 2.

156 *Table 2. Block matrices of the discretized Biot's system of equations.*

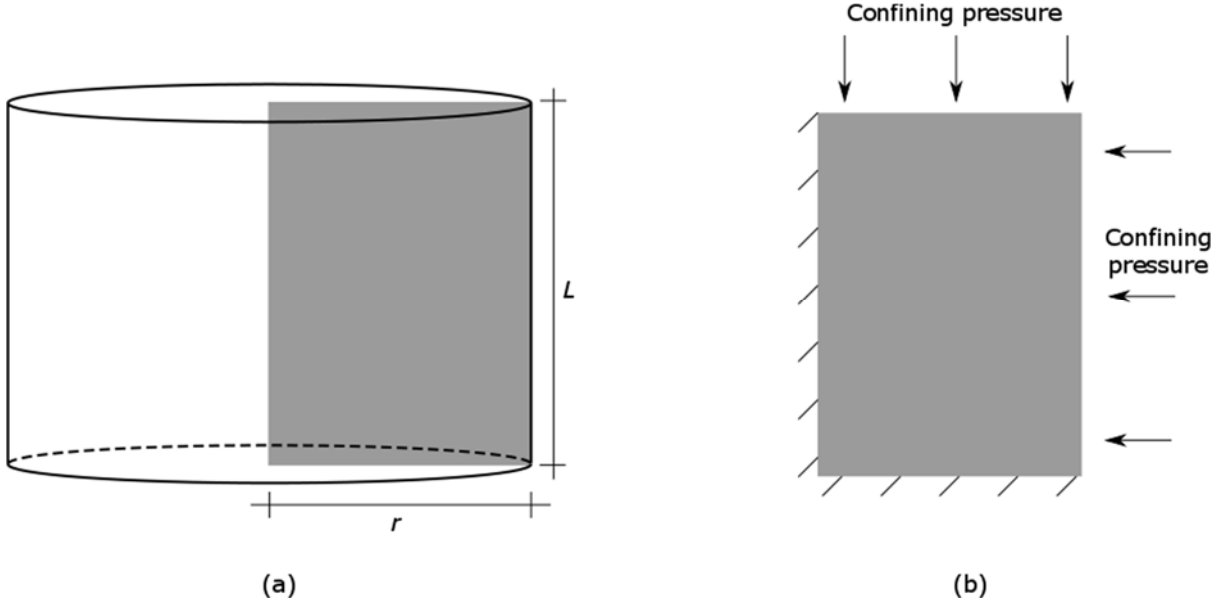
Matrix	Symbol	Expression
Flux matrix	$\mathbf{F}_p$	$\mathbf{F}_p = \int_{\Gamma_p} \mathbf{N}^T \mathbf{q} \, d\Omega$ , where $\mathbf{q}$ is the flux vector prescribed on the boundary $\Gamma_p$ .
Load matrix	$\mathbf{F}_u$	$\mathbf{F}_u = \int_{\Gamma_u} \mathbf{N}^T \mathbf{t} \, d\Omega$ , where $\mathbf{t}$ is the traction vector prescribed on the boundary $\Gamma_u$ .
Permeability matrix	$\mathbf{H}$	$\mathbf{H} = \int_{\Omega} (\nabla \mathbf{N})^T \frac{k}{\mu} \nabla \mathbf{N} \, d\Omega$
Soil stiffness matrix	$\mathbf{K}$	$\mathbf{K} = \int_{\Omega} \mathbf{B}^T \mathbf{C} \mathbf{B} \, d\Omega$ , where $\mathbf{B}$ is the matrix of shape function derivatives and $\mathbf{C}$ is the elastic stiffness tensor.
Coupling matrix	$\mathbf{L}$	$\mathbf{L} = \int_{\Omega} \mathbf{B}^T \mathbf{m} \mathbf{N} \, d\Omega$ , where $\mathbf{m} = [1, 1, 1, 0, 0, 0]^T$ .
Compressibility matrix	$\mathbf{S}$	$\mathbf{S} = \int_{\Omega} \mathbf{N}^T \phi \beta \mathbf{N} \, d\Omega$

157

### 158 **3.3 Model parametrization: Young's modulus and permeability estimation**

159 The coupled system of equations (9) can be solved to estimate the rock properties (namely the  
 160 hydraulic permeability and the Young's modulus) of geological materials subjected to a  
 161 consolidation test. As shown by Horseman et al. (2005), Biot's model is unable to represent  
 162 multiple testing stages from a single set of material values. Thus, for each material, each  
 163 consolidation stage is treated here as a separate test and the fitting procedure of Table 3 is  
 164 applied. Due to the nature of this problem, a two-dimensional axisymmetric finite element  
 165 model is used here (Figure 2).





166

167 *Figure 2. Schematic diagram of the main elements of (a) the axisymmetric plane for the*  
 168 *numerical calculations and (b) the prescribed boundary conditions.*

169

170 *Table 3. Iterative algorithm to fit the Young's modulus and the permeability, given experimental*  
 171 *outflow curves.*

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**Algorithm 1:** Fitting procedure to determine  $E$  and  $k$ , assuming a linear elastic skeleton deformation

---

**Requires:** experimental data (outflow  $f_{\text{exp}}$  versus time curve)

- 1: generate a two-dimensional finite element mesh (radius and length of the sample are required).
- 2: prescribe the *fixed* material parameters: that is, the Poisson's coefficient ( $\nu$ ), the dynamic viscosity ( $\mu$ ) and the specific storage ( $S_s$ ).
- 3: define initial values  $E_0, k_0$  for the two fitting parameters.
- 4: define a time discretization.
- 5: compute the constant block matrices of Table 2 (flux, load and coupling matrices).
- 6: compute the initial permeability matrix  $\mathbf{H}_0 = \int_{\Omega} (\nabla \mathbf{N})^T \frac{k_0}{\mu} \nabla \mathbf{N} d\Omega$ .
- 7: compute the initial stiffness matrix  $\mathbf{K}_0 = \int_{\Omega} \mathbf{B}^T \mathbf{C}_0 \mathbf{B} d\Omega$ , with  $\mathbf{C}_0 = \mathbf{C}(E_0, \nu)$ .
- 8: compute the initial compressibility matrix  $\mathbf{S}_0 = \int_{\Omega} \mathbf{N}^T (\phi\beta)_0 \mathbf{N} d\Omega$ , with

$$(\phi\beta)_0 = \frac{S_s}{\rho_w g} - \alpha_0$$

where  $\rho_w$  is the pore-water density [ $\text{kg/m}^3$ ],  $g$  is the gravitational acceleration ( $=9.81 \text{ m/s}^2$ ) and  $\alpha_0 = 3 \frac{1-2\nu}{E_0}$  is the initial solid-phase compressibility [ $\text{Pa}^{-1}$ ].

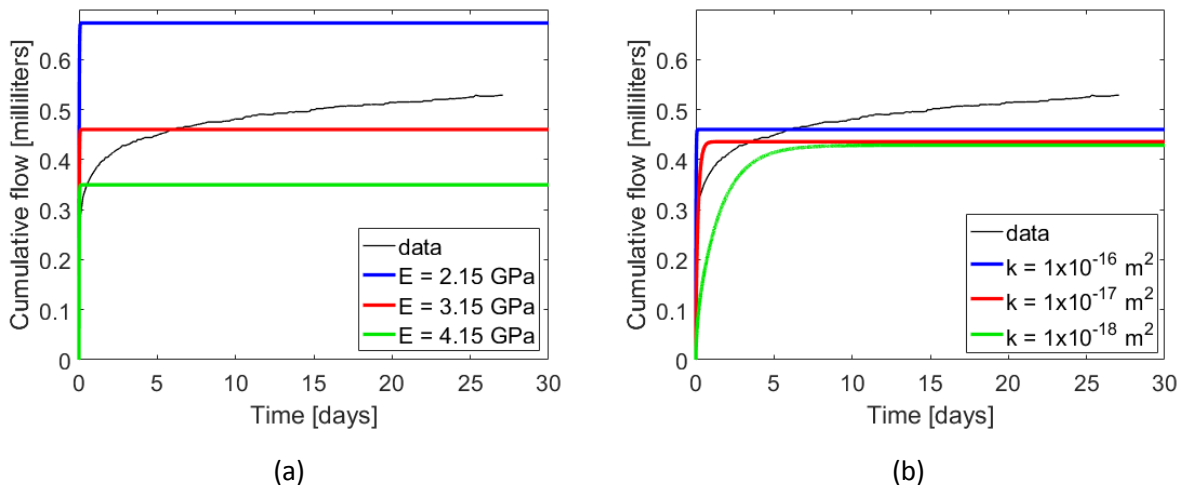
- 9: solve coupled system of equations (9).
  - 10: compute the numerical outflow  $f_{\text{num}}^0$ .
  - 11: iteratively update  $E$  in such a way as to minimise the difference between the numerical and the experimental outflows at time  $t = t_{\text{end}}$ .
-

12: iteratively update  $k$  in such a way as to minimise the difference between the numerical and the experimental transient phases of the outflow versus time curves.

172

173 Note that in Algorithm 1 the facts that (a) the Young's modulus mainly determines the total  
174 volume of fluid expelled (line 11) and (b) the permeability mainly determines the rate at which  
175 fluid is expelled (line 12) have been used, (Figure 3).

176



177

178 *Figure 3. Synthetic example: numerical outflow versus time curves obtained with (a) three*  
179 *different Young's modulus and the rest of parameters kept constant and (b) three different*  
180 *permeability values and the rest of parameters kept constant. In black, experimental data from*  
181 *Harrington et al. (2018) measured for the Mercia Mudstone Group sample is shown.*

179

180 This fitting algorithm provided reasonable theoretical flow-time curves for three Mercia  
181 Mudstone Group samples, see Harrington et al. (2018). However, as highlighted there and as  
182 seen in the synthetic example of Figure 3, Biot's model is unable to reproduce the time-  
183 dependent behaviour of the flow versus time curve. Thus, a more complex deformation model,  
184 allowing for this creep effect needs to be considered.

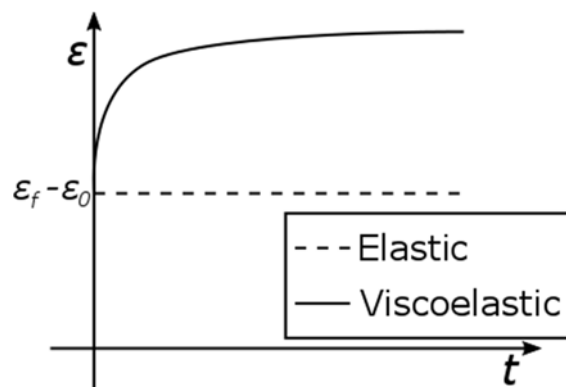
185

186 **4. Numerical model: time-dependent skeletal deformation**

187 In each consolidation stage, the stress is raised abruptly and then kept constant. The initial  
188 instantaneous increase in the confining stress (from  $\sigma_0$  to  $\sigma_f$ ) leads to an immediate increase  
189 of the strain (from  $\epsilon_0$  to  $\epsilon_f$ ) causing an instantaneous flow expulsion. This initial behaviour is  
190 observed in the experimental flow versus time curves (Figure 3) and can be successfully  
191 described with a linear elastic behaviour. However, a further flow ejection period also occurs  
192 once the confining stress is kept constant. This results from strain increasing further with time,  
193 despite the constant stress. This time-dependent behaviour is shown by viscoelastic materials  
194 and can be modelled by assuming a time-dependent Young's modulus at constant stress.  
195 Particularly, in a one-dimensional setting,

196 
$$E(t) = \frac{\sigma}{\epsilon(t)} \quad (10)$$

197 where the deformation (and thus  $E(t)$ ) approaches a constant value when the loading time  
198 becomes large, see Figure 4.



200 *Figure 4. Strain versus time curve at constant stress for an elastic and a viscoelastic material.*

201

202 **4.1. Creep modulus: standard solid element**

203 Different mathematical expressions for the time-dependent Young's modulus  $E(t)$  may be  
204 employed. However, classical linear viscoelastic models assume that materials behave as one-  
205 dimensional spring-dashpot systems (Figure 5). For the sake of simplicity, two-element models

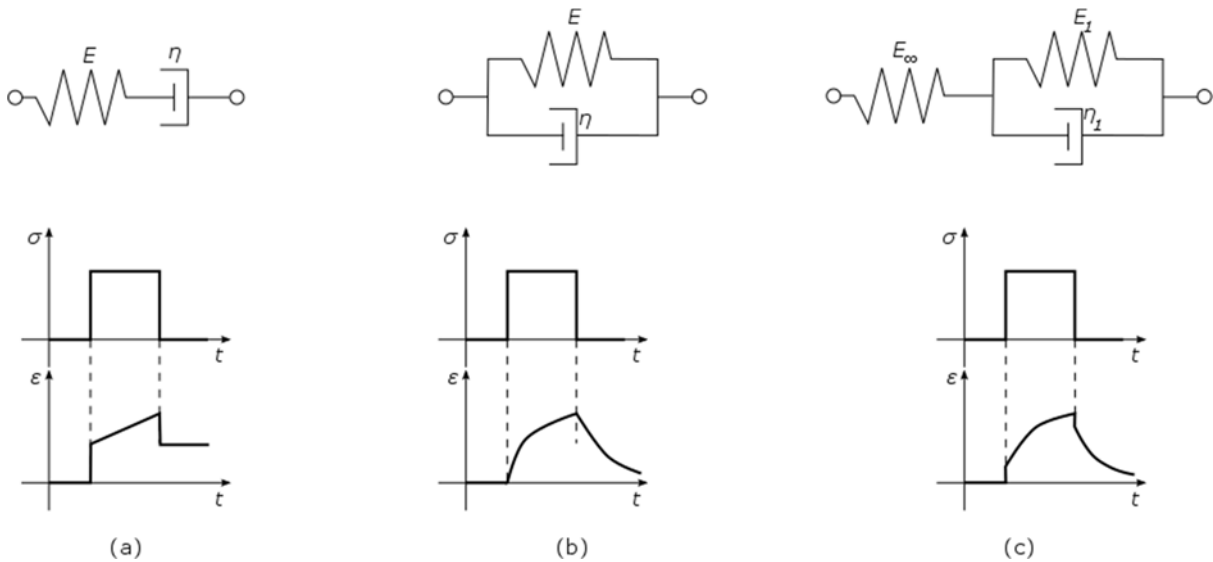
206 are preferred here. Nevertheless, the Maxwell model does not describe an anelastic recovery  
 207 (Figure 5a), whereas the Kelvin-Voigt model (Figure 5b), does not predict an instantaneous  
 208 strain. Thus, the standard solid model (Figure 5c) is adopted here, which leads to a creep  
 209 compliance

$$210 \quad J(t) = \frac{1}{E_\infty} + \frac{1}{E_1} \left(1 - e^{-\frac{\eta_1 t}{E_1}}\right) \quad (11)$$

211 and to a time-dependent elastic modulus

$$212 \quad E(t) = \frac{1}{\frac{1}{E_\infty} + \frac{1}{E_1} \left(1 - e^{-\frac{\eta_1 t}{E_1}}\right)} \quad (12)$$

213



215 *Figure 5. Time response of the strain in a creep experiment for a (a) Maxwell model, (b) Kelvin-*  
 216 *Voigt model and (c) standard solid.*  
 217

218 Note that by means of this viscoelastic model, the fitting complexity increases: in the elastic  
 219 model, one mechanical parameter ( $E$ ) is required while in the linear viscoelastic model, three  
 220 new mechanical values ( $E_\infty$ ,  $E_1$  and  $\eta_1$ ) need to be estimated. In order to reduce this complexity,

$$221 \quad E := E_\infty = E_1 \quad (13)$$

222 is here prescribed. Hence, the time-dependent Young's modulus (12) may be expressed as

223 
$$E(t) = \frac{E}{2 - e^{-at}} \quad (14)$$

224 where  $a := \eta_1/E$  [s<sup>-1</sup>] is here used.

225 This enhancement in the mechanical properties of the material leads to a time-dependent elastic  
 226 stiffness tensor and to a time-dependent fluid storage coefficient. Thus, the coupled system (9)  
 227 is now

228 
$$\begin{bmatrix} \mathbf{K}^{n+1} & \mathbf{L} \\ \mathbf{L}^T & -\Delta t \mathbf{H} \mathbf{S}^{n+1} \end{bmatrix} \cdot \begin{bmatrix} \mathbf{u}^{n+1} \\ \mathbf{p}^{n+1} \end{bmatrix} = \begin{bmatrix} \mathbf{F}_u \\ \mathbf{L}^T \mathbf{u}^n - \mathbf{S}^{n+1} \mathbf{p}^n + \Delta t \mathbf{F}_p \end{bmatrix} \quad (15)$$

229 with

230 
$$\mathbf{K}^i = \int_{\Omega} \mathbf{B}^T \mathbf{C}^i \mathbf{B} \, d\Omega \quad (16a)$$

231 
$$\mathbf{S}^i = \int_{\Omega} \mathbf{N}^T (\phi\beta)^i \mathbf{N} \, d\Omega \quad (16b)$$

232 where  $\mathbf{C}^i = \mathbf{C}(E(t^i), \nu)$  and  $(\phi\beta)^i = \frac{S_s}{\rho_w g} - 3 \frac{1-2\nu}{E(t^i)}$  stand for the elastic stiffness and the fluid  
 233 storage coefficient at time step  $i$  respectively.

234

#### 235 4.2. Model parametrization: spring stiffness, dashpot viscosity and permeability

236 The coupled system of equations (15) can be solved to estimate the new rock properties. If the  
 237 viscoelastic model with the time-dependent Young's modulus (14) is assumed, three  
 238 parameters are needed:  $E$ ,  $a$  and  $k$ . Here, the fitting procedure of Table 4 is proposed. Again,  
 239 as done in Section 3.3 for the elastic material, each consolidation stage is treated as a separate  
 240 test.

241

242 *Table 4. Iterative algorithm to fit the time-dependent Young's modulus and the permeability,*  
 243 *given experimental outflow curves.*

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**Algorithm 2:** Fitting procedure to determine  $E$ ,  $a$ ,  $k$  assuming a viscoelastic skeleton deformation

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**Requires:** experimental data (outflow  $f_{\text{exp}}$  versus time curve)

- 1: generate a two-dimensional finite element mesh (radius and length of the sample are required).
-

- 2: prescribe the *fixed* material parameters: that is, the Poisson's coefficient ( $\nu$ ), the dynamic viscosity ( $\mu$ ) and the specific storage ( $S_s$ ).
  - 3: define initial values  $E_0, a_0, k_0$  for the three fitting parameters.
  - 4: define a time discretization.
  - 5: compute the constant block matrices of Table 2 (flux, load and coupling matrices).
  - 6: compute the initial permeability matrix  $\mathbf{H}_0 = \int_{\Omega} (\nabla \mathbf{N})^T \frac{k_0}{\mu} \nabla \mathbf{N} d\Omega$ .
  - 7: compute the initial stiffness matrix  $\mathbf{K}_0 = \int_{\Omega} \mathbf{B}^T \mathbf{C}_0^0 \mathbf{B} d\Omega$ , with  $\mathbf{C}_0^0 = \mathbf{C}^0(E(E_0, a_0, t^0), \nu)$ .
  - 8: compute the initial compressibility matrix  $\mathbf{S}_0^0 = \int_{\Omega} \mathbf{N}^T (\phi\beta)_0^0 \mathbf{N} d\Omega$ , with  $(\phi\beta)_0^0 = \frac{S_s}{\rho_w g} - 3 \frac{1 - 2\nu}{E(E_0, a_0, t^0)}$
  - 9: solve coupled system of equations (15). Thus, at each time step, the stiffness and compressibility matrices are updated.
  - 10: compute the numerical outflow  $f_{\text{num}}^0$ .
  - 11: iteratively update  $E$  in such a way as to minimise the difference between the numerical and the experimental outflows at time  $t = t_{\text{end}}$ .
  - 12: iteratively update  $a$  in such a way as to minimise the difference between the numerical and the experimental outflow slopes (after the initial instantaneous flow expulsion).
  - 13: iteratively update  $k$  in such a way as to minimise the difference between the numerical and the experimental transient phases of the outflow versus time curves.
- 

244

245 Note that now, the total volume of fluid expelled is controlled by both the spring stiffness  $E$   
 246 and the numerical parameter  $a$ . However, as seen in Figure 6, each of these two parameters has  
 247 a different physical value. Indeed,  $E$  mainly determines the total volume of fluid expelled after  
 248 the instantaneous flow expulsion whereas  $a$  controls the slope of the time-dependent branch.  
 249 This has been taken into account in Algorithm 2 (see lines 11 and 12, respectively).

250

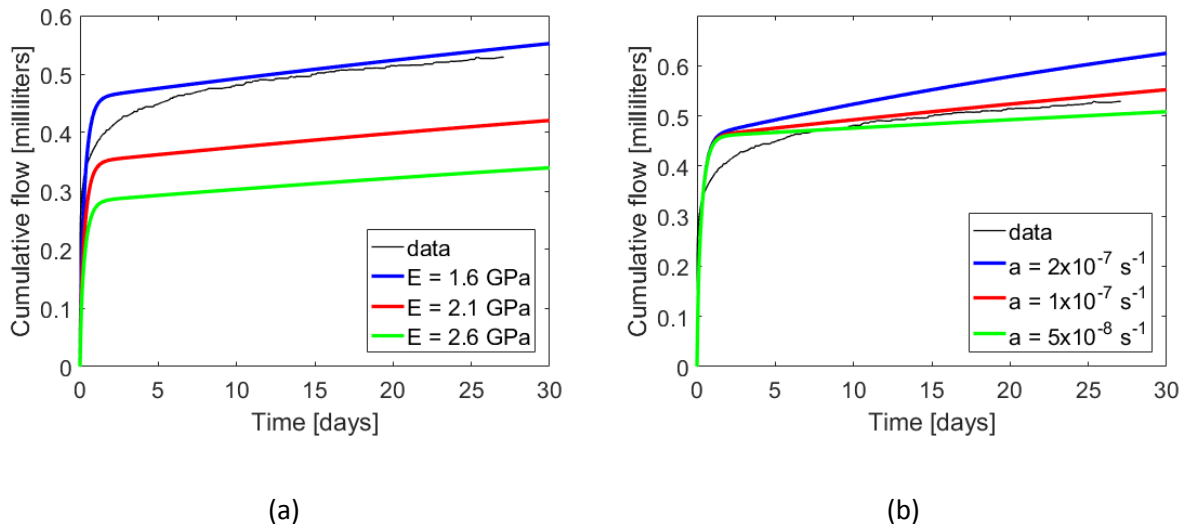


Figure 6. Synthetic example: numerical outflow versus time curves obtained with (a) three different values for the spring stiffness and the rest of parameters kept constant and (b) three different values of  $a$  and the rest of parameters kept constant. In black, experimental data from Harrington et al. (2018) measured for the Mercia Mudstone Group sample is shown.

251

## 252 5. Results: validation of the numerical model

253 The new numerical model is validated against different experimental results conducted at the  
 254 British Geological Survey (BGS). To illustrate the generality of the strategy, three different  
 255 materials are here analysed: (a) a Boom Clay sample extracted from the High Activity  
 256 Experimental Site (HADES) Underground Research Laboratory (URL) at Mol in Belgium, (b)  
 257 a Callovo-Oxfordian claystone (COx) specimen taken from the Meuse/Haute Marne URL in  
 258 France and (c) a mudstone sample of the Mercia Mudstone Group collected from a halite mine  
 259 in Northern Ireland. These materials differ in their clay content and thus, their physical  
 260 properties such as rock porosity and permeability are significantly different: clay-rich samples  
 261 are characterised by smaller pore-throats and thus by lower permeability values.

262

### 263 5.1 Boom Clay specimen

264 The first test relates to a Boom Clay sample extracted from the HADES Underground Research  
 265 Laboratory (URL) at Mol in Belgium (Figure 7). This specimen was taken from a location 223

266 m below surface within the research facility. At depth this material can be described as a hard,  
267 high plasticity clay, see Horseman et al. (1987). It is of interest in Belgium and the Netherlands  
268 as potential host formation for a radioactive waste disposal facility.

269



270

271 *Figure 7. Isotropic test assembly containing the Boom Clay sample.*

272 Following assembly of the apparatus, an initial equilibration period of 8 days was applied to  
273 the cylindrical sample, with confining pressure held constant at 4.4 MPa. The pore pressure  
274 within the sample was then allowed to equilibrate, with both the injection and backpressure  
275 ends being held at a constant condition of 2.2 MPa. These conditions were selected to return  
276 the clay to those experienced *in situ* prior to exhumation. Once the equilibration stage (stage  
277 [0]) was complete, a ten-step consolidation test was performed, see Table 5 and Figure 8. As  
278 seen, the injection and backpressure were held constant at 2.7 MPa and 2.2 MPa respectively  
279 during the entire consolidation period, ensuring a constant flow of water across the sample.  
280 Instantaneous flow rate and net cumulative flow volume data were collected, with the latter  
281 equating to volumetric strain. Estimated specific storage values, see Table 5, are here used to  
282 validate the proposed strategy.

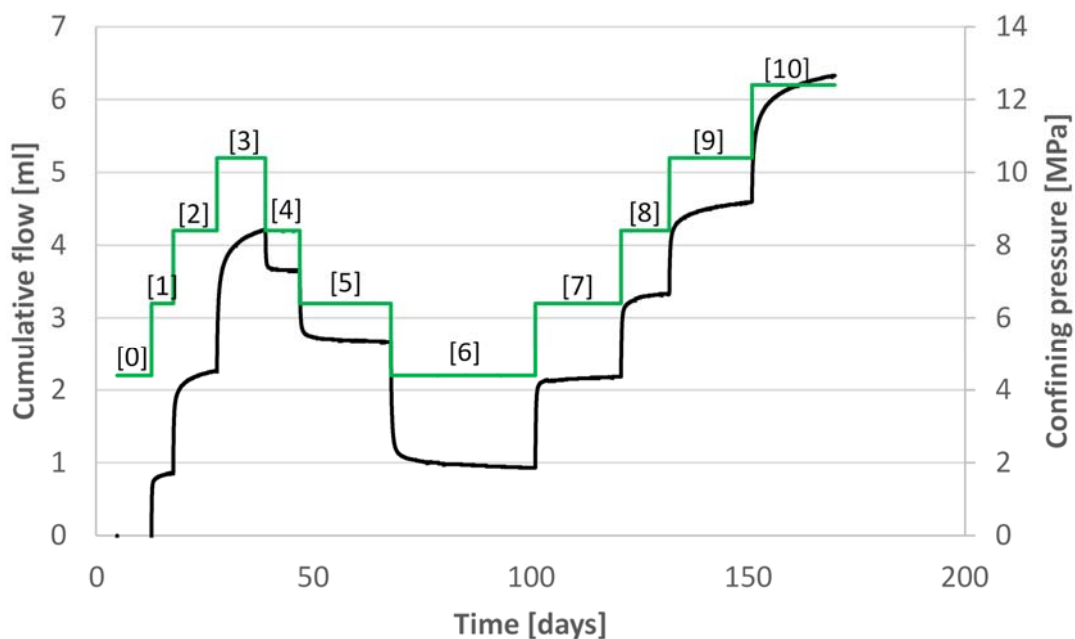
283



284 *Table 5. Summary of experimental histories for the Boom Clay sample.*

BOOM CLAY SAMPLE				
Stage number	Confining pressure [MPa]	Injection pressure [MPa]	Backpressure [MPa]	Specific storage [m <sup>-1</sup> ]
1	6.4	2.7	2.2	5.1 x 10 <sup>-5</sup>
2	8.4	2.7	2.2	13.7 x 10 <sup>-5</sup>
3	10.4	2.7	2.2	27.0 x 10 <sup>-5</sup>
4	8.4	2.7	2.2	3.5 x 10 <sup>-5</sup>
5	6.4	2.7	2.2	6.0 x 10 <sup>-5</sup>
6	4.4	2.7	2.2	10.9 x 10 <sup>-5</sup>
7	6.4	2.7	2.2	7.8 x 10 <sup>-5</sup>
8	8.4	2.7	2.2	7.2 x 10 <sup>-5</sup>
9	10.4	2.7	2.2	7.6 x 10 <sup>-5</sup>
10	12.4	2.7	2.2	12.7 x 10 <sup>-5</sup>

285



286

287 *Figure 8. Boom Clay sample: cumulative flow (in black) and confining systems (in green) from*  
 288 *test stages [1]-[10].*

289

290 Analysis of the consolidation data is here performed by assuming both an elastic and a  
 291 viscoelastic skeletal deformation. Here, the geometrical and material parameters of Table 6 are  
 292 used. As seen, the Poisson's coefficient reported by Barnichon and Volckaert (2003) and  
 293 Bésuelle et al. (2013) is used here.

294

295 *Table 6. Geometrical and material parameters used in the numerical fittings for the Boom Clay*  
 296 *specimen.*  
 297

<b>BOOM CLAY SAMPLE</b>		
<b>Meaning</b>	<b>Symbol [units]</b>	<b>Value</b>
Radius of the sample	$r$ [mm]	24.96
Length of the sample	$L$ [mm]	42.67
Poisson's coefficient	$\nu$ [-]	0.125
Dynamic viscosity	$\mu$ [Pa · s]	$2.32 \times 10^{-3}$

298

299 As done in Horseman et al. (2005), each consolidation stage is treated here as a separate test.  
 300 The fitting results are shown in Figure 9. As seen, the elastic model (blue-dotted curve) is not  
 301 able to represent the time-dependent behaviour observed experimentally in some consolidation  
 302 stages, whereas laboratory data fit better with the proposed viscoelastic model (red-dashed  
 303 curve). The fitted parameters obtained with both models are listed in Table 7 and Figure 10.  
 304 As seen, the two models lead to a similar Young's modulus. This is in agreement with the  
 305 suggested definition of the time-dependent Young's modulus, see Equation 14, since the  
 306 evolving Young's modulus tends to the elastic one when the loading time becomes large  
 307 enough. However, the new model does lead to significantly improved permeability value  
 308 predictions, especially for those stages where the confining pressure decreased.

309

310

311

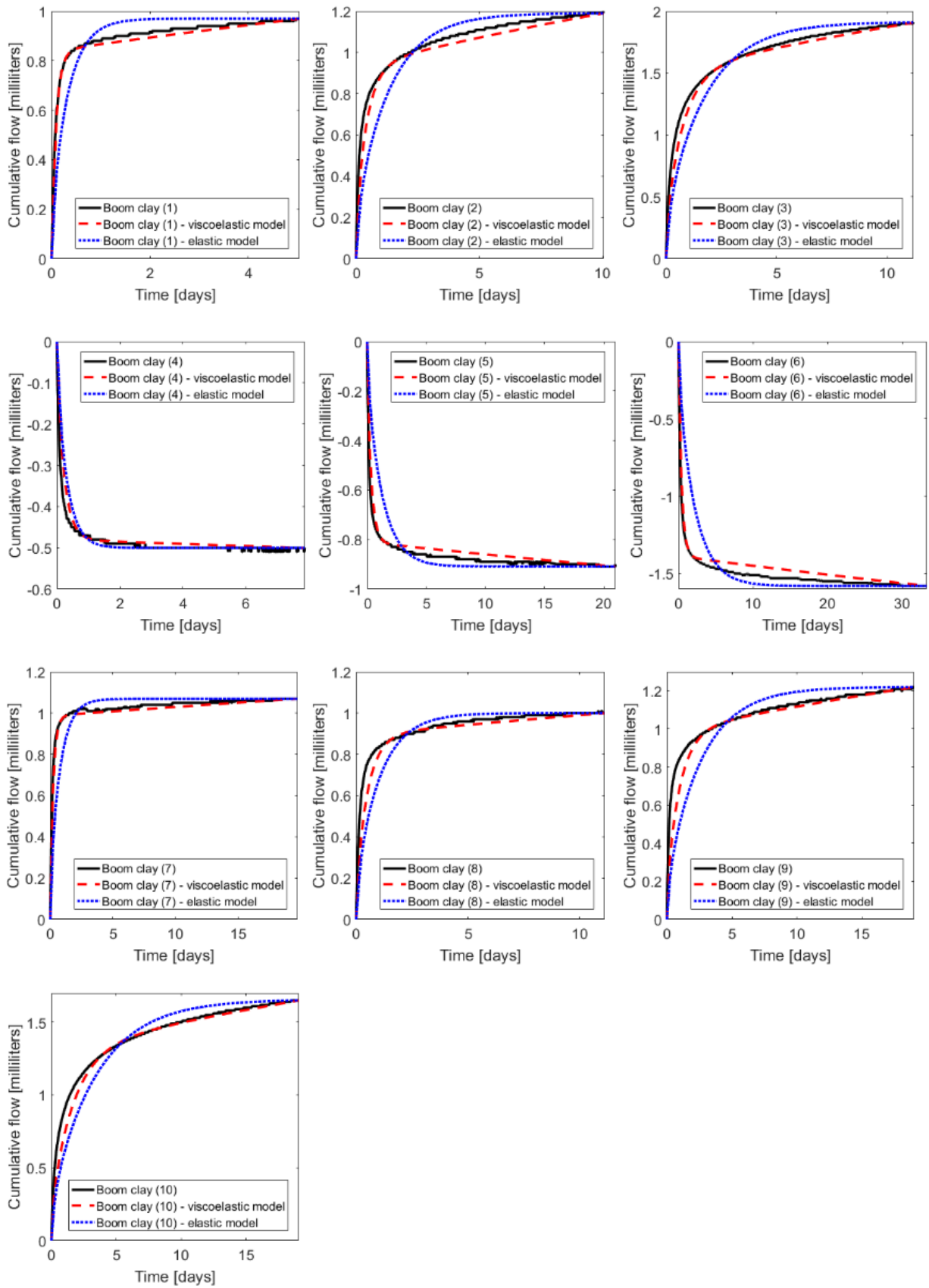


Figure 9. Boom Clay specimen: comparison of model to flow data.

312 Table 7. Boom Clay specimen: parameter values determined for each stage.

BOOM CLAY SAMPLE						
Stage number	EXPERIMENTAL		FITTED			
			ELASTIC SKELETAL DEFORMATION		VISCOELASTIC SKELETAL DEFORMATION	
	Permeability [m <sup>2</sup> ]	Young's Modulus [MPa]	Permeability [m <sup>2</sup> ]	Young's Modulus [MPa]	Permeability [m <sup>2</sup> ]	(Averaged) Young's Modulus [MPa]
1	1.44 x 10 <sup>-19</sup>	581.82	7.73 x 10 <sup>-20</sup>	351.10	3.44 x 10 <sup>-19</sup>	413.29
2	1.17 x 10 <sup>-19</sup>	221.01	4.92 x 10 <sup>-20</sup>	276.83	1.71 x 10 <sup>-19</sup>	321.85
3	8.33 x 10 <sup>-19</sup>	114.84	7.34 x 10 <sup>-20</sup>	171.15	1.77 x 10 <sup>-19</sup>	192.93
4	9.04 x 10 <sup>-20</sup>	846.02	5.77 x 10 <sup>-20</sup>	908.58	8.65 x 10 <sup>-20</sup>	950.87
5	1.03 x 10 <sup>-19</sup>	486.29	2.31 x 10 <sup>-20</sup>	467.43	1.00 x 10 <sup>-19</sup>	533.69
6	1.25 x 10 <sup>-19</sup>	263.08	2.43 x 10 <sup>-20</sup>	268.06	1.60 x 10 <sup>-19</sup>	316.09
7	1.11 x 10 <sup>-19</sup>	385.61	4.91 x 10 <sup>-20</sup>	315.51	1.94 x 10 <sup>-19</sup>	357.87
8	9.59 x 10 <sup>-20</sup>	413.78	3.37 x 10 <sup>-20</sup>	334.20	7.43 x 10 <sup>-20</sup>	363.93
9	8.09 x 10 <sup>-20</sup>	393.46	1.37 x 10 <sup>-20</sup>	262.60	3.73 x 10 <sup>-20</sup>	292.92
10	5.70 x 10 <sup>-20</sup>	236.86	1.76 x 10 <sup>-20</sup>	192.40	3.62 x 10 <sup>-20</sup>	211.45

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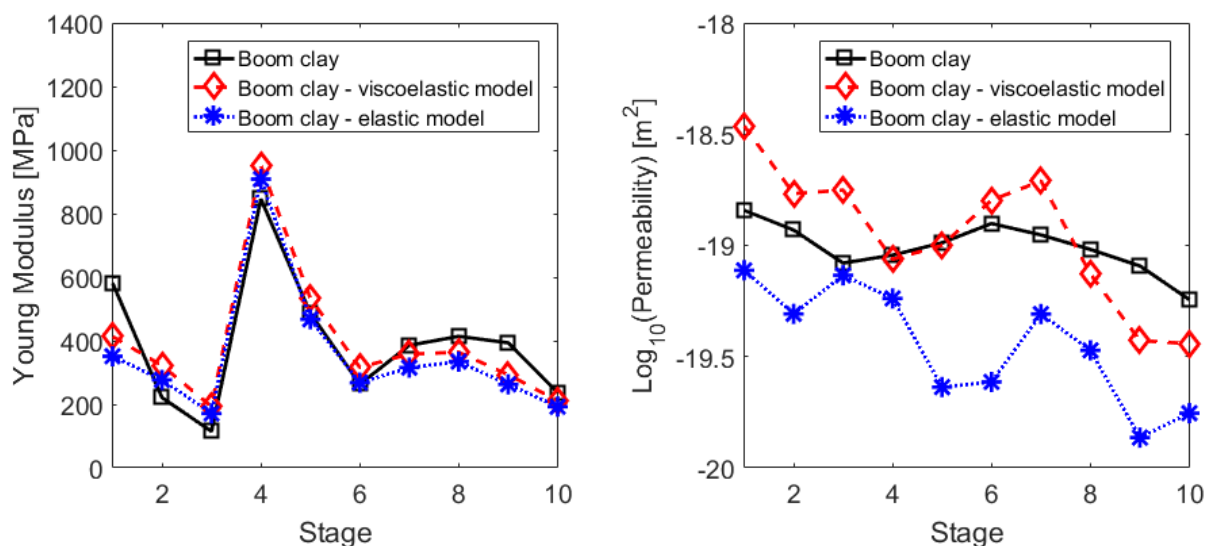


Figure 10. Boom Clay specimen: parameter values determined for each stage.

320 **5.2 Callovo-Oxfordian claystone**

321 The second test was conducted on a sample of the Callovo-Oxfordian claystone (COx)  
322 collected from a location 450 m below surface at the Meuse/Haute Marne URL (France), see  
323 Figure 11. The COx is of interest in France as a candidate host formation for a radioactive  
324 waste disposal facility. A six-step consolidation test was carried out after an initial equilibration  
325 period of 62 days, with the confining pressure held at 9 MPa, see Figure 12, and the pore  
326 pressure at 1.0 MPa. As seen in Table 8, the injection and backpressure during the entire  
327 consolidation period were held constant at 4.0 MPa and 1.0 MPa respectively, leading to a pore  
328 pressure gradient and continuous flow of water across the sample. Estimated specific storage  
329 values are here prescribed. For a detailed description of the test, see the report by Harrington  
330 and Tamayo-Mas (2016).

331



332

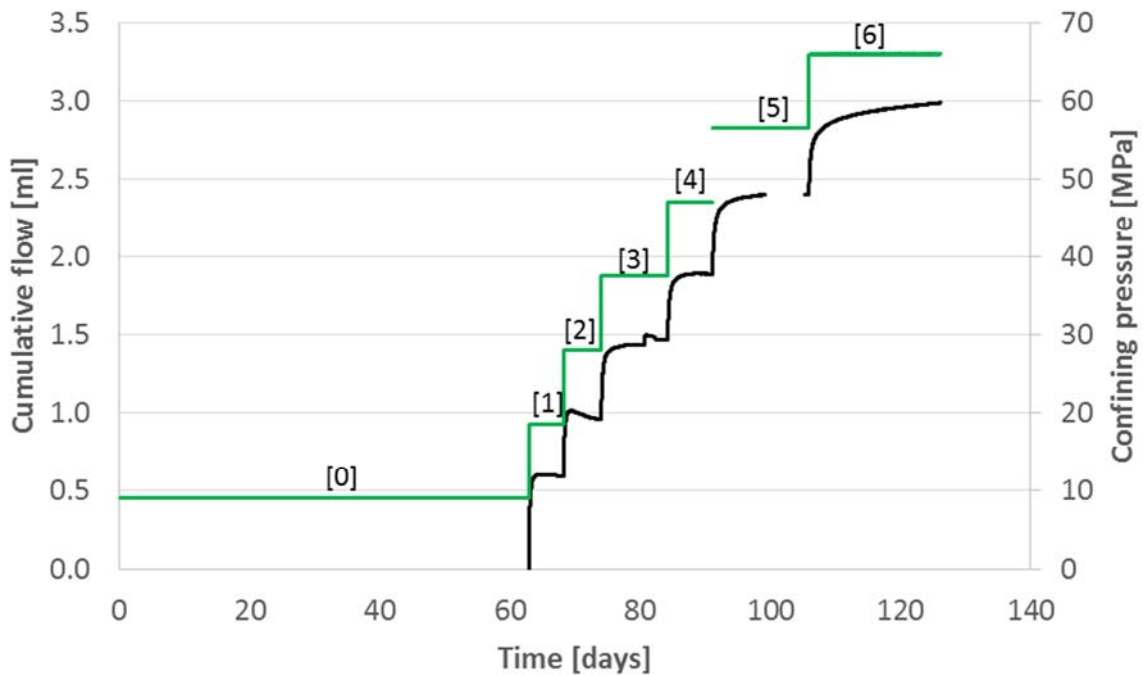
333 *Figure 11. Sample of the Callovo-Oxfordian claystone.*

334 *Table 8. Summary of experimental histories for the COx sample.*

CALLOVO-OXFORDIAN CLAYSTONE				
Stage number	Confining pressure [MPa]	Injection pressure [MPa]	Backpressure [MPa]	Specific storage [m <sup>-1</sup> ]
1	18.5	4.0	1.0	4.6 x 10 <sup>-6</sup>

2	28	4.0	1.0	$3.8 \times 10^{-6}$
3	37.5	4.0	1.0	$5.0 \times 10^{-6}$
4	47	4.0	1.0	$4.7 \times 10^{-6}$
5	56.5	4.0	1.0	$6.4 \times 10^{-6}$
6	66	4.0	1.0	$6.5 \times 10^{-6}$

335



336

337 *Figure 12. COx: cumulative flow (in black) and confining systems (in green) from test stages*  
 338 *[1]-[6].*

339

340 As for the Boom Clay sample, the consolidation data is analysed here by means of the elastic  
 341 and viscoelastic models. The geometrical and material parameters used in the numerical  
 342 simulations are shown in Table 9. Here, as with Harrington et al. (2018), the Poisson's ratio  
 343 value reported by Wileveau and Bernier (2008) is used. The fitting results obtained with both  
 344 models are shown in Figure 13. As seen, enhancing the elastic bulk with a dashpot viscosity  
 345 leads to better fitting in those cases where the elastic model is not appropriate (see consolidation  
 346 stages 3 and 6) and provides a very similar solution when yield has been reached and thus, the  
 347 traditional model is acceptable (see consolidation stages 1, 4 and 5). As observed for the Boom

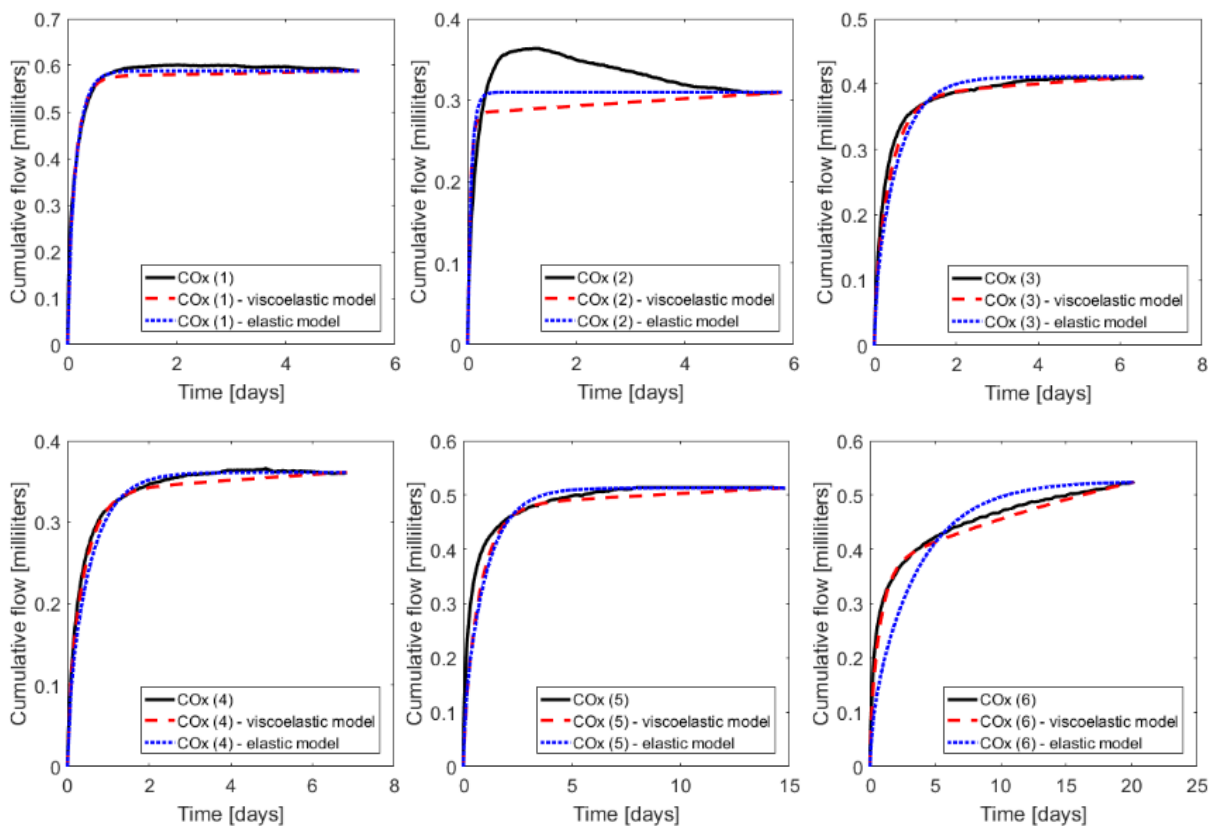
348 Clay sample, this improvement is especially significant for the permeability parameter, see  
 349 Table 10 and Figure 14. The unusual form of the experimental data obtained for stage 2 (Figure  
 350 13), is due to a mismatch between in- and outflow values and hence, the fitting is done at the  
 351 end of this test stage.

352

353 *Table 9. Geometrical and material parameters used in the numerical fittings for the COx*  
 354 *specimen.*

CALLOVO-OXFORDIAN CLAYSTONE		
Meaning	Symbol [units]	Value
Radius of the sample	$r$ [mm]	25.09
Length of the sample	$L$ [mm]	48.38
Poisson's coefficient	$\nu$ [-]	0.3
Dynamic viscosity	$\mu$ [Pa · s]	$1.00 \times 10^{-3}$

355



356

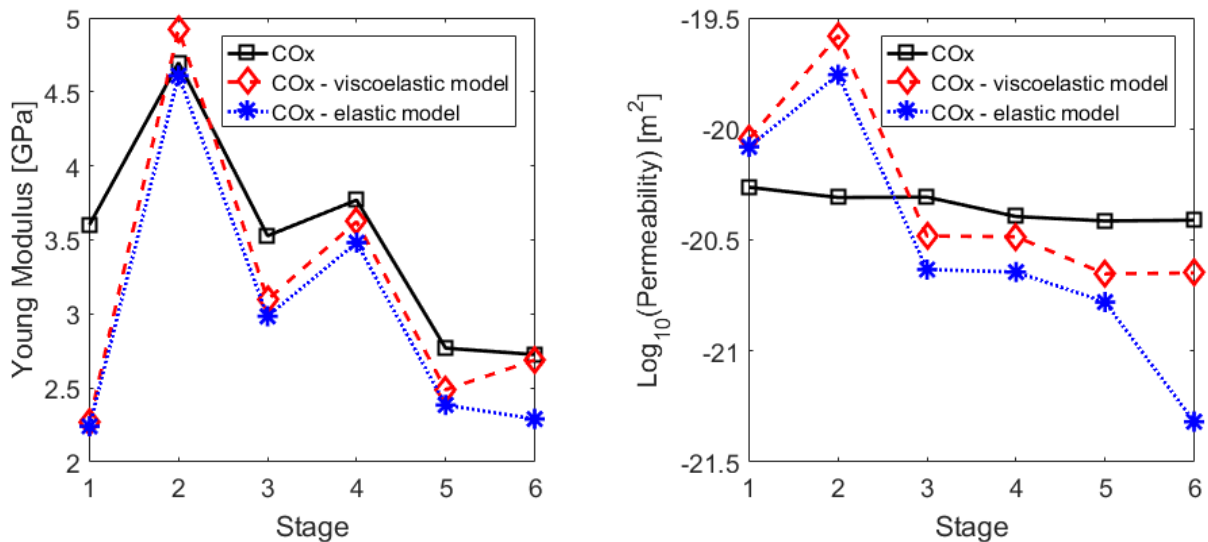
*Figure 13. COx specimen: comparison of model to flow data.*

357

358 Table 10. COx specimen: parameter values determined for each stage.

CALLOVO-OXFORDIAN CLAYSTONE						
Stage number	EXPERIMENTAL		FITTED			
			ELASTIC SKELETAL DEFORMATION		VISCOELASTIC SKELETAL DEFORMATION	
	Permeability [m <sup>2</sup> ]	Young's Modulus [GPa]	Permeability [m <sup>2</sup> ]	Young's Modulus [GPa]	Permeability [m <sup>2</sup> ]	(Averaged) Young's Modulus [GPa]
1	5.46 x 10 <sup>-21</sup>	3.60	8.30 x 10 <sup>-21</sup>	2.24	9.02 x 10 <sup>-21</sup>	2.27
2	4.91 x 10 <sup>-21</sup>	4.70	1.75 x 10 <sup>-20</sup>	4.61	2.63 x 10 <sup>-20</sup>	4.92
3	4.93 x 10 <sup>-21</sup>	3.53	2.33 x 10 <sup>-21</sup>	2.98	3.29 x 10 <sup>-21</sup>	3.10
4	4.03 x 10 <sup>-21</sup>	3.77	2.27 x 10 <sup>-21</sup>	3.48	3.27 x 10 <sup>-21</sup>	3.62
5	3.85 x 10 <sup>-21</sup>	2.77	1.66 x 10 <sup>-21</sup>	2.38	2.22 x 10 <sup>-21</sup>	2.49
6	3.88 x 10 <sup>-21</sup>	2.73	4.80 x 10 <sup>-22</sup>	2.29	2.25 x 10 <sup>-21</sup>	2.69

359



360 Figure 14. COx specimen: parameter values determined for each stage.

361

362 **5.3 Mercia Mudstone formation sample**

363 Thirdly, the approach is validated against a consolidation experiment conducted on a well-  
 364 preserved sample recovered from the Knocksoghey Formation in the Larne Basin (Figure 15).



365 This sample was collected during excavation of a new mine drift in Northern Ireland within  
366 the Mercia Mudstone Group (MMG), which is of interest as a caprock for potential CO<sub>2</sub> storage  
367 sites in the North and Irish Seas, Armitage et al. (2013). This material can be described as a  
368 fine-grained mudstone to microsparstone, but it should be noted that at the microscopic scale  
369 it is highly heterogeneous. For a detailed description of the material, we refer to Harrington et  
370 al. (2018).

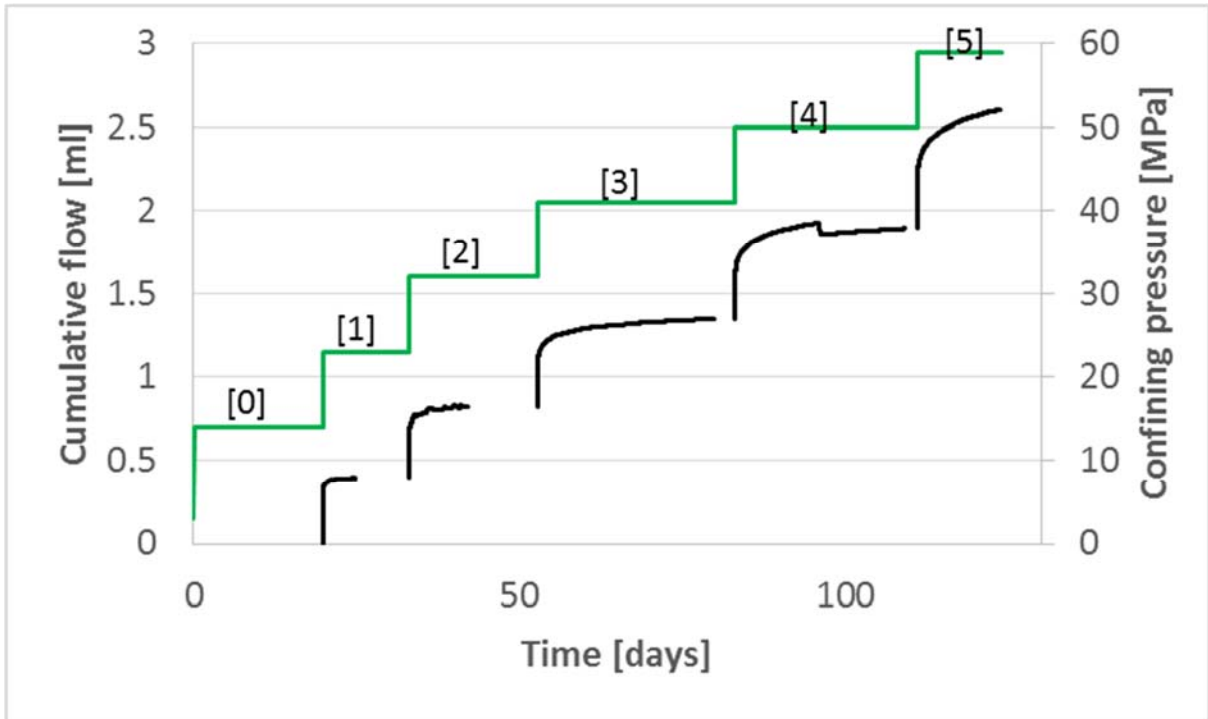
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372



*Figure 15. Sample from the Mercia Mudstone Group (left) and arranged with the isotropic test assembly (right).*

373 After the initial equilibration period (confining stress and pore pressure were 14.0 MPa and 1.0  
374 MPa respectively), the cylindrical specimen was subjected to a five-step consolidation test, see  
375 Figure 16 and Table 11. Here, no pore pressure difference across the sample was prescribed  
376 during consolidation.



377

378 *Figure 16. Mercia: cumulative flow (in black) and confining systems (in green) from test stages*  
 379 *[1]-[5].*

380

381 *Table 11. Summary of experimental histories for the Mercia sample.*

MERCIA MUDSTONE FORMATION SAMPLE			
Stage number	Confining pressure [MPa]	Injection pressure [MPa]	Backpressure [MPa]
1	23.0	1.0	1.0
2	32.0	1.0	1.0
3	41.0	1.0	1.0
4	50.0	1.0	1.0
5	59.0	1.0	1.0

382

383 As with the previous samples, the two suggested algorithms are here employed to derive the  
 384 hydraulic and mechanical parameters. Here, the geometrical and material parameters of Table  
 385 12 have been used. As reported by Hobbs et al. (2002), Poisson's ratios for the MMG were  
 386 found to vary from 0.2 and 0.4. Hence, an intermediate value  $\nu = 0.25$  is considered here for

387 all the numerical simulations. As seen, in this example, the specific storage has been considered  
 388 constant during the entire consolidation process.

389

390 *Table 12. Geometrical and material parameters used in the numerical fittings for the Mercia*  
 391 *specimen.*

<b>MERCIA MUDSTONE FORMATION SAMPLE</b>		
<b>Meaning</b>	<b>Symbol [units]</b>	<b>Value</b>
Radius of the sample	$r$ [mm]	27.21
Length of the sample	$L$ [mm]	48.76
Poisson's coefficient	$\nu$ [-]	0.25
Dynamic viscosity	$\mu$ [Pa · s]	$2.32 \times 10^{-3}$
Specific storage	$S_s$ [m <sup>-1</sup> ]	$4.5 \times 10^{-6}$

392

393 The fittings are shown in Figure 17 and listed in Table 13. As seen, the new method is able to  
 394 describe the experimental time-dependent behaviour also with this new material. Here, due to  
 395 the high heterogeneity of the material, direct measurements of the permeability should be  
 396 considered as indicative only. Thus, as with Harrington et al. (2018), numerical permeability  
 397 values are compared here with the derived values

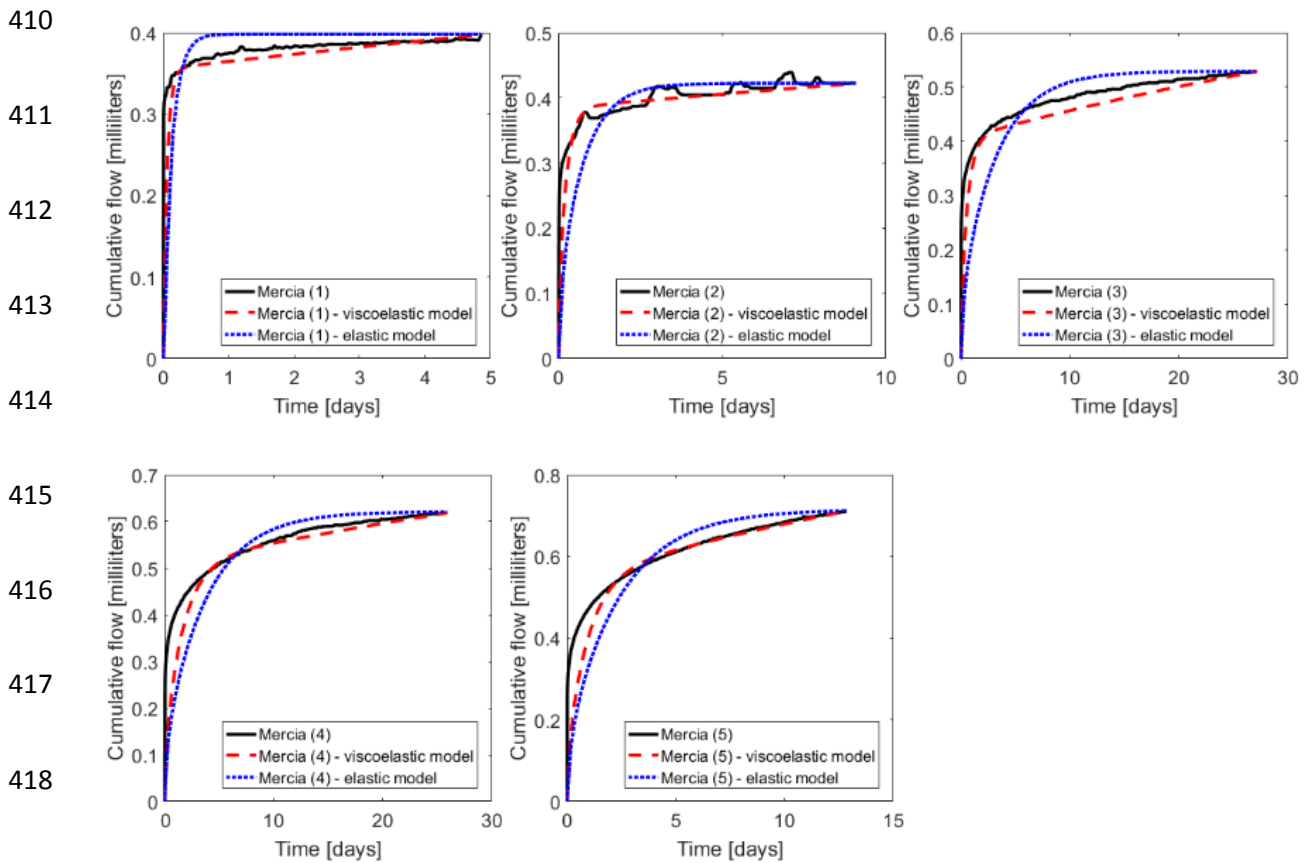
$$398 \quad k = K \frac{\mu}{\rho_w g} \quad (17)$$

399 where  $\mu$  is the dynamic viscosity of the fluid [Pa·s],  $\rho_w$  is the pore-water density [kg/m<sup>3</sup>],  $g$  is  
 400 the gravitational acceleration (=9.81 m/s<sup>2</sup>) and  $K$  is the hydraulic conductivity [m/s]. Here, this  
 401 value is estimated using the simple relationship

$$402 \quad K = m_v c_v \gamma_v \quad (18)$$

403 where  $\gamma_v$  is the unit weight of water (=9.81 N/m<sup>3</sup>),  $m_v$  is the coefficient of volume  
 404 compressibility [Pa<sup>-1</sup>] and  $c_v$  is the coefficient of consolidation [m<sup>2</sup>/year], computed here by  
 405 means of the Taylor's square root of time method, as described by Scott (1980). As seen in  
 406 Figure 18, the proposed viscoelastic model leads to more accurate rock properties. However,

407 as highlighted in Harrington et al. (2018) the experimental permeability values obtained for  
 408 some stages (3,4,5) should be treated as indicative only as outflow had not fully asymptoted by  
 409 the end of the stage.



419 *Figure 17. Mercia specimen: comparison of model to flow data.*

420

421 *Table 13. Mercia specimen: parameter values determined for each stage.*

MERCIA MUDSTONE GROUP SAMPLE						
Stage number	EXPERIMENTAL		FITTED			
			ELASTIC SKELETAL DEFORMATION		VISCOELASTIC SKELETAL DEFORMATION	
	Permeability [m <sup>2</sup> ]	Young's Modulus [GPa]	Permeability [m <sup>2</sup> ]	Young's Modulus [GPa]	Permeability [m <sup>2</sup> ]	(Averaged) Young's Modulus [GPa]
1	7.85 x 10 <sup>-19</sup>	3.937	7.06 x 10 <sup>-20</sup>	3.698	1.48 x 10 <sup>-19</sup>	3.731

2	$1.33 \times 10^{-19}$	3.472	$1.21 \times 10^{-20}$	3.058	$3.56 \times 10^{-20}$	3.276
3	$3.53 \times 10^{-19}$	2.758	$3.44 \times 10^{-21}$	2.355	$1.36 \times 10^{-20}$	2.813
4	$4.25 \times 10^{-20}$	2.326	$3.41 \times 10^{-21}$	1.972	$4.49 \times 10^{-21}$	2.156
5	$5.00 \times 10^{-19}$	2.007	$6.10 \times 10^{-21}$	1.732	$7.39 \times 10^{-21}$	1.881

422

423

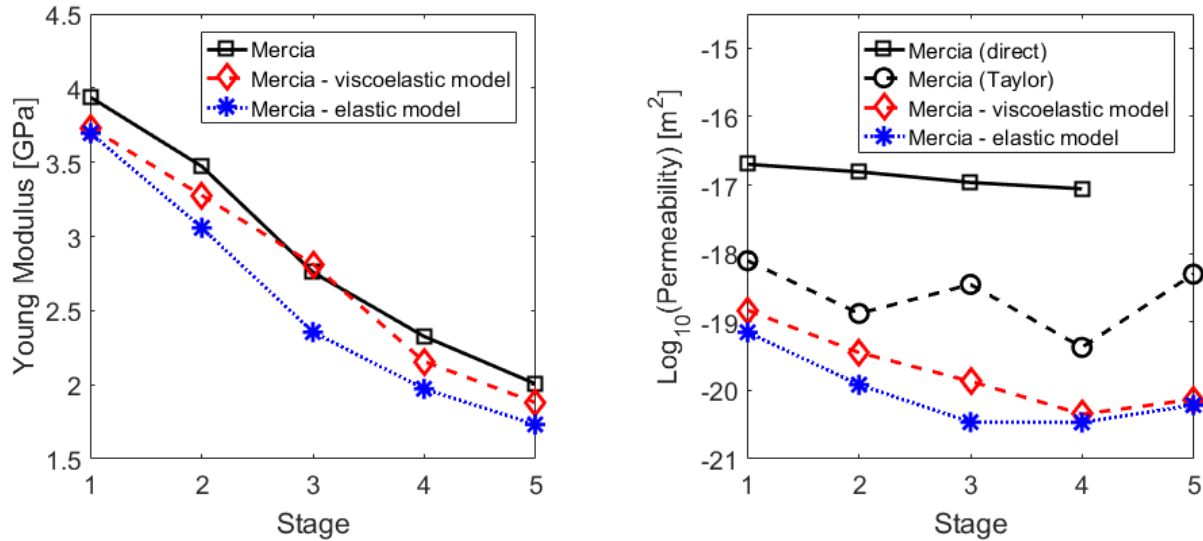
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428



429

Figure 18. Mercia specimen: parameter values determined for each stage.

430

## 431 6 Conclusions

432 Biot's general consolidation theory is here enhanced to include the creep effect observed in

433 experimental tests. The presented model assumes that the fluid flows through a viscoelastic

434 medium, which has been modelled as a purely elastic spring connected in series with a Kelvin-

435 Voigt model (another elastic spring connected in parallel with a dashpot). This is one of the

436 simplest models that predicts an anelastic recovery together with an instantaneous strain. For

437 the sake of simplicity, the elastic moduli of the two springs are here assumed to be equal thus

438 leading to a minimal parametric uncertainty. Indeed, compared to the standard Biot's

439 consolidation model, where two parameters need to be fitted from experimental observations,

440 here three parameters are needed to describe the hydro-mechanical model:

- 441 • Two different parameters control the mechanical response of the material:

- 442           • The elastic modulus of the two springs,  $E$ , which mainly determines the total  
443           volume of fluid expelled after the instantaneous flow expulsion.
- 444           • The dashpot viscosity coefficient,  $\eta$ . The ratio  $\eta/E$  mainly controls the slope of  
445           the time-dependent branch.
- 446           • One parameter (the hydraulic permeability,  $k$ ) controls the transient phase of the  
447           outflow versus time curve. This parameter has the same physical meaning as in the  
448           standard Biot's consolidation model.

449 The clear physical meaning of these parameters has been used here to derive two fitting  
450 algorithms: the former assumes the standard Biot's consolidation model whereas the latter,  
451 with only one extra line of pseudocode, is used for the viscoelastically-enhanced model.

452 The equations for this new model have been presented and implemented here within a finite  
453 element framework. As detailed, the proposed enhancement in the mechanical properties of the  
454 material leads to a time-dependent elastic stiffness tensor and to a time-dependent fluid storage  
455 coefficient. This procedure is thus computationally more demanding, but results in a more  
456 accurate hydro-mechanical model according to the experimental observations from different  
457 consolidation tests performed at the British Geological Survey:

- 458           • The enhanced model is able to better represent the consolidation behaviour of a Boom  
459           Clay sample extracted from the HADES URL at Mol (Belgium). In this particular  
460           example, the standard and new model lead to similar fitted Young's modulus. However,  
461           viscoelasticity leads to significantly improved predicted permeability values, especially  
462           for those stages where the confining pressure decreased.
- 463           • Similar results are obtained when validating against a specimen of the Callovo-  
464           Oxfordian claystone collected from the Meuse/Haute Marne URL (France). The  
465           proposed enhancement leads to better fitting in those cases where the elastic model is  
466           not appropriate and provides a very similar solution when the traditional model is

467 accurate enough. As observed for the Boom Clay sample, this improvement is  
468 especially significant for the permeability parameter.

- 469 • The consolidation experiments conducted on a sample recovered from the Upper  
470 Mercia Mudstone Group formation in the Larne Basin (Northern Ireland) are also better  
471 described with the proposed viscoelastic model rather than the standard Biot's model.  
472 Despite the high heterogeneity, better approximations of the Young's modulus and the  
473 permeability values are obtained if the medium is enhanced with a dashpot viscosity.

474 In all cases considered, this simple approach leads to an improved ability to predict the  
475 mechanical response of clay-based porous materials during loading and unloading. As such,  
476 incorporation of this visco-elastic component to deformation may result in improved  
477 predictions when assessing mechanical performance of natural and engineered barrier materials  
478 in geological applications such as the disposal of radioactive waste and the subsurface storage  
479 of CO<sub>2</sub>.

480

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488

489

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