Gas migration in pre-compacted bentonite under elevated pore-water pressure conditions

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Abstract

Pre-compacted bentonite has long been proposed as a primary component of an engineered barrier system for the safe geological disposal of radioactive waste. Selection of properties such as the clay composition, compaction-state and clay-to-sand ratio vary in different disposal concepts. However, a sound understanding of the gas transport properties of the barrier material is often considered a necessary part of safety case development for a geological disposal facility. In this study, results are presented from two gas injection experiments conducted in Mx80 bentonite, under elevated pore-water pressure conditions. Test observations indicate that the conditions necessary for gas to enter this material are remarkably consistent, irrespective of the applied water pressure. As expected, an association is noted between the total stress experienced by the clay and the gas pressure at the moment of entry. Gas migration is interpreted as occurring by the formation and propagation of dilatant pathways within the bentonite. Local pore-pressure and stress measurements indicate that significant reworking of the clay can occur, resulting in meta-stable episodes of 'pressure-cycling', as gas seeks a stable escape pathway. These findings demonstrate the potential for 'phases' of pathway development and propagation

within the buffer, resulting in successive migration episodes over the repository lifetime. Experiments also show the potential for gas entry into the buffer to occur as a result of declining pore-water pressure conditions. As such, the influence of significant deviations from hydrostatic conditions (for example, resulting from glacial loading) should not be neglected when considering gas interaction with the buffer over long timescales.

Keywords: Bentonite; gas- flow; experiment; pore-water pressure; gas entry/breakthrough; compressibility

1 Introduction

In the geological disposal of high level radioactive waste (HLW), the possibility of gas generation (e.g., due to corrosion of container material/metallic waste) and radiolysis of water, is a common consideration during safety case development. In some cases, the potential for gas generation rates to exceed the rate of dissipation by diffusion must be considered (Horseman et al., 1999; SKB, 2006). Gas pressures may continue to elevate in such a scenario, until either (i) gas generation reduces sufficiently or (ii) a free gas phase is able to enter the surrounding engineered clay or host formation, in which case advective transport may occur (Sellin and Leupin, 2013). It is important to ensure that gas pressure build-up will not result in undesired effects, such as the transportation of radionuclides or mechanical damage to repository infrastructure (SKB, 2006). Given this context, a thorough understanding of the conditions under which advective gas transport will occur and the resulting evolution of such a phenomenon is a necessary consideration in the planning process for a Swedish radioactive waste repository.

The Swedish concept for geological disposal of radioactive waste involves the use of Mx80 bentonite as part of an engineered barrier system (SKB, 2011). Vitrified waste material is enclosed within copper canisters, which are emplaced into a deposition hole within a crystalline host-rock. The void between the host-rock and the canister is then filled with blocks and pellets of pre-compacted bentonite, which provide a number of functions, including: (i) acting as a diffusional barrier surrounding the canister, and (ii) swelling on hydration (leading to the closure of voids and joints within the deposition hole).

In the Swedish radioactive waste disposal concept, the primary pathway for escaping gas will be through fractures present in the hostrock wall of a deposition hole. Characterisation of the granodiorite at the Äspö hard rock laboratory (and during site characterisation at Forsmark and Oskarshamn in Sweden) has shown a proportion of these fractures may be conductive (Nordqvist et al., 2008; Cuss et al., 2010). Such fractures will also enable hydration of the bentonite by the *in situ* groundwater.

Advective migration of a free gas phase through a porous media must involve one of two possible modes of transport: either (i) the gas passes through the original porosity of the clay, causing little disturbance to the surrounding matrix, or (ii) the gas generates new voidage through which it can migrate. The former is generally termed 'visco-capillary flow'. It is controlled by pore throat radii within the clay and the degree of water present within the pores, which must progressively be displaced in order for gas to enter (Marschall et al., 2005). The latter requires an opening (or pathway) to form resulting from applied gas pressure, accommodated for by localised deformation of the surrounding clay matrix (Marschall et al., 2005). This second mechanism is therefore generally termed 'pathway dilatancy'. Since this process essentially involves the separation of two parts of the clay

matrix under the action of an applied stress, it might also reasonably be described as a type of microfracture formation.

In the case of an unsaturated or partially saturated bentonite, a free gas phase is generally expected to migrate by conventional visco-capillary two-phase flow (Villar et al., 2012; Sellin and Leupin, 2013), since higher pressures are not required to mobilise the gas. In such cases, laboratory testing demonstrates that the applied pressure gradient will directly control the resulting gas flow rate (Villar et al., 2012). It has also been suggested that, for sand bentonite mixtures with a suitably high sand content, similar behaviour may also occur in reasonably well-saturated bentonite (Sellin and Leupin, 2013). However, in the case of fully saturated, pure bentonite, water is less mobile and pathway dilatancy is expected to occur instead (Sellin and Leupin, 2013). In such cases, gas migration at *in situ* conditions is not thought to occur unless the applied gas pressure, P_g, exceeds the total stress, σ, experienced by the clay (resulting from the sum of the applied water pressure and the swelling pressure, Π) (Horseman et al., 1999; Harrington and Horseman, 2003). Above this critical value, a mechanical interaction between the gas and clay will begin (Horseman et al., 1999; Harrington and Horseman, 2003; Birgesson and Karnland, 2013; Sellin and Leupin et al., 2013; Tawara et al., 2014), leading to the generation of pathways which allow gas transport. In order to assess the dominant form of advective gas flow in bentonite, it is therefore necessary to conduct laboratory experiments on the selected clay under the relevant boundary conditions for a given repository concept.

There is some degree of inconsistency in the use of certain terminology in relation to gas injection testing and care must be taken to be certain of the chosen terminology when making comparisons between studies. As such, the following definitions given for gas

89 entry, gas breakthrough and capillary entry pressure will be used for the remainder of this
90 paper. Laboratory testing generally comprises of the application of a pressure gradient
91 across the sample, until advective gas outflow is observed at the downstream end of the
92 sample, at some critical value of pressure. This critical threshold is generally known as the
93 'gas breakthrough pressure'. A further critical threshold also exists, defined as the 'gas
94 entry pressure', which is the value at which gas first enters (or is, at least, observed to enter)
95 the material. However, as experimental determination of gas entry can be challenging,
96 laboratory studies often test for the 'gas breakthrough pressure' instead. The influence of
97 declining gas pressure on gas flow is often quantified in the laboratory in terms of the
98 'capillary threshold pressure', which is generally considered as the minimum pressure with
99 which gas can remain mobile within the clay.

100 Experiments conducted at the laboratory scale (Horseman et al., 1997; Harrington and
101 Horseman, 1999; Harrington and Horseman, 2003; Birgesson and Karnland, 2013; Noseck, et
102 al., 2013) demonstrate that for pure, saturated Mx80 bentonite under expected *in situ*
103 conditions, the primary mode of gas transport is dilatant pathway formation. Significant
104 findings that support this conclusion include: (i) a lack of evidence for water displacement
105 resulting from gas entry into the clay, (ii) a strong association between the stress state of
106 the sample and the pressure required for gas to enter the clay (iii) a strong coupling evident
107 between σ, Π and P_g after gas breakthrough has occurred, (iv) localised changes in σ, Π and
108 p_p, associated with the migration of gas, (v) localised outflows during gas breakthrough,
109 which appear non-unique, (vi) the spatial evolution of gas pathways with time, evidenced by
110 unstable flow and, (vii) no measurable desaturation in any samples (post-gas testing),
111 indicating that gas has not passed through the bulk of the clay.

A number of studies also highlight evidence for gas flow by pathway dilatancy in tests involving other argillaceous materials (Ortiz et al., 2002; Angeli, et al., 2010; Cuss et al., 2010; Skurtveit et al., 2011; Harrington et al., 2012a; 2012b), as well as other studies in bentonite (Pusch et al., 1985; Namiki et al., 2014). Wiseall et al. (2015) used a specially designed pressure vessel, with a fused silica window, to directly observe the formation of discrete gas pathways during injection of Helium into kaolinite pastes. They found that these pathways were spontaneously induced above a threshold gas pressure and were observed to propagate and dilate further as gas pressure was increased. While attempts have been made to lay out a theoretical framework for this mechanism of gas flow in bentonite (Horseman et al., 1999), the capacity to generate numerical models which fully reflect the observed behaviours currently remains elusive. To begin to address this issue, two recent numerical approaches have been proposed, one using embedded fractures as conduits for gas flow (Gerrard et al., 2014) and the other linking permeability to void ratio, providing an indirect link to volume change and the stress field (Senger et al., 2014).

The Constant Volume Radial Flow (CVRF) apparatus used in this study was designed to simulate the generation of gas from a canister in the Swedish KBS-3 concept, with the injection rod representing a canister and the additional filter arrays simulating sinks, such as fractures, in the deposition-hole environment. Given that the primary phase of any potential gas generation is expected to occur post-thermal loading (SKB, 2006), clay samples are fully saturated before gas injection testing, as is anticipated in the Swedish concept. Results from previous CVRF experiments provide the basis for a well-developed conceptual model for gas flow in bentonite. Key features of this conceptual model are described in the following section, and are also covered by Harrington and Horseman (2003) and Graham et

al. (2012). In accordance with expected conditions during the most relevant scenario for gas pressure build-up in the current Swedish disposal concept, this behavioural description relates specifically to pure, fully saturated Mx80 bentonite (SKB, 2006).

1.1 GAS MIGRATION IN BENTONITE UNDER EXPECTED HYDROLOGICAL CONDITIONS

For a high-swelling clay such as bentonite, the effective stress, σ_{eff}, can be directly equated with the swelling pressure, Π (Horseman et al., 1996). In a clay–water system with the pore-water in thermodynamic equilibrium with an external reservoir of water at pressure, p_w, the total stress, σ, acting on a constant volume pressure vessel can therefore be expressed in terms of the effective stress law such that:

$$\sigma = \Pi + p_w \quad (1)$$

This is the definition for total stress and swelling pressure used in this study and throughout the remainder of this paper.

1.1.1 Gas entry ($p_g > \sigma$)

In this paper we describe the gas entry phase as the point at which gas is first observed to become mobile within the clay. During laboratory testing, the exact moment of gas entry can be difficult to detect. However, a number of observations may provide evidence that entry has taken place. In particular, a notable deviation in increasing gas pressure from expected ideal gas behaviour must occur, once gas is present in the clay. Evidence from testing conducted by Harrington and Horseman (2003), and published in Graham et al. (2012) clearly demonstrates that during gas injection testing under a constant backpressure condition, gas entry is only detected after the applied gas pressure, p_g, exceeds the total stress, σ, experienced by the bentonite (Figure 1). In reality, this relates specifically to the

157 local stress experienced by the clay at the point of gas entry. However, even in a relatively
158 heavily-instrumented laboratory experiment, total stress measurements may differ
159 somewhat from those precisely where gas entry occurs. Nevertheless, the association is
160 strong enough that it is generally apparent, even when using an average of the total stress
161 values measured within the clay (making this value a useful comparator). The importance of
162 locally measured stresses is more apparent at the larger scale and the association between
163 these and gas entry has been demonstrated at the Lasgit full scale gas injection test at Äspø,
164 Sweden (Cuss et al. 2014).

165 At the laboratory scale, observations also indicate that even, if significant quantities of gas
166 are passed through the clay over long periods of time (and at relatively high pressure), very
167 little water is displaced out of the bentonite during gas flow testing (Harrington and
168 Horseman, 2003). These findings are also consistent with the assertion that advective
169 migration of gas occurs through localised pathways, leading to little or no desaturation and
170 a strong association between the excess gas pressure at entry and the average total stress
171 experienced by the bentonite, which must be exceeded in order for dilatant pathways to
172 propagate (Pusch and Forsberg, 1983; Harrington and Horseman, 1999).

173 Where the bentonite is constrained under a constant volume boundary condition, pathway
174 propagation should result in significant perturbation of monitored stresses and pore-water
175 pressures (Fig. 1) and such changes have, indeed, been observed during this phase of gas
176 migration in CRVF experiments (Harrington and Horseman, 2003; Graham et al., 2012).
177 These perturbations display time-dependent behaviour, characteristic of pathway
178 propagation. Monitored total stresses may also display step-like responses, as pathways
179 grow during the approach to breakthrough (Graham et al., 2012). Localised consolidation

may be ongoing in the near-field of opening gas pathways, which would explain observations of small-scale hydrodynamic effects that perturb the local pressure distribution within the clay.

While the point of gas entry in all tests displays a predictability in relation to stresses within the clay, the pressure at which gas breakthrough may occur is highly variable in comparison. This is not unexpected, given the constant volume boundary condition and the moment of breakthrough should intuitively be related to the number/geometry (or 'availability') of sink filters and the compressibility of the clay. Such effects are exacerbated in the laboratory, which may lead to the observation of high gas pressures before breakthrough (Graham et al., 2012). However, at the field scale the greater compressibility of the clay, combined with the presence of interfaces and heterogeneities, is likely to moderate extremes in behaviour and markedly reduce the likelihood of higher gas pressures, as is observed at the Lasgit field-scale gas injection experiment (Cuss et al., 2011; Graham et al., 2012). Nevertheless, the influence of sink availability and homogeneity/compressibility of the clay on gas migration behaviour should be noted. Such factors have the potential to significantly impact the maximum gas pressure likely to be sustained by the buffer at the full canister scale.

1.1.2 Gas breakthrough ($p_g \geq \sigma$)

Gas breakthrough occurs once a gas conducting pathway comes into contact with a sink (at laboratory scale this is a filter, but could represent a void, opening, or fracture at field scale) and is manifested by a notable outflow of gas at the backpressure pump. Gas may be mobile within the clay, but until breakthrough is reached gas pressure may continue to rise, whilst a stable outflow path is sought. Once this occurs, gas injection pressure is seen to

spike (Fig. 1b), displaying the characteristic first breakthrough peak in pressure reported in earlier studies (Harrington and Horseman, 1999; Horseman et al., 1999; Horseman et al., 2004). These observations can be explained by a breakdown in the tensile strength of the bentonite, leading to the sharp pressure drop after the peak (Fig. 1).

After peak pressure has been reached, p_g is observed to drop close to σ, such that;

$$p_{g\ (post-breakthrough)} = \sigma \approx \Pi + p_{wi} \quad (2)$$

In the post-gas breakthrough phase, multiple observations demonstrate a pronounced coupling between p_g, σ and internal water pressure within the clay, p_{wi} (Harrington et al., 1999; Harrington and Horseman, 2003; Cuss et al., 2011; Graham et al., 2012). Likewise, monitored outflow of gas at this stage is often localised, with a non-uniform outflow distribution along the sample length. Furthermore, the distribution of flow is often seen to change abruptly and spontaneously, as localised flow alternates from one location to another (Harrington and Horseman, 2003). These findings imply an inherent instability in the nature of these gas pathways, which results in dynamic flow behaviour as their spatial distribution evolves.

1.1.3 Shut-in response

After gas breakthrough, the shut-in response of the clay can be determined by stopping the injection pump and allowing the gas pressure and resultant outflow to decay with time. As in- and out-flow cease, p_g approaches an asymptotic value that is must relate to the minimum pressure at which gas is mobile in the clay (Fig. 1c). Horseman et al. (1997) and Harrington and Horseman (2003) attributed observations during shut-in to the collapse of conducting pathways in response to declining pressure, leaving a series of discontinuous,

'remnant' gas-filled zones within the clay (Fig. 1c). At this stage, the capillary pressure, p_c, can be considered as the difference between the residual gas pressure, p_g, and the externally applied water pressure, p_w:

$$p_c = p_g - p_w \quad (3)$$

1.2 GAS MIGRATION IN BENTONITE UNDER ELEVATED PORE-WATER PRESSURE CONDITIONS

Under normal hydrostatic conditions, a strong association is apparent between the pressure at which gas entry occurs and the sum of the p_w and Π. However, a number of possible mechanisms may lead to noticeable deviations from such conditions during the lifetime of a repository. For example, in more Northern regions of Europe the influence of glacial loading must be considered, as it is known to influence subsurface pore-water pressure (Wildenborg, et al. 2003). Graham et al. (2014) presented experimental data indicating that such deviations may lead to a small persistent elevation in swelling pressure, even when returned to hydrostatic conditions. The cause of this hysteric behaviour remains unclear, though two possibilities include: (i) heterogeneity in the hydration behaviour of Mx80 bentonite, limiting the rate at which hydraulic equilibrium is reached (Cuss et al., 2011), and (ii) permanent alteration of the clay from a double-structured to single structured microfabric, as a fully saturated state is reached (Seiphoori et al., 2014). An improved understanding of the likely impacts of this phenomenon on gas-transport characteristics of the bentonite would therefore be beneficial in assessing gas flow behaviour during pore-pressure excursions. To examine this aspect of gas flow in bentonite, the experiments presented in this study were conducted at high pore-water pressure conditions on samples previously subjected to a series of load-unload cycles (Graham et al., 2014).

Most gas injection experiments reported in the literature generally involve applying an excess gas pressure to a bentonite sample and increasing the pressure gradient until gas migration by advection takes place. However, given the association between gas entry pressure and total stress (itself related to the applied water pressure), it is also conceivable that gas entry may be instigated by a local decline in pore-water pressure. The experiments presented in this study were, therefore, conducted so as to explore the potential for gas entry and breakthrough to occur as a result of declining pore pressure and any associated differences in the resulting flow behaviour.

Data were collected from two constant volume laboratory tests investigating this scenario, as part of the EC FP-7 project Fate of Repository GasEs (FORGE). Further description of these experiments and the wider test programme are given by Graham and Harrington (2014). In contrast to the conceptual model described in Section 1.1, gas injection was carried out at elevated pore-pressure conditions and breakthrough was instigated by dropping the applied backpressure, simulating a decline in pore pressure after prior elevation. In this paper we summarise the gas migration behaviour observed and make comparison to that previously reported at expected pore-water conditions.

2 Methodology

2.1 EXPERIMENTAL APPARATUS

In order to mimic realistic down-borehole conditions, this study was focussed on laboratory testing of bentonite under a constant volume boundary condition (as would be expected in a granodiorite deposition-hole). The experiments described here were carried out using a bespoke constant volume and radial flow (CVRF) apparatus, based on a design initially

produced to examine the sensitivity of gas flow in compacted bentonite to the test boundary conditions (Harrington and Horseman, 2003). The CVRF apparatus consists of: (1) a thick-walled stainless steel pressure vessel, (2) a fluid injection system, (3) three independent backpressure systems, each consisting of an array of four filters acting as fluid sinks, (4) five total stress sensors to measure radial and axial stress and (5) a logging system. The steel vessel mimics the rigid deposition-hole, while the filter arrangement provides the opportunity for fluid escape, as with natural fractures cross-cutting the deposition-hole.

The pressure vessel (Figure 2) comprises a stainless-steel, dual-closure, tubular vessel whose end-closures are secured by twelve, high tensile cap-screws that can also apply a small pre-stress to the specimen if required, using a gauged torque-wrench. A central injection rod allows fluid to be injected directly into the middle of the test sample. Filters are also housed in the end-closures at both the injection rod end of the vessel and the downstream end. The position of the sink arrays (p_p[1], [2] and [3]) and the stress sensors ($\sigma_{Ax\ Injection}$, $\sigma_{Ax\ Backpressure}$, σ_{rad1}, σ_{rad2}, σ_{rad3}) are shown in Figure 2. The radial arrays contain sintered, high density, polyethylene plugs and are each connected to a separate pressure transducer. Each array can also be isolated from the backpressure system and used to provide an independent measure of the local pore fluid pressure, or can be connected to the backpressure pump in order to monitor outflow. Stress measurements are made using push-rods that are each in direct contact with a miniature load cell. The central filter is embedded at the end of a 6.4 mm diameter stainless steel tube which can be used to inject the permeant, for gas flow testing purposes. The swelling of the clay samples on hydration is sufficient to seal the injection rod, which is clearly demonstrated during testing by the

ability to maintain high applied gas pressures with no inflow, when gas testing is initiated (see Sections 2.1 and 2.2)

The pressure and flow rate of test fluid is controlled and monitored using two, high-precision syringe pumps. Volume, flow rate and pressure data are then transmitted from each pump to a bespoke National Instruments logging system. Additional test parameters are logged simultaneously by the same system and the typical acquisition rate for all data is one scan every two minutes. All stress and pore pressure sensors were calibrated against laboratory standards by applying incremental steps in pressure, from atmospheric to a pre-determined maximum value, greater than that intended during experimentation. This was followed by a descending history to quantify any hysteresis, which was negligible.

Before injection, gas is allowed to equilibrate with water in an upstream interface vessel, ensuring that it is saturated before injection. During gas injection testing, helium was used as the permeant, due to its non-reactivity and similar molecular size to hydrogen. Because of the need to measure very small flows, testing was carried out under room temperature conditions maintained within an environmentally-controlled room, in order to minimise thermal fluctuations (± 1 °C). It should be noted that a greater degree of noise is generally apparent for upstream flow measurements, compared to those at the downstream end. This is to be expected and reflects the large gas volume being regulated by the upstream pump, in comparison to a water reservoir at the downstream pump. Often, the downstream pump exhibits a higher degree of noise once gas breakthrough has taken place and this is apparent in some of the findings presented later in this paper (Section 2.5).

2.2 SAMPLE PREPARATION AND PROPERTIES

Mx80 bentonite is a fine-grained sodium bentonite, from Wyoming (United States of America), which contains around 90% montmorillonite. Blocks of pre-compacted Mx80 bentonite were manufactured by Clay Technology AB (Lund, Sweden), by rapidly compacting bentonite granules in a mould under a one dimensionally applied stress (Johannesson et al., 1995). Two cylindrical specimens with a diameter of 60 (± 0.5) mm and length of 120 (± 0.5) mm were sub-sampled from the pre-compacted blocks. The samples were turned on a lathe, in order to provide consistent dimensions along their entire length. A 6.35 (± 0.5) mm diameter hole was then drilled into the centre of the material to accommodate the stainless-steel injection rod. The injection filter imbedded in the rod tip was machined to match the drill bit used, in shape and dimension, so as to achieve a tight fit. Care was taken to calculate the loss in clay volume as a result. Standard geotechnical properties for both samples were then calculated (pre- and post-test) and are shown in Table 1, alongside values from previous test programmes. The water content of the specimens was determined by weighing pre-test, then oven-drying (to greater than 105 °C for more than twenty-four hours) post-testing and weighing a second time. The void ratio, porosity and degree of saturation are based on an average grain density for the bentonite of 2.77 $Mg.m^{-3}$ (an average value, as measured by Karnland et al., 2010).

Samples Mx80-13 and Mx80-14 were tested under elevated pore-water pressure conditions, so as to compare behaviour with previous gas tests conducted under expected hydrostatic conditions (Harrington and Horseman, 2003; Graham et al., 2012). Geotechnical properties for these previous tests (Mx80-8 and Mx80-10) are given in Table 1, for easy comparison with the samples used in this study.

2.3 PORE PRESSURE CYCLING

Bentonite samples Mx80-10, Mx80-13 and Mx80-14 were first subjected to a pore-pressure cycling phase, in advance of gas injection testing. Hydration behaviour, swelling response and additional findings from these tests are presented by Graham et al. (2014). For reference, this part of the test history is briefly given in Table 2. This paper is focussed on the subsequent gas testing phase for samples Mx80-13 and Mx80-14.

2.4 MX80-13

Sample Mx80-13 was of notably higher density than the other bentonite samples (Table 1), most likely as a result of its slightly lower saturation state on receipt, pre-sample preparation. This initial state is consistent with a significantly higher than normal cumulative inflow observed during initial hydration of the sample (~9.5ml) and led to the generation of much larger total stresses during sample swelling as a result (Graham et al., 2014). Before gas testing, Mx80-13 was subject to incremental pore-pressure loading up to 42MPa, then unloaded to 1MPa (Table 2).

The sample was then reloaded and allowed to equilibrate at an elevated applied water pressure of 42MPa. Monitored data for the subsequent gas testing stage are shown in Figure 3. All filters were isolated from the backpressure pump, except the central filter array (p_p [2]), allowing the local evolution of pore fluid pressure to be monitored at separate locations within the clay. Water was then swapped for helium gas as the permeant and extreme care was taken to flush any residual water from the injection rod filter, whilst at pressure. At day ≈138 an initial gas pressure was applied to the sample through the central

injection rod and maintained at a constant value of 42MPa (so as to allow equilibration) for a period of approximately 11 days.

The injection pump was then switched to introduce gas at a constant flow rate of 40µl/hr (day 149), in order to generate a gradually increasing excess gas pressure with time. Over the following 18.6 day period (Figure 3a) a steady increase in locally measured pore fluid pressures was detected in the two isolated arrays (p_p [1] and p_p [3], as well as at the downstream (backpressure) end-closure. This gradual increase was also observed in the associated stresses along the length of the sample, though close inspection of the data at this time indicated that changes in pore pressure were the driving force.

In order to establish the nature of this flow, the injection pump was returned to constant pressure as it reached a value of 52MPa. At this stage, pore-water pressure was seen to continue to increase for several days after gas pressure was held constant. A gradual decline in pore-pressure was then observed over a period of more than 60 days, at a rate approximately three times faster than a much smaller associated decline in monitored total stresses. Since such behaviour is not expected for gas flow, this instead implies a hydro-dynamic cause and is consistent with the slow progression of a small amount of water being forced from the injection filter and into the body of the clay (despite careful flushing). These observations highlight the importance of hydrodynamic effects when monitoring such small changes in flow. In this case, the effect seems likely to be the result of 'slug flow', where small amounts of residual water remaining in the injection filter are displaced into the clay as gas pressure increases. Such behaviour would explain the observation of declining pore pressures, whilst a constant gas pressure is maintained.

379 After slug flow had ceased, the pump was returned to a constant flow of 40µl/hr (day 247)
380 and excess gas pressure began to increase until it had just superseded the monitored
381 average total stress (at around day 270). At this stage gas pressure was close to the
382 maximum specifications for the CVRF apparatus and the rate of increase in p_g could no
383 longer be maintained (Figure 3a). The injection pump was then reduced (at day 291.8) to a
384 constant gas pressure of 64MPa (Fig. 3b). The outflow response observed during this initial
385 gas stage was seen to be very small (40 µl in around 155 days) and indicates that, even at
386 these high pressures, gas did not enter the clay up to the value of the total stress.

387 In order to induce gas flow, the applied water pressure (shown here at the central array, p_p
388 [2]) was dropped from 42MPa to 37MPa at this stage (day 293.2) (Fig. 3b). The resulting
389 pore pressure decline in the rest of clay was seen to be exceptionally slow and the filters in
390 array 1 (p_p [1]) and both end-closure filters were opened to the backpressure pump at day
391 325, to speed the equilibration process (p_p [3] was kept isolated so as to continue
392 monitoring pore pressure evolution within the clay). This resulted in a small outflow in the
393 downstream pump as the system re-equilibrated, followed by a gradual decline in
394 monitored pore pressure within array p_p [3]. In spite of this, twenty days after the applied
395 water pressure was dropped, no notable outflow had been observed, although monitored
396 total stress did decay somewhat in response to the reduction in applied water pressure (Fig.
397 3d). As such, the applied backpressure was dropped a further 5MPa at day 325.3 (Fig. 3c).
398 The result was an almost immediate gas breakthrough (monitored pore-water pressure, p_p
399 [3], had only fallen by ~0.4MPa), indicating that the clay had already been very close to gas
400 entry under these conditions.

401 In response to the constant gas pressure condition, gas flow was rapid after breakthrough
402 with 25.3 litres of gas (at standard pressure and temperature) being injected into the clay
403 over 15.7 hours. As a result, the injection pump ran out of fluid at day 325.9, causing inflow
404 to cease and leading to a drop in the applied excess gas pressure. However, significant
405 outflow continued until around day 330 (Fig. 3c). This behaviour would not be expected for
406 standard visco-capillary flow, but is instead consistent with the closure of gas-filled
407 pathways as internal p_g declines. Gas pressure was 64MPa (~8.5 MPa above the average
408 total stress) at the point of breakthrough, but by the time outflow had ceased, the final gas
409 pressure at the end of the test stage had declined to around 58 MPa (<2MPa above the
410 average total stress) (Fig. 3d). Given an applied water pressure of 32MPa, this equates to an
411 apparent capillary threshold pressure of ~26MPa.

412 Whilst the sink geometry at breakthrough makes the path taken by outflowing gas unclear,
413 a number of observations suggest that the primary mechanism for gas migration during this
414 test was by pathway dilation. In particular, the estimated post-test saturation of the sample
415 Mx80-13 (Table 1) shows that the bentonite experienced very little desaturation during gas
416 flow, in spite of more than 25 litres passing through the clay during breakthrough. Given an
417 estimated pore volume for sample Mx80-13 of 128 µl (>0.5% of the total outflow volume
418 detected at the downstream filter), it is exceptionally difficult to explain the saturated
419 sample state, post-testing, using standard concepts of visco-capillary flow (which require a
420 significant desaturation of the clay in order for gas to migrate). These findings instead
421 indicate that highly localised gas flow must have occurred, thereby limiting the volume of
422 clay exposed to gas during transport through the sample. This finding and the observation
423 that gas entry occurred relatively close to the value of the average total stress and decayed

close to this value during shut-in, directly contradicts behaviours expected for visco-capillary flow, where hydromechanical coupling (excluding poroelasticity) is not involved. Instead, such behaviour is highly consistent with gas flow along localised, propagating pathways, which will interact with the local stress field. Given that the former mechanism is associated with gas flow through less than fully saturated bentonite or clay/sand mixtures, whilst the latter is associated with pure, fully saturated bentonite (see Section 1), this finding is entirely as might be expected. It is, therefore concluded that pathway dilation was the primary mode of gas migration in this experiment.

2.5 MX80-14

As with sample Mx80-13, the gas injection phase of testing for Mx80-14 was carried out following hydration and a programme of pore-pressure cycling (Table 2; Graham et al., 2014). After two load-unload pore-pressure cycles, the applied water pressure was taken once more to an elevated value (41MPa) and allowed to equilibrate. Monitored data for the gas testing stage of the experiment are shown in Figure 4. An excess helium gas pressure of 1MPa (42MPa in total) was then applied through the injection rod, directly into the middle of the bentonite sample (Fig. 4a, stage (1)). So as to provide observations of the pore pressure evolution during this period, all filters were isolated from the backpressure pump except those within the central filter array (monitored by p_p [2] and plotted as applied water pressure), which were used to maintain the applied water pressure boundary condition. At this stage the applied gas pressure was 5MPa below the average total stress measured within the sample. These conditions were maintained for a period of more than 50 days, during which time the monitored outflow response was negligible. No other detectable

446 signs of gas entry were observed over this period of testing, in spite of the high gas pressure
447 condition.

448 In order to determine the impact of pore pressure decay, the applied backpressure was next
449 reduced to 32MPa at day 225 (Figure 4a, stage (2)). Independent pore-pressure
450 measurements were seen to decay as a result, as were the monitored total stresses within
451 the clay. There was no evidence of gas entry into the clay during this time, including no
452 signficant outflow being detected. At day 244.4, the average total stress fell below the
453 value of the applied gas pressure. However, clear evidence of gas entry was not apparent
454 until p_g had fallen to the value of the highest monitored total stress (at the backpressure
455 end-closure), which occurred on day 253. Two days subsequent to this, the presence of gas
456 within the bentonite was indicated by a sudden increase in locally measured pore pressure
457 at the injection end-closure filter (Fig. 4a). The measured pressure rose to a value close to
458 the applied gas pressure at this time, and was approximately equal in magnitude to the
459 average total stress in the clay (Figure 4a and b). Local hydrodynamic responses were also
460 observed in the pore pressures monitored at all other local filters at this time. The pressure-
461 pulse at the injection filter was then seen to decay over the subsequent day, demonstrating
462 the capacity for self-sealing within bentonite.

463 This initial pressure-pulse was then followed by a series of further pulses of decreasing
464 maximum amplitude (Fig. 4b). An associated, but less pronounced, response was also
465 observed in the monitored total stresses within the clay, indicating that these fluctuations
466 were pressure driven. These signals were most prominent for the sensors closest to the
467 injection end-closure filter (σ_{Ax} injection and also σ_{rad1}), where full gas pressure had
468 previously been observed. At the same time, very small inflows were seen to correlate with

469 the onset of the pressure pulses (Figure 4b), though no comparable outflows were detected.
470 This phase of 'quasi-stable' pressure cycling continued for over 30 days and is interpreted as
471 a series of pathway opening events, which were unsuccessful in reaching a sink (Fig. 4c,
472 stage (2)). Such prominent perturbation of the stress field during gas flow is entirely
473 inconsistent with visco-capillary flow, but is well explained by the formation of dilatant
474 pathways (Section 1). The frequency of these oscillations remained unstable, whilst the
475 peak amplitude was seen to reduce with each successive event, until a phase of negligible
476 observable activity was reached.

477 As such, a series of decreasing steps in applied water pressure (each a 5MPa reduction)
478 were instigated (Fig. 4c, stages (3) to (5)) in order to encourage gas breakthrough. Each step
479 was held constant until the monitored local pore pressures had fully equilibrated (a
480 minimum of 3 weeks in each case), with the final step being to an applied water pressure of
481 17MPa. However, the system displayed remarkable quiescence during this test stage,
482 highlighting the ability of bentonite to sustain significant pressure gradients (25MPa for
483 stage (5)) for substantial periods of time without major outflows and breakthrough
484 occurring. This observation is consistent with other recent findings demonstrating the
485 potential to sustain large pressure differentials in a range of clay-rich porous materials (Cuss
486 et al., 2012; Cuss et al., 2014). At approximately day 392 (stage (5)), after 15 days with no
487 change in boundary condition and apparent system quiescence, the monitored pore
488 pressure at the injection end-closure was seen to increase sharply to the value of the
489 applied gas pressure. At the same time, a small inflow was noted at the upstream pump.
490 The inherent time-dependency of this behaviour can be explained by the development of a
491 gas-filled pathway/s within the sample. Unlike the previous event, this pressure elevation

492 remained close to p_g, indicating the presence of an open and quasi-stable gas pathway,
493 close to the injection end-closure filter. However, no notable change in outflow was
494 observed to correlate with this event, suggesting that any gas-filled pathways had yet to
495 intersect a filter in communication with the downstream pump (Figure 4d).

496 After a further period of negligible system activity, a significant outflow was detected at the
497 central array on day 413.8 (Figure 4d). Local pore-pressure in the other two isolated filter
498 arrays (p_p [1] and p_p [3]) was seen to elevate to applied gas pressure at this time, indicating
499 the development of a gas flow pathway along the length of the sample. This highlights the
500 potential for the clay to sustain high pressure gradients, as it implies that the majority of the
501 pressure drop along this transmissive pathway must occur close to the point where drainage
502 takes place. The detected outflow from the mid-array was observed to reduce dramatically
503 soon after and this behaviour also correlated with an associated decay in pore-pressure at
504 the neighbouring sink arrays, which was particularly pronounced for sink array p_p [1]. These
505 observations are consistent with the opening and fragmentation/closure of localised gas-
506 filled pathways within the clay and highlight the rapid self-sealing response of bentonite
507 under these conditions.

508 This event was followed by a series of highly repeatable pressure pulses, measured in filter
509 array p_p [3], as well as associated outflows (Figure 5a). Over time, the period between
510 pressure pulses was observed to increase and outflow reduced (Fig. 5a). The clear
511 repeatability in the onset pressure of each pulse demonstrates a pressure dependency to
512 this phenomenon and suggests an ongoing working of the clay, resulting in a reduction in
513 compressibility with time. Such behaviour appears to be the result of ongoing gas-driven
514 deformation of the clay, as an exit from the system is sought. Figure 5b shows the

515 associated changes in locally measured stresses during this episodic behaviour and indicates
516 that whilst, soon after gas entry, stresses were strongly coupled with the local pore-pressure
517 within the clay, by day 410 only very small perturbations were observed in relation to
518 comparatively large changes in the local pore pressures. As with observations at the field-
519 scale (Cuss et al., 2014), this decreased sensitivity of measured stresses to pressure
520 development suggests a transition to a regime controlled by the local gas pressure and is
521 highly indicative of a network of transient fractures, allowing accommodation of pressure
522 variations. At approximately day 465, pressure in the filter array p_p [3] returned to the gas
523 injection pressure and remained at this value, indicating a stable pathway from the injection
524 point to a filter in this array had formed.

525 Approximately 10 days later, two successive, quasi-stable periods of outflow were observed,
526 indicating that the ability of the clay to maintain open pathways was developing further.
527 Smaller fluctuations in the monitored pore pressure in filter array p_p [1], in association with
528 these events, suggest a gas path to the neighbouring sink array p_p [2] had formed, most
529 likely by way of array p_p [3]. However, the clay was still observed to self-seal effectively not
530 long afterwards and steady-state flow was never achieved, despite the sample being
531 subjected to significantly high gas pressures for more than 300 days. From the first
532 observations of gas entry, perturbations within the clay were seen to be highly localised and
533 seem to indicate changes in the gas distribution within the clay, as pathways formed and
534 closed. This and other observations relating to gas migration processes within this test are
535 not well explained by standard visco-capillary flow concepts, in particular the evolving
536 periodicity noted during episodes of pressure cycling within the clay. However, findings are
537 highly consistent with behaviours expected during the advective transport of gas by dilatant

538 pathway formation. The instability and episodic nature of gas migration observed in this
539 experiment is discussed further in the following section.

540 3 Discussion

541 **3.1.1 Pre-gas entry ($\sigma > P_g$)**

542 Testing was carried out at elevated water pressure for samples Mx80-13 and Mx80-14,
543 resulting in high total stresses before the injection of gas began. During the pre-gas entry
544 phase no notable outflows were detected that could be attributed to displacement of pore
545 water by visco-capillary action, in either test. Observations from these experiments also
546 highlight the exceptionally high pressure gradients that Mx80 bentonite is capable of
547 sustaining, without hydraulic outflow or gas breakthrough occurring.

548 **3.1.2 Gas entry ($\sigma \leq P_g$)**

549 Gas entry was reached in both tests by applying a constant gas pressure at the injection rod
550 filter and reducing the applied backpressure to the sample. The resulting decline in locally
551 measured pore pressures within the clay led to an associated reduction in total stress. In
552 both experiments, gas entry was only observed to occur once the measured average total
553 stress fell below the applied gas pressure. In this respect, the behaviour of saturated Mx80
554 bentonite at elevated pore-water conditions appears to be similar to findings under
555 hydrostatic conditions (Harrington and Horseman, 2003).

556 As in previous tests, observations after gas entry also display characteristics of a highly
557 localised process, especially for Mx80-14. During this period of testing, typical observations
558 include marked deviations from p_w of the monitored local pore pressures, which are
559 interpreted as being perturbations in the vicinity of propagating pathways. As with previous

experiments, geotechnical measurements were made for both samples, before and after testing (Table 1). The resulting data show that, in spite of the high applied gas pressures and extensive periods of gas flow during later stages of gas testing (see Section 2.4), no significant desaturation of the samples occurred. Such behaviour is in-line with observations that relatively little free water is likely to be present in bentonite under these conditions (Bucher and Müller-Vonmoos, 1989). These observations are also consistent with localised, as opposed to distributed, flow through the bulk of the clay and provide further explanation for the lack of significant water outflow before gas is seen to enter the clay. The lack of desaturation and localised changes in pore fluid pressures and stresses within the sample are inconsistent with expectations for visco-capillary flow, but are well-explained by gas migration via the formation of dilatant pathways.

3.1.3 Gas breakthrough ($\sigma \leq P_g$)

During the gas breakthrough phase, a number of important differences were observed between the two experiments. For sample Mx80-13, breakthrough occurred soon after a step down in the applied backpressure and directly resulted in a continuous outflow from the clay. Unlike more conventional tests, the characteristic peak in total stress and local pore pressure was absent, though a marked peak in flow rate did take place at this time. The form of this response appears to be characteristic for gas breakthrough instigated by declining pore-water pressure, under a constant p_g condition (Fig. 6). In the case of Mx80-13, breakthrough occurred seemingly spontaneously, after a significant period with boundary conditions held constant. The resulting breakthrough led to a small, transient outflow, followed by relatively rapid self-sealing (on the order of days). The differences between these tests could be the result of (i) the notable discrepancy in dry density

583 between the two samples (which would influence system compressibility), (ii) small
584 differences in sample responses during the pore-pressure cycling phase (Graham et al.,
585 2014), (iii) the selected geometry of the hydration and outflow sinks, (iv) pre-existing
586 weaknesses or heterogeneities within the clay, or (v) a combination of several of these
587 factors. However, an alternative explanation is that these differences simply reflect the
588 innate variability in the gas breakthrough process. Whilst findings from all CVRF tests to
589 date suggest that gas entry is a highly predictable process (see Section 3.1.4), repeatability
590 of gas breakthrough pressures has yet to be demonstrated. Nevertheless, it seems
591 intuitively likely that less working of the clay would occur, in advance of gas breakthrough,
592 for the higher density (and less compressible) sample, Mx80-13.

593 Although clear differences were seen between these two experiments, very high differential
594 pressures were required in order to instigate breakthrough in both tests. This is intuitively
595 likely, in part, to be the result of the selected sink geometry (and the associated distance to
596 the nearest sink), but could also be a consequence of breakthrough having been instigated
597 by a reduction in a previously elevated applied water pressure. Nevertheless, when gas
598 pressure was allowed to decay after breakthrough of sample Mx80-13, it was observed to
599 asymptotically approach a value close to the average total stress. This behaviour is
600 consistent with the conceptual model for gas flow at expected hydrostatic conditions
601 (Section 1.1.3) and implies that gas breakthrough at elevated pressures is similarly closely
602 linked with the stress state of the sample.

603 Steady-state flow proved considerably more difficult to achieve for sample Mx80-14. Here
604 gas flow was observed to be highly episodic and unstable in nature, alternating from one
605 state to another in an apparently spontaneous fashion. A high propensity to self-seal was

606 also evident in the post-entry behaviour of the clay. This phase of episodic events displayed
607 a strong cyclicity, with the pressure-controlled behaviour of the system being highly
608 apparent (Fig. 5). Despite this seemingly repetitive behaviour, an overlying temporal
609 evolution in response was also observed, which displayed characteristics consistent with a
610 continued working of the clay, until a more stable outflow could finally take place.

611 However, since this behaviour was not observed in both tests it seems likely that the
612 elevated pore-water pressure condition is not the primary cause of this behaviour. A more
613 plausible control is the influence of sink filter geometry in these tests, as in both cases only
614 one filter array was made available as a sink for gas escape, reducing the likelihood of a gas
615 pathway intersecting a sink and resulting in an inherently unstable system. Similar
616 observations have been made at the Lasgit field test, where gas was detected migrating
617 around the canister interface before it could eventually escape (Cuss et al., 2014). The
618 reduced compressibility of sample Mx80-13 might also explain the lack of a similar
619 reworking occurring before major breakthrough, resulting from the notably higher initial
620 density in this test.

621 The cyclic behaviour observed in test Mx80-14 would, therefore, represent an 'end-
622 member' phenomenon; the consequence of a semi-compressible system where gas escape
623 has not yet become possible. If correct, this implies that such behaviour may well also occur
624 on long timescales in a repository, for example in a situation where flow pathways are so
625 long that they are inherently unstable and unable to maintain a continuous pathway for
626 drainage of gas. Nevertheless, the controls influencing the duration and likelihood of this
627 form of behaviour and the approach to steady-state flow, require further investigation.

3.1.4 Implications

Dilatancy has been included in some recent numerical models, in order to represent the processes involved more accurately (Gerard et al., 2014; Senger et al., 2014, Tawara et al., 2014). However, there is currently a paucity of data relating to the spatial distribution and temporal evolution of gas pathways involved in this process (SKB, 2006), which is limiting further numerical model development at present (Harrington et al., 2012). Given the requirement to image gas pathways within a representative volume whilst under pressure, progress is challenging. An alternative approach is to utilise the observed threshold dependency of gas migration during repository design, so as to mitigate against this eventuality. High precision experiments, monitoring multiple parameters independently, have been used to develop a detailed conceptual model relating gas entry to total stress within the clay (Section1.1; Harrington and Horseman, 2003). This model exhibits a high degree of predictability and is consistent with findings in this and previous studies (Pusch et al., 1985; Pusch and Forsberg, 1983). A cross-plot showing key parameters for several Mx80 bentonite gas injection tests is shown in Figure 7. Data here are compiled from this study, from previous work of Harrington and Horseman (2003) and from the full-scale gas injection test Lasgit, at the Äspø Hard Rock Laboratory in Sweden (Cuss et al., 2014). Despite the different test conditions (and dry densities) covered by the experimental data used, an association between gas entry and stress state is highly apparent.

A significant amount of experimentally-generated data and associated process understanding is now available to numerate this conceptual model. Such a model provides a sound foundation from which to draw on when attempting to understand and the conditions under which gas migration might occur in a repository. Evidence from laboratory

testing indicates that, were such migration to occur, it is unlikely to lead to the displacement of large volumes of water (which would be unfavourable should soluble radionuclide release from the wasteform have taken place).

There is, however, one potential exception to this, which cannot be excluded at the present time. It is conceivable that the application of high gas pressures, over significant periods of time, could possibly lead to bulk compression and drainage of the clay (Harrington and Horseman, 2003; Sellin and Leupin, 2013). The potential for this phenomenon is currently not well understood, especially since the effect is only likely to be of significant magnitude over long timescales. However, one favourable aspect of the material performance over the long term is its propensity to 'self-seal' after a gas migration event. The laboratory experiments outlined in this paper demonstrate this capacity, with clay seemingly recovering its barrier characteristics rapidly, even between individual gas migration events. Harrington and Horseman (2003) also observed that this behaviour was further assisted by rehydration of the clay, which notably reduces prior 'memory' of gas migration and restores the clay close to its original state.

Findings presented here suggest that the relationship between gas entry pressure and locally measured total stresses is consistent with the conceptual model for gas entry given at lower pore-pressures (Section 1.1), irrespective of previous test history. Observations in both these experiments indicate that the subsequent advective transport of gas is also consistent with this model, and occurs via the propagation of localised, dilatant pathways. It, therefore, seems unlikely that excursions from expected hydrostatic conditions in the repository will impact on the current model for gas entry behaviour of bentonite. One consequence of this which should be noted is that during such an excursion, a bentonite

674 barrier will be capable of sustaining higher gas pressures before entry takes place. This
675 capacity is likely to continue over the timescales necessary for the clay to return to hydraulic
676 equilibrium. However, it is much less trivial to draw conclusions on the influence of prior
677 pore-pressure cycling upon the behaviour of migrating gas, once it has entered the clay.
678 The differences in the approach to gas breakthrough in these two tests make comparison
679 particularly difficult, especially considering repeatability of behaviour at this stage of gas
680 flow is not easy to achieve, even in tests where initial conditions and materials are
681 comparable. However, it is conceivable that any change in compressibility of the clay
682 resulting from loading may influence the approach from gas entry to breakthrough.

683 Observations relating to the post-gas entry phase provide insight into the evolution of the
684 system as it approaches breakthrough. In particular, a progressive time-dependent
685 evolution is observed during this phase, resulting from a re-working of the clay until a more
686 stable flow regime can be established. If the clay is relatively incompressible under the
687 applied pressure conditions, this phase may be short and less apparent before breakthrough
688 occurs. However, if the clay is able to accommodate a degree of compression and gas is
689 unable to find a sink, a constant pressure condition alone may be enough to energise the
690 system sufficiently for continued evolution. This behaviour may be sustained for significant
691 periods of time, while gas continues to seek a sink, and can result in seemingly spontaneous
692 changes (though it is likely this is a result of limitations in the number of points of
693 observation within the material). This alternation in system behaviour, from one meta-
694 stable state to another, is clearly gas pressure-driven and appears to be highly unpredictable
695 in nature. Observations suggest that any subsequent decline in ambient pore-water
696 pressure also has the potential to further destabilise the system and lead to additional

episodes of pathway propagation, though this is not a necessary condition for such behaviour. It is not yet clear whether repeated breakthrough events may lead to progressive reworking of the clay over long timescales. Findings from these experiments and those of Harrington and Horseman (2003) indicate that the presence and distribution of sinks (ie. conductive fractures/faults) within the deposition-hole will, therefore, be of primary importance to the duration and nature of this phase of gas migration, both under normal and elevated p_w conditions.

4 Conclusion

A primary objective of this work is to examine the influence of sustained periods of pore pressure elevation and consequent decline upon the swelling and gas flow properties of a bentonite buffer. Pore pressure cycling experiments were carried out on two Mx80 bentonite samples under constant volume conditions. Both samples were subjected to gas injection at an elevated pore pressure condition, following a phase of pore-pressure cycling. The consequent observations indicate that characteristics of the gas entry process are similar to those observed in comparable experiments conducted at lower applied water pressures. As with tests conducted under more conventional boundary conditions (see Section 1.1), there is an association between gas pressure and total stress after gas breakthrough. Unlike the majority of gas injection experiments on bentonite, gas entry was instigated by maintaining a constant gas pressure and reducing the applied water pressure incrementally. The pressure at which the onset of gas entry occurred was consistent with previous observations at hydrostatic conditions and appears to be relatively predictable.

As with previous studies conducted at lower pore pressures, the findings from these experiments are best explained by dilatant pathway formation as the primary mechanism

720 for gas flow through the clay. In contrast, standard concepts of visco-capillary flow cannot
721 explain observed hydromechanical coupling or a lack of desaturation of the clay after
722 testing.

723 Observations from both tests highlight the capacity of Mx80 bentonite to sustain very large
724 stress gradients over significant periods of time. In one test, gas entry and breakthrough
725 occurred close together, with very little evidence of reworking of the clay being apparent.
726 In the other test, highly episodic and unstable gas flow was observed, displaying meta-stable
727 qualities. This behaviour is interpreted as a consequence of gas being unable to escape,
728 whilst the system remains energised. Such behaviour is consistent with the development of
729 temporally evolving and potentially spatially unique gas pathways, suggesting that the time
730 to breakthrough and the associated breakthrough pressure may not be repeatable, even for
731 the same test methodology. Whilst, this behaviour was not observed for the other test, it is
732 proposed that these observations result from ongoing deformation of the clay, whilst gas is
733 unable to escape the system, rather than being a consequence of the elevated pore
734 pressure history. This hypothesis is an intuitive one, since meta-stability is a likely feature of
735 such conditions in bentonite, where the propagation front of a gas pathway is the most
736 unstable part of the system (Harrington and Horseman, 2003). This would seem to indicate
737 that the degree of compressibility of the clay may be an important control on the approach
738 to gas breakthrough in the buffer.

739 Findings from this study highlight the remarkable consistency of gas flow behaviour for
740 pure, saturated bentonite, with stress conditions playing a crucial role irrespective of pore-
741 water pressure conditions. Laboratory evidence also indicates the potential for gas entry to
742 occur as a result of declining pore-water pressure in a repository environment. Whilst the

743 controls on this behaviour appear no different from a more conventional approach to
744 breakthrough, the form of the resulting response differs somewhat, with the characteristic
745 break-through 'peak-pressure' no longer apparent. Perhaps more crucially, these findings
746 demonstrate that deviations in pore water pressure (for example, resulting from glacial
747 unloading) represent a potential mechanism for gas entry into the buffer and must
748 therefore not be neglected when considering gas generation scenarios in a repository
749 environment.

750

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876 **Tables:**

877 **Table 1** – Geotechnical properties of the test specimens, before and after testing.

Sample		Before testing					After testing
	Study	Water content (%)	Bulk density ($Mg.m^{-3}$)	Dry density ($Mg.m^{-3}$)	Void ratio	Saturation (%)	Saturation (%)
Mx80-8	*Harrington and Horseman (2003)*	26.7	1.997	1.577	0.756	97.6	≥100[1]
Mx80-10	*Harrington and Horseman (2003); Harrington and Birchall (2007); Graham et al. (2014)*	26.7	2.005	1.582	0.751	98.6	≥100[1]
Mx80-11	*Harrington and Birchall (2007); Graham et al. (2014)*	25.6	2.016	1.605	0.726	97.6	≥100[1]
Mx80-13[2]	*This study; Graham et al. (2014)*	20.1	2.064	1.718	0.612	91.1	≥100[1]
Mx80-14	*This study; Graham et al. (2014)*	26.6	1.999	1.579	0.754	97.7	≥100[1]

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[1] Measured value indicates sample was fully saturated, within uncertainty limits of the measurement.
[2] Mx8-13 was sub-sampled from a pre-compacted bentonite block with a notably lower initial saturation, leading to a higher starting dry-density than the other test specimens.

Table 2 – Test history for the samples subjected to pore-pressure cycling (reported by Graham et al., 2014), before gas injection testing.

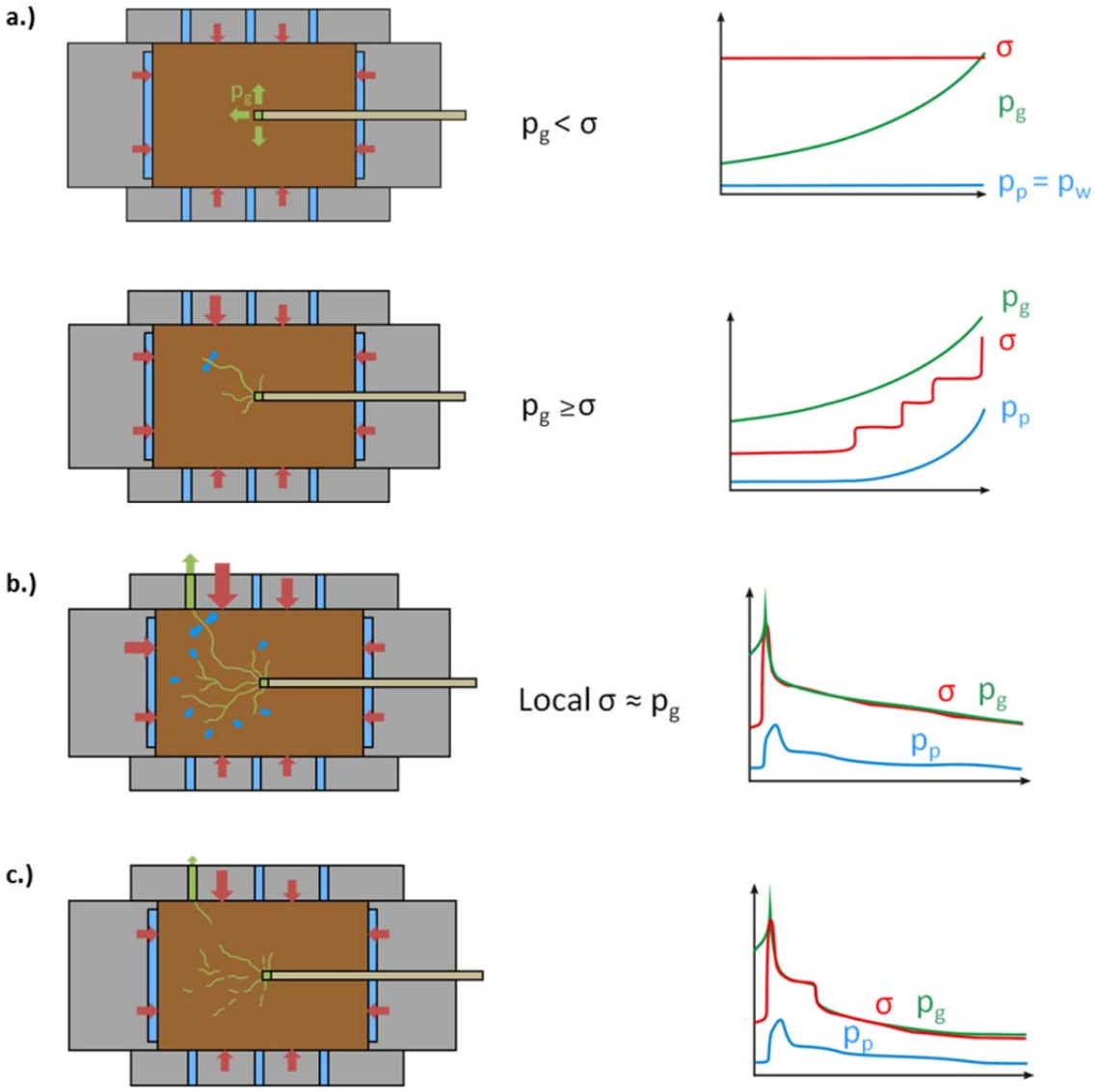
Sample	Mx80-10		Mx80-13		Mx80-14	
Stage	Applied p_w (MPa)	Ave. total stress (MPa)	Applied p_w (MPa)	Ave. total stress (MPa)	Applied p_w (MPa)	Ave. total stress (MPa)
1	1.0	6.3	1.0	22.1	0.9	6.7
2	2.0	7.0	6.0	26.6	11.0	16.2
3	3.0	7.9	24.1	43.1	20.9	25.8
4	4.0	8.7	42.2	60.0	10.8	16.0
5	5.0	9.7	24.1	45.7	1.0	6.9
6	6.0	10.6	6.0	29.4	20.7	25.5
7	7.0	11.5	1.0	25.2	30.7	35.1
8	4.0	9.6	6.0	27.9	40.7	45.0
9	1.0	7.4	24.1	43.3	30.8	36.0
10	---	---	42.2	59.6	20.8	26.4
11	---	---	---	---	1.3	7.0

Table 3 – Gas injection test history for samples Mx80-13 and Mx80-14. EQ = equilibration, CP = Constant pressure, CF=Constant flow rate, SI = gas injection stopped and pressure allowed to decay.

Sample	Mx80-13				Mx80-14			
Stage	Stage type	Applied p_g (MPa)	Gas flow rate (µl/h)	Applied p_w (MPa)	Stage type	Applied p_g (MPa)	Gas flow rate (µl/h)	Applied p_w (MPa)
Equilibration	EQ	---	---	42	EQ	---	---	41
1	CP	42	---	42	CP	42	---	41
2	CF	---	40	42	CP	42	---	32
3	CP	52	---	42	CP	42	---	27
4	CF	---	40	42	CP	42	---	22
5	CP	64	---	42	CP	42	---	17
6	CP	64	---	37	---	---	---	---
7	CP	64	---	32	---	---	---	---
8	SI	57[3]	---	32	---	---	---	---

[3] At end of shut-in, once further pressure decay was negligible.

900 **Figures:**



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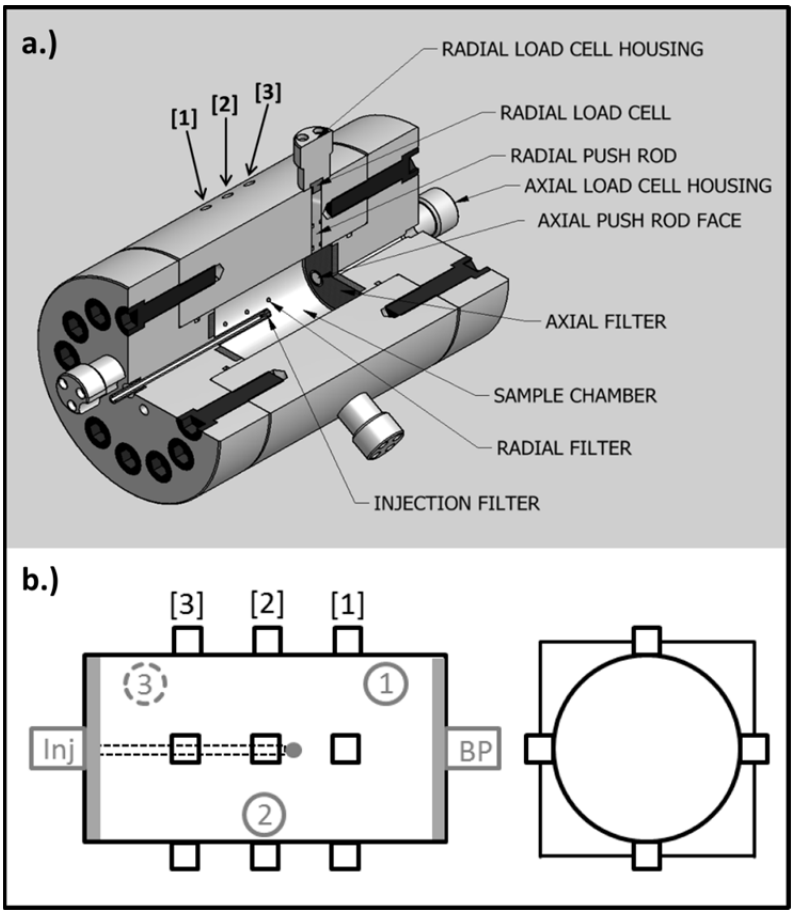
902 **Figure 1** – Schematic representation of gas injection in Mx80 bentonite under a constant volume
903 boundary condition.

904 a.) Gas entry under a constant flow rate gas injection. The internal pore-pressure, p_p, is initially
905 equal to applied water pressure, p_w. Entry only occurs once gas pressure exceeds the total
906 stress, σ, experienced by the bentonite. Consolidation may also occur in the near-field of
907 propagating pathways, leading to small-scale hydrodynamic effects in their vicinity.

b.) Gas breakthrough response under constant flow rate gas injection. Breakthrough occurs when a propagating gas pathway intersects a sink, allowing major outflow to occur and causing the applied gas pressure to peak. This characteristic response is followed by gas pressure falling to the value of the locally measured total stress, as gas pathways stabilise and steady-state flow is achieved.

c.) Shut-in response as constant gas flow is halted and pressure allowed to decline. As gas pathways collapse, outflow downstream is seen to fall, whilst declining stresses and gas pressure asymptote towards a value which is close to the sum of the swelling pressure and the applied water pressure. Residual gas is left in localised regions of the bentonite and may well impact the peak pressure required to achieve breakthrough in following gas injection episodes.



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924 **Figure 2** – The Constant Volume Radial Flow (CVRF) cell in a.) cutaway, showing the two end-closures
925 and drainage filters, the central fluid injection filter, the input ports for the radial sink arrays and the
926 push rod arrangement, and b.) sensor locations: (i) axial total stress at the injection and
927 backpressure end-closures (greyrectangles) – σ_{Ax} Injection, σ_{Ax} Backpressure, (ii) radial total
928 stressesin three independent locations (grey circles) – σ_{rad1}, σ_{rad2}, σ_{rad3}, (iii) porous filters embedded
929 in the injection and backpressure end-closures (shaded grey rectangles) and the tip of the gas
930 injection rod (shaded grey circle) are connected to independent pore pressure transducers – p_p
931 Injection, p_p Backpressure and p_g (gas) and (iv) three sink arrays (black squares), each linking four
932 porous filters to a pore pressure transducer – Pp[1], [2], [3].

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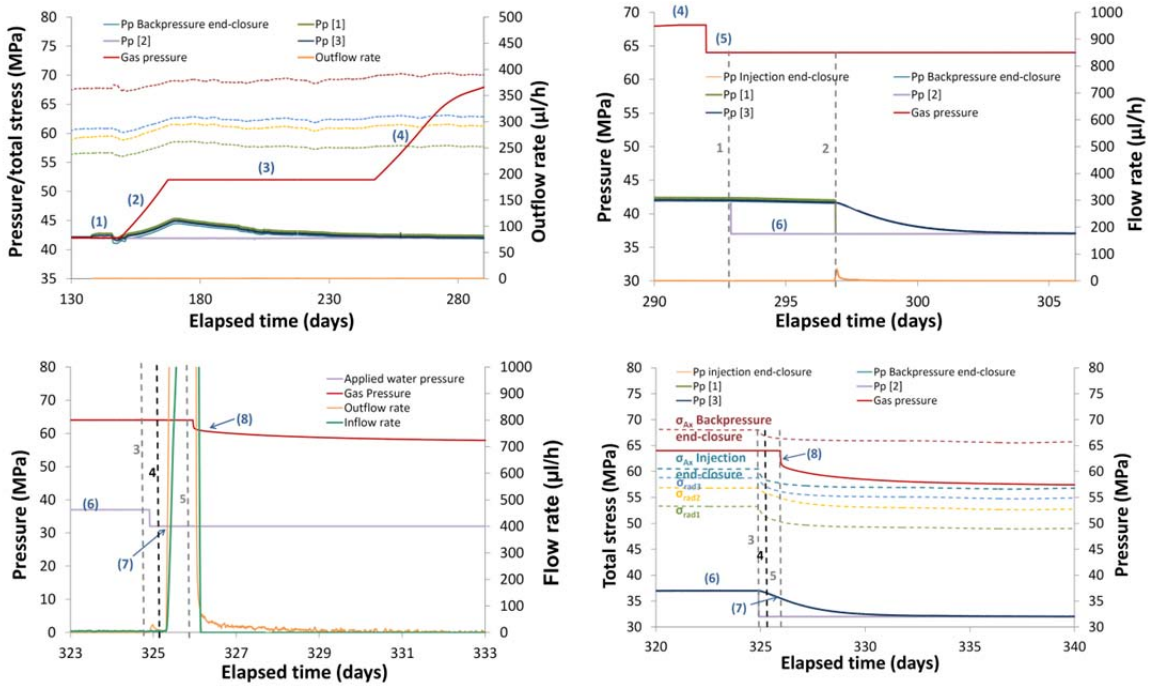

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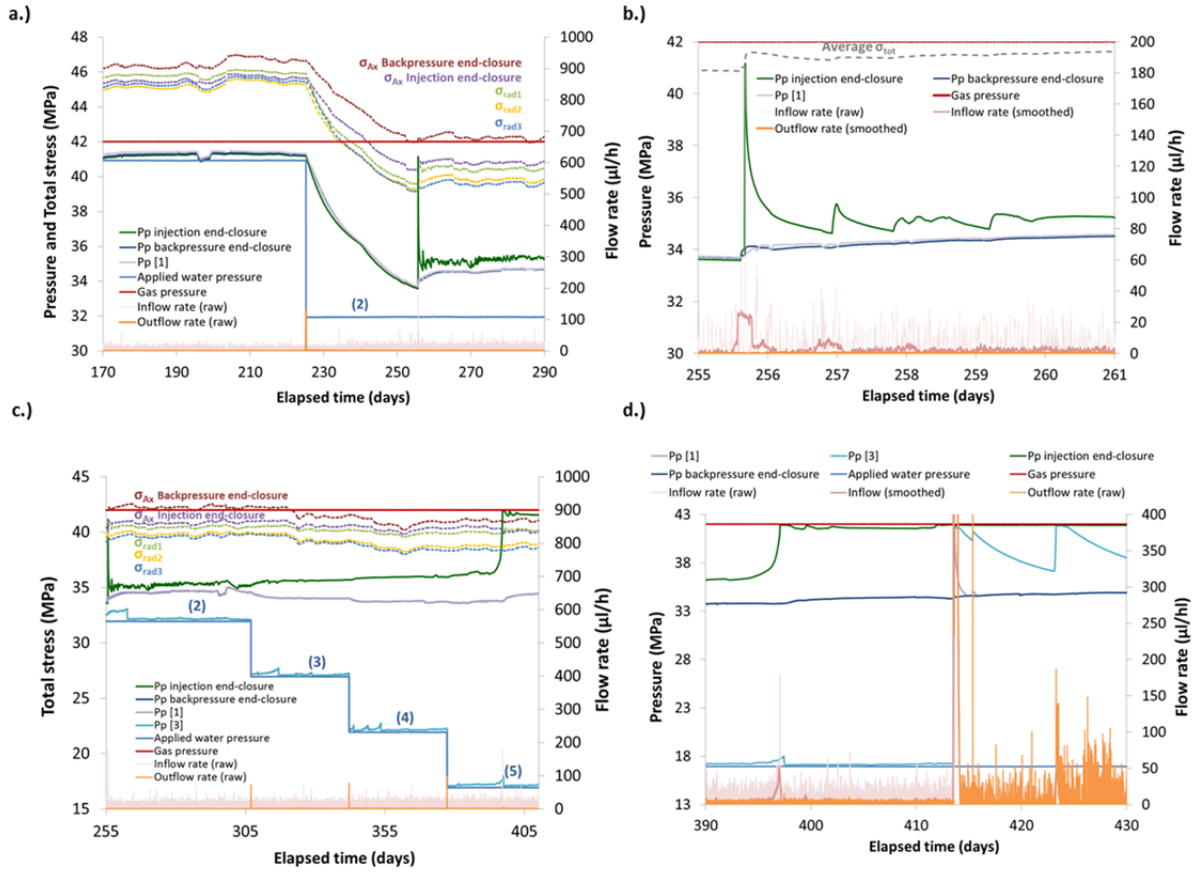
935 **Figure 3** – Sample Mx80-13. a.) System response to an applied gas pressure ramp. An increase in
936 locally measured pore pressures is attributed to the occurrence of 'slug flow'. Pore pressures slowly
937 returned to the value of the applied water pressure (p_p 2), after gas pressure was held constant. A
938 second pressure ramp, led to a decline in the rate of pressurisation, as gas pressure reached the
939 maximum specifications of the apparatus. b.) Local pore pressure decline, resulting from a reduction
940 in the applied water pressure (p_p [2], dashed line 1) was slow and the filters in array 1 (p_p [1]) and
941 both end-closure filters were opened to the backpressure pump, sepeeding equilibration (dashed
942 line 2). c.) Gas breakthrough occurred by day 325 (dashed line 4), after applied water pressure was
943 reduced (dashed line 3). The injection pump ran out of fluid at day 325.9 (dashed line 5) andinflow
944 ceased, resulting in a drop in applied gas pressure and a reduction in outflow. d.) Stress decline
945 following a step down in applied water pressure (dashed line 3), leading to gas breakthrough
946 (dashed line 4). Once gas pressure was allowed to decay (dashed line 5), it approached a value close
947 to the average total stress in the clay.



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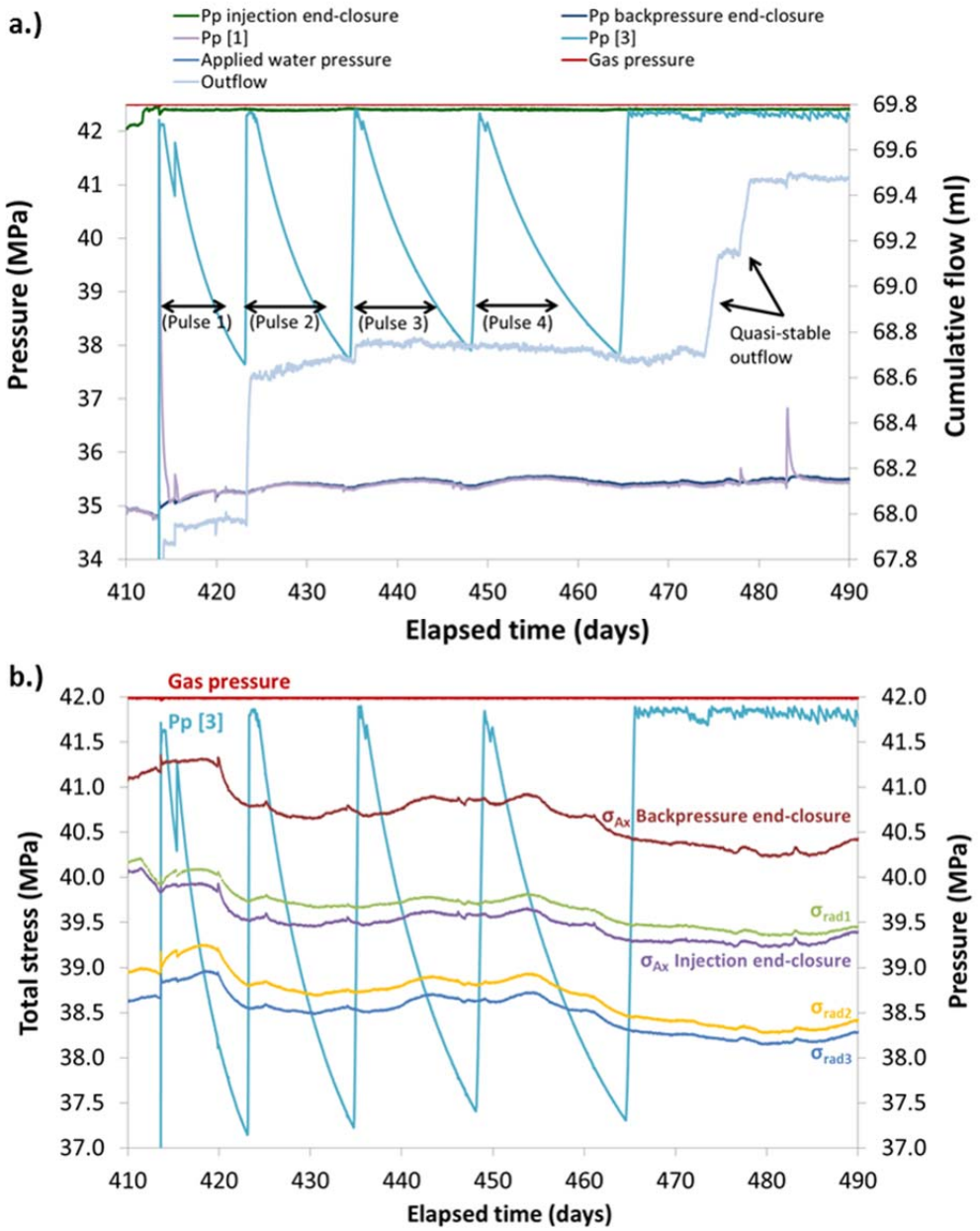
949 **Figure 4** – Sample Mx80-14. a.) Test history for the first 80 days of gas injection. Gas entry was first
950 detected when gas reached the injection end-closure filter (day ~ 255), b.) Gas injection history,
951 post-entry into the bentonite. The clay sustained significant pressure gradients (up to25MPa for
952 stage (5)) with no major outflow occurring, c.) At gas entry, small inflows correlate with pressure-
953 pulses, d.) Pore-pressure in arrays p_p1 and p_p3 reach gas pressure, before decaying.



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955 **Figure 5 -** Sample Mx80-14. a.) The initial gas breakthrough event. A transient outflow occurred
956 apparently spontaneously (day 413.8). The clay quickly began to self-seal and outflow ceased. A
957 phase of pressure cycling followed, with associated transient outflows and subsequent self-sealing.
958 Gas migration during this phase was highly episodic, displaying extreme cyclicity and temporal. b.)
959 Variation in local stresses during pressure cycling.

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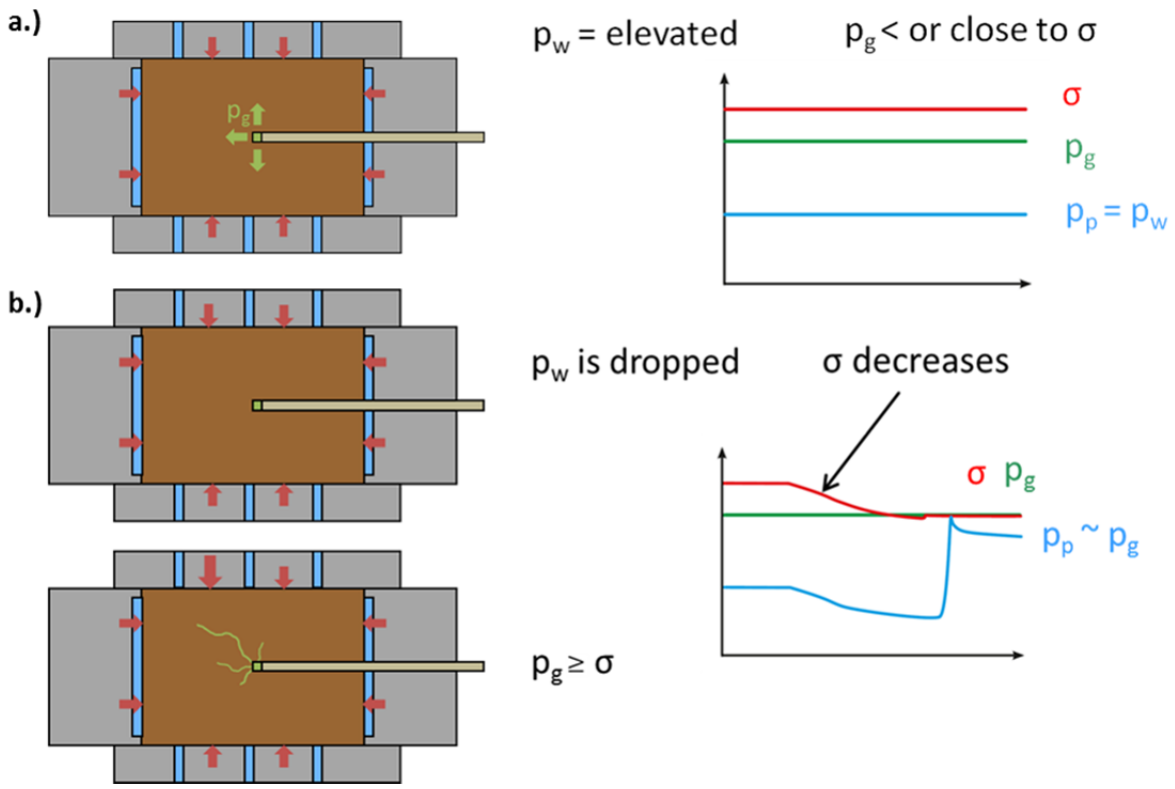

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963 **Figure 6** – Schematic representation of gas entry under a constant applied p_g condition and at
964 elevated p_w conditions. a.) Initial state with gas pressure held constant, at or below the total stress.
965 b.) Gas entry instigated by dropping p_w and allowing stresses to decline. Entry only occurs once
966 local total stress falls below the applied gas pressure. Pressure cycling and episodic behaviour may
967 occur during this phase as the clay is 'worked' to form stable pathways. No evidence was found for
968 elevated pore-water conditions impacting on the gas entry behaviour of the bentonite.

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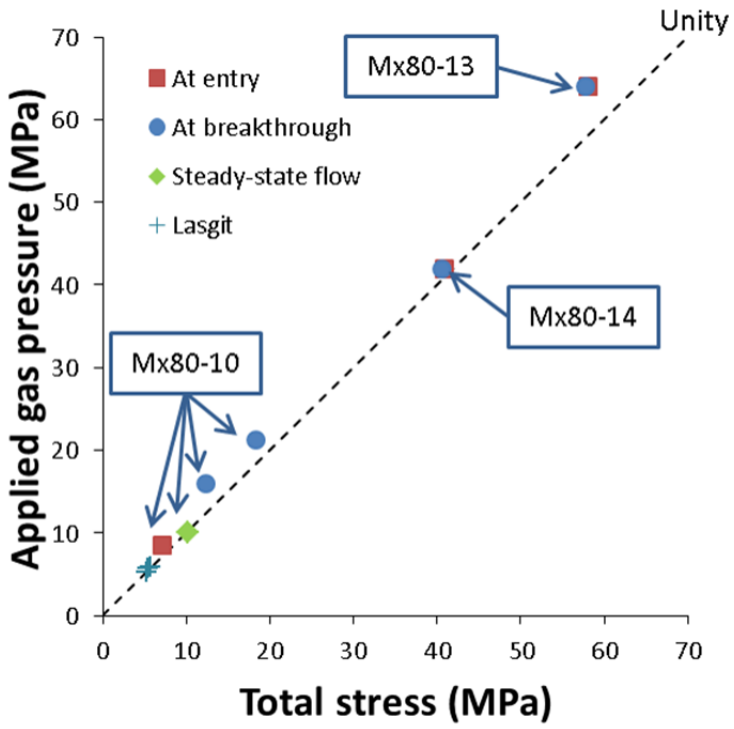

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976 **Figure 7** – Cross-plot showing key parameters for several gas injection tests. The general coupling

977 between gas pressure and total stress is clearly evident. Once steady-state flow is achieved, p_g falls

978 closer to σ. The dotted line represents unity.