ARTICLE IN PRESS

Applied Clay Science xxx (xxxx) xxx–xxx

Contents lists available at ScienceDirect

Applied Clay Science

journal homepage: www.elsevier.com/locate/clay

Research paper

Gas network development in a precompacted bentonite experiment: Evidence of generation and evolution

Jon F. Harrington[a,*], Caroline C. Graham[a], Robert J. Cuss[a], Simon Norris[b]

[a] British Geological Survey, United Kingdom
[b] Radioactive Waste Management Ltd., United Kingdom

ARTICLE INFO

Keywords:
Bentonite
Advection
Multi-phase flow
Deformation
Permeability
Radioactive waste

ABSTRACT

In a deep geological disposal facility for radioactive waste, precompacted bentonite is proposed as a sealing material for the isolation of boreholes, disposal galleries and deposition holes. The advective movement of repository gas in bentonite has been linked to the development of new porosity and propagation of dilatant pathways. For the first time we present a detailed analysis of stress field data during the generation and evolution of a gas network. A new experimental dataset, from a highly instrumented test, clearly shows the strong coupling between stress, gas pressure and flow in bentonite. Multiple discrete propagation events are observed, demonstrating spatial variability and time-dependency as permeability within the clay develops. Analysis of the stress data before, during and after gas entry indicates a heterogeneous stress field initially develops, resulting from the development of these pathways. The flow network is dynamic and continues to spatially evolve after gas entry, such that permeability under these conditions must be time-dependent in nature. Perturbation of the stress field is significant before all major gas outflow events, presumably resulting from the requirement to propagate an effective gas network before outflow is possible. In contrast, no major flow perturbations are detected which did not correlate with fluctuations in the stress field. The controls on the distribution and geometry of the resulting flow network are unclear, as well as its long-term evolution and stability. These will be beneficial in the assessment of gas pressure evolution as part of safety case development.

1. Introduction

The deep geological disposal of radioactive waste presents a number of significant engineering challenges, not least understanding the fate and impact of waste-package derived gas on the engineered barrier systems (EBS) and host rock, which form an integral part of a geological disposal facility (GDF). Corrosion of ferrous materials under anoxic conditions, combined with the radioactive decay of waste and radiolysis of water, lead to the formation of hydrogen, carbon dioxide, hydrogen sulphide and methane within a repository depending on the waste composition, availability of water and disposal concept. Determination of the primary mode of gas migration is a complex issue, dependent on both repository concept and evolution. As such, there remains a degree of uncertainty as to the relative importance of diffusion versus advection. However, in scenarios where the rate of gas production exceeds the rate of gas diffusion through the EBS or host rock, a discrete gas phase will form (Weetjens and Sillen, 2006; Ortiz et al., 2002; Wikramaratna et al., 1993, Sellin and Leupin, 2013; SKB, 2006; Norris, 2015). Under these conditions, a free gas phase begins to accumulate until its pressure becomes sufficiently large for it to move, through advection, in the surrounding material (Sellin and Leupin, 2013; Graham et al., 2012; Harrington and Horseman, 1999, 2003; Horseman et al., 1999). Previous studies (Angeli et al., 2009; Skurtveit et al., 2012; Harrington et al., 2009, 2012a,b; Cuss et al., 2014a,b; Gerard et al., 2014; Rodwell, 2000) indicate that in the case of plastic clays (Whitlow, 2001) and in particular precompacted bentonite, advective gas flow is associated with the development of new pressure induced pathways leading to a complex coupling between gas pressure, stress state and volumetric strain (Gensterblum et al., 2015; Amann-Hildenbrand et al., 2015; Cuss et al., 2014a,b; Harrington et al., 2012a, 2012b; Graham et al., 2012; Horseman and Harrington, 1994; Horseman et al., 1999; Harrington and Horseman, 2003; Romero et al., 2012; Marschall et al., 2005; Wiseall et al., 2015).

The phenomenon of dilatant flow is not new to geoscience. In early 1971, two French researchers Tissot and Pellet (1971) examining the mechanisms controlling primary hydrocarbon migration stated “The displacement of an oil or gas phase from the centre of a finely grained argillaceous matrix goes against the laws of capillarity and is in

---

[*] Corresponding author.
E-mail address: jfha@bgs.ac.uk (J.F. Harrington).

http://dx.doi.org/10.1016/j.clay.2017.07.005
Received 23 January 2017; Received in revised form 3 July 2017; Accepted 5 July 2017

Please cite this article as: Harrington, J.F., Applied Clay Science (2017), http://dx.doi.org/10.1016/j.clay.2017.07.005

principle impossible. The barrier can, however, be broken in one way. The pressure within the fluids formed in the pores of the source-rock increases constantly as products of the evolution of kerogen are formed. If this pressure comes to exceed the mechanical resistance of the rock, microfractures will be produced which are many orders of size greater than the natural (pore) channel of the rock, and will permit the escape of an oil or gas phase, until the pressure has fallen below the threshold which allows the fissures to be filled and a new cycle commences." This hypothesis was supported by Mandl and Harkness (1987) who, independently of Tissot and Pellet (1971), suggested hydrocarbon migration only occurs through thick, continuous water-wet rocks of low permeability through a process of fracturing, forming what they call 'dykelets'. Studies on subsea hydrocarbon seepages by Clayton and Hay (1992) and Judd and Sim (1998) suggest capillary displacement pressures are often so large, that the gas pressure required to initiate flow can approach or even exceed the local stress. These observations were supported by Donohew et al. (2000) who, examining gas migration processes in unconfined clay pastes of varying moisture content and mineralogy, observed the creation of dilatant, preferential pathways, the morphology of which was related to the plasticity and density of the clay.

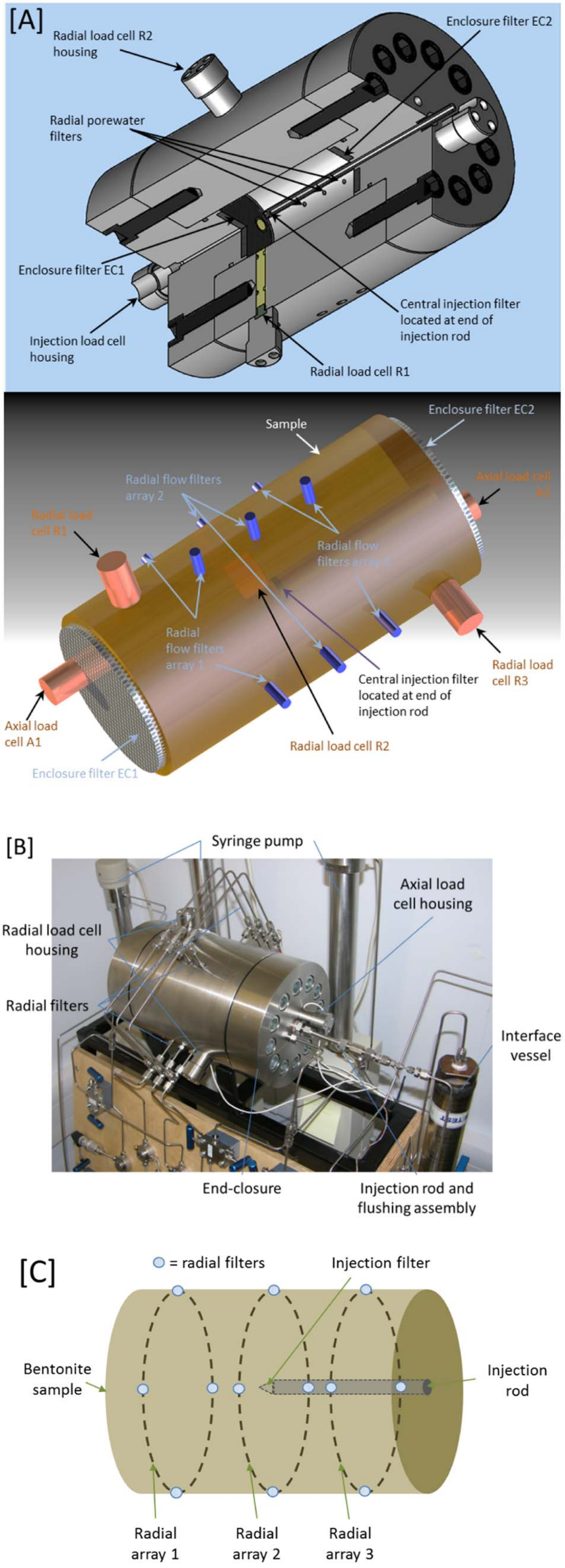
Despite the evidence for pathway dilation and sealing, the exact mechanisms controlling gas entry and flow in clay-rich media remain poorly understood and the memory[1] of such pathways and their potential impact on barrier performance is uncertain.

This paper describes a highly instrumented and detailed test examining the interaction between gas pressure and stress during initial pathway generation and the development of permeability in an EBS consisting of saturated, precompacted bentonite. A detailed analysis of the stress field during gas network development is presented and the implications for radioactive waste disposal discussed.

2. Apparatus

Conceptually, the apparatus reproduces some of the main features of the repository near-field within a hard host rock, including the deposition hole, a corroding canister generating gas and a number of conductive fractures in the host rock. Tests are performed in a constant volume apparatus which is a direct analogue for a radioactive waste repository within a hard (e.g. crystalline) host rock. The unyielding walls of the host rock confine the EBS which is used to encapsulate high-level radioactive waste containers (Sellin and Leupin, 2013). This boundary condition was selected for this test programme as it represents the favoured disposal concept in both Finland and Sweden, the two countries most advance in Europe in their development of an operational GDF.

In the configuration used in this study, there are five main components: (1) a thick-walled dual-closure stainless steel pressure vessel (representing the walls of the deposition hole), (2) a fluid injection system (simulating the generation of gas within the bentonite), (3) three independent backpressure systems (simulating conductive features intersecting the deposition hole), (4) five stress sensors to measure radial and axial stress and (5) a LabView™ based data acquisition system. Fig. 1A is a cut-away section showing both end-closures with their embedded drainage filters, EC1 and EC2 and axial stress sensors A1 and A2, the central fluid injection filter, the twelve radial sink filters, the three radial stress sensors (R1, R2 and R3) and the porewater pressure sensor. The central or "source" filter is mounted at the end of a 6.4 mm diameter stainless steel tube and is used to inject the permeant at the mid-point of the sample, either helium[2] or distilled water



(caption on next page)

---

[1] The term memory is used to describe a propensity for the re-establishment of a pathway at the same location despite prior closure.

[2] While hydrogen will be the primary gas generated in a GDF for high level waste and/or spent fuel, helium was selected as a safe substitute based on its inert nature and similar molecular diameter.