Applied Clay Science 129 (2016) 47–53

Contents lists available at ScienceDirect

Applied Clay Science

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

Research paper

Gas permeability evolution mechanism during creep of a low permeable claystone

Z.B. Liu [a,b], J.F. Shao [a,b,*], T.G. Liu [a,b], S.Y. Xie [a,b], N. Conil [c]

[a] University of Hohai, College of Civil Engineering and Transportation, Nanjing, China
[b] University of Lille, Laboratory of Mechanics of Lille, 59650 Villeneuve d'Ascq, France
[c] Andra, Chatenay Malabry, France

ARTICLE INFO

Article history:
Received 11 January 2016
Received in revised form 25 April 2016
Accepted 25 April 2016
Available online 10 May 2016

Keywords:
Gas permeability
Creep
Claystone
Clayey rock
Radioactive waste disposal

ABSTRACT

Clayey rocks, due to its low permeability and self-sealing ability, have been selected as a privilege candidate host rock for underground radioactive waste repository in many countries. The gas permeability evolution is very sensitive to the deformation process in such rocks and can be used as a good indicator of microstructure changes such as the growth of micro-cracks. In this work, the mechanism of gas permeability evolution in the Callovo-Oxfordian (Cox) claystone during a creep deformation is investigated. Firstly, multi-step creep tests with different confining pressures are carried out to characterize gas permeability evolutions under different levels of the deviatoric stress. Secondly, in order to minimize effects of multiple deviatoric loading steps, one-step creep tests are also realized. Throughout all creep tests, the gas permeability is measured by a transient pulse decay method in together with the axial and radial strains. It is first found that the gas permeability of the claystone significantly decreases with the confining pressure or hydrostatic stress. The gas permeability in multi-step creep tests can exhibit a four-stage evolution with the progressive increase of deviatoric stress level, composed of a rapid decrease, a gentle decrease, a gentle increase and a rapid increase. However, the gas permeability is continuously decreasing during one-step creep tests. The permeability change in a one-step creep test can be well correlated with the volumetric strain variation by a logarithmic function. Effects of loading orientation with respect to claystone bedding planes are also investigated.

1. Introduction

The geological disposal is considered as one of feasible solutions for the long term disposal of radioactive waste. Clayey rocks, due to their low permeability and self-sealing ability and the absence of major natural fractures, have been selected as a privilege candidate host rock in many countries. In this context, the Callovo-Oxfordian (Cox) claystone, has been extensively studied during the last decades in France in the framework of the underground research laboratory CIGEO (Andra, 2005a, 2012).

The long term safety analysis of the geological radioactive waste repository requires a complete investigation of the time-dependent hydro-mechanical behaviors of the geological barrier. The mechanical behaviors of the Cox claystone in the excavation near field have been investigated and a large number of results have been reported on its instantaneous behaviors (Hu et al., 2013; Huang et al., 2013; Masri et al., 2014; Yang et al., 2013; Zhang et al., 2013; Zhang et al., 2012) as well as time-dependent behaviors (Auvray et al., 2015; Bérest, 1987; Bérest et al., 2001; Fabre and Pellet, 2006; Gasc-Barbier et al., 2004; Ghoreychi, 1999; Gratier et al., 2004; Liu et al., 2015b; Valère et al., 2002; Yang et al., 2011). Those works have well addressed some of the mechanical problems in the construction stage especially the excavation damage zone.

However, behind the mechanical behaviors, a crucial issue to be addressed is the evolution of transport properties in particular the water and gas permeability. Complementary studies are still needed on this issue. In a long period, gases will be accumulated abundantly in the underground radioactive waste repository due to the erosion of metallic wastes and containers, the aerobic and anaerobic microbial activity, the degradation of cellulosic materials and other organic materials in the waste, and the gas release of small livings in the barrier rock formation (Voinis et al., 1992; Yang et al., 2010). The release of these gases from the barrier rock will induce leakage of nuclides in the life cycle, which will cause severe consequences to the life cycle. Thus, it is very necessary to characterize the gas permeability of the barrier rocks, especially the excavation damaged zone (EDZ), for sealing and more important its evolution in a long period. Further, the gas permeability is much sensitive than the water permeability to microstructure changes of rocks such as growth of micro cracks. The gas permeability was used as a good indicator to detect the induced damage and sealing capacity of the intact Cox claystone by gas injection in hydrostatic and/or triaxial tests (Billiotte et al., 2008; Zhang and Rothfuchs, 2008) under several stress states. The gas permeability of the intact Cox claystone under a high level deviatoric creep stress was observed with a decreasing

---

[*] Corresponding author at: Ecole Polytech Lille, University of Lille, 59655 Villeneuve d'Ascq France

http://dx.doi.org/10.1016/j.clay.2016.04.021
0169-1317/

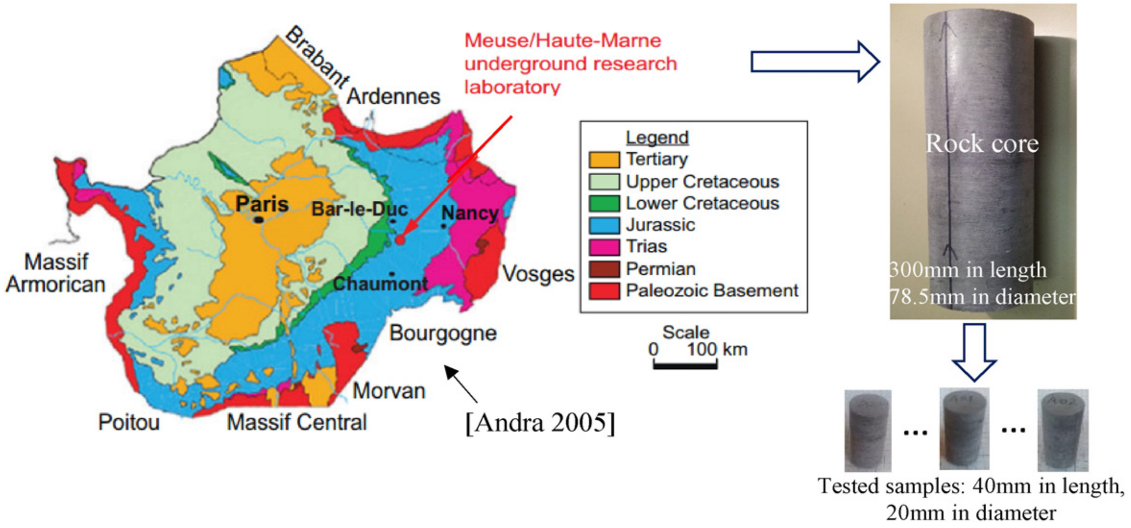

Fig. 1. Overview of the sample location and preparation.

phase (Liu et al., 2015a). Those works confirmed the sealing ability of the intact Cox claystone due to compressive stresses, but the complete characterization of the transport properties in the low-permeable Cox claystone in concurrence with the time-dependent deformation is still an open issue due to technical difficulties to realize such tests. In particular, the permeability evolution mechanism during the creep process is still unclear due to the lack of experimental data. Such experimental data are mandatory for a better understanding of transport properties in clayey rocks.

The present paper presents an in-depth complementary study to the previous ones and aims to investigate the gas permeability evolution in the Cox claystone subjected to different levels of deviatoric stress and confining pressure, by a series of creep tests with gas permeability measured throughout the creep strain process. Effects of initial anisotropy on the gas permeability evolution are also studied.

2. Tested material and sample preparation

The tested samples are cored from the Meuse/Haute-Marne underground research laboratory in France (see Fig. 1). The samples used for multi-step creep tests are cored from a depth about 467.2 m and those for one-step creep tests are cored from a depth about 489.85 m. The original rock core of 300 mm in length and 78.5 mm in diameter is drilled in laboratory in its natural saturation degree to minimize the impact of water content during sample preparation. The mineralogical composition as well as the connected porosity of the Cox claystone vary with the depth (Andra, 2005b). In mesoscopic scale, the Cox claystone is composed of clay minerals, carbonate and quartz grains. The average proportion of each constituent phase is about 40% (±15%) quartz and other hard minerals, 34% calcite and 26% clay minerals. The composition of clay phase (<2 μm) approximately includes 40% illite, 30% kaolinite, 5% chlorite, and 25% swelling minerals (smectite and interstratified) (Liu et al., 2015b). The porosity of the claystone is 17 ± 4% with a pore size ranging from 2 nm to 50 nm and a surface area of 32 ± 8 m^2 g^{-1}. The pH value of the pore water in the Cox claystone is 7.2. The chemistry components of ions in the pore water are Cl^-, SO_4^{2-}, HCO_3^-, Na^+, K^+, Ca^{2+}, Mg^{2+} and Sr^{2+} with concentrations respectively as 1460, 1820, 255, 1290, 35, 305, 140, 15 mg/L (Altmann et al., 2012; Andra, 2005b).

In order to investigate the effect of structural anisotropy, samples are drilled in both perpendicular and parallel direction of the bedding planes of the Cox claystone stratum. The drilled cylindrical samples for the hydro-mechanical tests are about 40 mm in length and 20 mm in diameter. Once drilled, each sample end is polished to be perpendicular to the sample axis. Then, all samples are conserved in a closed container in which relative humidity is maintained as 59% by a chemical solution, such as sodium bromide, and the temperature around the container is set as 23 °C controlled by an air conditioner. The relative humidity is selected to simulate a desaturation state of the Cox claystone in the EDZ in the underground repository. The samples are kept in the container for at least 2 weeks with mass monitored and will not be used for hydro-mechanical tests until their mass values are stable. The details of the geometry and testing conditions of the tested samples are given in Table 1.

3. Experimental apparatus and method

The time-dependent hydro-mechanical tests with Nitrogen gas as injected fluid are realized in an autonomous and auto-compensated hydro-mechanical testing system (see Fig. 2) (Liu et al., 2016) developed at the Laboratory of Mechanics of Lille (LML). The Nitrogen gas is chosen due to its applicability and chemical stability with respect to the minerals constituting the Cox claystone. The testing system consists of three independent components for hydro-mechanical loading, respectively for deviator stress loading, confining pressure application, and interstitial pressure generation and monitoring, which are all assembled independently in the triaxial cell. The acquisition of pressure/stresses, fluid pressure and/or flow rate, displacements or deformation is realized by some specific transducers and recorded by a data acquisition center.

All the tests are carried out in a thermally isolated small room with temperature and relative humidity respectively maintained as 23 °C and 59% by a central air-conditioner in order to exclude the influence of temperature fluctuations. The axial strain (ε) and radius strains (ε) of the samples are measured respectively by two pairs of local strain gages in the series of tests. The volumetric strain (ε_v) are obtained by

$$\varepsilon_v = \varepsilon_1 + 2\varepsilon_3. \tag{1}$$

The tests are executed mainly in the following steps:

(a) Check each part of the testing system and make sure it functions well.
(b) Install the sample at the right position with a filter paper and a filter plate at each sample end and seal the sample in a plastic jacket from the pressure chamber of the triaxial cell.
(c) Apply the confining pressure with the rate 0.5 MPa/min to a given level such as $\sigma_3 = 6$MPa and maintain it.