Physics and Chemistry of the Earth 33 (2008) S186–S195

Contents lists available at ScienceDirect

Physics and Chemistry of the Earth

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

Multiphase flow and multicomponent reactive transport model of the ventilation experiment in Opalinus clay

L. Zheng [a,1], J. Samper [a,*], L. Montenegro [a], J.C. Mayor [b]

[a] E.T.S. Ingenieros de Caminos, Canales y Puertos, Universidad de Coruña, Campus de Elviña s/n, 15192 La Coruña, Spain
[b] ENRESA, Emilio Vargas 7, 28043 Madrid, Spain

A R T I C L E I N F O

Article history:
Available online 15 October 2008

Keywords:
Ventilation experiment
Multiphase flow
Reactive transport
Opalinus clay
Mont Terri
Pyrite oxidation

A B S T R A C T

During the construction and operational phases of a high-level radioactive waste (HLW) repository constructed in a clay rock, ventilation of underground drifts will cause desaturation and oxidation of the rock. The ventilation experiment (VE) was performed in a 1.3 m diameter unlined horizontal microtunnel on Opalinus clay at Mont Terri underground research laboratory in Switzerland to evaluate these phenomena and their impact on rock properties. A multiphase flow and reactive transport model of VE is presented here. The model accounts for liquid and gas flow, evaporation/condensation and multicomponent reactive solute transport. Model results reproduce measured vapor flow, liquid pressure and chemical data and capture the trends of measured relative humidities, although such data are slightly overestimated near the rock interface due to uncertainties in the turbulence factor. Chloride exhibits anion exclusion and accesses only 54% of total porosity. Rock desaturation allows oxygen to diffuse into the rock and triggers pyrite oxidation, dissolution of calcite and siderite, precipitation of ferrihydrite, dolomite and gypsum and cation exchange. pH in the unsaturated rock varies from 7.8 to 8 and is buffered by calcite. Computed changes in the porosity of Opalinus clay in the unsaturated zone caused by oxidation and mineral dissolution/precipitation are smaller than 5%. Therefore, rock properties are not expected to be affected significantly by ventilation of underground drifts during construction and operational phases of a HLW repository in clay.

1. Introduction

Clay formations have been selected as candidate host rocks for high-level radioactive waste (HLW) disposal in deep geological repositories. Underground research laboratories are being operated by several countries to demonstrate the feasibility of HLW disposal, carry out research and provide data and knowledge on relevant phenomena.

Underground drifts will be subjected to ventilation during the construction and operational phase of a HLW repository. If carried out over significant periods of time, ventilation could cause desaturation of the rock adjacent to the drifts which in turn could have a detrimental effect on physical and geochemical rock properties. The ventilation experiment (VE) test was performed at the Mont Terri underground research laboratory (Switzerland) to investigate and evaluate the potential impact of ventilation on hydraulic, mechanical and chemical conditions of consolidated Opalinus clay (Mayor et al., 2005). Mayor et al. (2007) presented a hydromechanical model of VE using CODE_BRIGHT (Olivella et al., 1996). This model which accounts for evaporation at the rock–air interface led to the conclusion that thermal and hydraulic rock characteristics will not be affected significantly by ventilation except in a narrow ring around the surface of the tunnel having a thickness smaller than 30 cm where saturation degree of the rock is smaller than 95%. Mayor et al. (2007) documented uncertainties in the turbulence coefficient used to quantify vapor flux at the rock–air interface. Mayer et al. (2007) presented a two-phase flow model of VE which was solved with TOUGH2 (Pruess et al., 1999) and used it to derive hydraulic properties of properties of the excavation damaged (EDZ) and Opalinus clay. They found that rock desaturation extends up to a radial distance of about 1.35 m from the tunnel wall. The driest zone having saturation degrees smaller than 95% has a thickness of 40 cm measured from tunnel wall. Mayer et al. (2007) determined also that EDZ matrix permeability does not differ strongly from that of undisturbed rock. Fernández-García et al. (2007) presented a hydrogeological model of VE using MODFLOW (McDonald and Harbaugh, 1984) to estimate the effective regional-scale hydraulic conductivity of Opalinus clay taking into account EDZ and rock desaturation.

There are numerous studies of pore water chemistry in Opalinus clay at Mont Terri (Pearson et al., 2003). Their results show

* Corresponding author.
E-mail address: jsamper@udc.es (J. Samper).
[1] Present address: Lawrence Berkeley National Laboratory, 1 Cyclotron Road, Berkeley, CA 94720, USA

1474-7065/$ - see front matter © 2008 Elsevier Ltd. All rights reserved.
doi:10.1016/j.pce.2008.10.058

that desaturation of Opalinus clay by ventilation causes pyrite oxidation and dissolution/precipitation of mineral phases. However, there are no reported models of desaturation and geochemistry in Opalinus clay. A multiphase flow and multicomponent reactive transport model of VE is presented here. The multiphase flow model is based on the model of Mayor et al. (2007) and is calibrated with relative humidity, vapor flow and liquid pressure data. The reactive transport model is based on the conceptual geochemical model of Pearson et al. (2003) and is calibrated using mineralogical and aqueous extract data from Fernández et al. (2006). The inverse hydrochemical model of Samper et al. (2008c) is used to estimate the chemical composition of pore water of clay samples from aqueous extract data. The model is used to evaluate changes in porosity and permeability of the rock caused by mineral dissolution/precipitation.

The paper starts by describing the main features of VE. Conceptual and numerical multiphase flow and reactive transport models of VE are then presented. Finally, model results are described and main uncertainties and conclusions are discussed.

2. Ventilation experiment

VE was carried out in a 1.3 m diameter unlined horizontal microtunnel at Mont Terri underground laboratory in Switzerland. The microtunnel was excavated in 1999 within the shaly facies of the Opalinus formation. It runs sub-horizontal in a NW–SE direction (with a dip of about 2° towards SE) and is oriented perpendicular to the rock bedding strike (Mayor et al., 2007). A 10 m long section of the tunnel was sealed by two doors and subjected to several desaturation phases by prolonged ventilation (Fig. 1; Table 1).

The main objectives of VE include: (1) estimation of desaturation and resaturation times in Opalinus clay by drift ventilation; (2) estimation of saturated hydraulic conductivity of the rock; (3) estimation of EDZ and its time evolution in terms of changes in hydraulic conductivity and displacements induced by drying; (4) characterisation of hydraulic properties of the rock under saturated and unsaturated conditions; (5) geochemical characterisation of desaturated zone; and (6) calibration and validation of hydromechanical and geochemical models (Mayor et al., 2005).

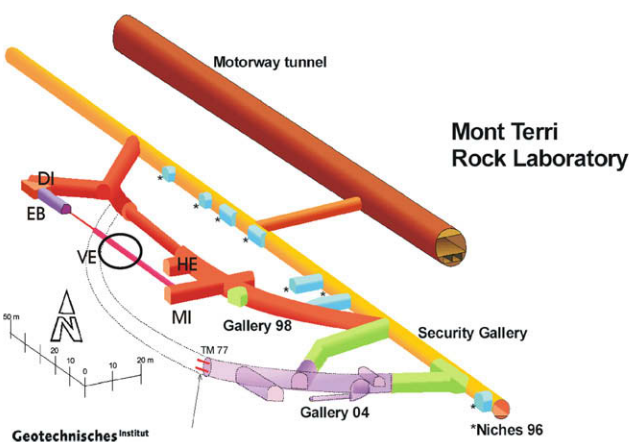

Fig. 1. Layout of Mont Terri URL and location of ventilation experiment (Fernández et al., 2006).

Hydraulic and mechanical data were monitored during desaturation and saturation phases while pore water chemical composition was measured at the end of desaturation phase 6 and resaturation phase 8 by aqueous extract tests (Fernández et al., 2006).

Ventilation was performed by circulating air with specified relative humidity, RH_{in} and temperature, T_{in} at a flux rate Q_{in}. Air was injected at one of the ends of the VE tunnel through an inflow pipe and evacuated at the other end of the test section with an outflow pipe. Values of Q_{out}, RH_{out} and T_{out} were measured in the outflow pipe by flow meters and temperature and relative humidity sensors. Relative humidity and temperature of the air inside the test area were recorded with number format should be consistent, either all are Arabic number like 4 hygrometers or all are English word like sixteen capacitive hygrometers Mayor et al. (2005, 2007).

VE involved several ventilation and resaturation phases (Table 1). The 3 first phases included the background period which goes from microtunnel excavation in 1999 to the beginning of the controlled ventilation in June 2003. The following 3 phases corresponded to the first ventilation period which was divided into three stages with decreasing relative humidities (phases 4, 5 and 6). Phase 4 was used to achieve initial quasi-saturated conditions in the rock while phases 5 and 6 were intended to desaturate the rock with strong ventilation with dry air (Table 1). Rock resaturation took place during phases 7 and 8. Phase 9 of VE started on July 1, 2005 and corresponded to a second ventilation period. Values of Q_{in}, RH_{in} and RH_{out} in each phase are listed in Table 1. Air temperature in the VE tunnel remained nearly constant around 15–16 °C.

Several boreholes (BVE) were drilled on the rock around the VE for geochemical characterisation at different phases. Boreholes BVE-85 and BVE-86 were drilled at the end of the first ventilation period (phase 6). After the resaturation period (phase 8) 5 boreholes BVE-96, BVE-97, BVE-98, BVE-99 and BVE-100 were drilled inside the VE area while BVE-101 and BVE-102 were drilled outside the VE area (Fig. 2). Most of these boreholes are horizontal and subparallel to the bedding. Only BVE-85 has a vertically down orientation and BVE-100 is perpendicular to bedding. Borehole lengths range from 1.45 to 2 m and have a diameter of 8 cm (Fernández et al., 2006).

Samples of the Opalinus clay from drillcores of boreholes BVE-97, BVE-99, BVE-100, BVE-101 and BVE-102 were collected at different depths to determine clay physico-chemical parameters. After drilling, drillcores were wiped to remove any drilling fluid on the core surfaces. Unaltered core samples were immediately packed in aluminum-foil bags, flushed with argon gas to displace atmospheric gases and sealed after applying vacuum. A second layer of aluminum-foil was placed and, finally, core samples were wrapped with air-evacuated and durable plastic bags to ensure full protec-

Table 1
Description, starting date, duration and RH of different phases of the ventilation experiment.

Phases	Period	Description	Starting date	Duration	RH[a]	β_g(m/s) Mayor et al. (2005)	β_g(m/s) base run
1st	Background	Microtunnel excavation	February 1999	~3.4 years	~90%	1.25×10^{-4}	1.25×10^{-4}
2nd		Test section sealing	1st August 2002	~8 months	~93%	1×10^{-5}	1×10^{-5}
3rd		Equipment tests	1st April 2003	72 days	~95%	2×10^{-4}	2×10^{-4}
4th	First ventilation	$Q_{in} \sim 20$ m^3/h $RH_{in} \sim 80\%$	12th June 2003	21 days	~84%	8×10^{-5}	8×10^{-5}
5th		$Q_{in} \sim 30$ m^3/h $RH_{in} \sim 30\%$	3rd July 2003	~2 months	~47%	1×10^{-4}	1×10^{-4}
6th		$Q_{in} \sim 30$ m^3/h $RH_{in} \sim 1$–3%	4th September 2003	~5 months	~15%	1×10^{-4}	7×10^{-5}
7th	Resaturation	$Q_{in} \sim 20$ m^3/h $RH_{in} \sim 100\%$	29th January 2004	~1 month	~92%	5×10^{-4}	4×10^{-4}
8th		Without ventilation	1st March 2004	~16 months	~95%	Not reported	5×10^{-5}
9th	Second ventilation	$Q_{in} \sim 40$ m^3/h $RH_{in} \sim 1$–3%	1st July 2005	~18 months	~15%	–	–

[a] RH = estimated mean relative humidity of the air in the test section.