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Influence of freeze–thaw cycles on the performance of covers with capillary barrier effects made of crushed rock–bentonite mixtures to control oxygen migration

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Abstract

In temperate climates, covers with capillary barrier effects (CCBEs) are successfully used to control oxygen flux from reaching the covered potentially acid mine drainage (AMD) generating mine tailings. In northern climate, the more attractive option for mine site reclamation is insulation covers, which are designed to keep reactive materials frozen. This article suggests that CCBEs can simultaneously control oxygen migration and mine waste temperature to inhibit AMD generation. However, in northern conditions, where natural fine-grained materials needed for the CCBE moisture-retaining layer are not always available, soil–bentonite mixtures could be used instead. This laboratory study assessed in instrumented columns the effects of freeze–thaw cycles on the performance of three CCBEs made with crushed rock–bentonite mixtures. An oxygen diffusion test was developed to determine the effective diffusion coefficient of oxygen (D_e) and its sensitivity to freeze–thaw cycles. The results show good initial performance for the saturated CCBEs. However, the tested CCBEs are significantly affected by freeze–thaw cycles and have limited oxygen limiting ability.

Key words: Covers with capillary barrier effects, crushed rock–bentonite mixtures, oxygen diffusion, acid mine drainage, northern climate.

Résumé

En climat tempéré, les couvertures avec effets de barrière capillaire (CEBC) sont utilisées avec succès pour contrôler les flux d'oxygène vers les résidus miniers potentiellement générateurs de drainage minier acide (DMA). En climat nordique, une option attrayante est le recouvrement isolant, qui est conçu pour maintenir les résidus miniers réactifs gelés. Cet article suggère que les

CEBC peuvent être utilisées pour contrôler simultanément la migration de l'oxygène et la température des résidus miniers pour limiter la génération de DMA. Toutefois, en conditions nordiques, les matériaux fins naturels nécessaires à la construction de la couche de rétention d'eau des CCBE ne sont pas toujours disponibles; des mélanges sol-bentonite pourraient être utilisés en remplacement. Cette étude de laboratoire vise à évaluer en colonnes instrumentées les effets des cycles de gel-dégel sur la performance de trois CEBC faites de mélanges matériaux granulaires-bentonite. Un test de diffusion de l'oxygène a été développé pour déterminer le coefficient effectif de diffusion de l'oxygène (D_e) et pour évaluer sa sensibilité aux cycles de gel-dégel. Les résultats montrent une bonne performance initiale des CEBC mais également une dégradation de la performance des recouvrements à contrôler migration d'oxygène suite aux cycles de gel-dégel.

Mots clés : Couverture avec effets de barrière capillaire, mélanges matériaux granulaires-bentonite, diffusion de l'oxygène, drainage minier acide, climat nordique.

1 Introduction

Acid mine drainage (AMD) is one of the most important environmental challenges for today's mining industry. This well-known environmental problem is caused by the oxidation of sulfide minerals contained in mine wastes when exposed to air and water (e.g., Aubertin et al. 2002). In recent decades, many engineered cover designs have been successfully developed to control water infiltration, oxygen migration, and/or mine waste temperature (e.g., Aubertin et al. 1995; Koerner and Daniel 1997; MEND 2001, 2004a, 2004b). In temperate climates, oxygen barriers are one of the most effective methods to control AMD generation (Collin and Rasmuson 1988; Nicholson et al. 1989). Among these barriers, covers with capillary barrier effects (CCBEs) have been demonstrated a practical option to prevent oxygen fluxes from reaching reactive mine tailings (Dagenais 2005; Bussière et al. 2003, 2006, 2007). In CCBEs, the oxygen barrier is created when a high air-entry value material (i.e., the moisture-retaining layer) overlays a lower air-entry value material (i.e., the capillary break layer). Capillary barrier effects occur between the two materials due to the contrast in hydrogeological properties, resulting in high saturation of the moisture-retaining layer (e.g. Nicholson et al. 1989; Aubertin et al. 1997, 1999; Bussière et al. 2003, 2006). Higher than 85–90% saturation of the moisture-retaining layer was reported to significantly reduce the oxygen flux that reaches the base of the cover, thus controlling AMD generation from sulfidic tailings (e.g., Yanful 1993; Aubertin et al. 1999).

In northern regions, AMD generation is generally limited by the presence of permafrost and low temperatures, which reduce contaminant migration (by limiting water transport due to freezing) through the environment and slow down the geochemical and biochemical reaction kinetics of sulfide minerals (MEND, 1996a, 1996b, 2009, 2010, 2012; Elberling et al. 2000; Elberling 2005). However, AMD can occur in northern mine sites (e.g., Elberling 1998; Godwaldt 2001), although

mainly in the permafrost active layer (MEND 1996a). The active layer is the layer of ground (soil or rock) that is affected by seasonal freezing and thawing in permafrost areas (French 2007).

So far, only a few reclamation cover options have been investigated for AMD mine tailings storage facilities located in such regions. The main reclamation strategy has been to encapsulate reactive tailings in permafrost beneath an insulation cover (MEND 2004a; Coulombe et al. 2012). The use of a CCBE combined with an insulation mechanism is a promising option for the reclamation of tailings storage facilities located in northern regions. This original approach could both reduce the time that the tailings are above freezing temperature and control oxygen fluxes when the tailings are unfrozen. CCBEs could also provide a more robust cover option in the climate change context, because sulfide oxidation could be limited year-round by controlling either mine waste temperatures and/or oxygen fluxes. However, the effectiveness of this approach has yet to be clearly demonstrated in a controlled laboratory environment.

Soil–bentonite mixtures have long been used as impervious materials in engineered structures such as dam cores, landfill covers, and impervious liners. Those mixtures provide good alternatives when natural fine-grained soils are not available close to a construction site, as is the case for many northern regions. Previous laboratory work conducted by the authors suggested that crushed rock–bentonite mixtures (CRBMs) provide a good potential alternative to fine-grained soils as moisture-retaining layer material in a CCBE. However, their capacity to act as an effective long-term oxygen barrier remains to be assessed (Boulanger-Martel et al. 2014).

This article presents a performance assessment using laboratory experiments of three CCBEs made of 5.0%, 6.5%, and 8.0% bentonite by dry weight of CRBMs exposed to cold climatic conditions. The main objectives were to 1) assess the influence of the bentonite content on the performance of CCBE configurations with and without freeze–thaw cycles, and 2) determine the effect of the initial degree of saturation of CRBMs on CCBE performance. This study was inspired by a real case: the Raglan Mine (a Glencore property), located in northern Quebec,

which is currently weighing the option of using a CRBM CCBE to reclaim their potentially AMD-generating tailings storage facility.

2 Oxygen diffusion in soil covers

The main mechanism responsible for oxygen transport in fine-grained (or small pore size) unsaturated materials, such as those used as a moisture-retaining layer in CCBEs, is molecular diffusion (Collin 1987; Collin and Rasmuson 1988). When the saturation of these materials is below 85–90%, oxygen diffusion is controlled mainly by the air filled porosity, represented by the volumetric air content (θ_a). In this case, O_2 diffusion is controlled by the oxygen concentration in the air (C_a). However, at higher saturations, the air phase becomes discontinuous (Corey 1957), and oxygen transport is controlled by diffusion in the water-filled voids, represented by the volumetric water content (θ_w). In this case, oxygen diffusion is limited by the oxygen concentration in the water (C_w), which is approximately 30 times lower than C_a (Mbonimpa et al. 2003).

For non-reactive unsaturated cover materials, the 1D oxygen diffusion can be described by Fick's first and second laws (Glauz and Rolston, 1989; Hillel 1998):

$$[1] \quad F(z, t) = -D_e \frac{\partial C(z, t)}{\partial z}$$

$$[2] \quad \frac{\partial}{\partial t}(\theta_{eq} C) = \frac{\partial}{\partial z} \left(D_e \frac{\partial C}{\partial z} \right)$$

where $F(z, t)$ is the diffusive flux of oxygen, D_e is the effective oxygen diffusion coefficient, and $C(z, t)$ is the interstitial oxygen concentration at time t and depth z . The equivalent porosity (θ_{eq}) is a parameter that was introduced to account for the oxygen transport that occurs in both the air and water phases (Aubertin et al. 1999, 2000), defined as:

$$[3] \quad \theta_{eq} = \theta_a + H\theta_w$$

where H is the dimensionless Henry's equilibrium constant (~ 0.03 at 20°C). The above equations show that the oxygen flux through a soil cover is strictly related to the D_e of the cover materials and the oxygen concentration gradient. Because the performance of an oxygen barrier is usually assessed by measuring the O_2 flux that reaches the reactive tailings (e.g., Mbonimpa et al. 2003; Bussière et al. 2004, 2007), reliable D_e values are necessary for cover performance assessment and optimal design.

Laboratory studies of various soil types have demonstrated that D_e can be estimated with semi-empirical models (Millington and Shearer, 1971; Collin 1987; Aachib et al. 2004). Based on simple geotechnical parameters such as porosity (n) and volumetric water and air contents, D_e can be estimated with the following equation (Aachib et al. 2004):

$$[4] \quad D_e = \frac{1}{n^2} (D_a^0 \theta_a^{p_a} + H D_w^0 \theta_w^{p_w})$$

where D_a^0 ($1.8 \times 10^{-5} \text{ m}^2/\text{s}$) and D_w^0 ($2.5 \times 10^{-9} \text{ m}^2/\text{s}$) are the free oxygen diffusion coefficients in air and in water, and p_a and p_w are parameters related to the tortuosity of the air and water phases, expressed as:

$$[5] \quad p_a = 1.201\theta_a^3 - 1.515\theta_a^2 + 0.987\theta_a + 3.119$$

$$[6] \quad p_w = 1.201\theta_w^3 - 1.515\theta_w^2 + 0.987\theta_w + 3.119$$

In the laboratory, the D_e value for inert cover materials is often determined at small scale using oxygen diffusion cells (e.g., Shackelford 1991; Aachib et al. 2004), but this testing method is limited to relatively fine-grained materials, and is not designed to assess the effects of freeze–thaw cycles on D_e . Accordingly, the authors developed an original column experiment, described in this paper, to determine the D_e of moisture-retaining layers made of crushed rock–bentonite mixtures and its sensitivity to freeze–thaw cycles.

3 Materials and Methods

3.1 Main physical and geotechnical properties

The granular materials used in the column tests were crushed rock (0–20 mm) and three crushed rock–bentonite mixtures. The crushed rock was obtained from a stockpile at the Raglan Mine in northern Quebec, Canada. The average natural gravimetric water content of the crushed rock was 4.4%. Three different CRBMs were made in the laboratory by hand mixing a commercially available PDSCo Grout bentonite powder and crushed rock at 5.0%, 6.5%, and 8.0% bentonite content by dry weight. The liquid used for wetting, hydration, and permeation throughout the experiment was tap water from the city of Rouyn-Noranda (Quebec, Canada). The main physical and geotechnical properties of the materials were determined prior to column testing, as summarized in Table 1. The grain size distribution of the crushed rock was determined according to ASTM (2007) standard D422 for particle sizes larger than 425 μm , and with a laser diffraction analyzer for finer grain sizes. Grain size distribution of bentonite was obtained by laser diffraction in isopropanol to prevent bentonite swelling during analysis. The grain size distributions of the three mixtures were obtained by combining the grain size distributions for crushed rock and bentonite. The specific gravity (G_s) was measured with a helium pycnometer.

Insert Table 1 about here

Geotechnical properties of the crushed rock were provided by Coulombe (2012). The average saturated hydraulic conductivity (k_{sat}) was measured at 2.6×10^{-1} cm/s. Based on the water retention curve, the air-entry value (ψ_a) is 0.5 kPa, and the residual suction (ψ_r) is 7.0 kPa. For crushed rock–bentonite mixtures, the saturated hydraulic conductivity was determined using a flexible wall permeameter according to methods A and D, ASTM (2010) standard D5084 for constant head and constant flow rate saturated hydraulic conductivity tests. Flexible

permeameters are preferred over rigid wall permeameters due to the shorter time to achieve full bentonite hydration and consolidation (3–7 days vs. 1–4 weeks) (Chapuis 1990). A long-term (1000h) initial permeability test conducted on an 8.0% bentonite mixture showed that the maximum time to reach minimum and stable saturated hydraulic conductivity values was approximately 80 hours. Hence, permeability tests were conducted for approximately 100 h under low hydraulic gradients (<8). Measured k_{sat} values ranged from 3.5×10^{-7} cm/s for the 5.0% CRBM to 4.8×10^{-8} cm/s for the 8.0% CRBM, and are a function of the bentonite void ratio within the mix (Boulanger-Martel et al. 2014).

The water retention curves were obtained by performing drainage tests using a pressure plate apparatus equipped with a high air-entry value cellulosic membrane and vapor desiccators with saturated salt solutions. Nine pressure increments between 10 and 2000 kPa were achieved with the pressure plate apparatus, and five different vapour equilibrium tests were performed for suctions ranging from 3.5 MPa to 3250 MPa. Saturated salt solutions and their corresponding suctions can be found in the literature (e.g., Delage and Cui 2000; Cui et al. 2008; ASTM 2012a). The obtained water retention curves showed air-entry values ranging from 8.0 to 20.0 kPa. These curves also showed low desaturation rates for high suction values, trending towards the behaviour of bentonite alone. The 6.5% and 8.0% bentonite mixtures showed similar behaviour, whereas the 5.0% mixture showed lower water retention capacity. Hydraulic tests (permeability and suction) were performed on samples from the three CRBMs moulded at a target porosity of 0.25. Detailed testing procedures and geotechnical properties are provided in Coulombe (2012) for the crushed rock and in Boulanger-Martel et al. (2014) and Boulanger-Martel (2015) for the crushed rock–bentonite mixtures.

Based on the air-entry values for the tested mixtures, a CRBM could provide sufficient hydraulic property contrasts to induce capillary barrier effects if placed over a very low air-entry value material such as the crushed rock. When the crushed rock reached its residual suction value, the

air-entry values for the three tested CRBMs were not yet reached. Therefore, saturations higher than 85–90% are expected in a moisture-retaining layer made of a CRBM overlaying a crushed rock capillary break layer.

3.2 Column design

3.2.1 *Materials preparation*

Column instrumentation and configuration are shown in Figure 1. Three column set-ups were designed to examine three different 2.0 m thick CCBE configurations. The tested CCBEs consisted of a 0.4 m capillary break layer, a 0.6 m moisture-retaining layer, and a 1.0 m crushed rock protective layer. Both the capillary break and protective layer were made of crushed rock alone, whereas the moisture-retaining layer was made of a CRBM at bentonite content of 5.0%, 6.5%, and 8.0% for columns 1, 2, and 3, respectively. A porosity of 0.25 was targeted for all cover layers. This porosity corresponds to a compacted dry density of approximately 90% of the Proctor maximum dry density (ASTM, 2012b) for the three CRBMs (Boulanger-Martel et al. 2014). Granular materials were placed in compacted layers of 7 kg of dry material (corresponding to a 5 cm lift thickness) at the required effort to reach the desired dry density. To control for CRBM bentonite content, the CRBMs were made in the laboratory with oven-dried and homogenized materials. All mixtures were prepared in small batches by hand mixing to the desired weight percentages of dry bentonite and dry aggregate. Tap water was then added to reach the target water content, and the mixtures were mixed to achieve homogeneity with no excess water remaining. The 5.0% and 8.0% CRBM moisture-retaining layers were compacted at a gravimetric water content of 10.0%, whereas the 6.5% bentonite CRBM moisture-retaining layer was compacted at a gravimetric water content of 5.0%. The corresponding initial saturations for

the moisture-retaining layers were 91.0%, 38.7%, and 91.5% for columns 1, 2, and 3, respectively. The initial gravimetric water content for the column 2 CRBM corresponded to the weighted average of the natural gravimetric water contents of the crushed rock (4.4%) and the bentonite (8.1%). This initial water content was used to simulate the construction of the moisture-retaining layer without adding water to the available materials, as used previously at the Raglan Mine to produce a CRBM. The crushed rock protective and capillary break layers were homogenized and moulded at their natural water contents.

Insert Fig. 1 about here.

3.2.2 Column description

The columns were made of a 22.3 mm thick, 2.0 m tall high-density polyethylene (HDPE) cylinder. In order to prevent preferential fluid and gas flow paths along the column wall, an inner diameter (D) of 28.0 cm was chosen, based on the geometric considerations of ASTM standard D-2434 (ASTM, 2006) for permeability testing of granular soils in rigid wall permeameters, which recommends a minimum D/d_{100} ratio of 8 to 10 (a ratio of 14 was present in the tested columns). The experimental column instrumentation and hardware were designed to conduct oxygen diffusion tests in the moisture-retaining layer materials directly in the columns. Each column was equipped with a bottom drain and a purge valve located just below the interface between the capillary break and retaining layers. A total of 8 Decagon Devices 5TM ($\pm 0.02 \theta_w$ and $\pm 1.0^\circ\text{C}$ accuracy) sensors were installed at the center of each column to measure variations in volumetric water content and temperature at various depths into the CCBE (Fig. 1). The sensor located at the center of the moisture-retaining layer was duplicated in case of failure (depth 1.30m). Three gas sampling ports were also drilled through the column, and custom-made plastic vials with a septum stopper were installed in the holes and then sealed. Finally, one Apogee SO-

110 (sensitivity~ 1.1% O₂) oxygen sensor was installed in the capillary break layer close to the interface between the capillary break and moisture-retaining layers to monitor interstitial oxygen concentration variations during oxygen diffusion tests. The experimental columns were set up in a controlled atmosphere room where air temperatures can range from 20°C to -20°C so that the influence of freeze–thaw cycles on the performance of the tested CCBEs could be determined.

3.3 Oxygen diffusion test

In this study, the performance assessment of the tested CCBEs is based mainly on the results of oxygen diffusion tests conducted in the experimental columns. The column oxygen diffusion test set-up and procedure were adapted from the single reservoir with constant source concentration method (Shackelford, 1991), as follows. An oxygen concentration gradient was induced between the protective layer and the capillary break layer to force oxygen diffusion through the moisture-retaining layer. To do so, a nitrogen tank was connected to the base drain and the purge valve was opened (Fig. 1). A low nitrogen flow was then induced through the capillary break layer until most of the interstitial O₂ was purged. Purge completion was confirmed by stable (near zero) signal readings on the Apogee SO-110 oxygen sensor. Average purge time was 30 minutes. The base drain and purge valve were then closed. Oxygen migration from the protective layer to the capillary break layer was strictly controlled thereafter by molecular diffusion. At test start, the initial gaseous oxygen concentration profile of the moisture-retaining layer was sampled through the vials located at 1.10 m and 1.50 m depth. Initial O₂ concentrations sampled at 1.50 m depth confirmed that the nitrogen purge had no effect on O₂ concentrations at the base of the moisture-retaining layers. Interstitial gas samples were analyzed to obtain oxygen concentration using an Agilent Technologies Micro GC gaseous phase chromatograph. The change in gaseous O₂ concentration in the capillary break layer was measured with the Apogee SO-110 sensor and

recorded with a data logger during testing. In order to reliably interpret the Apogee sensor signal, a calibration equation was obtained from measured oxygen concentrations (using the chromatograph) in the capillary break layer as a function of the sensor signal (see Boulanger-Martel 2015 for details). These results were further interpreted to obtain a bulk oxygen diffusion coefficient (D_e) for the moisture-retaining layer using a POLLUTE v6 numerical model (Rowe et al. 1994). The modelling procedure is discussed in detail in Section 5.

3.4 Experimental approach

Prior to column testing, an 84-day pre-testing phase was conducted for bentonite hydration, saturation, and thermal conditioning. In order to produce a high initial degree of saturation for the crushed rock–bentonite mixtures in columns 1 and 3, a water head of 1.75 m was gradually applied through the base drain and maintained for 52 days. The columns were then drained for 21 days and the temperature of the controlled atmosphere room was set to 5 °C for thermal conditioning of the materials. Column 2 was set up simultaneously with the two other columns. Because the objective for column 2 was to assess the performance of the cover system starting at the natural water content, no water head was applied to the base drain. Column 2 was maintained airtight until the beginning of the first diffusion test to prevent evaporation from the CRBM.

The general experimental approach comprised a sequence of oxygen diffusion and infiltration tests conducted on each column, as described in Table 2. An initial oxygen diffusion test was performed to obtain an initial D_e value. An infiltration test was then conducted to simulate water infiltration and assess the CRBM's ability as a moisture-retaining layer to recharge after drainage. Infiltration tests were performed by adding 5 cm of water twice (10 cm of water total) to the top of each column and then monitoring the water mass exiting the base drain over time. The first water mass was added at the beginning of the test and the second 24 hours later. Another oxygen

diffusion test was then conducted to quantify the change in oxygen diffusion behaviour attributed to the water recharge in the CRBM layer. Both oxygen diffusion and infiltration tests were conducted at an ambient temperature of 5 °C, which is a realistic estimate of the average maximum temperature for a moisture-retaining layer in a similarly configured CCBE located in northern Quebec. After the diffusion–infiltration–diffusion cycle, freezing conditions were applied to the column by setting the controlled atmosphere room to a temperature of -13 °C for 72 hours. This temperature is a typical average minimum moisture-retaining layer temperature for a CCBE located in northern Quebec. The room temperature was then reset to 5 °C for 72 hours of thawing, and another freeze–thaw cycle or diffusion test was initiated (diffusion tests were performed after 1, 3, and 5 freeze–thaw cycles). Diffusion tests were conducted only in unfrozen state in order to assess the worst-case scenario in terms of oxygen diffusion (maximum D_e value). Unfrozen volumetric water contents and temperatures were also monitored throughout the experiment. In order to obtain reliable volumetric water content values, the raw sensor signals were interpreted using a specific volumetric content calibration equation (see Boulanger-Martel, 2015 for details). System equilibrium (θ_w stability) was monitored during diffusion testing, and the average measured θ_w values were used for data interpretation.

At the end of testing, the three columns were dismantled to obtain the total porosity and volumetric water content at different depths in each column. Eleven days after the last diffusion test, the columns were frozen at -13 °C in the controlled atmosphere room and cut into 10 cm thick slabs with a diamond blade saw. The slabs were oven-dried for 7 days and the gravimetric water content of each slab was calculated. The total porosity and volumetric water content were then estimated based on the mass and volume measurements. The volumetric water contents measured at dismantling were compared to the final sensor readings to validate the calibration used for data interpretation. Initial calculated porosities were also checked against the measured dismantling values.

Insert Table 2 about here.

4 Column test results

4.1 Changes in volumetric water content profiles

Figure 2 summarizes the θ_w profiles measured during oxygen diffusion testing before (BI) and after (AI) infiltration under various testing conditions for column 1 (5.0% bentonite), column 2 (6.5% bentonite), and column 3 (8.0% bentonite). The θ_w values represent the average volumetric water contents measured during oxygen diffusion testing. Variations in θ_w below $0.02 \text{ m}^3/\text{m}^3$ were typically observed during diffusion testing. These profiles are based on θ_w values in completely thawed state, and were used to assess the unfrozen hydrogeological behaviour of the tested CCBEs. Moisture-retaining layers are positioned between the dashed lines.

Insert Fig. 2 about here.

The θ_w values measured in the protective layers of all columns were typically close to residual values for the whole testing period. Saturations for these layers were generally below 8%. However, a relatively high θ_w value (0.32) was recorded at the base of the protective layer during the first infiltration test in column 1 (slightly higher than the porosity), which can be attributed to variations in local porosity and sensor accuracy. Thus, a temporary accumulation of water at the interface between the moisture-retaining and protective layers occurred during the first infiltration test only. The θ_w values measured in the capillary break layers were slightly higher than those in the protective layers, and ranged from 0.02 to 0.04 for columns 1 and 2 and from 0.04 to 0.06 for

column 3. Assuming the porosities presented in Figure 1, the saturations measured in the capillary break layers for columns 1 and 2 ranged from 6% to 14%. A maximum saturation of 24% was observed in column 3 under initial conditions, but saturation remained mainly in the 15% to 17% range after the first freeze–thaw cycle.

The θ_w profiles presented in Figure 2 for columns 1 and 3 show that the contrast in hydraulic properties between the crushed rock and the CRBM at the beginning of the test was sufficient to induce capillary barrier effects. Similar hydrogeological behaviours were observed for columns 1 and 3, although column 3 showed slightly better water retention capacity. Before the first freeze–thaw cycle, the average θ_w measured by the two sensors located at the bottom of the moisture-retaining layers in columns 1 and 3 ranged from 0.22 to 0.25. Considering the average porosities presented in Figure 1, this part of the moisture-retaining layers was near saturation in both columns. The θ_w values measured for the initial conditions at the top of the moisture-retaining layers, before and after infiltration, were 0.06 and 0.29 for column 1 and 0.23 and 0.21 for column 3, respectively. Here again, a measured θ_w value was slightly higher than the average porosity, which is attributed to local porosity variation and sensor accuracy in column 1. In the initial conditions, saturations greater than 85% were observed before and after the infiltration test in the moisture-retaining layer of column 3, whereas these saturation conditions were observed only after the infiltration test in column 1. In the other testing conditions, the infiltration tests produced only limited CRBM recharge in columns 1 and 3. With freeze–thaw cycles, θ_w measured in the top portion of the two moisture-retaining layers dropped to the residual value in column 1 and to values from 0.08 to 0.13 in column 3. Similarly, the sensors located at the center of the moisture-retaining layers showed rapid desaturation to values ranging from 0.11 to 0.13 and from 0.13 to 0.18 in columns 1 and 3, respectively. At the base of the moisture-retaining layers, measured θ_w presented lower, more gradual desaturation tending toward 0.16 and 0.19 in these two columns. Hence, the 8.0% bentonite CRBM CCBE was better able to withstand freeze–

thaw cycles compared to the 5.0% bentonite CRBM CCBE by maintaining a minimum saturation of 82% (vs. 63%) in this part of the moisture-retaining layer.

Column 2 showed different hydrogeological behaviour. Note that this column was prepared with CRBM at its natural water content. Figure 2 shows no significant variations in θ_w for the moisture-retaining layer. The θ_w values ranged from 0.10 to 0.13 at the base (at 1.5 m depth). These measured θ_w values correspond to saturations ranging from 38% to 52%. Residual volumetric water contents were observed in the middle and at the top of the moisture-retaining layer. Independently of the number of freeze–thaw cycles, infiltration had very little effect on θ_w measured in the moisture-retaining layer in this column. Based on the obtained θ_w profiles, it can be concluded that the saturation and hydration of the 6.5% CRBM with initial natural water content were very low, and insufficient to ensure the nearly full saturation of the moisture-retaining layer required for an effective CCBE.

4.2 Oxygen diffusion tests

The results of the column oxygen diffusion tests conducted in experimental columns 1, 2, and 3 are presented in Figure 3, showing the change in interstitial O_2 concentration (%) with time measured at the top of the capillary break layer. Oxygen diffusion tests lasting approximately 10 days were conducted before (BI) and after (AI) infiltration testing in initial conditions and after 1, 3, and 5 freeze–thaw cycles.

Insert Fig. 3 about here.

In column 1, interstitial oxygen concentration increased by 4.4% in the capillary break layer over the testing period in initial conditions before infiltration, and dropped below sensor sensitivity

after infiltration. In column 3, oxygen concentration in the capillary break layer increased slightly ($<1.0\%$) after 10 days in these conditions. These results are in agreement with the θ_w observed near the top of the moisture-retaining layers and near the base of the protective layers (see Fig. 2). The nearly saturated moisture-retaining layers fulfilled their purpose, and only insignificant quantities of oxygen (approaching zero) diffused through these layers. After the first freeze–thaw cycle, the moisture-retaining layer in column 1 maintained fairly good oxygen barrier capacity before and after the infiltration test, and limited oxygen ingress ($<4.4\%$) in the capillary break layer over the testing period (10 days). In the same conditions, column 3 showed a decrease in oxygen barrier capacity for the moisture-retaining layer compared to initial conditions (oxygen concentration in the capillary break layer increased to 6.7 % and 9.7 % before and after infiltration, respectively). Final interstitial oxygen concentrations from 16% to 20.9% were measured in the capillary break layer in columns 1 and 3 during the diffusion tests (10 days) conducted after 3 and 5 freeze–thaw cycles. Slight improvements in oxygen diffusion controlling capacity were observed after compared to before infiltration tests due to the slightly higher θ_w in the moisture-retaining layers.

The results obtained for column 2 show that all diffusion tests tended to obtain a final interstitial oxygen concentration in the capillary break layer close to atmospheric O_2 concentration after 10 days. Hence, water infiltration did not allow the moisture-retaining layer to recharge, which would help control oxygen diffusion. The scattered curve fitting was attributed mainly to the variations in initial oxygen concentration (0.75–2.07%) in the capillary break layer.

4.3 Column dismantling results

The average dismantling porosities obtained for the moisture-retaining layers were 0.24, 0.25, and 0.24 for column 1, 2, and 3, respectively (Fig. 4). A more compact zone was observed in column

1 at 1.20 m depth. Average porosities measured in the protective and capillary break layers were 0.25 and 0.26, respectively, for the three columns. Considering the accuracy of the method used to estimate the porosity (estimated at ± 0.02), these measurements confirm that the total porosity of the materials tested did not change significantly during the column tests.

Insert Fig. 4 about here.

The final dismantling θ_w values are presented in Figure 2 and compared to the monitored θ_w values. At dismantling, residual θ_w values were observed in the protective layers, whereas higher values (0.06–0.10) were observed in the capillary break layers. Values ranging from 0.06 to 0.15 were obtained for the moisture-retaining layers. From the comparison of the θ_w monitored during the last diffusion test (cycle 5 after infiltration) with the dismantling θ_w , it is reasonable to conclude that the sensors provided reliable measurements during the test.

5 Discussion

The obtained θ_w profiles showed that high initial saturations (>85%) were obtained in the moisture-retaining layers of columns 1 and 3, suggesting good initial cover performance. This good performance was confirmed by the low oxygen concentrations measured in the capillary break layers after 10 days of oxygen diffusion tests. However, the θ_w measurements and diffusion test results obtained after freeze–thaw cycles 1, 3, and 5 suggest a significant influence of freeze–thaw cycles on the performance of the tested CCBEs. Hence, the above obtained results were further analyzed for performance assessment. This assessment includes a determination of D_e for the layered systems and oxygen flux calculations.

5.1 Effective oxygen diffusion coefficient determination

5.1.1 Numerical interpretation of oxygen diffusion tests

A one-dimensional interpretation of the oxygen diffusion tests was performed using the POLLUTEv6 numerical model (Rowe et al. 1994). This program was initially developed for modelling solute transport in saturated media, but can also be used for gas diffusion modelling in unsaturated materials if certain parameters are adjusted to allow solving Fick's first and second laws (Rowe and Booker 1985). More specifically, in the case of unsaturated non-reactive materials, oxygen diffusion can be modelled if parameters such as the porosity ($n=\theta_{eq}$), Darcy's velocity ($v=0$), and the coefficient of hydrodynamic dispersion ($D^*=D_e\theta_{eq}^{-1}$) are adjusted. This software has often been used to determine the oxygen diffusion coefficient and oxygen flux through various soil cover systems (e.g., Yanful 1993; Aubertin et al. 1995, 2000; Mbonimpa et al. 2003, 2011; Aachib et al. 2004).

In this study, a general conceptual model consisting of a five-layer system where the moisture-retaining layer is separated into three distinctive sublayers (20 cm thick each) was used to represent the various column saturation conditions (Fig. 2). A boundary condition (BC) of constant atmospheric O_2 concentration (20.9%) was set at the top of the column and a boundary condition of zero flux was imposed at the bottom. The sampled initial oxygen concentration profiles confirmed that an initial oxygen concentration of 20.9% could be used for the protective layer, and the three moisture-retaining sublayers for all numerical analyses. The initial oxygen concentration in the capillary break layer was set equal to the measured value at the beginning of each test (0.4%–2.1% after the nitrogen purge). For a given test interpretation, a model layer θ_{eq} was calculated based on the average measured θ_w and θ_a (see Eq. [3]) during the testing phase with a Henry's constant value of 0.04 (calculated value at 5 °C). The D^* parameters were

determined using POLLUTE or calculated from estimated values of the D_e parameter using Aachib et al. (2004) semi-empirical model (see Eq. [4]). The conceptual model and the various modelling parameters are schematized and described in Figure 5.

Insert Fig. 5 about here.

For the protective and capillary break layers, the calculated θ_{eq} (sensor at 0.5 m depth for the protective layer) and the estimated D^* values (using Eq. [4]) were used as model parameters. Estimated D^* values are indicated as E_{eqn4} in Figure 5. The obtained θ_w profiles for the moisture-retaining layers highlighted three typical saturation conditions: 1) the whole moisture-retaining layer is uniformly and highly saturated, 2) the bottom 40 cm of the moisture-retaining layer is significantly more saturated than the top 20 cm, and 3) the base 20 cm of the moisture-retaining layer is significantly more saturated than the top 40 cm. For saturation condition 1, a single average mean θ_{eq} was used as input for the three moisture-retaining layer sublayers and a single moisture-retaining layer D^* value was obtained with POLLUTE. For saturation condition 2, the modelling parameters for the moisture-retaining layer's top 20 cm were set equal to the calculated θ_{eq} and the estimated D^* (Eq. [4]). For the bottom 40 cm, the average θ_{eq} (center and base of the moisture-retaining layer) was used to fit a D^* with POLLUTE. For saturation condition 3, the calculated θ_{eq} for each part (top, middle, and base) of the moisture-retaining layer were used to calculate the analogous D^* for the two top sublayers (Eq. [4]) and to determine the base D^* with POLLUTE. In all cases, the POLLUTE-modeled D^* was determined by iterative solution and by comparing the experimental and modelled oxygen concentrations over time. POLLUTE-modeled D^* were also compared with their corresponding estimated values (using Eq. [4]) and showed good agreement (see Boulanger-Martel 2015 for details). The final selection of D^* was based on the best-fit curves. Finally, the POLLUTE-obtained D^* values were converted to their analogous

D_e values. The interpretation method is described in detail in Boulanger-Martel (2015) and Boulanger-Martel et al. (2015).

5.1.2 D_e modelling inputs and results

The θ_w values monitored in the protective (0.0–1.0 m) and capillary break (1.6–2.0 m) layers remained close to residual values, and showed only very low variation throughout the testing conditions. Therefore, high and almost constant estimated D_e values were obtained ($\sim 2.5 \times 10^{-6} \text{ m}^2/\text{s}$). These two relatively dry layers had an insignificant impact on oxygen diffusion through the moisture-retaining layer, and could have been neglected in the numerical calculations, as suggested by Mbonimpa et al. (2003). The estimated and modelled D_e of the moisture-retaining layers are presented in tables 3, 4, and 5 as a function of depth and saturation test conditions for columns 1, 2, and 3, respectively. Depending on the saturation conditions of the moisture-retaining layer (1, 2, or 3), the D_e was estimated or modelled with POLLUTE. In tables 3, 4, and 5, the POLLUTE-modelled D_e are the framed values. When the whole moisture-retaining layer controlled oxygen diffusion (saturation conditions 1), D_e values in the range of $10^{-11} \text{ m}^2/\text{s}$ to $10^{-9} \text{ m}^2/\text{s}$ were computed. For cases where the bottom 40 cm of the moisture-retaining layer controlled oxygen diffusion (saturation conditions 2), D_e values from $10^{-7} \text{ m}^2/\text{s}$ to $10^{-8} \text{ m}^2/\text{s}$ were obtained. Typical D_e were in the $10^{-8} \text{ m}^2/\text{s}$ range when the bottom 20 cm was more saturated (saturation conditions 3). Considering that the D_e design criterion is typically on the order of $1.0 \times 10^{-8} \text{ m}^2/\text{s}$ (Bussière 1999) for CCBE configurations similar to saturation conditions 1, the 5.0% and 8.0% CRBMs showed good performance in the initial conditions. However, because only saturation conditions 2 and 3 were observed for the 6.5% CRBM CCBE and after the first freeze–thaw cycle for columns 1 and 3, D_e values much lower than $1.0 \times 10^{-8} \text{ m}^2/\text{s}$ should be required to significantly reduce oxygen ingress to the reactive tailings.

Insert Tables 3, 4, and 5 about here.

5.2 Steady-state oxygen flux through the cover

One of the most effective ways to assess a CCBE's capacity to limit oxygen diffusion is to calculate the oxygen fluxes that pass through the cover and reach the reactive tailings. For an effective CCBE, the targeted O_2 flux is typically less than $2 \text{ mol } O_2/\text{m}^2/\text{year}$ (e.g., Bussière 1999; Dagenais 2005; Nastev and Aubertin 2000). Because the saturation measured in the moisture-retaining layers was constant during the oxygen diffusion tests, θ_{eq} and D_e can be considered independent of time and depth. Assuming a rapid and complete consumption of O_2 by the reactive tailings ($C_{z=L}=0$), Equation [1] can be solved to estimate the oxygen flux ($F_{s,L}$) at the base of the moisture-retaining layer for steady state conditions ($t>0$) (e.g., Mbonimpa et al. 2003):

$$[7] \quad F_{s,L} = \frac{C_0 D_e}{L}$$

POLLUTE-modelled D_e were obtained for layers of different thicknesses (0.2 m, 0.4 m, and 0.6 m). Because the contrast in D_e between the oxygen-controlling portion and the more unsaturated portion of the moisture-retaining layer for saturation conditions 2 and 3 was usually greater than one order of magnitude, the desaturated top portion had a lower impact on the CCBE's capacity to limit oxygen transport. Thus, for saturation conditions 2 and 3, a harmonic average effective oxygen diffusion coefficient ($\overline{D_e}$) was calculated for the moisture-retaining layer to account for the contribution of the less saturated portion to control oxygen fluxes. $\overline{D_e}$ was calculated using the following expression (Aubertin et al. 2000):

$$[8] \quad \overline{D_e} = \frac{\sum_{j=1}^m L_j}{\sum_{j=1}^m \frac{L_j}{D_{e,j}}}$$

where m is the number of layers, and L_j and $D_{e,j}$ are the thickness and the effective diffusion coefficient, respectively, of layer j . For the calculation of a given $\overline{D_e}$, the POLLUTE-modelled D_e

value was used for the base layer of the moisture-retaining layer and the estimated D_e values were used for the top layers of the moisture-retaining layer. Equation [7] is then expressed as follows:

$$[9] \quad F_{s,L} = \frac{C_0 \overline{D_e}}{L}$$

For each column and oxygen diffusion test, the oxygen fluxes were calculated using Equation [9], with a moisture-retaining layer thickness (L) of 0.60 m, the corresponding calculated $\overline{D_e}$, and an initial O_2 concentration of 9.151 mol/m³ (at 5 °C) at the top of the moisture-retaining layer. The flux calculation results are presented in Figure 6. Based on a performance criterion of 2 mol O_2 /m²/year, column 3 showed effective initial performance with calculated O_2 flux less than 1 mol O_2 /m²/year. Column 1 had an initial flux of approximately 8 mol O_2 /m²/year but dropped to 0.01 mol O_2 /m²/year after the infiltration test. After one freeze–thaw cycle, the calculated O_2 flux reached 9 mol O_2 /m²/year and 18 mol O_2 /m²/year for columns 1 and 3, respectively. Beyond that point (for more than one freeze–thaw cycle), fluxes reached values ranging from 71 mol O_2 /m²/year to 138 mol O_2 /m²/year for column 1, and from 57 mol O_2 /m²/year to 90 mol O_2 /m²/year for column 3. High O_2 fluxes (>100 mol O_2 /m²/year) were observed for all testing conditions for column 2. The results obtained for columns 1 and 3 indicate a strong effect of freeze–thaw cycles on the oxygen fluxes, with a significant increase in O_2 fluxes during the three first cycles. The increase in O_2 fluxes is attributed mainly to the desaturation of the CRBM moisture-retaining layers. The overall performance of the 8.0% CRBM CCBE is slightly better than that for the 5.0% CRBM CCBE. Results on column 2 also show that the initial water content has a significant impact on the CCBE's capacity to control oxygen migration. Even when infiltration water was added at the top of the column, no significant volumetric water content increase was observed in the moisture-retaining layer for the duration of the experiment.

Insert Fig. 6 about here.

5.3 Freeze–thaw influence on CRBMs

Based on the test results obtained for the two initially saturated CRBM CCBs (columns 1 and 3), the tested CRBMs appear to have been significantly affected by freeze–thaw cycles. The tested columns reproduced well the bottom hydrogeological boundary conditions generally found in conventional CCBs, where the capillary break layer prevents water from being transmitted upward from the water table to the moisture-retaining layer. However, due to the column's large height-to-width ratio, three-dimensional instead of one-dimensional freezing was simulated, and faster cooling rates compared to field conditions were applied. To validate the experimental column results, closed-system uniaxial vertical freeze–thaw cycle permeability tests were performed on a 6.5% bentonite CRBM. Due to sample manipulation problems, freeze–thaw permeability tests were conducted on a different sample for each selected number of freeze–thaw cycles. For a given sample, the initial saturated hydraulic conductivity was first measured in a triaxial cell according to ASTM standard D5084 (ASTM 2010). After measurement, the sample was removed from the triaxial cell along with its flexible membrane and set up in a polystyrene–fiberglass insulated mould (to allow uniaxial vertical heat transfers). The sample was then subjected to the desired number of freeze–thaw cycles and the final saturated hydraulic conductivity was measured in the triaxial cell. This procedure is described in detail in Boulanger-Martel et al. (2014). The results presented in Figure 7 indicate that a 20-fold increase in the initial saturated hydraulic conductivity (initial k_{sat} of 2×10^{-7} cm/s) occurs after the first freeze–thaw cycle, and k_{sat} reaches a maximum increase at around 3 to 5 cycles (169 to 735 times the initial k_{sat}). Based on the test performed after 10 cycles, the CRBM k_{sat} value would be expected to stabilize in the long term. Because similar conclusions can be drawn from the O_2 diffusion column test results, we may conclude that the proposed test procedure obtains an acceptable

estimate of the influence of freeze–thaw cycles on the performance of this type of CCBE for design purposes.

During dismantling, the visual examination of frozen and thawed cross-sections showed no macroscopic changes to the CRBM texture due to freeze–thaw cycles. The bentonite grains were homogeneously distributed in the crushed rock matrix, and there was no evidence of ice lens formation in the frozen slabs. The same observations were made for the samples used for freeze–thaw saturated hydraulic conductivity tests. In view of these observations and the fact that no significant changes in total porosity were observed during testing, the changes in hydrogeological properties and the capacity to control oxygen migration of the tested CCBEs due to freeze–thaw cycles can be attributed mainly to micropore structure changes, which increased the interconnectivity of the CRBM pore system.

Insert Fig. 7 about here.

6 Conclusion

This article presented the results of a laboratory investigation of the performance of three CCBEs made with CRBMs at 5.0%, 6.5%, and 8.0% bentonite to control oxygen migration when exposed to freeze–thaw cycles. The initial saturation conditions of the moisture-retaining layer were close to saturation for the 5.0% and 8.0% bentonite CRBM CCBEs and at natural water content for the 6.5% bentonite CRBM CCBE. These different saturation conditions allowed assessing the performance of ideal (saturated) and unsaturated (as would normally occur in the field) CCBEs. The experimental procedure consisted of a sequence of oxygen diffusion and infiltration tests conducted in initial conditions and after 1, 3, and 5 freeze–thaw cycles. The measured θ_w profiles indicated that, in initial conditions, the capillary barrier effects allowed fairly high saturation in

the moisture-retaining layers for the 5.0% and 8.0% bentonite CRBM CCBEs. However, these high saturations could not be maintained after applying freeze–thaw cycles. The θ_w profiles monitored for the 6.5% bentonite column showed that the CRBM moisture-retaining layer failed to recharge following water infiltration, and saturation remained below 52%. Oxygen diffusion tests were performed on the experimental columns in various conditions in order to estimate D_e for all moisture-retaining layers and saturation conditions. Based on the obtained D_e , steady-state oxygen fluxes $F_{s,L}$ through the cover were calculated. The results for D_e and $F_{s,L}$ were consistent, indicating good initial cover performance for the 5.0% (after infiltration only) and 8.0% bentonite CRBM CCBEs, with D_e values below $1 \times 10^{-8} \text{ m}^2/\text{s}$ and oxygen fluxes below $1 \text{ mol O}_2/\text{m}^2/\text{year}$. However, the capacity of the tested covers to act as oxygen barriers was strongly reduced by freeze–thaw cycles, as oxygen fluxes exceeding $71 \text{ mol O}_2/\text{m}^2/\text{year}$ and $57 \text{ mol O}_2/\text{m}^2/\text{year}$ were observed after the third freeze–thaw cycle for columns 1 and 3, respectively. This loss of performance is mainly due to changes in the internal pore structure and the increased pore interconnectivity after the freeze–thaw cycles: k_{sat} values increased by nearly three orders of magnitude after 5 cycles. The overall performance of the 8.0% bentonite CRBM CCBE was slightly higher than that for the 5.0% bentonite CRBM CCBE. In the case of the 6.5% CRBM CCBE placed at its natural water content, the results indicate that poor cover performance would be expected in the initial conditions, and no significant recharge was observed during the testing period.

This study was inspired by a case study at the Raglan Mine in northern Quebec, where the potential use of a CRBM CCBE is under study. For the present laboratory column experiments, the materials used to construct a CRBM CCBE at the Raglan Mine were used, with no changes in parameters or properties. The tested CCBE materials could therefore be optimized in terms of gravel grain size distribution and bentonite type in order to achieve acceptable oxygen barrier performance. Finally, the results of this study suggest that the laboratory approach used is suitable for performance assessments of these CCBE configurations and for assessing the effects of

freeze–thaw cycles on water and gas transport in these layered systems. The proposed approach is not site-dependent, and it could be adapted for performance assessments of other CCBE configurations.

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Table 1. Main physical and geotechnical properties of tested materials.

Parameter	Crushed rock	Bentonite	5.0% CRBM	6.5% CRBM	8.0% CRBM
d_{10}^1 (mm.	0.38	0.05	0.20	0.18	0.16
d_{50}^1 (mm.	7.05	0.27	6.527	6.368	6.203
d_{90}^1 (mm.	17.20	0.50	16.98	16.92	16.86
d_{100}^1 (mm.	20.00	1.00	20.00	20.00	20.00
C_U^2 (-.	24.00	6.02	42.73	47.67	52.73
G_s (-.	3.04	2.59	3.02	3.01	3.00
k_{sat} (cm/s.	2.6×10^{-1} *	-	3.5×10^{-7}	2.2×10^{-7}	4.8×10^{-8}
ψ_a (kPa.	0.5*	-	8	10 - 20	10 - 20
ψ_r (kPa.	7*	-	15 000	15 000	15 000
θ_r (m ³ /m ³ .	0.02*	-	0.02	0.02	0.02

¹ d_x : diameter corresponding to x w/w % passing on the cumulative grain-size distribution curve

²: Coefficient of uniformity (d_{60}/d_{10}).

*: data from Coulombe (2012).

-: not tested

Table 2. Detailed experimental stages with their corresponding duration and characteristic testing temperatures.

Testing conditions	Stages*	Duration (days)	T _{average} (°C)	T _{freezing} (°C)	T _{thawing} (°C)
Pre-testing phase	H	11	-	-	-
	H+S	52	-	-	-
	TCD	21	5.0	-	-
Initial conditions	DT ₁	13.9	5.2	-	-
	IT ₁	19.9	5.1	-	-
	DT ₂	10.1	5.1	-	-
1st freeze–thaw cycle		6.0	-	-13.2	5.0
1	DT ₃	10.1	5.1	-	-
	IT ₂	10.0	5.1	-	-
	DT ₄	9.0	5.0	-	-
2nd freeze–thaw cycle		6.0	-	-11.6	5.0
3rd freeze–thaw cycle		6.0	-	-11.5	5.0
3	DT ₅	10.8	5.0	-	-
	IT ₃	10.0	5.2	-	-
	DT ₆	10.3	4.3	-	-
4th freeze–thaw cycle		6.0	-	-12.7	5.0
5th freeze–thaw cycle		6.0	-	-12.7	5.0
5	DT ₇	7.0	5.0	-	-
	IT ₄	10.0	5.1	-	-
	DT ₈	10.0	5.4	-	-
6	DIS	11.0	5.0	-	-
		>3.0	-	-13.0	-

*: H (hydration., S (saturation., TCD (thermal conditioning and drainage., DT_i (diffusion test_i., IT (infiltration test_i., DIS (Dismantling.

-: not applicable

Table 3. Applied saturation conditions and column 1 moisture-retaining layer estimated and modelled D_e (framed values..

Depth (m.	Initial conditions		Cycle 1		Cycle 3		Cycle 5	
	BI	AI	BI	AI	BI	AI	BI	AI
D _e (m ² /s.								
1.0–1.2	1.1×10 ⁻⁶	3.0×10 ⁻¹¹	2.3×10 ⁻⁶	2.1×10 ⁻⁶	2.6×10 ⁻⁶	2.4×10 ⁻⁶	2.7×10 ⁻⁶	2.4×10 ⁻⁶
1.2–1.4	1.1×10 ⁻⁸		3.1×10 ⁻⁷	3.1×10 ⁻⁷	5.2×10 ⁻⁷	3.9×10 ⁻⁷	5.1×10 ⁻⁷	3.8×10 ⁻⁷
1.4–1.6			1.6×10 ⁻⁹	6.6×10 ⁻⁹	1.2×10 ⁻⁷	5.8×10 ⁻⁸	9.3×10 ⁻⁸	6.4×10 ⁻⁸

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Table 4. Applied saturation conditions and column 2 moisture-retaining layer estimated and modelled D_e (framed values..

Depth (m.	Initial conditions		Cycle 1		Cycle 3		Cycle 5	
	BI	AI	BI	AI	BI	AI	BI	AI
	D_e (m^2/s .)							
1.0 – 1.2	2.1×10^{-6}	2.3×10^{-6}	2.6×10^{-6}	2.4×10^{-6}	2.9×10^{-6}	2.6×10^{-6}	2.9×10^{-6}	2.6×10^{-6}
1.2 – 1.4	3.7×10^{-6}	3.7×10^{-6}	3.7×10^{-6}	3.6×10^{-6}	3.1×10^{-6}	3.7×10^{-6}	3.7×10^{-6}	3.7×10^{-6}
1.4 – 1.6	8.3×10^{-8}	9.2×10^{-8}	7.5×10^{-8}	7.5×10^{-8}	1.2×10^{-7}	8.0×10^{-8}	1.0×10^{-7}	7.2×10^{-8}

Table 5. Applied saturation conditions and column 3 moisture-retaining layer estimated and modelled D_e (framed values..

Depth (m.	Initial conditions		Cycle 1		Cycle 3		Cycle 5	
	BI	AI	BI	AI	BI	AI	BI	AI
D_e (m^2/s).								
1.0 – 1.2	1.1×10^{-9}	2.0×10^{-9}	5.3×10^{-7}	4.1×10^{-7}	9.0×10^{-7}	3.3×10^{-7}	7.2×10^{-7}	3.3×10^{-7}
1.2 – 1.4			1.9×10^{-7}	1.6×10^{-7}	3.7×10^{-7}	9.1×10^{-8}	1.4×10^{-7}	9.6×10^{-8}
1.4 – 1.6			7.1×10^{-9}	1.4×10^{-8}	7.3×10^{-8}			

Figure Captions

Fig. 1. Instrumentation, stratigraphic configurations, and laboratory set-up for the three experimental columns.

Fig. 2. Average volumetric water contents measured during the various oxygen diffusion tests and at the dismantling stage for column 1, column 2, and column 3.

Fig. 3. Change in interstitial O_2 concentration (%) with time measured in the top of the capillary break layer during the oxygen diffusion tests conducted in various testing conditions for experimental columns 1, 2, and 3.

Fig. 4. Initial average porosity and final dismantling porosity for columns 1, 2, and 3.

Fig. 5. One-dimensional column oxygen diffusion test conceptual model and POLLUTE modelling parameters.

Fig. 6. Stationary oxygen flux at the base of the tested CCBs based on a moisture-retaining layer thickness of 0.60 m and the corresponding calculated $\overline{D_e}$.

Fig. 7. Increase in hydraulic conductivity in the 6.5% bentonite CRBM due to cyclic freezing and thawing.

Fig. 1. Instrumentation, stratigraphic configurations, and laboratory set-up for the three experimental columns.
159x121mm (300 x 300 DPI)

Fig. 2. Average volumetric water contents measured during the various oxygen diffusion tests and at the dismantling stage for column 1, column 2, and column 3.
106x79mm (600 x 600 DPI)

Fig. 3. Change in interstitial O₂ concentration (%) with time measured in the top of the capillary break layer during the oxygen diffusion tests conducted in various testing conditions for experimental columns 1, 2, and 3.
148x263mm (600 x 600 DPI)

Fig. 4. Initial average porosity and final dismantling porosity for columns 1, 2, and 3.
95x78mm (600 x 600 DPI)

oxygen diffusion test

Fig. 6. Stationary oxygen flux at the base of the tested CCBEs based on a moisture-retaining layer thickness of 0.60 m and the corresponding calculated $\overline{D_e}$.

Fig. 7. Increase in hydraulic conductivity of a 6.5% bentonite CRBM due to cyclic freezing and thawing.

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