Prediction of Drainage Water Quality from Mining Waste with Carbon Sequestration Potential

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ABSTRACT

Fixation of CO_2 by brucite or serpentine is widely recognized as a passive way to reduce carbon footprint. Ultramafic rocks are rich in minerals with high potential to sequester CO_2 in ambient conditions and are sometimes associated with base metals, such as Ni. Extraction of these base metals can lead to exposure of ultramafic wastes to atmospheric conditions, such as in the case of the Dumont deposit (Royal Nickel Corporation) near Amos, Quebec, Canada. The wastes of this future open pit mine are low in sulfide content (<1 %) and rich in ultramafic minerals with significant potential to trap atmospheric CO_2. This natural process, leading to the precipitation of stable carbonates, can influence the long term drainage water quality of the wastes. The aim of this study is to predict the geochemical behavior of such wastes, as well as the impact of CO_2 sequestration on drainage water quality. The five main lithologies found in the Dumont deposit were sampled, and a combination of these lithologies was studied as waste rock, whereas a tailings sample was produced by pilot scale metallurgical testing. All samples were submitted to laboratory kinetic columns tests and to CO_2 consumption tests. The results suggest that dissolution of Magnesium silicates and precipitation of secondary Magnesium carbonates are the main geochemical processes controlling Mg, Ca, and Si concentrations in the leachates, and generating high alkalinity values. The ultramafic lithologies, which fix CO_2, show alkaline drainage, with pH values varying from 9 to 10, versus 7.0 to 8.5 for non-ultramafic samples. These results help understand the mechanisms of CO_2 sequestration by mine wastes and its impact on water quality.

Keywords: mine wastes, ultramafic rocks, carbon sequestration, kinetic columns tests, alkaline drainage

INTRODUCTION

Ultramafic rocks are considered to be good candidates as an anthropogenic carbon sink. Many studies highlight the use of ultramafic rocks with high concentrations of serpentine (lizardite, chrysotile) and brucite to react with CO_2 (Goff & Lackner, 1998; Lackner et al., 1995). There are large amounts of mafic and ultramafic rocks scattered in the earth's crust. These rocks are sometimes associated with base metals, and their extraction leads to the exposure of mine wastes at the surface. Even though the fixation of CO_2 by magnesium silicates is a slow process, the exposure of significant quantities of mining wastes with high concentrations of serpentine minerals can reduce the carbon footprint of the mining activities (Harrison et al., 2013). Carbon sequestration by mine waste is very attractive because of the quantities that are produced during mining. Furthermore, these materials are often finely ground, which increases the exposed surface area.

Existing literature on carbon sequestration by mining wastes focuses mainly on the estimation of their sequestration potential or on the evaluation of factors that potentially affect their sequestration potential, such as temperature, surface passivation, or degree of saturation (Assima et al., 2014; 2012). Many research projects also focused on techniques to accelerate the process, in order to fix more Carbon and to move to the industrial scale (Larachi et al., 2010). Fixation of CO_2 by Mg or Ca silicates, also called mineral carbonation, likely occurs in the mine tailings in the following steps: (1) dissolution of CO_2 into interstitial water; (2) acid release is consumed by silicates which release Mg^{2+} or Ca^{2+}; (3) reaction between Mg^{2+} or Ca^{2+} with HCO_3^- or CO_3^{2-} to form stables carbonates under ambient conditions (Harrison et al., 2013; Wilson et al., 2009a). Processes leading to the formation of stables carbonates can impact the quality of mine drainage. As reported by Bea et al. (2012), Carbon sequestration by ultramafic tailings at Mount Keith (Australia) leads to alkaline drainage with pH varying from 8 to 10. Wilson et al. (2009b) suggest that precipitation of nesquehonite in kimberlite processing wastes could increase their neutralization potential by 1-2 %. Rollo and Jamieson (2006) also report that dissolution of chrysotile and precipitation of Magnesium carbonates are an important process affecting pore water composition at the same site. Thus, it is important to consider the Carbon sequestration potential of ultramafic mine wastes in order to predict the drainage water quality.

The wastes studied here are those of the Dumont project of Royal Nickel Corporation, an open pit project located 25 km west of Amos in the Abitibi-Témiscamingue region, Québec, Canada. The project intends to mine and process a deposit of Nickel mineralization which contains 2 types of mineralization: Nickel sulfides (pentlandite, millerite) and an Iron-Nickel alloy (awaruite). The operation plans to treat up to 105 tons/day of ore at an average grade of 0.27 % Ni, over a lifetime of 31 years. The deposit lies within a mineralized zone of serpentinized dunite, composed mainly of serpentine, and minor minerals including magnetite, brucite, chlorite, diopside, and chrysotile. Waste rocks are composed mainly of peridotite, gabbro and basalts containing serpentine, chromite, clinopyroxene, , quartz, and plagioclase (RNC, 2012). Previous work on the Royal Nickel wastes have highlighted their ability to fix Carbon (Plante et al., 2014; Awoh et al., 2013; Pronost et al., 2011), due to high serpentine concentrations in the wastes. (Pronost et al., 2011). Eudiometer tests show that brucite and serpentine could react with CO_2 to form stable hydrated Magnesium carbonates (Pronost et al., 2011). The carbonates formed are dypingite $Mg_5(CO_3)_4(OH)_2$-$5H_2O$) and nesquehonite $Mg(HCO_3)(OH)$-$2H_2O$) (Pronost et al., 2011). Awoh et al. (2013; 2014) developed an innovative CO_2 consumption test in order to quantify the CO_2 flux sequestered by mine wastes. Results of the various laboratory and *in situ* CO_2 consumption tests suggest that the Dumont wastes sequester CO_2 fluxes of up to 1400 g/m^2/year in typical ambient conditions. These studies have also shown that the non-ultramafic lithologies of the Dumont deposit do not consume CO_2. CO_2 consumption tests also show that a specific water content is required for optimal carbonation (Assima et al., 2012; Awoh et al., 2013). Since CO_2 sequestration involves the precipitation of secondary carbonates that may affect water quality, the aim of this work is to predict water quality of the future Dumont wastes and the impact of Carbon sequestration on the quality of the drainage water from the Dumont wastes.

MATERIALS AND METHODS

Materials characterization

Different types of waste rocks from the Dumont project were sampled for the present study: one tailings sample, one ultramafic waste rock sample, and 5 samples of the main lithologies found in the Dumont project (low grade dunite, upper peridotite, footwall ultramafic (which is dominantly peridotite)gabbro, and volcanic basalt). The tailings sample was produced in a pilot-scale plant, while the ultramafic waste rock was taken from a bulk sample at the site consisting of dunite and peridotite. The five main lithologies were sampled from representative sections of drill core.

All samples were characterized physically and chemically. The chemical characterization was performed after an acid digestion (HNO_3-Br_2-HF-HCl) followed by ICP-AES analysis of over 20 elements. A whole-rock analysis of major elements of the Dumont samples was performed by X-ray fluorescence (XRF) following a Lithium borate fusion on pulverized sample aliquots (85 % < 200 mesh) by Acme Analytical Laboratories Ltd, Vancouver, Canada. The detection limits of the chemical characterizations of the oxides ranged between 0.001 and 0.1 %. Results of major elements are presented in Table 1. These results, combined with X-ray diffraction (XRD) analysis, allow identifying and quantifying minerals present in the samples. The tailings and ultramafic lithologies (low grade dunite, upper peridotite, footwall ultramafic) contain between 34.06 % and 42.68 % MgO. The gabbro and volcanic (which are not ultramafic materials) contain 9.96 % and 6 % MgO, respectively. All wastes contain significant amounts of SiO_2between34.17 (footwall ultramafic) and 49.27 % (gabbro). While the theoretical ratio of MgO/SiO_2 in serpentine equals 1.00, it is >1.00 for the tailings, low grade dunite, upper peridotite, and footwall ultramafic samples, suggesting an excess of Mg which can be attributed to brucite, $Mg(OH)_2$. Iron is present in all samples, with Fe_2O_3 concentrations between 5.82 % and 12.62 %. The gabbro and volcanic samples also contain more Al_2O_3 (13.41 and 14.07 %, respectively) and CaO (11.97 and 9.78, respectively) compared to the other samples (Al_2O_3: 0.22-3.08 %; CaO: 0.05-0.86 %), revealing the differences in mineralogical compositions between the non-ultramafic samples of gabbro and volcanic in comparison to the other materials. The initial total carbon and sulfur contents of samples were measured by a LECO carbon/sulfur analyzer (Maxxam Analytical, QC, Ca) with a reportable detection limit of 0.05 %.

The highest concentration of carbon was detected in the volcanic sample, due to the presence of calcite (0.8 %). The tailings, waste rock, low grade dunite, upper peridotite, gabbro, and footwall ultramafic contain similar carbon concentrations (between 0.12 and 0.17 %). The highest sulfur content was measured in the tailings sample with 0.2 %, while other samples contain less than 0.08 %.

X-Ray Diffraction results coupled with oxides and trace elements analysis indicate that the matrix of ultramafic samples such as tailings, waste rock, low grade dunite, upper peridotite, and footwall ultramafic is composed mainly of serpentine ($(Mg,Fe)_3Si_2O_5(OH)_4$). Other minerals are also, present such as brucite ($Mg(OH)_2$), magnetite (Fe_3O_4), and chlorite ($(Fe,Mg,Al)_6(Si,Al)_4O_{10}(OH)_8$). The waste rock sample also contains albite ($NaAlSi_3O_8$) and quartz (SiO_2). The gabbro and volcanic contain predominantly hornblende and epidote, with lesser quartz, albite, and diospide ($CaMgSi_2O_6$). The volcanic sample contains around 5 % calcite ($CaCO_3$). There is no serpentine detected in the gabbro and volcanic samples.

Table 1 Chemical composition (major elements) of the Dumont samples

	Ultramafic Waste rock	**Tailings**	**Gabbro**	**Low Grade Dunite**	**Upper Peridotite**	**Volcanic (Basalt)**	**Footwall ultramafic**
%SiO_2	41.40	35.10	49.27	34.53	35.83	47.93	34.17
%Al_2O_3	3.08	0.22	13.41	0.23	1.27	14.07	0.58
%Fe_2O_3	7.07	5.82	9.41	6.71	9.59	12.62	9.42
%CaO	0.86	0.05	11.97	0.15	0.69	9.78	0.16
%MgO	34.06	42.49	9.96	42.68	38.97	6.00	40.64
%Na_2O	0.74	<0.01	1.64	<0.01	<0.01	1.99	<0.01
%K_2O	0.25	<0.01	0.18	<0.01	<0.01	0.28	<0.01
%MnO	0.09	0.11	0.18	0.11	0.14	0.22	0.12
%TiO_2	0.13	<0.01	0.40	<0.01	0.05	0.87	0.02
%P_2O_5	0.02	0.05	0.03	<0.01	<0.01	0.08	<0.01
%Cr_2O_3	0.16	0.24	0.07	0.49	0.72	0.03	0.99
%LOI	12.31	16.15	2.72	15.41	12.94	5.30	14.11
total	100.18	100.23	99.22	100.30	100.20	99.18	100.21
MgO/SiO_2	0.82	1.21	0.20	1.24	1.09	0.13	1.19
% C	0.17	0.15	0.13	0.12	0.16	0.80	0.12
% S	0.04	0.17	0.02	0.04	0.03	0.06	0.02

The <2 cm fraction of the samples was selected for this study, and the grain size distribution was analyzed by sieving for the fraction between 2 cm and 355 µm. The tailings and the <355 µm fraction of the other samples was analyzed with a Malvern Mastersizer S laser grain size distribution analyzer. The main characteristics of the particle size distributions are shown in Table 2. The waste rock sample contains more fines (10% <355 µm) compared to the lithology samples (less than 4 %< 355 µm) because of the difference in the sampling method. This higher fine content in the waste rock sample could lead to a higher reactivity (due to the higher specific surface) compared to the lithology samples in the present study.

Table 2 Grain size distribution parameters of the samples

	% < 355 μm	D_{10} (μm)	D_{50} (μm)	D_{90} (μm)
	wt.%	wt.%	wt.%	wt.%
Tailings	99	2.2	32.4	191
Waste Rock	10	425	6700	13200
Low grade dunite	2	1400	9500	13200
Upper Peridotite	2	2800	11200	16000
Footwall ultramafic	2	1400	9500	16000
Gabbro	2	4750	11200	16000
Volcanic	2	2800	11200	16000

Kinetic column tests

Samples from the Dumont project were submitted to laboratory kinetic column tests. Approximately 20 kg of coarse material were deposited in plexiglass columns of 14 cm in diameter and 90 cm in height, while 8 kg of tailings were deposited in a column of the same diameter (14 cm) but lower height (60 cm). The interior of the tailings column was coated with vacuum grease in order to avoid preferential flow along the column wall. A porous ceramic plate was installed at the bottom of the tailings column in order to control the suction potential at a value of 1 m below the tailings. A volumetric water content probe and a dielectric water potential probe were installed 15 cm from the bottom of the tailings in the column. A geotextile was put at the base of the other columns in order to retain the fines within the column. The columns are flushed with 2 L of deionized water every two weeks for the waste rock and lithology samples, and once per month for the tailings column. The deionized water used has a pH between 5.5 and 6 and its carbonate content is negligible. The leachates were recovered after a contact time of a few hours. The column tests were run for over one year for coarse materials and for 2 years for tailings. Thermodynamical equilibrium calculations using Vminteq version 3.0 (USEPA, 307 1999) were performed on data from kinetic column tests in order to identify the different carbonates generated during kinetic tests. CO_2 consumption tests were performed on the columns each week. Detailed results of CO_2 consumption tests are not presented here, but they reveal that the gabbro and volcanic do not consume CO_2, while all other samples do (Plante et al., 2014; Awoh et al., 2014, 2013). The calculated fluxes for the tailings column vary between 50 and 800 g/m^2/year, depending on the interpretation method used and the water content. For coarse materials, the estimated fluxes reached up to 1400 g/m^2/year.

RESULTS AND DISCUSSION

Leachates recovered after each flush were analyzed for several parameters such as acidity, alkalinity, pH, electrical conductivity, and dissolved and total metals by ICP-AES and ICP-MS, and various anions by ionic chromatography. Only pH, sulfate, Mg, Si, and Ni are presented here. The pH of the leachates stabilized quickly for all materials and do not show a transitional phase, except for column tailings. The lithologies which do not sequester carbon (gabbro and volcanic) have a pH around 8.5, while other lithologies (low grade dunite, footwall ultramafic, upper peridotite) and the waste rock have a pH between 9.0 and 10. For tailings, the pH leachate remained near neutral (between 7.0 and 8.5) during the first 200 days, before gradually increasing and stabilizing between 9.0 and 9.5.

The lower pH observed during the first cycles for the tailings can be explained by the slower diffusion of CO_2 through the tailings than through the coarser waste rock and lithology samples, leading to a slower CO_2 sequestration and, therefore, a slower effect on leachate pH. The leachates of the ultramafic materials (tailings, ultramafic waste rocks, low grade dunite, upper peridotite, footwall ultramafic) contain high Mg concentrations (around 50-100 mg/L). In these types of wastes, Mg comes from the dissolution of serpentine and/or brucite with dissolved CO_2. For gabbro and volcanic, Mg in the leachates ranges from 1 to 5 mg/L, probably from the dissolution of gangue minerals such as hornblende and chlorite. The Si concentrations in the tailings leachates (which vary from 20 to 80 mg/L) are higher than in the other leachates. For gabbro and volcanic, Si varies from 1.7 to 5.7 mg/L. The other samples only show Si in some of the last leachates (0.1-1.0 mg/L) at the end of the kinetics test. The ratio of Mg/Si concentrations are very high, which suggest an incongruent dissolution of serpentine with a preferential release of Mg over Si and/or a fastest dissolution of brucite compared to serpentine (Assima et al., 2013; Harrison et al., 2013). All columns produce alkalinity and highest values were obtained for waste rock (275-300 mg $CaCO_3$/L) compared to other lithologies (<20 mg $CaCO_3$/L). The alkalinity in the tailings leachates increase with pH and stabilize around 200 mg $CaCO_3$/L after 400 days, probably due to the gradual buildup of secondary carbonates within the tailings. The metal concentrations remain very low during the column tests. For example, the Ni concentrations are shown in Figure 1; all leachates show very low Ni concentrations not exceeding 12 µg/l, and often below the detection limit (1 µg/L).

The sulfate release rates were calculated from the cumulative leached masses for the samples leaching regular detectable sulfate concentrations (Table 3). These results indicate an increase of sulfate release rate for tailings from 0.89 g/kg/d to 1.25 mg/kg/d at 290 days. Saturation indexes calculated indicate that a variety of calcium and magnesium carbonates may precipitate (Table 4). No carbonate is suggested to precipitate in the volcanic column and before day 290 in the tailings, when pH is near neutral. It suggests that carbonate precipitation occurs at alkaline pH values for tailings column. The most common magnesium carbonates to precipitate are magnesite and hydromagnesite.

At the end of the kinetics tests, all columns were dismantled and the final carbon content was determined for the first layer of the column, the first 5 cm for waste rock, tailings and footwall ultramafic, and for the first 10 cm of the others columns. The initial and final carbon contents are shown in Table 5. All materials show an increase in total carbon content except for the gabbro. The carbon enrichment for volcanic needs further investigation, since no CO_2 consumption is observed in that lithology. According to the carbon enrichment during the kinetic tests, the Dumont wastes can fix between 4 and 15 kg of CO_2 per ton of waste exposed at the surface, although the scale-up of these results to the large scale still needs to be investigated. Also, cementation was observed in the first 5 cm of the column for all samples that consume CO_2. Examples of cemented phases found in the columns are shown in Figure 2. The precipitation of secondary magnesium carbonates probably cemented the grains in the columns. The cementation of surface mining wastes due to mineral carbonation is already reported for chrysotile mine wastes in southern Québec, Canada (Beaudoin et al., 2008; Assima et al., 2012, 2013) and for ultramafic wastes of Cassiar and Clinton Creek in the Yukon Territory and British Columbia, Canada (Wilson et al., 2009a).

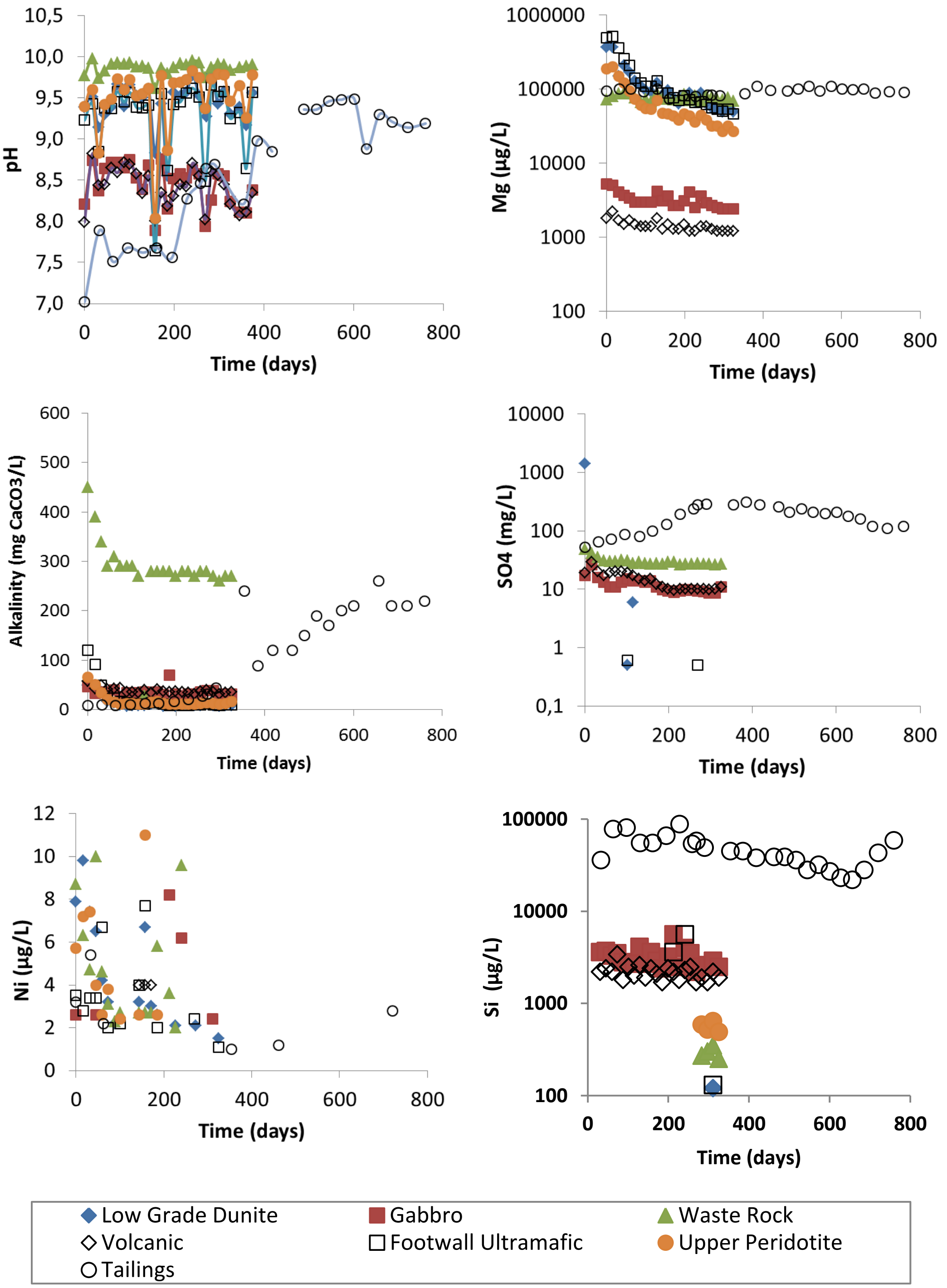

Figure 1 Evolution of pH, Mg, Alkalinity, Sulfates, Ni and Si concentration in kinetic column tests

Table 3 Sulfate release rates from kinetic column tests

	Ultramafic Waste Rock	Tailings (0 to 290 days)	Tailings (290 to 830 days)	Gabbro	Volcanic (Basalt)
Sulfate release rate (mg/kg/day)	0.204	0.894	1.252	0.083	0.099

Table 4 carbonates suggested to precipitate with Visual MINTEQ

	Low grade dunite	Footwall ultramafic	Upper peridotite	Ultra-mafic Waste rock	Tailings (0 to 290 days)	Tailings (290 to 830 days)	Gabbro	Volcanic (Basalt)
Magnesite	✓	✓	✓	✓		✓		
Hydromagnesite	✓	✓	✓	✓		✓		
Calcite	✓	✓		✓		✓	✓	
Dolomite	✓	✓	✓	✓		✓	✓	
Aragonite		✓	✓	✓		✓	✓	
Artinite	✓	✓	✓	✓		✓	✓	
Huntite	✓	✓	✓			✓		
Vaterite			✓	✓				
Magnesioferrite				✓				

Table 5 Initial and final carbon content of Dumont waste before and after kinetic column tests

	Tailings	Ultramafic Waste rock	Gabbro	Low Grade Dunite	Upper Peridotite	Volcanic (Basalt)	Footwall ultramafic
Initial % C	0.15	0.17	0.13	0.12	0.16	0.80	0.12
Final % C (first layer)	0.60	0.48	0.16	0.24	0.30	0.98	0.23

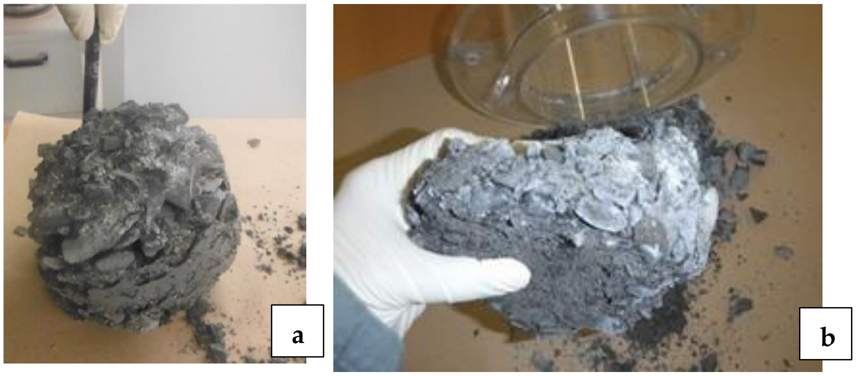

Figure 2 cementation on the first cm of columns a) Upper Peridotite, b) footwall ultramafic

CONCLUSION

Previous studies have confirmed the capacity of the Dumont wastes to sequester CO_2, and our investigations show that this process has an effect on the drainage water quality, as it leads to alkaline drainage with pH values up to 10. However, metal concentrations in the leachates remain low. The exact nature of the secondary carbonates formed will be investigated in the coming months. Cementation of the grains upon Carbon sequestration might reduce wind erosion and dust generation from the future waste rock piles and tailings ponds.

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