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Geochemical Study of Laterite and Its Use as Reactive Permeable Barrier for Responsible Management of Mine Tailings at Sabodala Gold Operations

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Abstract

The Sabodala mine uses laterite in the tailings and water dams as the main material for the dikes of the hydraulic structures and as a sealing layer (physical barrier) to limit the seepage of discharge water and protect groundwater from contamination, arsenic in particular. For the permanent storage of arsenic in mining environments and the protection of environmental matrices (soil, water and air), it is processed in the form of scorodite ($FeAsO_4 \cdot 2H_2O$) or arsenic-adsorbed ferrihydrite. Both minerals are ecologically stable under a wide range of physicochemical conditions. This study aims to investigate the geochemical behaviour of laterite and its use in a Reactive Permeable Barrier (RPB) for the responsible management of arsenic mine tailings in a sustainable development context. The methodology begins with a physico-chemical and mineralogical characterization of laterite and mine tailings. After characterization, two kinetic tests were carried out in a control Mini-Cell Alteration (MCA) and a laterite beneficiation test in a Reactive Permeable Barrier (RPB). Electrochemical parameters (pH, electrical conductivities), sulphates, calcium ions and metalloids (As and Sb) in the leachates were measured. The results of physico-chemical and mineralogical characterizations of laterite and mine tailings showed that: (i) laterites are composed of quartz (SiO_2), kaolinite ($Al_2Si_2O_5(OH)_4$), hematite (Fe_2O_3), goethite ($FeO(OH)$) and anatase (TiO_2); (ii) tailings contain high levels of arsenic and antimony; (iii) they also include calcite, ferrodolomite, quartz albite, muscovite and chlorite. Kinetic tests in MCA geochemical studies have shown that mine tailings are non-acid-generating, with low metalloid mobility (0.90% for arsenic and 0.86% for antimony). The study of laterite recovery in an RPB in MCA showed a high retention of metalloids (55.26% for arsenic and 57.14% for antimony) and sulphate and calcium ions by the laterite layer. This reactive property of laterite helps protect groundwater from pollutants leached from mine tailings and should enable the mine to develop an appropriate management approach as part of a sustainable development strategy.

Keywords

Laterite, Tailings, Contamination, Reactive Permeable Barrier, Sabodala Gold Operations

1. Introduction

Mining activities generate economic benefits for many countries around the world. The West African sub-region is currently experiencing a mining boom. Almost every country in the sub-region is in a frantic race to exploit gold, iron, diamond and oil mines. However, the mining industry generates large quantities of waste whose environmental impact, if not properly managed, can be devastating. Proper management to minimize the environmental impact of these discharges [1]-[4], particularly arsenic [5] [6], is one of the challenges for the mining industry. To store arsenic permanently or in an ecologically stable way in mining environments, the mineral is coprecipitated with iron (III) in the form of scorodite ($FeAsO_4 \cdot 2H_2O$) and arsenic-absorbed ferrihydrite, which are stable secondary minerals under a wide range of physicochemical conditions [7]-[10]. A Fe (III)/As(V) > 3 molar ratio has proved effective [11]. At neutral pH and a Fe (III)/As(V) = 4 molar ratio, poorly crystallized ferric arsenate ($FeAsO_4 \cdot H_2O$) was formed as the main arsenic phase, with arsenic-adsorbed ferrihydrite as the minor phase [12]-[14]. Over many years, the Vale plant (formerly known as lnco) in Sudbury, Canada, has deposited scorodite and arsenic-adsorbed ferrihydrite precipitates in its tailings management facility. No significant arsenic releases were observed [11] [15]. However, there is a possibility that arsenic may be released from ferrihydrite or ferric arsenate in reducing environments [16]. Kinetic testing is used to assess a material's behaviour in the face of environmental conditions. Kinetic testing is used to simulate accelerated or natural oxidation of mine tailings under controlled conditions [17] [18]. MCA tests have been used to predict Contaminated Neutral Drainage (CND) or Acid Mine Drainage (AMD). The work of Plante *et al.* (2011) on waste material from an Fe-Ti mine (Rio Tinto, Canada) predicted the CND of waste rock using MCA. The work of Chopard *et al.* [19] allowed the prediction of the AMD of various sulphide minerals and sulfosalts often encountered in mining tailings. Hakkou *et al.* [20] also used the method to show the AMD potential and estimate the mineral reaction rates of fine and coarse tailings from the abandoned pyrrhotite mine (Kettara site, Morocco). To predict the geochemical behaviour of waste rock and assess the factors controlling the mobility of trace metals, Edahbi *et al.* [21] (Kipawa project, Canada) used the MCA. The aim of this work is to study the geochemical behaviour of mine tailings from the Sabodala mine in the presence of laterite in a Reactive Permeable Barrier (RPB) for responsible management of arsenic mine tailings as part of sustainable development. The methodology of this work began with a characterization of laterite and mine tailings from the Sabodala mine using ICP-OES and x-ray diffraction. Then, kinetic tests in a Mini-Cell Alteration (MCA) were carried out. A control MCA to study the environmental behaviour (geochemistry) of mine tailings, and an MCA to study the recovery of laterite in a BPR were carried out. The electrochemical parameters (pH, electrical conductivities), sulphates and calcium ions in the MCA leachates were measured. Part of the leachate was filtered and then acidified for metals analysis by ICP-OES. Analysis of MCA leachates allows determination of the geochemical and environmental behaviour of the tailings. The comparative study of leachates from the two MCAs enables us to investigate the reactivity of laterite with respect to contaminants (arsenic, antimony, sulphates, calcium, electrical conductivity and pH).

2. Materials and Methods

2.1. Laterite Description

Laterite is a red or brown rock formed by weathering in tropical climates. In the broadest sense, it refers to all loose or indurated materials, rich in iron hydroxides or aluminum hydroxides, constituting soils, superficial horizons and deep horizons of weathering profiles. Laterites are found mainly in the intertropics. They cover almost 40% of the world's continents. Lateritic soils are lean, leached and depleted in silica and fertilizing nutrients. Oxidized iron gives the red color of a laterite. Iron minerals such as goethite (FeOOH); siderite ($Fe(CO)_3$); ankerite ($Ca(Fe,Mg)(CO)_3)_2$); chlorite [$(Mg,Fe)_5Al\ (Si_3Al)O_{10}\ (OH)_2$] and biotite K $(Fe,Mg)_3\ AlSi_3O_{10}\ (OH)_2$ can constitute metallotects hosting metalliferous concentrations (Al, Fe, Mn, Ni, Cu, Co, Cr, Ti, Au, Ag, Pt), some of which are of economic interest to countries in the intertropical zone [22].

2.2. Sampling Laterite

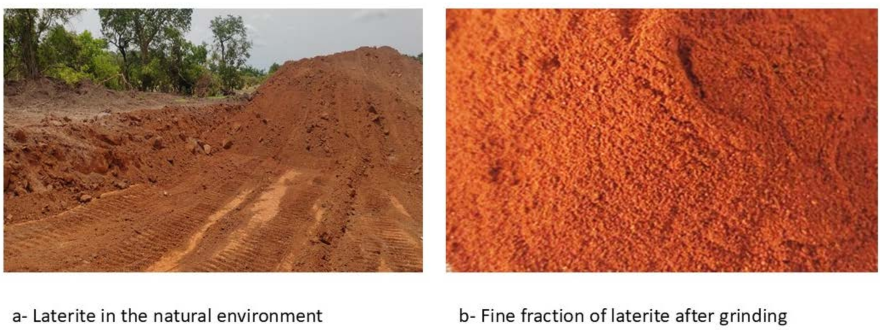

Figure 1. Laterite used.

The laterite used in this study was taken from the Sabodala mining complex (Massawa). Tailings were collected from the mine's first tailings storage pond. The lateritic armour was finely ground in an agate mortar and sieved to 80 µm (**Figure 1**). The fine fraction was used for the various laboratory experiments. Mineralogical and chemical characterization of laterites and tailings was determined by XRD and ICP-OES.

2.3. Analyses

2.3.1. DRX Analyses

Major minerals were identified with room temperature bulk-rock X-Ray Diffraction (XRD) analyses conducted at the "Institut des Matériaux de Nantes-IMN", using a Brüker "D8 Advance" powder diffractometer operated in Bragg-Brentano geometry with a Cu anode sealed X-ray tube and a focusing Ge (1 1 1) primary monochromator (selecting the Cu Kα1 radiation; λ = 1.540598 Å). The XRD patterns were obtained on bulk powders between 3.5 and 70˚ 2Θ with steps of 0.0157˚ and a counting time per step of 0.8 s.

2.3.2. ICP-OES Analyses

Alkaline fusion method was used to dissolve laterite samples and rock/soils reference materials. 100 mg of sample powder was stirred with 300 mg of lithium metaborate flux in a graphite crucible. The mixture was then heated at 1050˚C for 15 minutes and cooled to room temperature before being dissolved into a beaker filled with 100 mg of 5% HNO_3. Major and trace elements were analysed by ICP–OES (iCAP-6300, Thermo). The external calibration curves were constructed using the reference materials BIR-1, JA-1, AGV-2, Mica-Fe, FER-2 and the recommended values from Govindaraju K. [23] for BIR-1, JA-1, AGV-2 for Mica-FER and FeR-2. The boron derived from the 300 mg lithium metaborate flux was used as internal standard to monitor and correct for any instrumental drift.

2.3.3. Mini-Cell Alteration Tests

MCA tests are based on a methodology developed by Cruz *et al.* [24] modified by Villeneuve [25]. These are also kinetic tests involving wetting-drying cycles [20] [23]. To study the environmental behaviour of mine tailings and the Reactive Barrier effect of laterite, we will perform two kinetic tests in MCA (**Figure 2**). The first, known as the control test, consists of placing 100 g of mine tailings on 1.2 µm filter paper in a Büchner. In the second Büchner, the filter paper is topped with a 5 cm thick layer of laterite, and 100 g of residue is placed on top. Each Büchner is placed on a stand surmounting a 250 ml vacuum Erlenmeyer flask to collect the leachate (**Figure 2**). The device is brought to room temperature. Leaching is carried out twice a week (every 3 and 4 days alternately). During each leach, 100 ml of deionized water is introduced into each MCA and remains in contact with the residues for a period of 3 hours. The water is then removed from the cell and collected in the Erlenmeyer flask. Electrochemical parameters pH, redox potential and conductivity are measured after 3 hours of rinsing. Part of the leachate is filtered and acidified for metals analysis by ICP-OES.

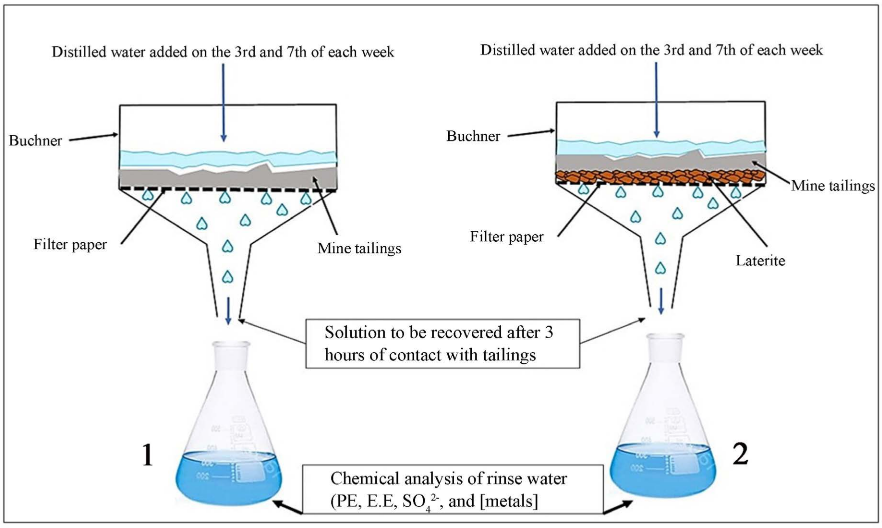

Figure 2. Schematic diagram of MCA trial set-up [26] adapted from de Cruz *et al.* [24]; 1) Geochemical study; 2) Study of laterite recovery in a BPR.

The retention rate of contaminants by laterite is calculated using the following relationship:

$$\text{Retention rate} = \frac{Mc - Ml}{Mc} \times 100 \quad (1)$$

1) Mc = Cumulative leached mass (mg) of the control MCA.
2) Ml = Cumulative leached mass (mg) of the MCA with laterite.

3. Results

3.1. Characterization of Laterite Soils

The results of chemical analysis of laterites by ICP-OES are presented in Table 1 and Table 2. The iron oxide level of the laterite studied represents 37.2%. This material is richer in ferric oxides than that from Dano (Burkina-Faso), *i.e.*, 20.4% [27]. However, they are less affluent than Ouagadougou (Burkina-Faso), *i.e.* 42.31% [28].

Table 1. Oxide composition of majors in laterites.

Lat.	Al_2O_3	CaO	Fe_2O_3	K_2O	MgO	MnO	Na_2O	P_2O_5	SiO_2	TiO_2	Total
%	18.4	0.1	37.2	0.5	0.2	0.1	0.1	0.3	33,4	1	91

Table 2. Trace element contents (mg/kg) in laterites.

Latérite	Cr	Cu	Ni	Sc	V	Zn
mg/kg	453	64	62	24	581	101

X-ray powder diffraction mineralogical analysis of laterites is shown in Figure 3. Mineralogical analysis by X-ray diffraction shows that the laterites studied are composed of quartz (SiO_2), kaolinite ($Al_2Si_2O_5(OH)_4$), hematite (Fe_2O_3) and goethite (FeO(OH)). This composition is similar to that of Burkina-Fasso laterites [27].

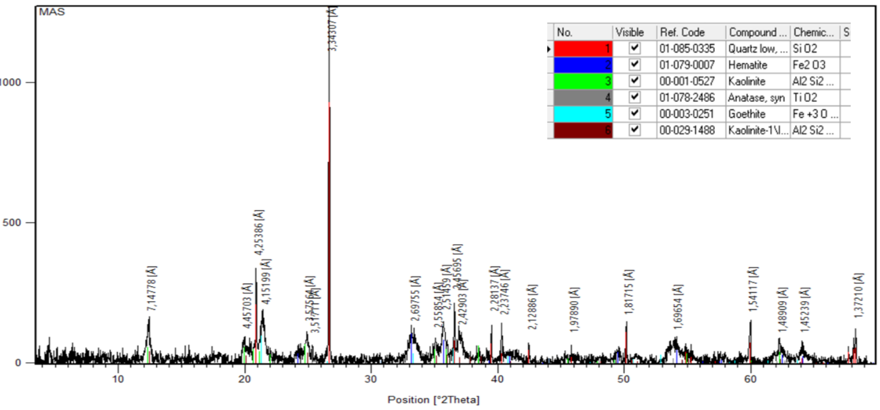

Figure 3. X-ray diffractogram (CuKα) of laterite.

3.2. Tailings Characterization

Chemical analysis of the tailings analyzed yields results presented in Table 3. The arsenic, cadmium, nickel and antimony contents are 420.16, 5.41, 55.33 and 70.72, respectively. These levels are higher than the geochemical background [29]. Mineralogical analysis has shown that the tailings consist of carbonates and silicates. Carbonates are composed of calcite, $CaCO_3$, and ferrodolomite, $CaMg_{0.6}Fe_{0.4}(CO_3)_2$. Silicates include quartz (SiO_2), albite ($NaAlSi_3O_8$), muscovite ($Kal_3Si_3O_{10}(OH)_2$) and chlorite $(Mg, Fe)_6(Si,Al)_4O_{10}(OH)_8$ (Table 4).

Table 3. Trace element contents (mg/kg) in laterite.

Tailings	As	Cd	Ni	Sb
mg/kg	420.16	5.41	55.33	70.72

Table 4. Mineralogical analysis of mine tailings by x-ray diffraction with semi-quantification using the Rietveld method.

Mineral names		Content (%)	Chemical formula
Silicates	Muscovite	21.2	$KAl_3Si_3O_{10}(OH)_2$
	Albite	17.7	$Na_{0.98}Ca_{0.02}Al_{1.02}Si_{2.98}O_8$
	Chlorite	12.9	$(Mg, Fe)_6(SiAl)_4O_{10}(OH)_8$
	Quartz	40.0	SiO_2
Carbonates	Ferrodolomite	6.5	$CaMg_{0.6}Fe_{0.4}(CO_3)_2$
	Calcite	1.7	$CaCO_3$

3.3. MCA Leaching Tests

3.3.1. Variation in Electrochemical Leachate Parameters

Samples were subjected to several rinses, in each of which physico-chemical parameters were measured to assess the rate of sulphide oxidation and carbonate mineral neutralization [25] [30]. The electrochemical parameters (pH and electrical conductivities) of the leachates are presented in **Table 5**.

Table 5. Mineralogical analysis of mine tailings by X-ray diffraction with semi-quantification using the Rietveld method.

Days	pH with laterite	Control pH	E.C with laterite	Control E.C
1	6.85	6.92	5600.0	12150.0
3	6.87	6.93	2810.0	1206.0
7	6.86	6.91	1707.0	801.5
13	6.95	7.25	674.9	499.9
17	6.75	7.15	339.8	191.4
20	7.13	7.81	319.9	287.3
24	7.39	7.40	248.8	248.8
27	7.26	7.30	512.2	512.2
30	7.16	7.30	483.3	483.3
34	7.33	7.34	237.1	237.1

1. pH

Leachate pH levels vary little overall, and fluctuate around the "natural" pH, *i.e.* from 6.91 to 7.81 for the geochemical study MCA known as the control MCA. For the laterite beneficiation study MCA, leachate pH values decrease and oscillate between 6.75 and 7.39 (**Figure 4**). This decrease is due to the presence of acid-generating iron (III) in the lateritic layer. At the start of the experiment, the pH of the leachates is slightly acidic, rising until the thirteenth day of leaching. This rise in pH is due to the dissolution of carbonates such as calcite, $CaCO_3$ and ferrodolomite ($CaMg_{0.6}Fe_{0.4}(CO_3)_2$). The drop in pH at day 17 shows dolomite exhaustion [31]. By the sixth wash (day 20), carbonate remobilization justifies the rise in pH. This remobilization is due to the dissolution of calcite, ($CaCO_3$), showing pH peaks. In the seventh wash, pH levels vary slightly but remain alkaline as the silicates dissolve. A comparative study of the leachates shows that the lateritic layer results in a slight decrease in pH. This lateritic influence has no environmental consequences, as pH values oscillate in the 6.5 - 9 range.

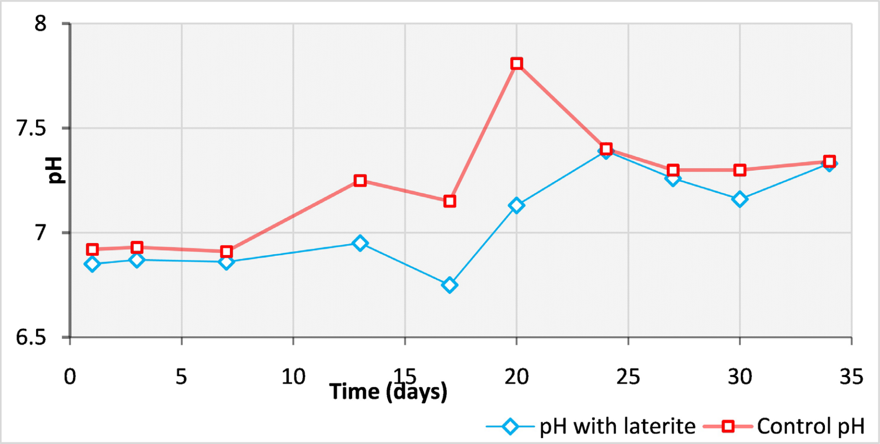

Figure 4. Evolution of pH versus time of leachates from tests in control MCA and with lateritic layer.

2. Electrical conductivity

The electrical conductivities of leachates are shown in Figure 5. At the start of the leaching process, we observed a high electrical conductivity (12,150 µs/cm) of the leach water for the control MCA compared with that with a lateritic layer (5600 µs/cm). This is due to pre-oxidation of the materials, as explained by Mayer *et al.* [32]. This is due to pre-oxidation of the materials, as explained by Mayer *et al.* This stability is due to a lack of release of ions retained by the lateritic mass, thus showing chemosorption as the mechanism for retaining the ions responsible for electrical conductivity. This variation in electrical conductivity shows that laterite can act as a reactive barrier for ions present in mine tailings.

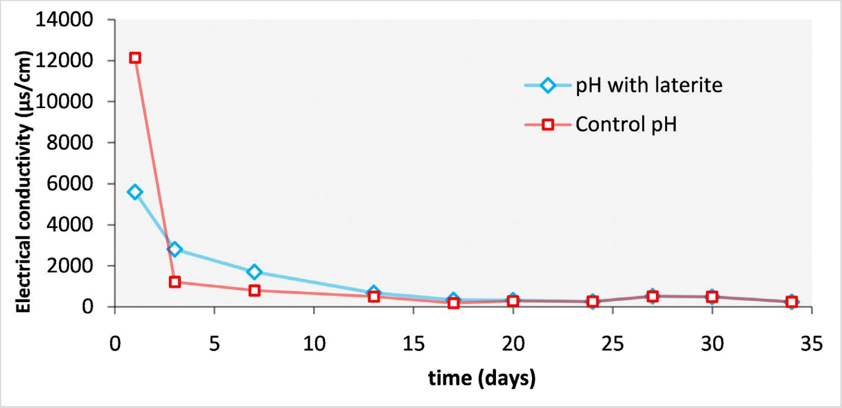

Figure 5. Evolution of electrical conductivities of leachates from tests in control ACM and with lateritic layer.

3.3.2. Variation in Sulphate Levels in Leachates

At the first wash, a high sulphate content was observed in leachates from the control MCA (3500 mg/l) compared to the MCA with lateritic layer (1202 mg/l) (Figure 6). This is due to pre-oxidation of sulphides in mine tailings, as Mayer *et al.* [32] indeed explain. It's interesting to note that pre-oxidation of a grain can modify the reactivity of sulphides, so the contact between our samples and water plus oxygen removed the oxidized surface part of the particles, taking 20 days to arrive at the unoxidised core of the mineral (sulphide passivation). The comparative study shows that the lateritic layer leads to a lowering of sulphate levels. The downward trend in sulphate levels in the leachate is due to the depletion of sulphates in the tailings and the non-release of sulphates retained in the laterite layer. The cumulative mass of sulphate leached from the control MCA is 412.6 mg greater than the 237.5 mg from the MCA with lateritic layer. The retention rate of the lateritic layer is 42.43%. The lateritic layer enables sulphates to be retained without being released.

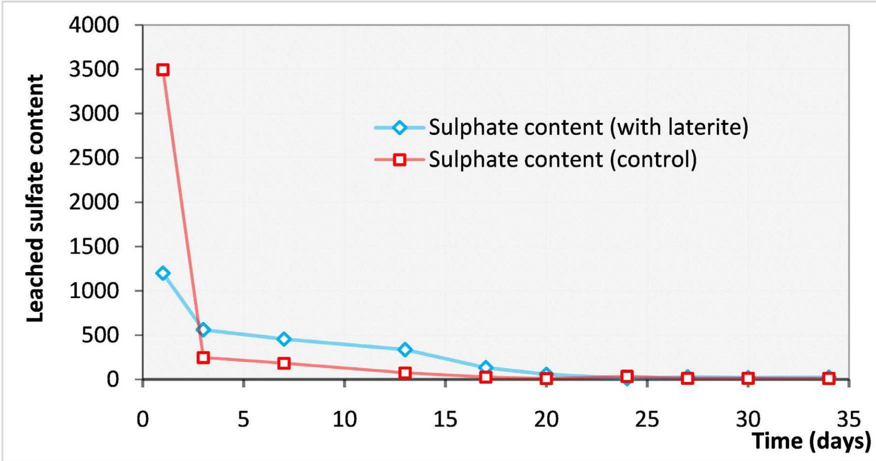

Figure 6. Variation in sulfate levels in leachates from tests with control ACM and with lateritic layer.

3.3.3. Variation in Arsenic Levels

The time trend in cumulative arsenic leaching from MCAs has not reached a plateau (**Table 6**, **Figure 7**). The absence of a plateau shows that mobilization kinetics are slow. Arsenic release from mine tailings into MCAs is low. In the control MCA, the cumulative mass of arsenic in the leachate is equal to 0.38 mg. That of the MCA containing laterite is equal to 0.17 mg. These leached masses are very low compared with the initial mass of arsenic in each mining residue, which is equal to 42.01 mg. The mobility factor for arsenic is low [22].

Table 6. Variation of Arsenic and antimony content.

Days	As content with laterite	As content control	Sb content with laterite	Sb content control
0	0	0.01	0	0.02
3	0.01	0.03	0	0.03
7	0.01	0.05	0	0.04
13	0.03	0.10	0	0.05
17	0.04	0.16	0.02	0.05

Continued

20	0.05	0.21	0.02	0.07
24	0.13	0.21	0.03	0.07
27	0.14	0.27	0.03	0.07
30	0.16	0.36	0.03	0.07
34	0.17	0.38	0.03	0.07

Arsenic is considered to be a rather immobile element because most of this metalloid is bound to the residual mineral fraction. The lateritic layer acts as a reactive barrier to arsenic, reducing the leached mass from 0.38 g to 0.17 g. In terms of the results of the environmental study, the results of the kinetic tests confirmed that arsenic concentrations are much lower in the MCA with a lateritic layer. This result shows that laterite can be used to manage arsenic discharges generated by CND.

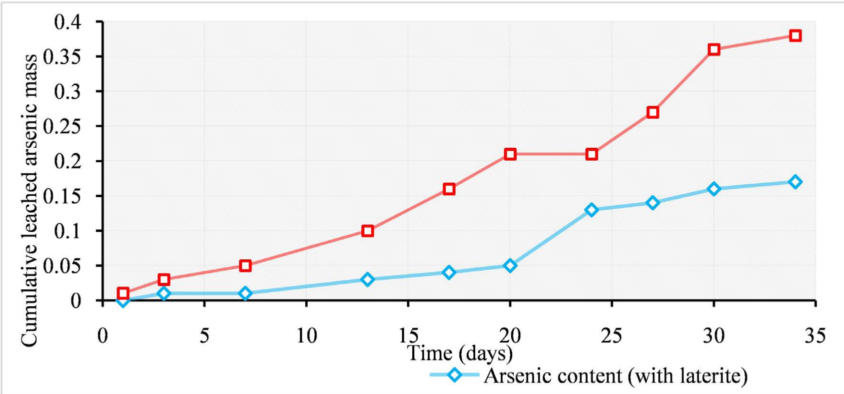

Figure 7. Variation curves for cumulative arsenic mass in rinsing solutions from ACM tests with and without a lateritic layer.

3.3.4. Variation in Antimony Levels

The time trend curves for the cumulative quantities of antimony leached from the test leachates are shown in **Figure 8**. The control curve increases until day 20, before reaching a plateau at 0.07 mg. The curve for the cumulative quantities leached from the laterite-containing MCA shows three phases. A first phase with zero content up to the fourth leach. A second phase of increasing levels up to the seventh wash cycle, with a maximum of 0.03 mg. After this stage, the curve reaches a plateau at 0.03 mg. The initial antimony mass in each tailing is 7.84 mg. The masses of antimony leached into the MCAs are low (0.07 mg for control MCA and 0.03 mg for MCA with lateritic layer). As with arsenic, the distribution of antimony in mine tailings indicates that this element was rather immobile [33] [34]. The presence of steps in the time curves of cumulative antimony leaching masses shows that all the mobile fraction is leached. For the control MCA, 0.89% of the antimony represents the mobile fraction. In the case of a lateritic layer, 0.38% passes into the leachate. This difference shows that 0.51% was retained by the laterite barrier. Lateritic soil is an excellent sorbent for metalloids, removing 55.26% arsenic and 57.14% antimony (Table 7).

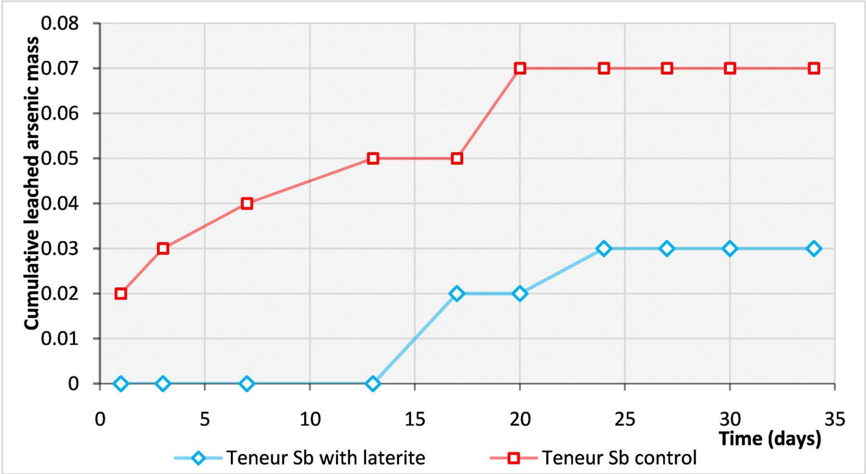

Figure 8. Variation curves for cumulative antimony mass in rinsing solutions from MCA tests with and without laterite layer.

Table 7. Trace metal leaching rates.

	As	Sb
Total initial mass (mg)	42.01	7.84
Cumulative leached mass (mg) of control ACM	0.38	0.07
Percentage of leaching of control ACM (%)	0.90	0.89
Cumulative leached mass (mg) of ACM with laterite	0.17	0.03
Percentage of leaching of ACM with laterite (%)	0.40	0.38
Retention rate of laterite layer	55.26%	57.14%

4. Conclusion

In this study, kinetic tests in Mini-Cell Alteration (MCA) were carried out to better determine the geochemical behaviour of mine tailings and to study mine lateritic soil reclamation in a Reactive Permeable Barrier. Concerning the results of the environmental study, kinetic tests by MCA confirmed that mine tailings do not produce AMD. The presence of carbonates and silicates neutralizes the acidity generated by sulphide oxidation. However, leached arsenic concentrations slightly exceed the limit value for industrial liquid effluent discharge parameters (NS 05 061), which could represent a risk of arsenic CND generation. The study shows that a lateritic layer behaves like a Reactive Permeable Barrier, retaining ions such as calcium, sulphates and metalloids (arsenic and antimony) in the leachate. In addition, there was no release of ions retained by the lateritic mass, pointing to chemosorption as the retention mechanism. This sorption method is a good indicator for the use of laterite as a Reactive Permeable Barrier (RPB). Laterite can be used in the design of tailings dams and sealing barriers to prevent contaminant migration, and in the construction of foundations under waste rock piles to limit leaching. As it is inexpensive and locally available, laterite is essential for the construction of Sabodala mine storage facilities (tailings ponds, waste rock piles, etc.). This study shows that laterite can replace the geomembranes and clay used by northern countries as waterproofing mats.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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