Strategies for Fixation of Leachable Ions from Bauxite Residue for use in Cementitious Materials

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Abstract

Driven by sustainability goals, the major players of the aluminum industry aim to eliminate new permanent storage of bauxite residue (BR) by 2050. Complete BR use is feasible through large material streams, with construction materials – particularly as a supplementary cementitious material (SCM) – being one of the most promising approaches due to the high demand for cement. However, BR's high alkalinity and low reactivity pose challenges for cement chemistry, alongside environmental concerns regarding leachable elements such as sodium, which can cause irritation and corrosion in human tissue, chromium, and other toxic metals with carcinogenic potential. Two main strategies have been developed to mitigate the negative effects of BR: thermochemical activation, which involves energy-intensive processes like calcination and vitrification, and chemical fixation, which incorporates BR into ternary cementitious systems that react with SCMs. This study presents strategies to immobilize the main deleterious elements in BR prior to its application through chemical fixation with reactive materials (such as slags, pozzolans, and other materials). These strategies will enable BR to be used both in the production of a co-product that can be marketed as an SCM and in direct cementitious applications (concrete and mortar) without the need for energy-intensive processes. Pure solutions of the target deleterious elements were simulated based on their concentrations in raw BR. These solutions were mixed with the reactive materials in varying proportions, and the solubilized extract was analyzed to assess ion fixation efficiency. Effective reactive materials were then tested in BR solutions to validate their ability to immobilize these ions. The results demonstrate that ion fixation is achievable without high-energy processing, thereby supporting the large-scale use of BR as a safe byproduct in the cement industry.

Keywords: Bauxite residue, Leaching, Ion fixation, Supplementary cementitious material.

1. Introduction

The aluminum industry faces a significant environmental challenge related to the disposal of bauxite residue (BR) generated by the Bayer process. It typically exhibits high alkalinity and contains residual NaOH and trace elements that can leach into the environment if not properly immobilized [1–3]. Therefore, its chemical stabilization is a key step toward expanding its potential for safe and effective reuse. Full-scale utilization of BR requires high stream material flows enabled by competitive, economically viable, and large-scale solutions. In this context, the construction sector emerges as a promising pathway for BR reuse, particularly in cementitious formulations where it can act as a supplementary cementitious material (SCM) [2, 4].

However, to ensure the safe use of BR for both end-users and handling personnel, certain technical challenges must still be addressed, particularly the need to stabilize potentially leachable deleterious elements. Two main strategies have been adopted to safely incorporate BR into

cementitious compositions. The first involves the thermochemical activation of the residue, employing thermal processes such as calcination and vitrification to stabilize BR, thereby reducing the leaching of harmful substances and improving its suitability for cement-based applications [5–7]. Nevertheless, these techniques are energy-intensive, leading to higher costs and environmental impacts, which hinder their large-scale feasibility. The second strategy focuses on direct chemical fixation, wherein BR is incorporated into ternary cementitious systems containing other supplementary cementitious materials. These systems promote reactions that immobilize leachable elements without requiring significant energy input [7–13].

Aiming to reduce the handling risks associated with BR without resorting to energy-intensive processes, this study evaluates the chemical fixation capacity of key deleterious elements in BR using reactive materials such as slags, pozzolans, and other compatible materials, before its application on cementitious materials. Once these ions are immobilized, BR can be used either in the production of a co-product marketable as an SCM or directly in cementitious applications such as concrete and mortars.

2. Materials and Methods

Three different Brazilian bauxite residues (BRs) were used in this study and were solubilized in water following the Brazilian standard procedure for obtaining solubilized extracts from solid waste (NBR 10006) [14]. This procedure consists of mixing the solid waste with deionized water at a 1:4 ratio, agitating for 5 minutes, letting it stand for 7 days, and then filtering the mixture through a 0.45-µm membrane filter to obtain the solubilized extract. The concentrations of various ions in the solubilized extract were determined using Inductively Coupled Plasma Optical Emission Spectroscopy (ICP – OES), from Thermo Scientific, iCap 6300 Duo, and the results are presented in Table 1. Two ions are highlighted due to their potential risk to human health when BR is used in cementitious materials: sodium (Na) and chromium (Cr). Their immobilization is therefore necessary prior to application. The highest concentrations of Na and Cr among the three BRs were selected as reference values. Chromium remained insoluble in BR1, which is an important observation. The chemical composition of the primary oxides, present in each BR, as determined by X-ray fluorescence (XRF), is shown in Table 2.

Table 1. Solubilized ions of each BR sample.

Soluble ions (in mg/L)	BR1	BR2	BR3	LQ
Al^{3+}	58.2	136	123	0.05
As^{3-}	< LQ	< LQ	< LQ	0.01
Cd^{2+}	< LQ	< LQ	< LQ	0.003
Cr^{tot}	< LQ	1.17	1.79	0.02
Fe^{tot}	1.28	< LQ	< LQ	0.04
Na^{+}	2359	880	1590	0.05
Pb^{2+}	< LQ	< LQ	< LQ	0.02
SO_4^{2-}	21.3	1.0	18.1	0.03
Se^{tot}	< LQ	< LQ	< LQ	0.01
pH	10.7	12.2	12.0	-

LQ = Limit of Quantification of the equipment.

Table 2. Chemical compositions of BRs.

Element (%)	BR1	BR2	BR3
Na_2O	5.23	9.72	7.1
MgO	0.15	< 0.10	0.11
Al_2O_3	24.7	22.1	21.7
SiO_2	19.8	16.3	16.5
P_2O_5	0.98	0.05	0.56
K_2O	1.52	0.03	0.12
CaO	2.64	1.15	4.26
TiO_2	3.11	5.27	3.62
Cr_2O_3	0.01	nd	0.03
MnO	0.29	0.07	0.18
Fe_2O_3	31.4	36.2	31.8
ZrO_2	0.67	0.62	0.27
LOI	8.39	8.40	13.8

LOI = Loss on ignition, nd = not detected.

Based on the data in Table 1, Na and Cr were selected as target elements for fixation/immobilization. To simulate their behavior in isolation, pure solutions containing equivalent ionic concentrations were prepared. Sodium hydroxide (LabSynth, P.A. – A.C.S) pellets were dissolved in deionized water to prepare a 0.1 mol/L Na solution, while a potassium chromate solution (SpecSol) was diluted to 1.0 mol/L in deionized water.

Six reactive materials, referred to as potential mitigators, were selected to test their ability to immobilize Na and Cr. Each mitigator was mixed separately with each solution using four different material-to-solution weight ratios: 1:40, 1:20, 1:8, and 1:4. Analogous to NBR 10006 [14], the mixtures were agitated for 5 minutes, rested for 7 days, and then filtered through a 0.45 μm membrane filter to obtain the solubilized extract, which was analyzed for Na and Cr concentrations. Blast furnace slag (BFS), steel slag (SS), two types of silica fume (SFB and SFD), magnesian hydrated lime (L) and bentonite (B) were the potential mitigators selected. Their chemical compositions, determined by XRF and expressed in terms of main oxides, are shown in Table 3.

The main physical characteristics of potential mitigators are in Table 4. Specific gravity was determined in a gas He pycnometer, Micromeritics – AccuPyc II 1340; specific surface area (SSA) was determined in a Belsorp-Max (Bel Japan) equipment according to the BET method, after sample treatment in a Belprep-vac II equipment at 40 °C and 10 Pa vacuum for 24 h; and particle size distributions was evaluated in a Helos (Sympatec) laser granulometry, with a detection range of 0.1–350 μm.

Mitigators that demonstrated effective fixation of Na and Cr in the simulated solutions were then tested using actual BR aqueous extracts. These extracts (originally described in Table 1) were mixed with the selected mitigators at a 1:8 material-to-solution weight ratio. Again, analogous to NBR 10006 [14], the mixtures were agitated for 5 minutes, allowed to rest for 7 days, and filtered. The resulting extracts were analyzed to verify the reduction of Na and Cr concentrations, confirming the ability of the mitigators to immobilize these ions in real BR leachates.

Table 3. Chemical characteristics of potential mitigators.

Element (%)	**BFS** Blast furnace slag	**SS** steel slag	**SFB** silica fume	**SFD** silica fume	**L** magnesian hydrated lime	**B** bentonite
Na_2O	0.13	0.07	0.61	0.15	nd	0.95
MgO	4.47	4.71	1.59	0.50	24.1	2.58
Al_2O_3	11.0	4.88	0.48	0.26	0.23	20.4
SiO_2	33.9	14.5	78.4	94.4	2.54	52.2
SO_3	3.50	0.58	0.23	0.12	0.02	0.03
K_2O	0.47	0.03	6.20	1.82	0.07	2.44
CaO	44.3	34.1	2.91	0.70	42.3	2.20
TiO_2	0.61	0.63	0.02	nd	nd	0.84
Cr_2O_3	nd	1.87	0.02	0.01	nd	0.03
MnO	0.75	4.95	1.35	0.06	0.02	0.06
Fe_2O_3	0.53	29.1	4.88	0.12	0.18	8.29
LOI	gof	3.13	1.94	1.52	30.5	9.57

LOI = Loss on ignition, nd = not detected, gof = gain on fire.

Table 4. Physical characteristics of potential mitigators.

Parameter	**BFS**	**SS**	**SFB**	**SFD**	**L**	**B**
Specific gravity (g/cm³)	2.99	3.56	2.69	2.33	2.59	2.39
SSA (m²/g)	0.96	3.30	16.23	16.5	8.78	78.9
d_{10} (µm)	2.31	1.63	1.59	3.85	1.70	1.40
d_{50} (µm)	14.5	12.5	8.56	23.5	25.75	6.20
d_{90} (µm)	47.5	62.8	19.5	49.2	71.94	35.3

3. Results and Discussion

3.1 Mixtures of Potential Mitigators and Pure Solutions

Figure 1a and Figure 3 present the immobilization performance of each potential mitigator for Cr and Na, respectively. The y-axis represents the percentage of immobilization, which indicates the extent to which each ion was retained by the material. A value of 100 % means the ion was completely removed from the solubilized extract, indicating full immobilization by the mitigator. The x-axis shows the material-to-solution ratio (by weight) used in each test.

Among the materials tested, hydrated lime showed the highest effectiveness in immobilizing Cr, achieving near-complete immobilization (> 70 %) even at low material-to-solution ratios (1:40 and 1:20) and complete immobilization at higher levels (1:8 and 1:4). This performance is likely due to the high pH environment created by lime (composed mostly of calcium and magnesium oxides), which reduces chromium leaching [15, 16]. Steel slag (SS) also exhibited good performance for Cr immobilization, particularly at higher dosages, with over 70 % immobilization at the ratios of 1:8 and 1:4. As seen in Figure1b, the mixtures with hydrated lime and steel slag reached the highest pH, being also the materials with higher immobilization effect. It is worth saying that the pH with BFS is also high, but the ion immobilization was not effective for the case analyzed.

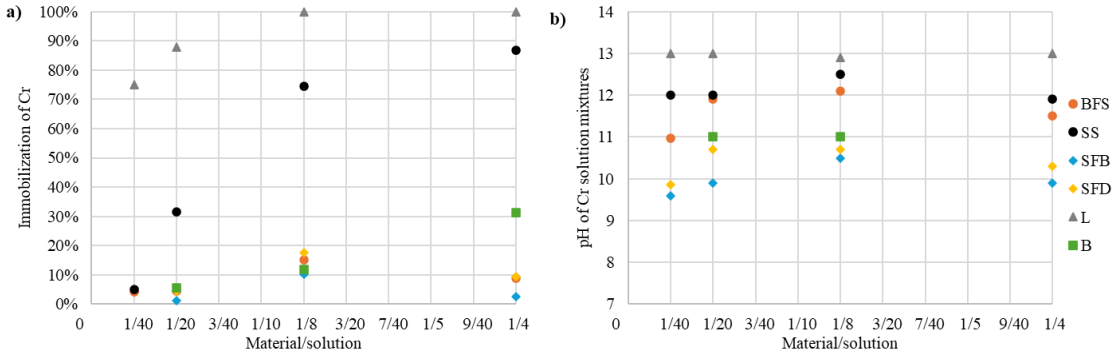

Figure 1. a): Immobilization capacity of potential mitigators for chromium, b): pH of Cr solution, at varying material-to-solution weight ratios.

However, for 1:20 ratio only 30 % of Cr was immobilized by SS and the immobilization was not effective for the lowest ratio. So, it is clear that pH is not the only factor responsible for Cr immobilization, since the level of immobilization does not follow the pH value of the mixtures. The other materials did not show effective immobilization of Cr at any ratio, except for 1:4 of bentonite, which fixed 30 % of Cr.

Another theory for the effectiveness of hydrated lime in the immobilization of Cr is the high amounts of calcium, present in the form of calcite (Ca) and portlandite (CH), as shown by its mineralogical composition represented in Figure 2Figure 2. A study about the immobilization of Cr(VI) by hydrated Portland cement pastes found that the peak intensities of portlandite in XRD patterns decrease with the increase of Cr(VI) amounts [15]. This could indicate a reaction between calcium hydroxide and Cr, by precipitation of Cr in less soluble forms, such as $Cr(OH)_3$ [17] and/or the formation of $CaCrO_4$ or $CaCrO_4.2H_2O$, which were identified in previous research as the reaction products responsible for Cr(VI) immobilization [18]. Further studies must be done to investigate and confirm the theory.

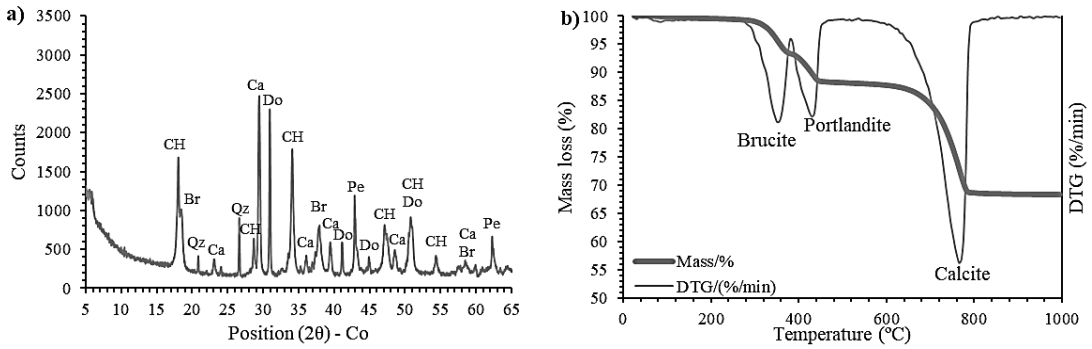

Figure 2. Mineralogical composition of hydrated lime.
a): obtained by X-ray diffraction (XDR), b): by thermogravimetric analysis (TGA).

Considering SS, previous studies have shown that certain Fe-treated materials can release Fe^{2+} into solution, which can act as a reducing agent for Cr(VI). This behavior has been demonstrated in Fe(II) modified zeolites and vermiculite, where Fe^{2+} drives the reduction of Cr(VI) [19]. If Fe^{2+} was released into solution from a metallic or reduced phase present in the steel slag, it could be able to reduce Cr(VI) to Cr(III), with the extent of reduction depending on the amount of Fe^{2+} available, following the stoichiometric ratio of $3Fe^{2+}$ to 1Cr(VI) ($3Fe^{2+} + Cr^{6+} \rightarrow 3Fe^{3+} + Cr^{3+}$). The removal mechanisms involve cation exchange between Fe^{2+} and K^+, followed by the reduction of Cr(VI) and subsequent precipitation of Cr(III) and Fe(III) [19]. These findings

support the hypothesis that similar redox interactions may contribute to Cr immobilization in the present work, although further analyses would be necessary to confirm the occurrence of this mechanism with steel slag.

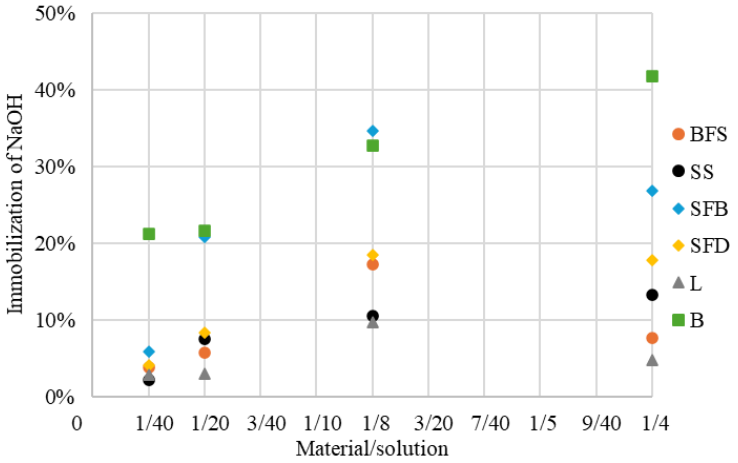

Figure 3. Immobilization capacity of potential mitigators for sodium, at varying material-to-solution weight ratios.

In contrast to Cr, sodium was more difficult to immobilize across all materials and ratios. None of the materials achieved complete immobilization of Na. Besides that, bentonite (B) consistently showed the highest immobilization capacity, reaching up to 30 % at the 1:8 ratio, > 40 % at the 1:4 ratio, and around 20 % of immobilization at lowest material-to-solution ratios (1:40 and 1:20). Silica fume B (SFB) also exhibited good results, with an immobilization capacity between 20 % and 35 % for ratios above 1:20. This may be attributed to high surface area of these materials (Table 4Table 4), which intensifies van der Waals and surface forces [20], and might lead to ion adsorption. In the case of bentonite, its ion exchange capacity may also explain sodium immobilization [21], for example, Na^+ replacing Ca^{2+}, Mg^{2+} and/or K^+, ions present in both bentonite and silica fume B. Some studies used sodium additivation in bentonites to expand their lamellae, enhancing their cation exchange properties [22, 23]. This makes it effective in adsorbing Na^+ ions, which are otherwise difficult to immobilize due to their high solubility and low reactivity.

Silica fume (D) and blast furnace slag demonstrated limited ability to reduce Na levels (~ 20 %) at 1:8 ratio. It was expected that BFS would be able to immobilize a greater amount of sodium, due to the ability of BR to activate it as a geopolymer [13, 24, 25]. However, the results show that this material has a limited capacity to reduce the sodium content in solution. Since the reactivity of the slag depends on its fineness [26], and the BFS used has a relatively small SSA and particles above 5 µm, it is believed that the immobilization capacity of this material could increase with grinding. However, this is a stage that is beyond the scope of the presented research, but it should be considered in the following steps.

Moreover, except for bentonite, it is noticed that after the material-to-solution ratio of 1:8, the immobilization capacity drops, probably due to saturation effect of sodium in solution.

3.2 Mixtures of mitigators and BR solutions

Following the preliminary screening with pure ionic solutions, the most effective mitigators identified for each ion were further tested using the actual aqueous extracts obtained from BRs. This step aimed to evaluate whether the selected materials could maintain their immobilization

performance under more complex conditions where multiple ions coexist and potential competitive interactions may occur.

Hydrated lime showed the best results for chromium immobilization, and bentonite for sodium. Therefore, these materials were selected to be mixed with aqueous extract from the three different BR samples, as illustrated in Figure 4. Figure 5a shows chromium immobilization efficiency, and Figure 5b presents the results for sodium.

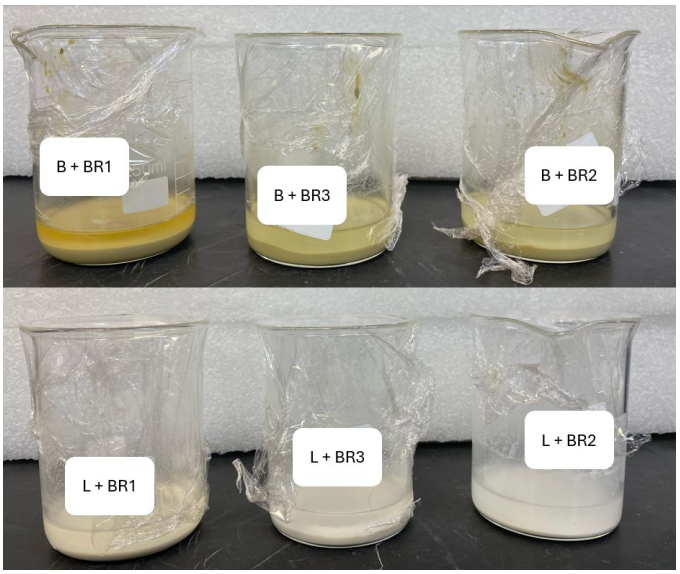

Figure 4. Mixtures of bentonite (above) and hydrated lime (below) in BR aqueous extracts after 7 days of rest.

In the case of BR1, the solution did not contain chromium (as shown in Table 1), therefore, all Cr values were below detection limits, and the immobilization efficiency appears as 100 %. However, this does not represent actual immobilization, rather than the absence of Cr, and this result should be interpreted with caution.

For the remaining samples, hydrated lime demonstrated consistent and effective Cr immobilization, achieving 100 % removal in BR2 and 78 % in BR3. This confirms lime's efficiency even under complex chemical conditions. The drop from 100 % to 78 % in BR3 suggests that the presence of competing ions may slightly interfere with the immobilization mechanism, mainly chemical precipitation of insoluble forms promoted by the pH increase.

Regarding sodium, bentonite, while less effective than in the pure solution, still demonstrated some ability to reduce Na concentrations. Its immobilization efficiencies ranged from 15 % to 26 % across the three BR extracts, with the best result seen in BR2.

Since Na is a highly soluble and non-reactive monovalent cation, its immobilization depends more on physical entrapment or cation exchange mechanisms than surface adsorption. Bentonite's moderate performance is likely attributed to its capacity of cation exchange, which allows partial substitution of interlayer cations by Na^+. The limited immobilization observed is consistent with the known difficulty of fixing monovalent alkali metals like sodium, which are not easily precipitated.

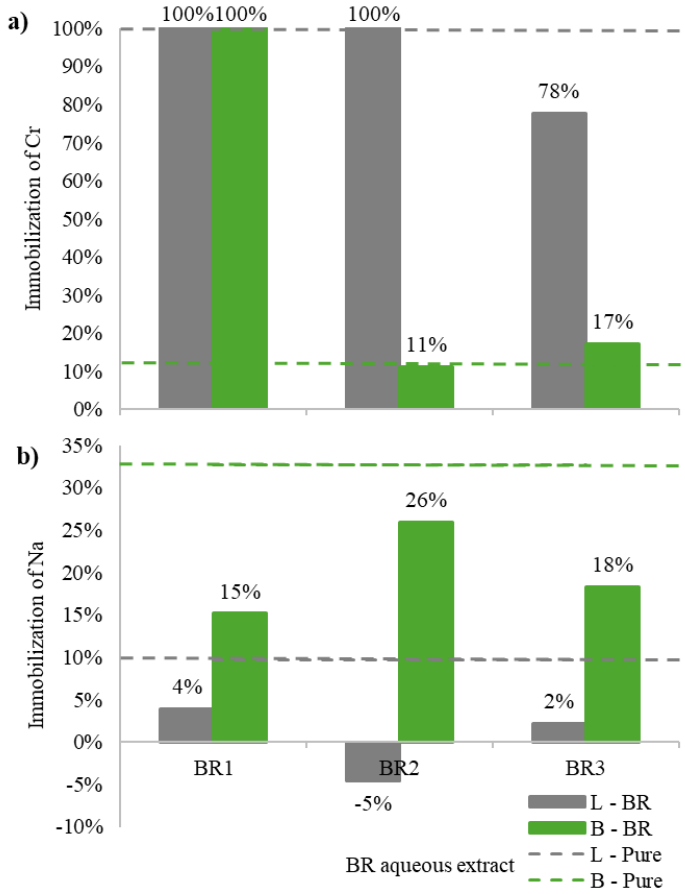

Figure 5. Immobilization efficiency of mitigators in different BR aqueous extracts compared to pure solutions: a) Cr and b) Na. Bars represent values in BR solutions; dashed lines represent performance in pure solutions.

These findings emphasize the need to optimize mitigation strategies for BR treatment, particularly when targeting multiple soluble ions with different chemical behaviors. Future studies will evaluate the performance of combined mitigators and explore different proportions between them and bauxite residue. This approach may enhance overall immobilization, even when a single material is not equally effective for both Cr and Na. Adjusting these parameters could allow a more balanced and efficient treatment, promoting safer reuse or disposal of bauxite residues.

4. Conclusions

This study evaluated the effectiveness of several materials in the immobilization of chromium and sodium from both simplified ionic solutions and real aqueous extracts derived from different BRs.

The results indicate that different mechanisms are responsible for the immobilization of each ion. Cr is more readily fixed through chemical precipitation under alkaline conditions, while Na, being a highly soluble alkali metal, requires physical adsorption or ion exchange processes, which tend to be less efficient in complex matrices.

Hydrated lime proved to be the most effective material for Cr immobilization, achieving 100 % removal in both the simulated solution and in a BR extracts. Its performance is attributed to the increased pH it induces, which favors the precipitation of Cr as insoluble hydroxides. Although

still high, a reduction in efficiency in BR3 suggests that competing ions or specific matrix characteristics can influence immobilization effectiveness.

Bentonite showed the most consistent results for Na immobilization, with moderate efficiency in both simulated and real solutions. Though the immobilization percentages in BR extracts were slightly lower than in the pure solution, it maintained a relatively stable behavior across different matrices, likely due to its ion exchange capacity.

The results indicate that while some mitigators perform well under controlled conditions, their effectiveness can be altered in real BR scenarios. Therefore, preliminary testing with real leachates is essential before implementing remediation strategies.

Further studies are needed to confirm the mechanisms that led each material to the immobilization of Cr and Na, particularly through analysis of the solid phases obtained after treatment. Additionally, although not required in the Brazilian standard, it would be relevant to quantify Cr in its different oxidation states, Cr(III) and Cr(VI), since only Cr(VI) poses significant risks to human health. This work focused on evaluating the chemical fixation capacity of deleterious elements in BR using accessible mitigators, showing the possibility of using BR safely without incorporating high amounts of energy. Studies will be continued to improve the understanding of physico-chemical fixation mechanisms, whether by chemical reaction, promoting crystal precipitation, or physical adsorption and ion exchange processes.

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