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Research

A Novel Rotational Limestone Treatment System for Effective Acid Mine Drainage Remediation

Un sistema novedoso de tratamiento rotacional de roca caliza para la remediación eficaz del drenaje ácido de minas

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Abstract

Context: The mining industry is the main culprit behind the generation of acid mine drainage (AMD). During coal extraction processes, sulfide minerals react with groundwater, releasing ions such as Fe^{2+} and Fe^{3+} , sulfates (SO_4^{-2}), and protonic acidity (H^+). The low pH of AMD can cause significant environmental damage. AMD remediation is usually achieved using alkaline systems, wherein the AMD passes through limestone to be neutralized. Nevertheless, this process requires prolonged treatment times and constant cleaning steps to remove the coating formed on the limestone, which reduces its effectiveness.

Method: This study evaluates a novel oxalic-limestone rotational system for the treatment of AMD produced by the coal industry. The AMD collected was characterized in terms of pH, dissolved oxygen, Fe (Fe total, Fe^{2+} , and Fe^{3+}), and SO_4^{-2} .

Results: The results demonstrate the optimal efficiency of the proposed system, reducing the treatment time from 120 h in conventional systems to 1.5 h when applying a ratio of 0.25k g of limestone per liter of AMD.

Conclusions: The rotational system enables the superficial degradation of the limestone, maintaining an active contact area for longer periods. This allows for optimized AMD remediation efficiency, reducing operating costs and necessitating fewer system cleanup steps.

Keywords: coal mining, acid mine drainage, rotational system, superficial degradation

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Resumen

Contexto: La industria minera es el principal responsable de la generación de drenaje ácido minero (DAM). Durante los procesos de extracción del carbón, los minerales sulfurados reaccionan con las aguas subterráneas, liberando iones como Fe^{2+} y Fe^{3+} , sulfatos (SO_4^{-2}), y acidez protónica (H^+). El bajo pH del DAM puede generar importantes daños medioambientales. La remediación del DAM generalmente se realiza a través de sistemas alcalinos, donde el DAM pasa a través de roca caliza para ser neutralizado. No obstante, este proceso requiere tiempos prolongados de tratamiento y etapas de limpieza constante para eliminar el recubrimiento formado en la roca, lo que reduce su eficacia.

Método: Este estudio evalúa un novedoso sistema rotacional de óxido-caliza para el tratamiento de DAM producido por la industria del carbón. El DAM recolectado se caracterizó en términos de pH, oxígeno disuelto, Fe (Fe total, Fe^{2+} y Fe^{3+}), y SO_4^{-2} .

Resultados: Los resultados demuestran la eficiencia óptima del sistema propuesto, que reduce el tiempo de tratamiento de 120 h con sistemas convencionales a 1.5 h al aplicar una relación de 0.25 kg de caliza por litro de DAM.

Conclusiones: El sistema rotacional facilita la degradación superficial de la caliza, manteniendo un área de contacto activa durante periodos más largos. Esto permite optimizar la eficiencia de remediación del DAM, reduciendo los costes de operación y las etapas de limpieza requeridas por el sistema.

Palabras clave: minería del carbón, drenaje ácido minero, sistema rotacional, degradación superficial

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Highlights

- A novel oxidic limestone rotational drainage system for the treatment of AMD is proposed in this study.
- The rotational system promotes a superficial degradation of the limestone, increasing its activity for longer periods.

- The novel system improves the efficiency process, cutting AMD treatment time by 98.7% over conventional static systems.

1. Introduction

The acid mine drainage (AMD) produced during mining activities is a polluting byproduct formed by the reaction between minerals, oxygen, and water (1). In the case of coal mining processes, AMD is characterized by a low pH and a high concentration of ions in the form of SO_4^{2-} , Fe^{2+} , and Fe^{3+} , produced by the oxidation of pyrite (FeS_2), in addition to other contaminating metal ions in lower proportions, such as Mn^{2+} and Al^{3+} , which are typical of AMD waste (2–5). The concentrations of these ions vary according to the geology of the area where the mining activity takes place (6).

In Colombia, the coal industry is one of the main mining activities, carried out mainly in the Boyacá region. This region contributes 38% of the national coal production, which represents 3.087 million metric tons destined for use in the thermal and metallurgical sectors (7, 8). Coal mining operations in the area are not efficient, producing a high amount of pollutants that threaten nearby tributaries (9, 10). This problem is exacerbated by stormwater runoff, which can carry large amounts of AMD along with particulates and dissolved materials, substantially reducing the oxygen and nutrient content of the soil, as well as degrading aquatic systems and dependent biota (11, 12).

The application of systems such as aerobic wetlands and limestone drainage are potential alternatives that allow treating AMD (13, 14). The application of an active system (oxic) enables a higher rate of neutralization and removal of metal contaminants (15, 16). The high concentrations of SO_4^{2-} depend on the mineralogy of the place (17, 18), which makes water treatment difficult. Furthermore, the efficiency of open limestone channel (OLC) systems is compromised by the formation of gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) on the limestone surfaces, acting as a barrier that reduces its effectiveness during AMD treatment (19).

Similarly, optimal Fe removal values can be obtained using these systems (3). Nevertheless, these systems require continuous maintenance stages for their proper functioning, in addition to prolonged periods of fluid retention (20).

Considering the above, this study aims to evaluate the application of a novel rotational oxic limestone system to optimize AMD treatment processes. As a control mechanism, a non-moving device is used, called a *static system*. Tests were carried out in a controlled manner, using AMD samples collected from the area of interest. The authors did not find literature related to the evaluation of the effect of alkalization and the treatment of AMD using similar devices, which reinforces the novelty of this research.

2. Experimentation

The AMD used in this study was supplied by Cooperativa Agro-Minera (Coagromin Ltda.), located at Km 5 of the Paipa-Tunja road in Boyacá, Colombia. The AMD was later processed by adding limestone with a particle size between 1.27 and 2.54 cm. The limestone used was collected from Cantera Metrópolis, located at Km 6.6 of the Moniquirá-Arcabuco road (Boyacá). The limestone used in this study was analyzed via X-ray diffraction (XRD) in a Pananalytical diffractometer (Co, $\lambda = 1.75 \text{ \AA}$). The analysis of the XRD pattern was carried out using the HighScore-Plus software, the Crystallography Open Database (COD), and the Inorganic Crystal Structure Database (ICSD).

3. Static tests

Static tests were performed on the system presented in Fig. 1. The proposed system was isolated during the analysis period in order to avoid agitation, favoring a greater sedimentation and settling of the compounds that may be generated. The tests were conducted under controlled conditions, *i.e.*, at 2800 meters above sea level (m.a.s.l.), with a pressure of 740 hPa and an average temperature of 17 °C (21). The limestone:AMD ratio was 2.5:1 (in weight), ensuring the total coverage and reaction of the AMD. In this study, we evaluated the concentration of Fe ions (*i.e.*, Fe^{2+} and Fe^{3+}) and sulfates (SO_4^{2-}), considering that these are the main dissolved ions in AMD from coal mining processes due to the presence of pyrite (22). Therefore, other trace metals in AMD samples were not characterized. The tests were carried out until an optimal removal of Fe (total, Fe^{2+} , and Fe^{3+}) and SO_4^{2-} was achieved. Fe and sulfate measurements were taken using the FerroVer (iron reagent, Hach, USA) and SulfaVer IV (sulfate reagent, Hach, USA) reagents along with a DR6000 spectrophotometer (Hach, USA). The dissolved oxygen (DO) content was obtained from a Hach-Flexi HQ30d oximeter, and pH measurements were carried out using a SCOTT HandyLab pH11. Alkalinity was determined using 25 mL of undiluted AMD samples mixed with phenolphthalein and bromocresol green. The AMD sample was titrated with 0.02 N H_2SO_4 (*i.e.*, $10 \text{ mol}\cdot\text{m}^{-3}$). Likewise, acidity was evaluated by adding phenolphthalein as an indicator solution. The mixture was also titrated with 0.02 N NaOH (*i.e.*, $20 \text{ mol}\cdot\text{m}^{-3}$) (23).

3.1. Dynamic tests using a rotational limestone system

Dynamic tests were performed in a two-reactor rotational mechanical system. The vessel had an internal length of 30 cm and a diameter of 12 cm, containing a maximum volume of 3393 cm^3 . The system was not operated at maximum capacity to preserve the functionality of the pressure gauges used to measure carbon dioxide (CO_2). We employed a vessel inclination of 6° and a rotation speed of 11 rpm. Although an angle of 10° is typically used, as has been reported by (3, 24, 25), we opted for a smaller inclination to ensure a greater retention of the liquid within the system. Fig. 1 shows a scheme of the devices used in this study. 0.25 kg of limestone were used for every 1 L of AMD (Limestone:AMD ratio of 0.25:1). The mixing process was carried out for 0.5, 1, and 1.5 h. pH, DO, alkalinity, acidity, Fe^{2+} , Fe^{3+} , and SO_4^{2-} measurements were made using the above-presented procedure.

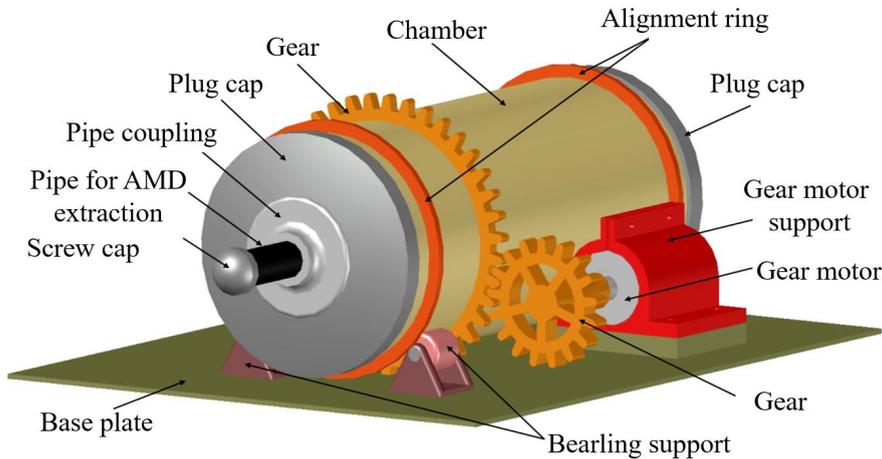


Figure 1. Schematic illustration of the AMD treatment device used in this study

4. Results

Fig. 2 shows the XRD pattern of the limestone used in this study. A semi-quantitative analysis, conducted using the Rietveld refinement method, revealed the presence of calcite— CaCO_3 , space group $P_{121/c1}$ (14), ICSD 150 (26)—, which was the main mineralogical compound (73.2%). Likewise, we observed the presence of dolomite—9.3%, $\text{CaMg}(\text{CO}_3)_2$, $R\bar{3}H$ (148), ICSD 10404 (27)—, magnesite—0.5%, MgCO_3 , $R\bar{3}cH$ (167), ICSD 10264 (28)—, and silicates—17%, mainly SiO_2 , P_{3221} (154), ICSD 16331 (29)—in a lower proportion. The composition of the limestone was similar to that found in other regions of Colombia, as reported by (30). A high content of carbonates in the limestone favors an optimal degree of alkalinity, as well as the limestone's ability to react with AMD (31). The presence of amorphous material can be observed in the first degrees of diffraction, which corresponds to organic material typical of sedimentary rocks.

Figs. 3 and 4 illustrate the behavior of the AMD when treated with a static and a rotational system, respectively. We followed the Colombian resolution no. 0631/2015 (32) to determine the efficacy of both systems. An initial characterization of the AMD is presented in Table I. Note that the values are outside the permissible values as per the aforementioned resolution, which was issued by the Colombian Ministry of the Environment and Sustainable Development.

Table I. Chemical characterization of the AMD sample used in this study and maximum permissible limit values for water resources linked to the extraction of coal and lignite according to Resolution 0631 of 2015 (32)

Parameter	Unit	This study	0631/2015 Legislation
pH	a.u.	2.5	6.0-9.0
DO	$\text{mg}\cdot\text{L}^{-1}$	7.09	-

Fe _{Total}	mg·L ⁻¹	228.75	2
Sulphates	mg·L ⁻¹	3300	1200
Acidity	mg SO ₄ ·L ⁻¹	1180	analysis and report
Alkalinity	mg CaCO ₃ ·L ⁻¹	0	analysis and report

5. Discussion

Limestone is an inexpensive, natural, and efficient raw material for the treatment of AMD from coal mining processes. Dolomitic limestone, *i.e.*, CaMg(CO₃)₂, is widely used within AMD systems due to its ease of reaction (33). Limestone reacts in the presence of hydrogen, enabling the release of carbonic acid, which can subsequently be converted into bicarbonate ions (3). The process is governed by the chemical reactions shown in Eqs. (1) and (2) (14,34). This dissociation generates a buffer effect within the system that maintains stable pH values during the formation of metal precipitates. The presence of bicarbonate ions and limestone also favors the removal of Fe, according to Eqs. (3) and (4) (35,36).



A lower rate of acidity reduction in AMD after rotational treatment for 0.5 h may be due to the release of acidity (H⁺) during the formation of siderite (FeCO₃). Nevertheless, part of the H⁺ produced can also react with the limestone, which generates CO₂, favoring carbonic acid formation (20,36).



Although the generation of acidity by hydrogen should lower the pH of the solution, it can also be slowed down when carbonic acids come into contact with the limestone, as shown in Eq. (1), as well as by the formation of CO₂ when reacting with part of the carbonic acids formed, *i.e.*,



Likewise, the pH and alkalinity of the AMD can be leveled by the formation of passive layers of gypsum and siderite, which can be deposited on the surface of the limestone rock (Fig. 5). These precipitates act as a barrier that reduces the reactivity between the AMD and the rock (35,37). Calcium

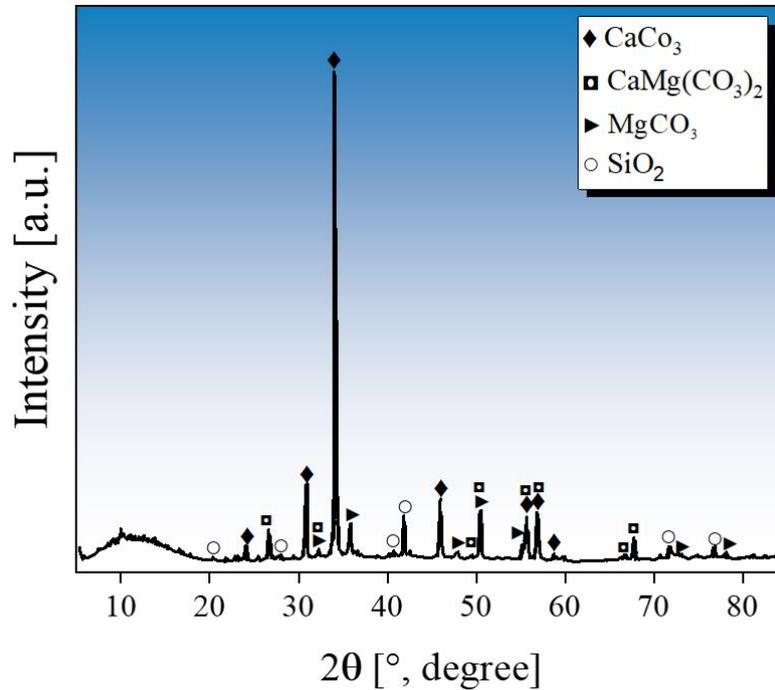
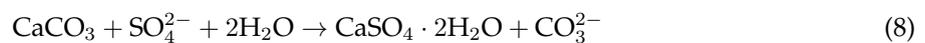


Figure 2. SXRD pattern of the limestone used in this study and collected in the region of Boyacá, Colombia

sulfate (CaSO_4) hydrates with two water molecules, forming gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$). Based on a stoichiometric analysis, for every 136.14 g of anhydrite, 36.03 g of water are required to form 172.17 g of gypsum. Therefore, no significant reduction in the volume of water in the system is expected due to this reaction. The volume of gypsum formed may be regarded as negligible, since these are very thin layers deposited on the limestone, formed to mitigate the rock's activity.



The formation of these passive layers allows explaining the low efficiency of the AMD treatment when occurring in a static system, which is unable to remove the precipitates deposited on the limestone surface (35,38). Although the limestone:AMD ratio is ten times lower in the rotational system, the latter favors an autogenous grinding effect of the limestone (39), degrading the particles and removing the gypsum and siderite coatings.

The delay in Fe^{2+} removal may be due to the low reactivity of this ion to generate compounds other than siderite within the AMD, so it is highly dependent on their reactivity with limestone. A DO concentration greater than $0.5 \text{ mg} \cdot \text{L}^{-1}$ favors the reaction of Fe^{2+} ions with AMD (35,37,40). The difficulties in creating other compounds, *e.g.*, $\text{Fe}(\text{OH})_2$, which is greenish, is impaired by the formation of limonite, $\text{Fe}(\text{OH})_3$, from Fe^{3+} , which has a 5/8 10YR (Munsell Soil Color Chart (41)) color, as seen

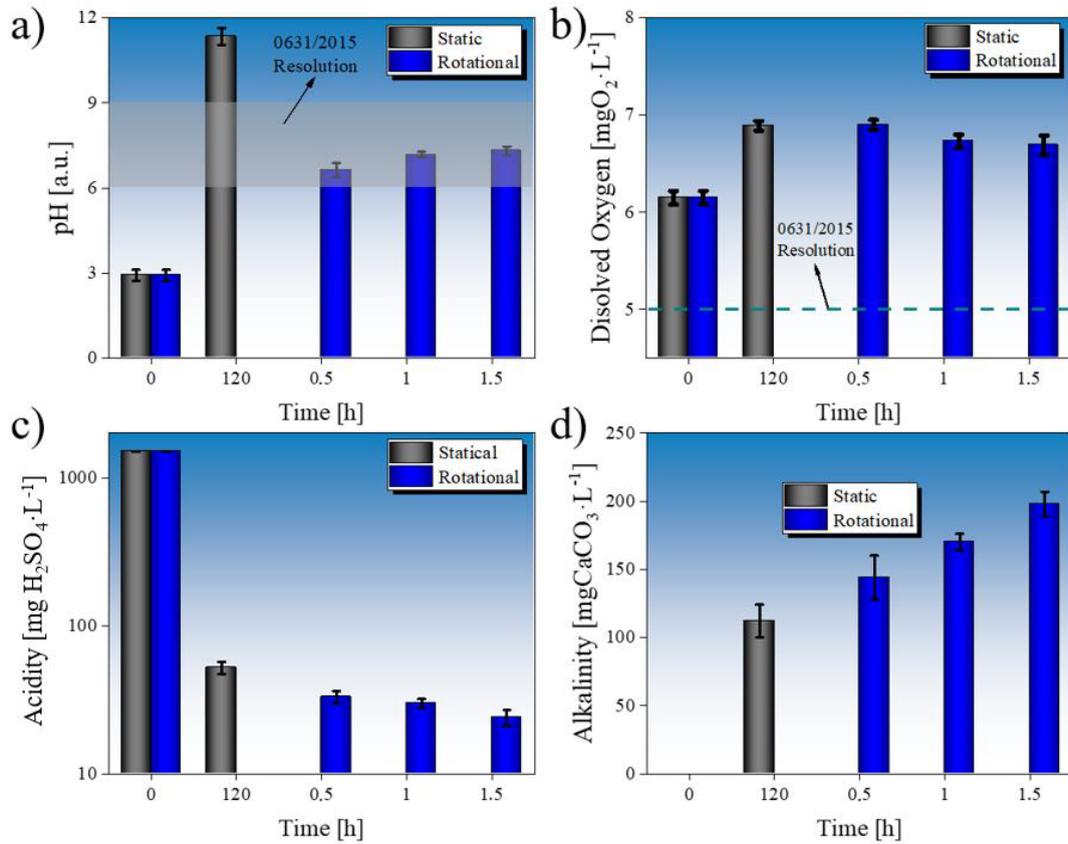


Figure 3. Values of a) pH, b) DO, c) acidity, and d) alkalinity for untreated and treated AMD in static and rotational systems. The pH and DO values obtained in this study were compared against those in resolution no. 0631/2015 for water quality (32)

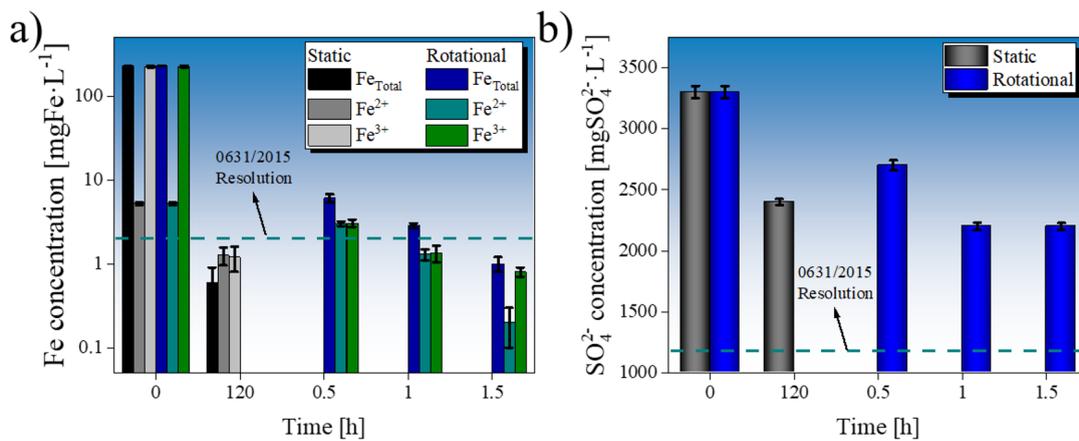


Figure 4. Concentration of a) Fe and b) sulfate ions in untreated and treated AMD after static and rotational treatment. The permissible values for pH and DO obtained in this study were compared against those in resolution no. 0631/2015 for water quality (32)

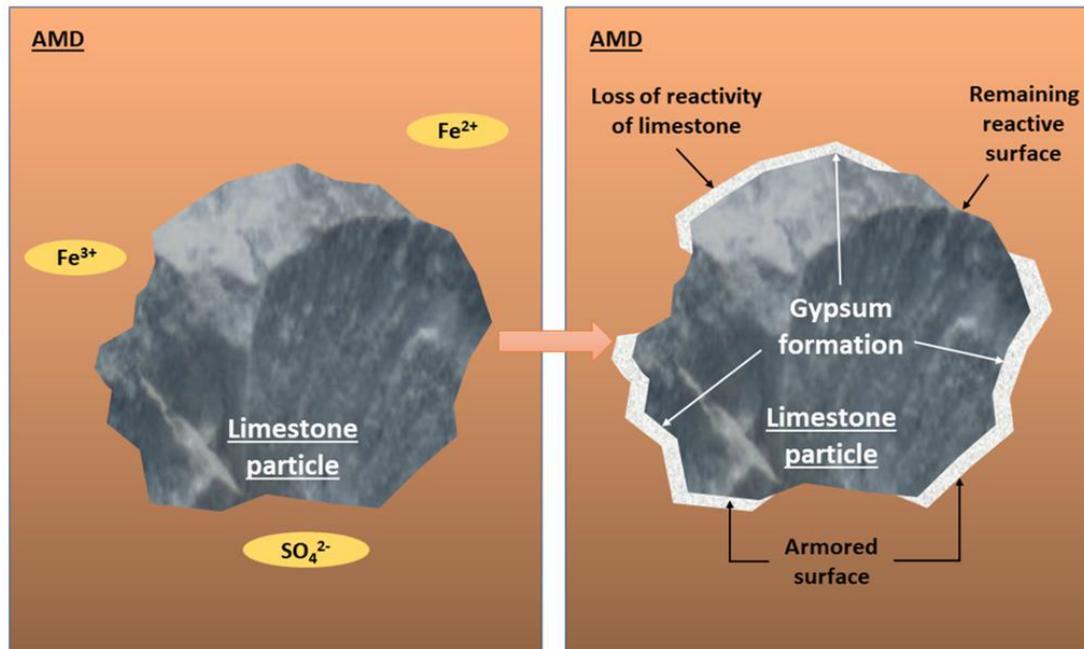


Figure 5. Schematic illustration of gypsum binder formation from the reaction between AMD and limestone

in Fig. 6. Likewise, the formation of $\text{Fe}(\text{OH})_3$ is favored by pH values between 6.5 and 8 (42–45). Our measurement of the potential in the static and dynamic systems showed values between 0.1 and 0.15 V. As shown in Fig. 7, the final conditions obtained in this study favor the formation of the $\text{Fe}(\text{OH})_3$ mineralogical phase.

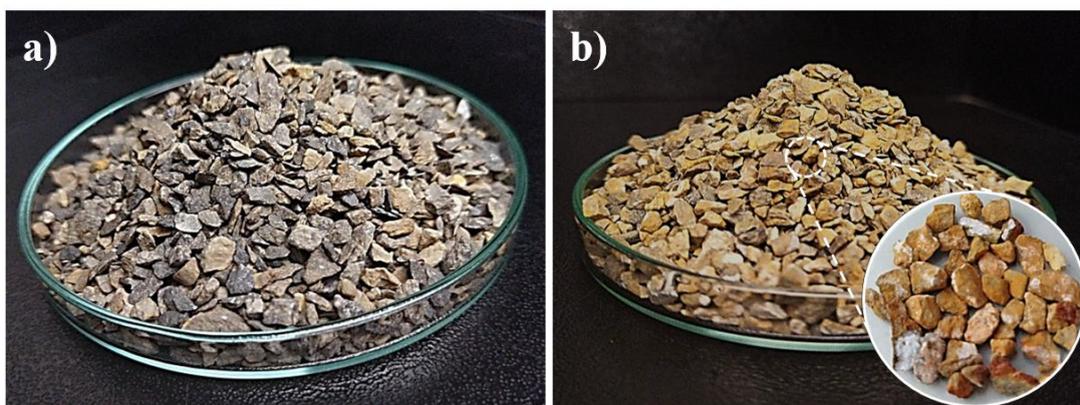


Figure 6. Photographs of the limestone samples used in this study a) before and b) after the AMD treatment. A change in the color of the limestone can be observed, which is due to the formation of precipitates on the rock surface

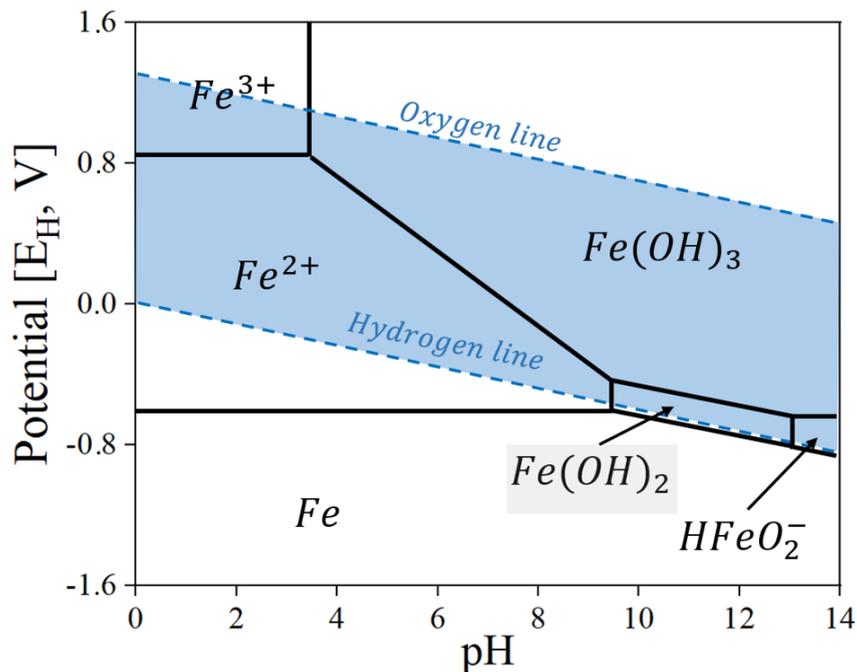


Figure 7. Generalized Pourbaix diagram for the Fe-H₂O system. The pH values obtained in this work show a greater tendency towards the formation of the Fe(OH)₃ phase

It has been reported that the Fe³⁺/Fe²⁺ ratio present in leachate from coal minerals can favor biodesulfurization through the reaction of sulfate ions, allowing for the formation of pyrite and, subsequently, the iron (II) (FeSO₄) and iron (III) (Fe₂(SO₄)₃) sulfate phases (20,46). A reduction of Fe³⁺ ions, in addition to the potential formation of gypsum precipitates adhered to the limestone surface, explains the blockage during sulfate removal from the AMD. Stagnation in the process of removing sulfate ions could be observed. This is because the increase in pH caused by the interaction between the limestone and the AMD inhibits the reactive capacity of the limestone particles with the sulfate and iron (II) ions, so their concentrations are generally not significantly affected (47). Although removal levels of 28 and 33% were obtained for the samples treated in the static and rotational systems, respectively, it is necessary to apply secondary stages in order to comply with Colombian legislation. Even so, the application of a limestone system still exhibits an economic advantage compared to other technologies for the treatment of AMD and the removal of sulfates (35).

A system's operation must be evaluated based on its working time, as well as on the cost of the raw materials. The application of our rotational system in this affected region is facilitated by the ease of obtaining limestone. The authors express their motivation for the further development of this project, which can help to address the effects generated by mining activities in the region. The application of raw materials from the affected area and the construction of passive treatment systems allows for significant cost reductions.

While these systems can be used in other regions and for different types of AMD, it is necessary to consider that the effectiveness of the treatment depends on the characteristics and initial conditions of the raw materials, *i.e.*, the AMD and the limestone. In this study, the feasibility of this system was supported by the nearby availability of limestone. Therefore, when considering implementation in other affected areas, the costs of transporting limestone—as well as sustainability and logistical factors related to mining and transportation—must be considered.

During the AMD treatment, some residual materials were produced in the form of precipitates and clays. The chemical and toxic complexity of the waste generated currently precludes direct use in industrial activities due to the high risk of contamination associated with its release into the environment (48). The authors hope to conduct a feasibility analysis of the waste generated in subsequent studies, hoping to increase the sustainability of the proposal through new industrial products made from said waste.

6. Conclusion

This study evaluated the applicability of oxic-limestone drainage for the treatment of AMD and its effectiveness in the removal of Fe^{2+} , Fe^{3+} and SO_4^{2-} using a novel rotational system.

The application of a static system, which was used for comparison, required longer treatment times and yielded a lower ion removal efficiency compared to the rotational system. This was mainly due to the formation of precipitates that served as a barrier between the limestone and the AMD, hindering the system's ability to react due to a reduction in the active area of the limestone. This barrier cannot be directly removed within the static system. In contrast, the dynamic behavior of our proposed solution generates an autogenous grinding process that favors the removal, by wear, of the precipitates deposited in the limestone, thus maintaining a continuous active contact area between the limestone and the AMD. The inability to remove sulfate ions was due to the premature depletion of Fe^{2+} ions and the formation of precipitates adhering to the limestone surface, which slowed the chemical reactions generated during the treatment of the AMD.

The authors would like to express their interest in the application of this type of system for the treatment of AMD and other leachates generated during different industrial activities in the region, as well as in the application of new raw materials from the area that allow for optimized industrial water treatment.

7. Acknowledgments

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8. Availability of data and materials

Data presented in this study are available from the corresponding author upon reasonable request. Data is not publicly available because they pertain to ongoing research.

9. Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could appear to have influenced the study reported in this paper.

10. Author contributions

C.R. Blanco-Zúñiga: conceptualization, methodology, validation, obtaining funds, writing (review and editing).

L. Ulloa-Amador: methodology, research, formal analysis, writing (review and editing).

N. Rojas-Arias: supervision, conceptualization, methodology, validation, writing (original draft, review, and editing).

11. Use of artificial intelligence

During this work, the authors did not employ any technologies associated with artificial intelligence.

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