RESEARCH ARTICLE

A COMPARATIVE STUDY OF CHEMICAL PRECIPITATION AND ELECTROCOAGULATION FOR CALCIUM AND MAGNESIUM REMOVAL FROM LANDFILL LEACHATE

Brahima Seyhi[1]*, Franck Maxime Gnamba[1], Georges Kouame Kouadio[2], Kassoum Sangare[3]

[1]Department of Geosciences, University Peleforo Gon Coulibaly, Korhogo, Côte d'Ivoire
[2]Department of Science and Technology, École Normale Supérieure, Abidjan, Côte d'Ivoire
[3]Department of Sciences and Agro-Industrial Technologies, University of San-Pédro, San-Pédro, Côte d'Ivoire

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ABSTRACT

This study evaluates and compares three physicochemical pretreatment processes: sodium hydroxide precipitation, , lime precipitation, and electrocoagulation (EC) for calcium and magnesium removal from landfill leachate to mitigate scaling and improve compatibility with downstream biological treatment. FTIR analysis of scale deposits collected from heat exchanger plates confirmed calcite ($CaCO_3$) as the dominant scaling compound, highlighting the need for calcium removal before biological treatment. Batch experiments conducted on real landfill leachate collected during the winter season showed that sodium hydroxide precipitation achieved the highest removal efficiencies, with simultaneous calcium and magnesium removals exceeded 96% under optimal conditions of pH 12, stirring speed of 50 rpm and reaction time of 1 min. This indicated extremely fast equilibrium-controlled kinetics. Lime precipitation provided moderate calcium removal (73.0%) and good magnesium removal (87.2%). However, it was limited by high sludge production and dosage sensitivity. Electrocoagulation achieved high calcium removal (92.7%) and moderate magnesium removal (64.1%) under optimal conditions of current density of 4.5 mA/cm^2 and treatment time of 30 min. EC avoided chemical addition and extreme pH adjustment. Although sodium hydroxide precipitation showed higher simultaneous removal performances, EC emerged as a promising alternative, offering reduced chemical consumption and greater operational flexibility. The results of the present study demonstrated the relevance of physicochemical pretreatment to enhance the reliability and sustainability of landfill leachate treatment systems.

INTRODUCTION

Management of municipal solid waste involves several treatment and disposal options, including recycling, reusing, incineration, energy recovery, composting, and landfilling. Among these methods, landfilling is widely used due to its technical and economic advantages (Drouin, 1995; Karimipourfard *et al*., 2020; Yatoo *et al*., 2024). However, a major environmental challenge associated with landfilling is the generation of landfill leachate, a complex and highly variable effluent that is difficult to treat. Landfill leachate contains a wide range of pollutants, including organic and inorganic compounds, ammonia nitrogen, phosphorus, heavy metals, mineral salts, micropollutants, and pathogenic organisms (Kjeldsen *et al*., 2002; Mojiri *et al*., 2016; Vaccari *et al*., 2019; Abdel-Shafy *et al*., 2023). Among the inorganic constituents, calcium and magnesium ions are often present at high concentrations, depending on the age of the leachate. This results in significant leachate hardness (Dang *et al*., 2014; Yang *et al*., 2022) and promotes scaling, which is directly responsible for fouling, clogging, and loss of process efficiency in landfill leachate treatment systems (Rowe *et al*., 1995; Rittmann *et al*., 2003; Fleming and Rowe, 2004; VanGulck and Rowe, 2004).

Numerous studies have demonstrated the critical influence of calcium concentration on the stability and efficiency of biological treatment processes (Yu *et al*., 2001; Liu *et al*., 2011; Dang *et al*., 2014; Yepez-Ceron *et al*., 2025).During the winter season, scaling of the heat exchangers used to preheat leachate before biological treatment poses a significant operational problem. Indeed, scaling of the heating equipment affects the maintenance of temperatures required for biological treatment (Renou *et al*., 2008). This significantly impacts treatment performances and compliance with discharge standards. Furthermore, cleaning the plates of the heat exchangers is a complex, costly, and time-consuming operation requiring considerable personnel. Scaling of the equipment is a limiting factor in the reliability, durability, and long-term performance of landfill leachate treatment plants. In this context, the implementation of decarbonation processes is essential. Several processes are commonly used, such as chemical precipitation (precipitation with lime or soda), adsorption, ion exchange, chemical coagulation, electrocoagulation (EC), CO_2 stripping, membrane processes and scale inhibitors. Chemical precipitation is widely used in industrial applications (Musvoto *et al*., 2000; Pakarinen and Paatero, 2011, Wang *et al*., 2019). They offer promising performances, but come with drawbacks, such as an

increase in the pH of the treated waters, significant consumption of chemical reagents (lime or caustic soda), and sludge production. In addition, subsequent pH neutralization and sludge management are necessary. They increase treatment costs. EC is an attractive alternative to conventional chemical coagulation. EC uses electrodes (generally made of aluminum or iron) and generates coagulants in situ by applying an electric current (Mollah *et al.*, 2001; Chen, 2004). Therefore, EC does not require the use of chemicals, as in chemical coagulation. Despite its advantages, the application of EC to landfill leachate treatment remains limited. The objective of the present study was to evaluate and compare the performance of three physicochemical methods for decarbonation of landfill leachate: chemical precipitation with sodium hydroxide, chemical precipitation with lime and electrocoagulation.

MATERIAL AND METHODS

Sanitary Landfill Site: The landfill leachate investigated was collected in a sanitary landfill site located in the province of Quebec (Canada). This facility receives approximately 40% of the municipal solid waste (MSW) generated in the Greater Montreal area, corresponding to nearly 1.2 million tonnes of waste per year. Landfill leachate was collected through a system of collectors and drains located on the walls and bottom of the cells. The collected leachate is conveyed to equalization and storage basins that allow temporary storage and ensure a regulated hydraulic feed to downstream treatment units. The pretreatment of the landfill leachate was carried out in the aerated ponds, and then the pretreated leachate was sent to moving bed bioreactors (MBRs). During the winter period, the leachate is preheated using plate heat exchangers to maintain favourable thermal conditions for optimal biological process performance within the MBRs.

Sampling: Landfill leachates were collected at the outlet of the aerated ponds, before the heat exchangers. During each sampling campaign, approximately 40 L of leachate were collected in clean, hermetically sealed polyethylene containers. The samples were stored at 4 °C. Three independent sampling campaigns were conducted during the study period. Physicochemical analyses were carried out as soon as possible after sampling to limit potential variations due to storage. The main characteristics of the raw leachate are summarized in Table 1. In addition, scale deposits formed on the heat exchanger plates were collected for characterization. Samples were collected in winter.

Table 1. Physicochemical characteristics of the landfill leachate

Parameter	Concentration
COD(mg/L)	2462± 501
BOD$_5$ (mg/L)	716± 339
Ca^{2+} (mg/L)	280.7±63.6
Mg^{2+} (mg/L)	258.8±29.4
pH	7.9 – 8.4
Conductivity (ms/cm)	14.6 – 15.7

Experimental Procedures: All experiments were conducted in batch mode at room temperature (20 ± 2 °C), using 500 mL of raw leachate per test. After each treatment, the suspensions were allowed to settle for 30 min to promote solid–liquid separation, and the supernatant was carefully collected for subsequent analyses.

Chemical precipitation with Sodium Hydroxide: Chemical precipitation with sodium hydroxide was carried out by adjusting the leachate pH using a 50% (w/w) NaOH solution. The tests were carried out in beakers with 500 mL of samples, mixed using a magnetic stirrer (Thermo Scientific, Super-Nuova Multi-Place). Three series of tests were performed to evaluate the effect of certain operating parameters on decarbonation performance:

• Effect of initial pH: The tests consisted of varying the initial pH of the leachate between 8 and 12, with stirring at 300 rpm for 5 min.

• Effect of stirring speed: The tests consisted of varying the stirring speed between 50 and 800 rpm at pH 12 for 5 min.
• Effect of reaction time: The tests consisted of varying the reaction time between 1 and 15 min at a fixed pH of 12 with stirring at 50 rpm.

Chemical Precipitation with Lime: Chemical precipitation using lime was carried out in a jar-test apparatus (Phipps & Bird, Jar Tester PB-700). Lime was added in the form of milk at 100 g/L. Three series of tests were conducted to evaluate the effect of certain operating parameters on decarbonation performance:

• Effect of lime dose: The tests consisted of varying the lime dose from 0 to 15 g/L with stirring at 300 rpm for 5 min.
• Effect of stirring speed: The tests consisted of varying the stirring speed between 50 and 300 rpm while maintaining a fixed lime dose of 7 g/L and a reaction time of 5 min.
• Effect of reaction time: The tests consisted of varying the reaction time between 1 and 15 minutes while maintaining a fixed lime dose of 7 g/L and a stirring speed of 50 rpm.

Electrocoagulation: Electrocoagulation (EC) experiments were performed in a Plexiglas reactor (10 × 10 × 10 cm) equipped with two vertically oriented aluminum electrodes (11 × 5 cm) with an inter-electrode distance of 1 cm. A DC power supply (BK Precision 1621A) was used to apply a constant current. Two series of tests were conducted to evaluate the effect of certain operating parameters on decarbonation performance:

• Effect of current density: the tests consisted of varying the current density from 2.3 to 11.4 mA/cm² for 30 min.
• Effect of treatment time: the tests consisted of varying the treatment time between 0 and 60 min while maintaining the current density at 4.5 mA/cm².

Analytical Methods: pH, temperature, and electrical conductivity (EC) were measured immediately after sample collection. pH was measured using a Fisher Scientific Accumet pH meter (model XL500) equipped with a Cole-Parmer double-junction electrode (Ag/AgCl reference). The temperature was measured with a calibrated thermometer. Conductivity was measured using a conductivity meter (Hach HQ1140) equipped with a four-pole graphite probe and an integrated temperature sensor. Chemical oxygen demand (COD) was analyzed by the closed reflux colorimetric method (HACH Method 8000; Standard Methods 5220 D) and UV–visible spectrophotometer (Evolution 201, Thermo Scientific). Absorbance readings were recorded using a DR1900 spectrophotometer (HACH, Canada). The five-day biochemical oxygen demand (BOD$_5$) was analyzed using the Standard Methods for the Examination of Water and Wastewater (5-day BOD method). Calcium ions (Ca^{2+}) and magnesium ions (Mg^{2+}) concentrations were analyzed by atomic absorption spectrometry (Shimadzu AA-7000). Scaling deposits collected from the plates of the heat exchangers were characterized using Fourier Transform Infrared (FTIR) spectroscopy. Before analysis, the samples were reduced to powder.

RESULTS

FTIR characterization of scaling deposits on heat exchanger plates: The results of the FTIR analysis of the scaling deposit collected from the plates of the heat exchangers are shown in Figure 1. The scaling deposit wasmainly composed of calcite (CaCO$_3$). Indeed, strong absorption bands observed at 1400 – 1450 cm^{-1}, 875 cm^{-1} and 710 cm^{-1}, correspond to carbonate groups.

Chemical precipitation with sodium hydroxide (NaOH): The influence of initial pH on calcium and magnesium removal by sodium hydroxide precipitation is presented in Figure 2. The increase of pH reduced the residual concentrations of both calcium and magnesium.

However, residual magnesium concentration was first stabilized (244.2 – 244.7 mg/l), as pH was increased from 8 to 9. At pH = 12, residual concentration reached 1.6 mg/L for calcium (99.2% of removal) and 1.3 mg/L for magnesium (99.5%).
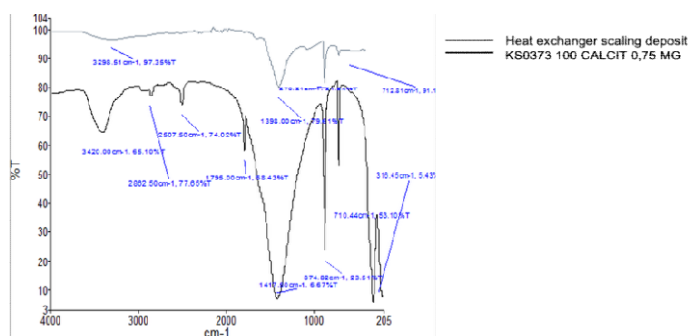

Figure 1. FTIR spectrum of scaling deposits collected from the plates of the heat exchangers

The effect of stirring speed on calcium and magnesium removal is shown in Figure 3. The concentration of calcium decreased from 280 mg/l to 0.6 mg/L at 50 rpm and then remained constant, regardless of the applied stirring speed (removal performance exceeded 99%). Also, residual concentration of magnesium decreased from 245.7 mg/L to 4.5 mg/L and did not significantly change (0.6 - 4.5 mg/L), regardless of the applied stirring speed. Removal performance of magnesium exceeded 98% for all stirring speeds applied. The effect of reaction time on calcium and magnesium removal is shown in Figure 4. Calcium and magnesium concentrations were instantaneously and significantly reduced when NaOH was added. Calcium concentration decreased from 280 mg/l to 0.4 mg/L at 1 min of reaction time, and remained in this range, regardless of the increasing of reaction time up to 15 min. The removal performance of calcium exceeded 99% regardless of the reaction time applied. For magnesium, residual concentration initially decreased significantly to 9.2 mg/L and then decreased slightly to 2.7 – 6.9 mg/l when the reaction time was increased beyond 1 min. The increase of the reaction time beyond 1 min did not result in any significant improvement in removal performances. Removal performance of magnesiumwas high and exceeded 96%, regardless of the reaction time applied. The optimal operating conditions for calcium and magnesium removal by sodium hydroxide precipitation were identified as follows: i) initial pH = 12, ii) stirring speed = 50 rpm, iii) reaction time = 1 min. Under these conditions, removal performance was 99.6 % of calcium and 96.2% for magnesium.
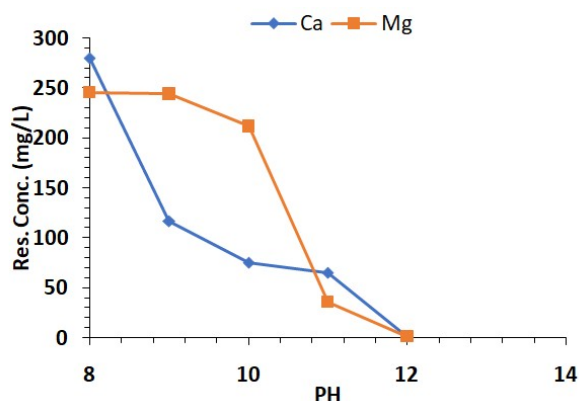

Figure 2. Effect of initial pH on calcium and magnesium concentrations (reaction time: 5 min, stirring speed: 300 rpm).

Chemical precipitation with lime: The effect of lime dosage on calcium and magnesium removal is shown in Figure 5. The results showed that lime dosage had a significant effect on calcium and magnesium removal. For calcium, two distinct phases were observed: phase I: characterized by a gradual decrease of calcium concentration, from 260.7 mg/L to 112.5 mg/L (56.8% of removal) when lime dose was increased up to 7 g/L; phase II: characterized by an increase of

calcium concentration from 112.5 mg/l to 779.8 mg/L, when lime dosage was increased up to 15 g/L. For magnesium, two phases were observed: i) a decreasing phase, characterized by a gradual reduction in magnesium concentration from 236.7 mg/L to 0.1 mg/L, when lime dosage was increased up to 9 g/L (99.9% of removal); ii) a stable phase,
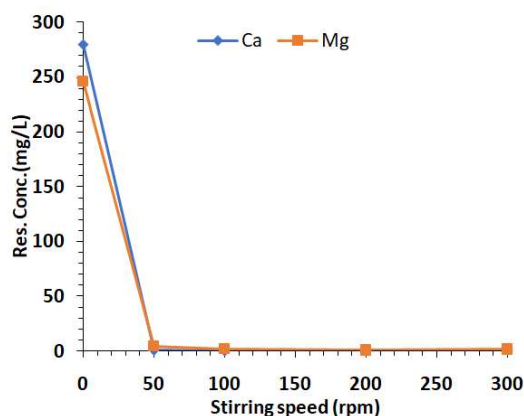

Figure 3. Effect of stirring speed on calcium and magnesium removal (pH = 12, reaction time: 5 min)
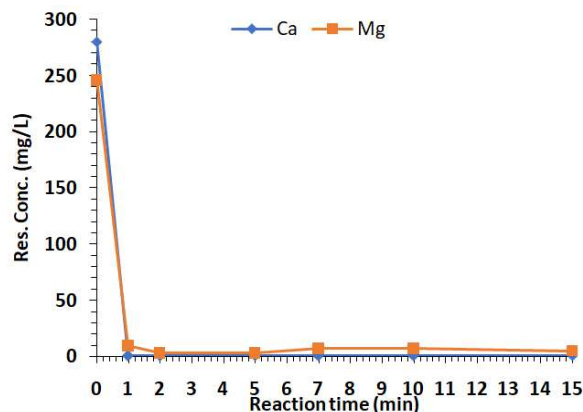

Figure 4. Effect of reaction time on calcium and magnesium concentrations (pH = 12, stirring speed: 50 rpm)

where an increase in lime dosage were not necessary because very low residual magnesium concentration was found in the leachate (0.0 – 0.2 mg/l). For better simultaneous calcium and magnesium removal, an optimal lime dose of 7 g/L was identified. At this dosage, residual calcium and magnesium concentrations were 112.5 mg/L (56.8% removal) and 109.5 mg/L (53.6% removal), respectively. The effect of stirring speed on calcium and magnesium removal is shown in Figure 6. The results indicate that stirring speed had a significant effect on precipitation performance. For both calcium and magnesium, two distinct phases were observed: i) phase I: characterized by a decrease of concentrations to 78.9 mg/L for calcium (71.2% removal) and 15.0 mg/L for magnesium (93.6% removal) up to 50 rpm; ii) phase II: characterized by an increase of calcium and magnesium concentrations, when stirring speed was increased up to 300 rpm. The effect of reaction time on calcium and magnesium removal is shown in Figure 7. Calcium and magnesium were instantaneously reduced when lime was added. Calcium concentration reduced from 260.7 mg/L to 70.3 mg/L (73.0% of removal) and magnesium from 237.7 mg/l to 30.3 mg/l (87.2% of removal), within the first minute of treatment. Increasing the reaction time beyond 1 min did not result in any improvement in removal performances. The optimal operating conditions for calcium and magnesium removal by lime precipitation were identified as follows: i) Lime dose = 7 g/L, ii) stirring speed = 50 rpm, iii) reaction time = 1 min. Under these conditions, removal efficiencies were 73.0% for calcium and 87.2% for magnesium.

Electrocoagulation: The effect of current density on calcium and magnesium removal by EC is shown in Figure 8. Current density plays a crucial role in calcium and magnesium removal. As the applied current density increased from 2.3 to 11.4 mA/cm², a decrease in calcium and magnesium residual concentrations was observed.
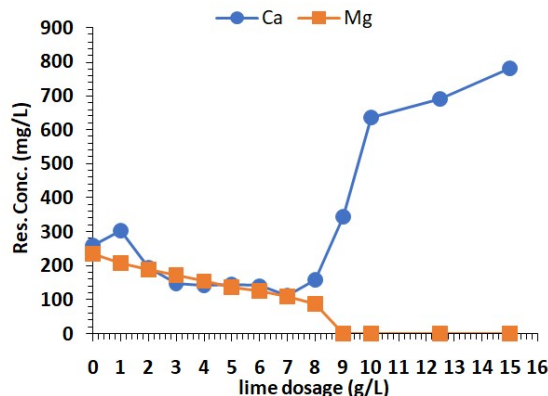

Figure 5. Effect of lime dosage on residual calcium and magnesium concentrations (stirring speed: 300 rpm, reaction time: 5 min)
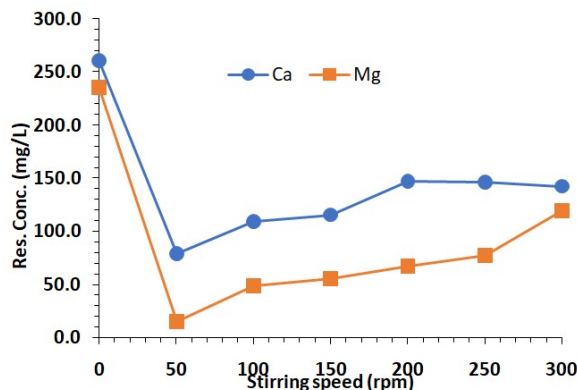

Figure 6. Effect of stirring speed on calcium and magnesium concentrations (lime dose of 7 g/L, reaction time: 5 min)
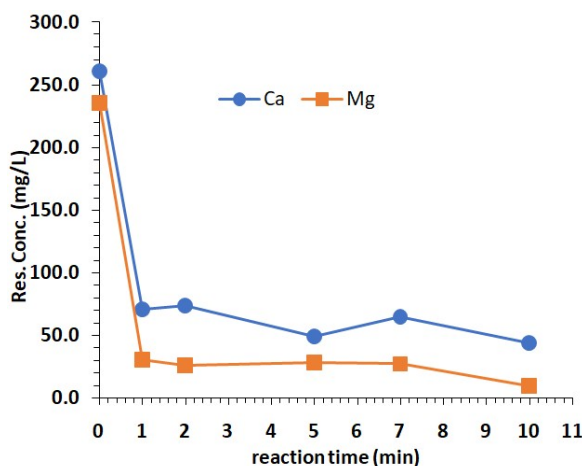

Figure 7. Effect of reaction time on calcium and magnesium concentrations (lime dose: 7 g/L, stirring speed:50 rpm)

Calcium concentration was 55.6 mg/L (82.7% of removal) at 2.3 mA/cm^2, 25.6 mg/L (92% of removal) at 4.5 mA/cm^2 and 3.2 mg/L (99.0% of removal) at 11.4 mA/cm^2. In comparison, magnesium concentration decreased slightly when current density increased. It was 210.2 mg/L (25.6% of removal) at 2.3 mA/cm^2, 172.3 mg/L (39.0% of removal) at 4.5 mA/cm^2 and 62.4 mg/L at 11.4 mA/cm^2 (77.9% of removal). Current density of 11.4 mA/cm^2 leads to the highest performances: 99% of removal for calcium and 77.9% of

removal for magnesium. However, beyond 4.5 mA/cm², the settling of the treated leachate is impossible. The effect of treatment time on calcium and magnesium removal is shown in Figure 9. For calcium, two distinct phases were observed: i) phase I : characterized by a rapid decrease in concentration, reaching a minimum value of 37.7 mg/L (88.3% of removal) at 20 min; ii) phase II: characterized by a slow decrease in concentration, reaching 23.5 mg/L (92.7% of removal) at 30 min, 9.1 mg/L (97.1% of removal) at 40 min and 3.2 mg/L (99.7% of removal) at 60 min. In comparison, magnesium reduction followed one phase: a decreasing phase, characterized by a gradual decrease in magnesium concentration, reaching 134.2 mg/L (52.5% of removal) at 20 min, 101.3 mg/L (64.1% of removal) at 30 min,75.3 mg/L (73.3% of removal) at 40 min and 35.6mg/L at 60 min (87.4% of removal). Treatment time of 60 min leads to the highest removal performance of calcium and magnesium. However, beyond 30 min, the settling of the treated leachate is impossible. The optimal operating conditions for calcium and magnesium removal by EC were then identified as follows: i) current density = 4.5 mA/cm², treatment time = 30 min. Under these conditions, removal efficiencies were 92.7% for calcium and 64.1% for magnesium.
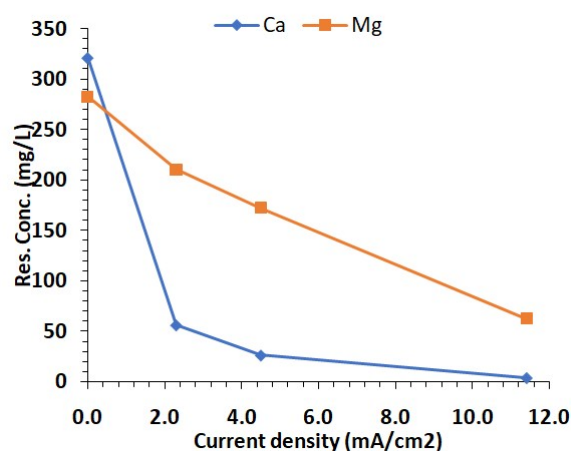

Figure 8. Effect of current density on calcium and magnesium concentrations (reaction time = 30 min).
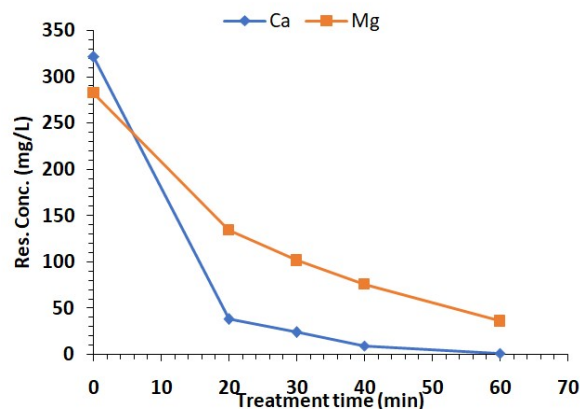

Figure 9. Effect of treatment time on calcium and magnesium concentrations (current density of 4.5 mA/ cm²)

DISCUSSION

The predominance of calcite in the scaling deposits can be directly related to the high calcium concentrations in the raw leachate. Calcite precipitation is thermodynamically favoured, particularly during leachate heating, which decreases CO_2 solubility and shifts the carbonate equilibrium toward $CaCO_3$ formation. The formation of calcite deposits on the plates of the heat exchangers leads to a significant reduction in heat transfer efficiency and contributes to a gradual fouling of the equipment. This phenomenon explains some

operational difficulties observed, particularly during the winter season, when leachate was heated to maintain suitable temperatures for the biological processes (Renou *et al*., 2008; Renou *et al*., 2009). Chemical precipitation by sodium hydroxide demonstrated high performance for the simultaneous removal of calcium and magnesium, which occurred under high pH. The optimal reaction time of 1 min indicates an extremely rapid kinetics, confirming that NaOH precipitation is primarily governed by chemical equilibrium rather than mass transfer limitations. Such behaviour is typical of hydroxide and carbonate-driven precipitation reactions, occurring at high pH, where solubility limits are rapidly exceeded (Metcalf & Eddy, 2014). The results of the present study are consistent with previous studies on highly mineralized wastewaters, such as landfill leachates. Liu *et al*. (2024) reported quasi-complete removal of calcium and magnesium (>97%) using a two-stage precipitation process combining lime and caustic soda for the treatment of reverse osmosis concentrate. Similarly, Shafer-Peltier *et al*. (2020) showed that adjusting produced water to pH 11 using NaOH efficiently removed cations such as Ca^{2+} and Mg^{2+}. Ayoub *et al*. (2014, 2024) also reported a quasi-complete removal of calcium and magnesium during seawater softening. Despite its high efficiency, NaOH precipitation presents significant operational limitations, such as high pH conditions, chemical consumption and the need for neutralization of treated wastewater. Moreover, large amounts of precipitated sludge are generated, requiring appropriate handling and disposal, which increases operational costs and may limit large-scale application (Shafer-Peltier *et al*., 2020). High pH conditions may also adversely affect the biological treatment.

Lime precipitation showed moderate removal performance of calcium and magnesium. These results are consistent with previous studies on lime precipitation (Renou *et al*., 2009). The optimal lime dosage obtained in this study is comparable to values reported in the literature for landfill leachates and industrial effluents. The increase of residual calcium concentration beyond the optimal lime dosage (7 g/L) was correlated to the excess of calcium introduced into the system through lime addition, which outweighs the precipitation effect. This highlights the need for precise dosage control. The rapid kinetics observed during lime treatment indicate that precipitation reactions are also governed by chemical equilibrium. Once supersaturation is reached, nucleation and crystal growth occur rapidly, leading to fast calcium removal (Metcalf & Eddy, 2014). The increase of the residual calcium concentration beyond a stirring speed of 50 rpm can be explained by the destabilization and breakage of formed precipitates because of the high mixing conditions, leading to the redispersion of calcium and magnesium into the liquid phase. Therefore, moderate mixing conditions are essential to promote effective precipitation with lime. The main limitations of lime precipitation remain the generation of important quantities of calcium carbonate sludge, which increases handling and disposal costs (Shafer-Peltier *et al*., 2020). Furthermore, lime treatment alone may be insufficient for the complete simultaneous removal of calcium and magnesium.

The electrocoagulation process proved to be an effective alternative for the removal of calcium and magnesium. Its performance was strongly dependent on current density and treatment time, confirming the critical role of electrochemically generated coagulants. Calcium and magnesium removal occurred mainly through coprecipitation and adsorption onto freshly formed aluminum hydroxide flocs (Chen, 2004; Mollah *et al*., 2001). Increasing the current density increased the dissolution rate of the aluminum anode, which generates more Al^{3+} ions and, consequently, more aluminum hydroxide flocs for adsorption and coprecipitation. However, high current densities or treatment time result in poor floc settleability and high energy consumption. Similar results have been reported in the literature (Mendez-Ruiz *et al*., 2023). Compared with chemical precipitation, electrocoagulation does not require chemical reagents or extreme pH. EC generally produces smaller amounts of sludge. Nevertheless, electrode passivation, energy consumption, and sludge management remain key challenges for large-scale implementation.

Among the three investigated processes, sodium hydroxide precipitation achieved the highest removal performances of calcium (99.6 % of removal) and magnesium (96.2% of removal) under optimal conditions. However, it required high chemical consumption, high sludge production, and post – adjustment of pH. Lime precipitation provided moderate removal performances for calcium (73.0% of removal) and high sludge production under optimal conditions. Concerning EC, it achieved high removal performance for calcium (92.7% of removal), but moderate performance for magnesium (64.1% of removal), under optimal conditions. EC minimize the use of chemicals and offers more operational flexibility. EC appears to be a promising complementary or alternative pretreatment option when reducing chemical consumption is a priority. Its integration with biological treatment may offer a balanced and cost-effective strategy for the management of landfill leachates.

CONCLUSION

This study demonstrated that scaling deposits formed on the plates of the heat exchangers in landfill leachate treatment facilities are predominantly composed of calcite ($CaCO_3$). This confirms the critical need for calcium removal before any landfill leachate heating. Among the three physicochemical pretreatment processes investigated for the removal of calcium and magnesium, sodium hydroxide precipitation achieved the highest removal performance under optimal conditions, exceeding 96% for both calcium and magnesium removal. However, its application is limited by the high chemical consumption, high pH and sludge production. Lime precipitation provided moderate removal performance for calcium (73.0% of removal) and significant sludge production. Electrocoagulation achieved high calcium removal and moderate magnesium removal, while minimizing chemical use and extreme pH adjustment of the treated water. Although high current densities and long treatment times enhanced removal efficiencies, they negatively affected floc settleability. Overall, sodium hydroxide precipitation remains the most efficient option, whereas electrocoagulation emerges as a promising alternative pretreatment because it reduces the chemical use and gives operational flexibility. The integration of physicochemical pretreatment with biological processes represents a viable strategy to mitigate scaling, improve process reliability, and enhance the long-term sustainability of landfill leachate treatment systems. This study provided practical guidance for selecting an appropriate pretreatment strategy to control scaling and enhance the long-term performance of landfill leachate treatment systems. Future work should focus on pilot-scale validation, techno-economic assessment, energy optimization, and sludge management to support the full-scale implementation.

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