

ORIGINAL RESEARCH

Climate Change

ENVIRONMENTAL PROGRESS
& SUSTAINABLE ENERGY

Direct mineral carbonation of fly ash under high pressure using acid mine drainage: Effects of solid-to-liquid ratio, stirring speed and CO₂ pressure

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Abstract

CO₂ emissions contributing to global warming and waste related to energy generation using coal pose an issue in developing countries. Mineral carbonation (MC) of fly ash (FA) using acid mine drainage (AMD) can reduce CO₂ and the negative environmental impact of FA including AMD from coal mining activity. The present study examined the direct carbonation (DC) of FA using pure water and AMD in a 600 mL autoclave pressure reactor. A preliminary study was conducted to observe the leaching behavior of FA in water and AMD. DC was conducted to determine the effect of solid-to-liquid ratio (0.2 and 0.5 g mL⁻¹), stirring speed (100 and 400 rpm) and CO₂ pressure (1–4 MPa) on the carbonation performance. The maximum calcium carbonate content in the carbonated FA was 4.33 wt% with a 60% conversion of calcium to CaCO₃ for DC with pure water. The electricity required by the process was 18.9 kWh t⁻¹-CO₂, corresponding to 0.019 t-CO₂ emitted t⁻¹-CO₂ fixed in FA. DC using AMD was effective and the maximum CaCO₃ content in FA was 6.68 wt%. This was due to the additional calcium content and enhanced calcium extraction provided by AMD. The actual CO₂ uptake capacity was 29.4 g-CO₂ kg⁻¹ fly ash. Studies on AMD as a reaction solvent for MC are few but can improve the carbonation performance of FA. Hence, the method can be viable for mining industries to mitigate negative environmental impacts and generate additional revenue through carbon credits.

KEYWORDS

calcium carbonate content; carbon capture, utilization, and storage; CO₂ uptake capacity; direct carbonation

1 | INTRODUCTION

The continued use of fossil fuels to meet energy demands worldwide contributes significantly to global warming.¹ Considering the economic benefit

Abbreviations: AMD, acid mine drainage; CCUS, carbon capture, utilization, and storage; CFA, coal fly ash; MC, mineral carbonation.

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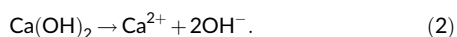
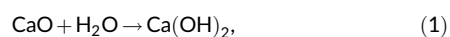
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of the coal mining industry in developing countries, fossil fuels such as coal are likely to continue to provide the primary energy needs before the full transition into renewable energy.² CO₂ sequestration is therefore currently amongst the practical methods sought for achieving the temperature goals reflected in the Paris Agreement (i.e., below 2°C above pre-industrial levels) and for reaching the net zero target by 2050.^{3,4}

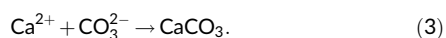
Carbon capture, utilization, and storage (CCUS) is a promising CO₂ sequestration method.⁵ CCUS through accelerated mineral carbonation (MC) is a promising solution to permanently store CO₂ in mineral carbonate form.⁶ Accelerated MC demonstrates considerably high CO₂ storage potential (i.e., >10,000 Gt of CO₂) due to the large availability of silicates worldwide and is considered a safer method.^{7,8} Natural MC occurs too slowly to achieve effective CO₂ storage.⁹ Hence, accelerated MC through the application of engineered processes (ex-situ MC) for more effective consumption of atmospheric carbon (CO₂) was first proposed by Seifritz.¹⁰

For accelerated MC, captured CO₂ from a process plant is stored permanently by reacting it with calcium and/or magnesium-bearing natural silicate minerals, often in the form of wollastonite (CaSiO₃), serpentine (Mg₃Si₂O₅(OH)₄), or olivine (Mg₂SiO₄),^{11,12} or by reacting the captured CO₂ with alkaline-rich waste materials such as concrete waste,¹¹ coal fly ash (CFA),¹³ steel slag,¹⁴ waste cement,¹⁵ and so forth, under controlled reaction conditions.¹⁶

Natural silicate minerals require mining and expensive pre-treatment measures (i.e., crushing, grinding, etc.) for effective reactivity; however, alkaline-rich industrial waste materials do not require such pre-treatment and offer higher reactivity because they are chemically less stable than natural silicate minerals.^{17,18} A previous study identified these waste feedstocks as viable alternatives to natural silicate minerals for MC, particularly in facilities producing both CO₂ and alkaline waste residues.¹⁹ Additionally, using waste feedstocks for MC eliminates the negative environmental impact arising from their disposal.¹¹ CaCO₃ and MgCO₃ are normally produced by reacting captured CO₂ with these alkaline-based materials, with CO₂ permanently stored in mineral carbonate form under the following reaction mechanism²⁰:



Calcium carbonate (CaCO₃) is then formed through the reaction between calcium ions (Ca²⁺) and carbonate ions (CO₃²⁻) formed from the interaction of CO₂ and H₂O^{21,22}:



Although MC has advantages, including high CO₂ storage potential and better safety due to the permanent storage of CO₂, the method is costly.²⁰ Hence, the focus has been on improving the rate of the carbonation process by determining the optimum set of process parameters, such as temperature, reaction time, particle size, S L⁻¹ ratio, partial pressure of CO₂, and so forth, to reduce the energy and costs associated with accelerated MC.^{18,23} Some studies have found that certain process parameters are critical in improving carbonation performance.²⁴

Research on the accelerated MC process using alkaline waste material such as fly ash has mostly focused on direct carbonation.^{25,26} The main advantage of direct carbonation is the simplicity of the process, as it is a single-step process, unlike indirect carbonation, which takes place in two or more steps.⁴ For the accelerated carbonation of alkaline-rich wastes, whether through the direct or indirect carbonation route, water has been used as the reaction medium, and brines have also been utilized.^{27–29} Given the high volumes of water required for MC processes, the potential application of this technology at a commercial scale in a water-scarce country like South Africa presents a serious challenge.³⁰ Wastewater reaction media are thus promising alternatives to water for accelerated MC.²⁸

The use of wastewater reaction media for CO₂ sequestration is advantageous, as effective CO₂ capture can be achieved due to the additional calcium from the wastewater, with the wastewater neutralized in the process.³¹ Previously, brine solutions were used to enhance the retention of CO₂ in fly ash (FA).^{32,33} A feasibility study on the CO₂ sequestration process using the neutralization process of AMD has also been investigated.³⁴ However, there are limited studies on the use of wastewater for CO₂ sequestration, particularly on the use of AMD.³¹ AMD is acidic wastewater that results from mining activity when the mining activity exposes sulfur-bearing minerals (mostly iron sulphides) to oxygen, thus causing these sulfur-bearing minerals to undergo oxidation. When sulfur-bearing minerals come into contact with oxygen and water, this results in an acidic pH solution that contains high concentrations of sulphates (SO₄²⁻), iron (Fe²⁺), aluminium (Al³⁺), calcium (Ca²⁺), manganese (Mn²⁺), magnesium (Mg²⁺), and so forth.³⁵ Based on the limited work conducted on CO₂ sequestration using AMD, this study aims to investigate accelerated MC using fly ash in the presence of pure water and AMD for CO₂ sequestration and to highlight the potential benefit of using AMD as a reaction medium.

Mineral carbonation of FA is an interesting research area, especially considering its laborious management, with high volumes of historical and fresh ash being produced each year.³⁶

The present work aimed to investigate the carbonation performance of a direct carbonation process of FA using water and AMD to compare the carbonation performance from the respective processes, which is a novel approach for CO₂ sink systems, given the limited work on CO₂ sequestration using AMD. Based on the present study, the management of two waste products, FA and AMD, generated from coal mines, can be addressed simultaneously, which contributes toward achieving sustainable development goals.

2 | EXPERIMENTAL

2.1 | Materials

The fly ash used in this study was Durapozz fly ash, which is processed and classified fly ash from Lethabo power station, South Africa. The fly ash contains 54.36 wt% SiO₂, 34.19 wt% Al₂O₃, 4.06 wt% CaO, and 3.12 wt% Fe₂O₃ expressed in oxide forms. 1.75 wt% TiO₂ and 0.90 wt % MgO was present in lower concentrations, with other elemental species contributing to the final elemental composition. According to the

American Society for Testing and Materials (ASTM), the fly ash can be classified as Class F fly ash due to the calcium content of 4.06 wt%, which is lower than 10%.³⁷ Penstock AMD was obtained from a coal mine in Mpumalanga, South Africa. Raw AMD had a pH of 2.63 and contained SO_4^{2-} . Fe, Ca, Mg, and Al were present as major elements.

2.2 | Methods

2.2.1 | Analytical methods

AMD samples including the leachate obtained after calcium extraction and carbonation were analyzed using inductive coupled plasma: optical emission spectroscopy (ICP-OES). The instrument used for ICP-OES analysis was a Varian 710. The instrument contains an axial torch, and the instrument uses Ar gas. The instrument was calibrated with a blank and 1, 5, 10, and 100 ppm LGC 1586 spectrascan multi-element standard. Mineralogical phases present in the fly ash samples were determined by x-ray diffraction (XRD). XRD data were recorded on a Bruker D2 Phaser Diffractometer using Cu K radiation ($\lambda = 1.5418 \text{ \AA}$) generated at 30 kV and 10 mA. Each sample was scanned from 4° to 80° in 2θ steps of $\sim 0.02^\circ$ with a 0.5-s exposure time per step.

The elemental phases were determined using x-ray fluorescence (XRF) spectroscopy on a PANalytical Axios Wavelength Dispersive Spectrometer. The spectrometer was fitted with an Rh tube with the following analyzing crystals: LIF200, LIF220, PE 002, Ge 111, and PX1. The instrument was fitted with a gas-flow proportional counter and then a scintillation detector. The gas-flow proportional counter used a 90% Argon-10% methane mixture of gas. Major elements were analyzed on a fused glass disk using a 2.4 kW Rhodium tube. The matrix effects in the respective samples were corrected.

This was done by applying theoretical alpha factors, as well as measured line overlap factors to the raw intensities measured by using the SuperQ PANalytical software.

2.2.2 | Procedure for preliminary test: Calcium extraction

A calcium extraction test was conducted using water and AMD with no CO_2 introduced into the reaction vessel. The calcium extraction test was used to identify the difference in the behavior of calcium extraction from fly ash while using water compared to AMD. The selected conditions were based on optimized parameters for the extraction of calcium from literature.^{24,38} A reaction mixture (i.e., fly ash with water or AMD) was prepared using a solid-liquid ratio of 0.2 g mL^{-1} and placed inside the reaction vessel, with the temperature and stirring rate set to 70°C and 400 rpm, respectively. The extraction times were 30, 60, 90, and 120 min. After extraction, the solution mixture was separated using a $0.45 \mu\text{m}$ pore filter paper. The recovered solids and liquids were analyzed accordingly.

2.2.3 | Procedure for carbonation experiments using autoclave reactor

Direct carbonation experiments were conducted using a 600 mL high temperature/high-pressure autoclave reactor (6.3 cm inner diameter) made of stainless-steel material (model series 4560, maximum temperature and pressure of 350°C and 345 bar) as shown in Figure 1.

The process conditions for direct carbonation were selected based on power consumption considerations and suitable conditions for calcium extraction, CO_2 dissolution, and CaCO_3 precipitation from the literature.^{24,39,40} A reaction mixture (i.e. fly ash with water or AMD) was prepared using a solid-liquid ratio of 0.5 g mL^{-1} and placed inside the reaction vessel, with the temperature and stirring rate set to 70°C and 100 rpm respectively. CO_2 was then introduced. The reaction time varied between 30, 90, and 120 min, while CO_2 pressure remained constant initially at 1 MPa. The same was done at 2, 3, and 4 MPa. Chittick tests were carried out to determine the CaCO_3 content in the carbonated fly ash. A diagram of the Chittick test apparatus is provided in Figure 2.

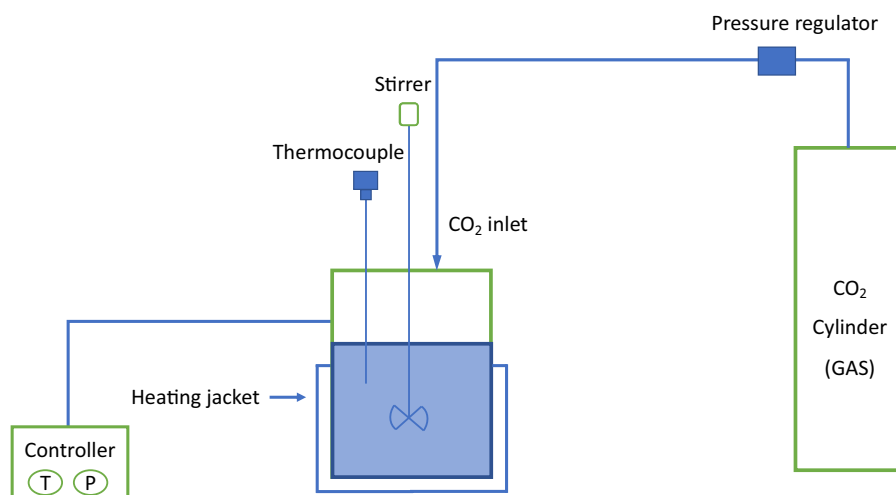


FIGURE 1 Schematic diagram of experimental setup.

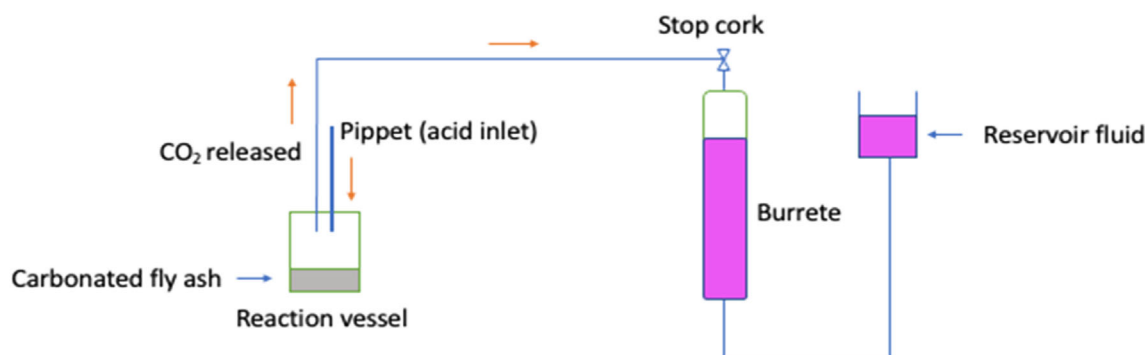


FIGURE 2 Schematic diagram of Chittick test apparatus.

The Chittick tests were done by reacting 1.70 g of carbonated fly ash solid residues with 20 mL of a hydrochloric acid (HCl) solution. The CO_2 evolved was determined by reading the displacement of the reservoir fluid in the burette, which was a 1-L solution with 100 g of sodium chloride (NaCl), 1 g sodium hydrogen carbonate (NaHCO_3), and 2 mL of methyl orange, where a sulfuric acid (H_2SO_4) solution was added enough to turn the solution dark pink. The testing time was 21 min.

3 | THEORY/CALCULATION

3.1 | Carbonation performance

The formulae used in this study were used to calculate the CaCO_3 content in the carbonated fly ash, the calcium conversion, the actual CO_2 uptake capacity ($\text{g-CO}_2 \text{ kg}^{-1}$ fly ash) of fly ash, and the power consumption.

The CaCO_3 content in the carbonated fly ash sample was determined from Equation (4).

$$\text{CaCO}_3 \text{ content} = \frac{M_{\text{CaCO}_3}}{M_{\text{CFA}}} \times 100, \quad (4)$$

where M_{CaCO_3} was the mass of CaCO_3 in the carbonated fly ash sample and M_{CFA} was the mass of the weighed carbonated fly ash sample. M_{CaCO_3} was calculated from the Chittick test.

It was based on the volumetric displacement of the reservoir fluid shown in Figure 2 when the carbonated fly ash was reacted with an HCl solution. M_{CFA} was 1.70 g. For the calculation, it was assumed that all generated gas was CO_2 due to the decomposition of CaCO_3 using an HCl solution.

Moreover, M_{CaCO_3} can be calculated using Equation (5).

$$M_{\text{CaCO}_3} = \frac{P(fV)}{RT} \times m_{\text{CaCO}_3}, \quad (5)$$

where P was the atmospheric pressure, 1 bar. $V(\text{L})$ was the generated volume of CO_2 during the test. The testing time 21 min. R was the ideal gas constant, $0.08314 \text{ L bar K}^{-1} \text{ mol}^{-1}$, and T was the temperature at which the acid test was conducted, which was

the room temperature, 298.15 K. m_{CaCO_3} was the molar mass of CaCO_3 .

To obtain the correction factor f , a calibration procedure was conducted using pure CaCO_3 (Kimix Chemicals, Cape Town, South Africa, purity: $\sim 98.8\%$).

The calcium conversion to CaCO_3 from fly ash for the $\text{Ca-H}_2\text{O-CO}_2$ system was calculated using Equation (6).

$$\text{Calcium conversion} = \frac{\text{Final CaCO}_3 \text{ content} \times \frac{m_{\text{Ca}}}{m_{\text{CaCO}_3}}}{w_{\text{CaO}} \times \frac{m_{\text{Ca}}}{m_{\text{CaO}}}} \times 100, \quad (6)$$

where CaCO_3 content was the calcium carbonate content in the carbonated fly ash, m_{Ca} , m_{CaO} , m_{CaCO_3} was the molar mass of calcium, calcium oxide, and calcium carbonate respectively, while w_{CaO} was the starting mass of CaO (wt%) in the reactor. The actual CO_2 uptake capacity, which provided an estimation of the CO_2 in CaCO_3 (solid phase) was calculated using Equation (7).

$$\text{CO}_2 \text{ uptake capacity} \left(\text{g-CO}_2 \text{ kg}^{-1} \text{ fly ash} \right) = \text{CaCO}_3 \text{ content} \times \frac{m_{\text{CO}_2}}{m_{\text{CaCO}_3}}, \quad (7)$$

where CaCO_3 content was the calcium carbonate content in the carbonated fly ash, m_{CO_2} , and m_{CaCO_3} was the molar mass of CO_2 and CaCO_3 respectively. Reactor carbonation efficiency was calculated using Equation (8).

$$\text{Reactor carbonation efficiency} = \frac{M_{\text{CO}_2}}{Vt}, \quad (8)$$

where M_{CO_2} were the grams of CO_2 from the CaCO_3 content in the carbonated fly ash, $V (\text{m}^3)$ was the reactant volume and $t(\text{h})$ was the total reaction time.

3.2 | Energy balance

The energy balance performed was used to determine the power requirement of the process from the general energy balance expressed by Equation (9).⁴¹

$$E_{\text{system}} = Q + W, \quad (9)$$

where Q for the $\text{Ca-H}_2\text{O-CO}_2$ system was the power required for heating. The power for heating was determined from the sum of the heat due to heating of water, fly ash, and CO_2 .

$$Q = Q_{\text{water}} + Q_{\text{fly ash}} + Q_{\text{CO}_2}, \quad (10)$$

where $Q_{\text{water}} + Q_{\text{fly ash}} + Q_{\text{CO}_2}$ were calculated using Equation (11).

$$Q = mc_p \Delta T, \quad (11)$$

where m was the mass of the substance used during the carbonation process, c_p was the specific heat capacity of the substance, and ΔT was the temperature difference between the starting temperature of the reaction and the temperature at which the carbonation reaction was conducted. No heat recovery was considered during the calculation.

W was the power required for stirring, W_s , and the power for pressurization, W_p . The power for stirring was estimated using Equation (12).⁴²

$$W_s = N_p \rho n^3 d^5, \quad (12)$$

where the term W_s was the overall power consumption from stirring, N_p was the power number (which was dimensionless), ρ was the density of the fluid (kg m^{-3}), n (rps) was the stirring rate per second (s^{-1}), and d was the diameter of the paddle (m).

The power number N_p was determined from Nagata's equation.^{42,43}

$$N_p = \frac{A}{Re} + B \left(\frac{1000 + 1.2 Re^{0.66}}{1000 + 3.2 Re^{0.66}} \right)^P \left(\frac{Z}{D} \right)^{0.35 + \frac{b}{D}} \sin \theta^{1.2}, \quad (13)$$

where $Z(m)$, $D(m)$, $b(m)$, $\theta(-)$, $\mu(\text{Pa s})$ was the reaction solution height, reactor diameter, paddle height, paddle gradient, and the viscosity of the fluid respectively. The constants A , B , and P were determined from Equations (14) through (18).⁴²

$$A = 14 + \frac{b}{D} \left(670 \left(\frac{d}{D} - 0.6 \right)^2 + 185 \right), \quad (14)$$

$$\log B = 1.3 - 4 \left(\frac{b}{D} - 0.5 \right)^2 - 1.4 \left(\frac{d}{D} \right), \quad (15)$$

$$P = 1.1 + 4 \left(\frac{b}{D} \right) - 2.5 \left(\frac{d}{D} - 0.5 \right)^2 - 7 \left(\frac{b}{D} \right)^4. \quad (16)$$

The Reynolds number was calculated using Equation (14).

$$Re = \frac{\rho n d^2}{\mu}. \quad (17)$$

The power for pressurization W_p was determined from Equation (15).

$$W_p = nRT \log \frac{P_2}{P_1}, \quad (18)$$

where n was the number of moles of CO_2 fixed in the solid phase (i.e., CO_2 in CaCO_3). R was the ideal gas constant, $0.08314 \text{ L bar K}^{-1} \text{ mol}^{-1}$, T was the temperature of the reaction, and P_1 and P_2 was the initial and final pressure of the reaction, respectively. The total power requirement was, therefore, the sum of the power due to heating, stirring, and pressurization.

4 | RESULTS AND DISCUSSION

4.1 | Preliminary test: Calcium extraction

A comparison was made on the extraction of calcium from fly ash using water and AMD. It was envisaged that this comparison would demonstrate better calcium extraction using AMD from fly ash⁴⁴ (Figure 3).

Figure 3 illustrates higher calcium concentration in solution with time when AMD was used for all cases considered. The higher calcium concentration from AMD extraction was due to the initial calcium concentration of 363 mg L^{-1} in AMD. Additional calcium extracted from the fly ash material can be attributed to the acidity of AMD, which possibly enhanced the leaching of calcium from fly ash.⁴⁴ The calcium extraction percentage from fly ash using AMD was 16%, while the extraction percentage using water was 7%. Figure 3 indicates that the extraction using AMD was relatively higher; however, further tests were conducted to examine the calcium extraction and precipitation process as presented in Section 4.2.

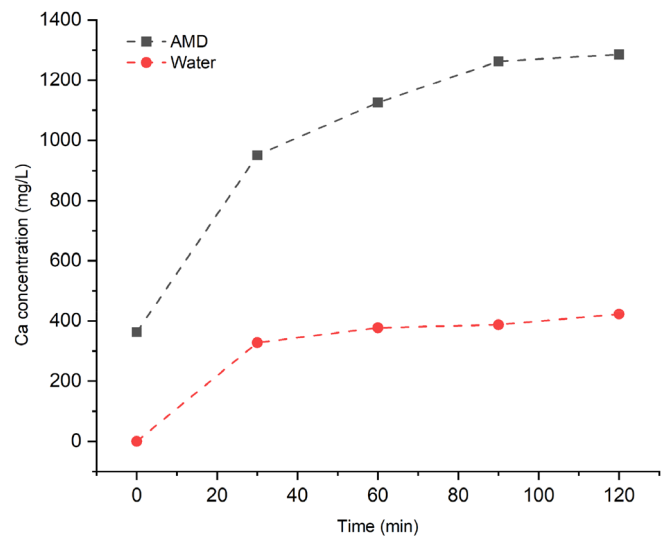


FIGURE 3 Calcium extraction with water and acid mine drainage (AMD) over time (70°C , 0.2 g mL^{-1} , 400 rpm).

4.2 | Carbonation experiments using autoclave reactor

4.2.1 | Direct carbonation with water

Direct carbonation with pure water was conducted at 70°C as this is the optimal temperature for carbon capture as per previous studies.³⁹ Previous studies found that the CO₂ sequestration capacity might be enhanced at higher solid dosages. Therefore, an initial S L⁻¹ ratio of 0.5 g mL⁻¹ was used for the experiments.^{9,40} The lower stirring speed of 100 rpm was initially selected based on power consumption considerations.

The CaCO₃ content in the carbonated fly ash was estimated using Equation (5). Figure 4 shows the CaCO₃ content in fly ash over 120 min of direct carbonation with water for different pressures (1–4 MPa). The CaCO₃ content showed an increase, with increasing CO₂ pressure from the initial CaCO₃ content found in the raw fly ash before carbonation in all cases. The initial CaCO₃ content of 0.75 wt% in the fly ash can be attributed to the uptake of CO₂ from the atmosphere during handling and bulk ash storage. The highest CaCO₃ content found in the carbonated fly ash over time was 4.33 wt% (5.08–0.75 wt%) for the initial CO₂ pressure of 4 MPa. The concentration of CO₂ molecules dissolved in the liquid phase was directly proportional to the pressure at a given temperature.^{25,45}

This means that for a higher CO₂ pressure, there was a higher concentration of CO₂ molecules available to react with calcium, resulting in a higher CaCO₃ content. The result was similar to the results obtained by Dananjayan et al.,⁴⁶ who found the net carbon gain in the coal fly ash (CFA) increased with increasing initial CO₂ pressure up to 10 bar (1 MPa). The increase in the CaCO₃ content with increasing CO₂ pressure can also be explained by the higher solubility of calcium for increasing CO₂ pressure owing to Le Chatelier's principle.⁹ The direct carbonation at higher pressure resulted in a

higher calcium concentration because as the pressure increased, the dissolution of fly ash increased, increasing the calcium concentration as shown in Figure 5.

According to Figure 5, there was an increase in the concentration of calcium with time and with increasing pressure in the autoclave system. Katsuyama et al.¹⁵ also observed an increase in the solubility of calcium with increasing pressure. A previous study obtained a similar trend for calcium concentration behavior during carbonation.⁹ In this case, more calcium becomes available, which helps in the formation of CaCO₃, as observed in Figure 4.

Results in Figure 5 seem to indicate a considerable increase in the calcium extraction and CO₂ dissolution during the initial 30 min of carbonation, which stabilized after 30 min. This explains the steep increase in CaCO₃ content during the initial phase of the experiment (30 min) and lower CaCO₃ content thereafter at 4 MPa (Figure 4).

Despite the higher Ca concentration obtained in the initial 30 min, the calcium concentration still increased after 30 min, meaning that even though some of the calcium precipitated as CaCO₃, the increase in the calcium concentration indicated that the extracted amount of calcium was more than the amount of calcium precipitated into CaCO₃ in all cases.

The amount of calcium that precipitated as CaCO₃ was 60%. Muriithi et al.⁴⁷ achieved a similar calcium conversion percentage using fresh FA, with some previous work achieving lower conversion of calcium to CaCO₃.⁴⁸ The conversion was based on the CaCO₃ content in the carbonated fly ash. Figure 5 illustrates that calcium concentration after carbonation with pure water was generally low at 310 mg L⁻¹, considering the use of high CO₂ pressure and reaction times (i.e., 4 MPa and 120 min respectively).

Furthermore, the calcium concentration obtained after carbonation was lower compared to the concentration obtained from the calcium extraction test (Figure 3) due to the parameters used, such as the lower S L⁻¹ ratio of 0.2 g mL⁻¹, which possibly improved the

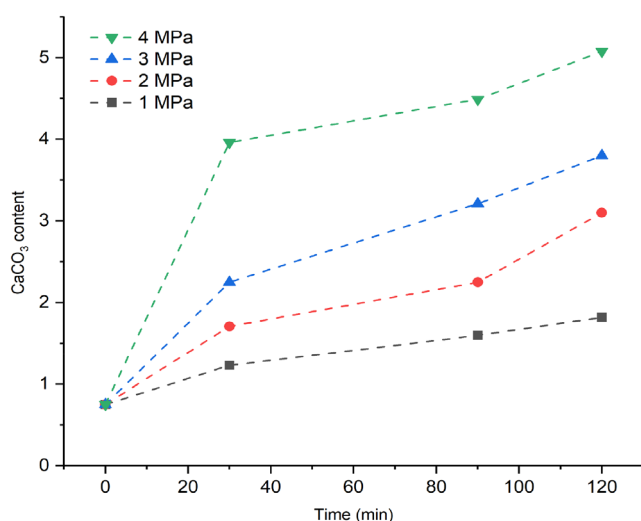


FIGURE 4 Effect of CO₂ pressure on CaCO₃ content using pure water (70°C, 0.5 g mL⁻¹, 100 rpm).

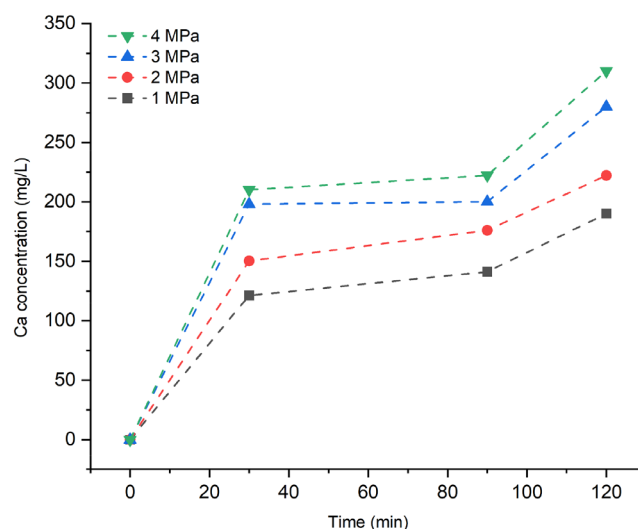


FIGURE 5 Calcium concentration after carbonation process with pure water (70°C, 0.5 g mL⁻¹, 100 rpm).

leaching of calcium during the calcium extraction with no CO_2 owing to the dilution effect. The decrease in calcium concentration after carbonation can also be ascribed to precipitation of calcium to CaCO_3 .¹⁴ The precipitation of calcium in the form of calcite (CaCO_3) can be observed from the XRD spectra presented in Figure 6. XRD analysis was carried out on solid residue samples collected after 30, 90, and 120 min reaction times, and the spectra are presented in Figure 6.

The XRD pattern presented in Figure 6 shows the mineral phases present in solid residues after direct carbonation using water. Strong intensity peaks were identified for quartz and mullite, which was expected because these mineral phases generally constitute the major crystalline phases for fly ash. Calcite was also identified, and its low peaks were found before 30, 40, and 65 (2θ). The peaks after 40 (2θ) were characterized by amorphous phases. XRD confirmed the transformation of the CaO fraction from uncarbonated fly ash to calcite in the carbonated fly ash. This was due to the interaction of calcium with CO_2 during the carbonation reaction.⁴⁵

4.2.2 | Direct carbonation with AMD

Direct carbonation with AMD was conducted using a lower S L^{-1} ratio of 0.2 g mL^{-1} and a higher stirring speed of 400 rpm as it was believed the carbonation performance might further be enhanced under these conditions.⁴⁹

According to Figure 7, the CaCO_3 content in the carbonated fly ash using AMD shows a similar comparison to that of pure water, where a rapid increase in the CaCO_3 content in the initial 30 min can

be observed. However, in the former case, the CaCO_3 content continued to increase even after 30 min, possibly due to the higher calcium concentration available while carbonating with AMD. The CaCO_3 content in the carbonated fly ash with AMD eventually reached a maximum of 6.68 wt% (7.43–0.75 wt%) after 120 min of carbonation at 4 MPa, and this was comparable to the % CaCO_3 formed (6.5 wt%) reported by Muriithi et al.⁴⁷ despite the higher CaO content (9.2 wt%) in the fly ash used in their study. We believe that higher % CaCO_3 conversion was due to the contribution of Ca from AMD and

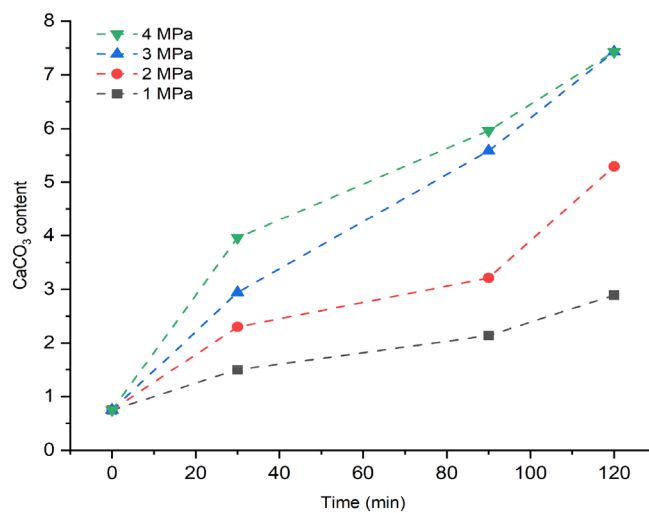


FIGURE 7 Effect of pressure on CaCO_3 content over time using acid mine drainage (AMD) (70°C , 0.2 g mL^{-1} , 400 rpm).

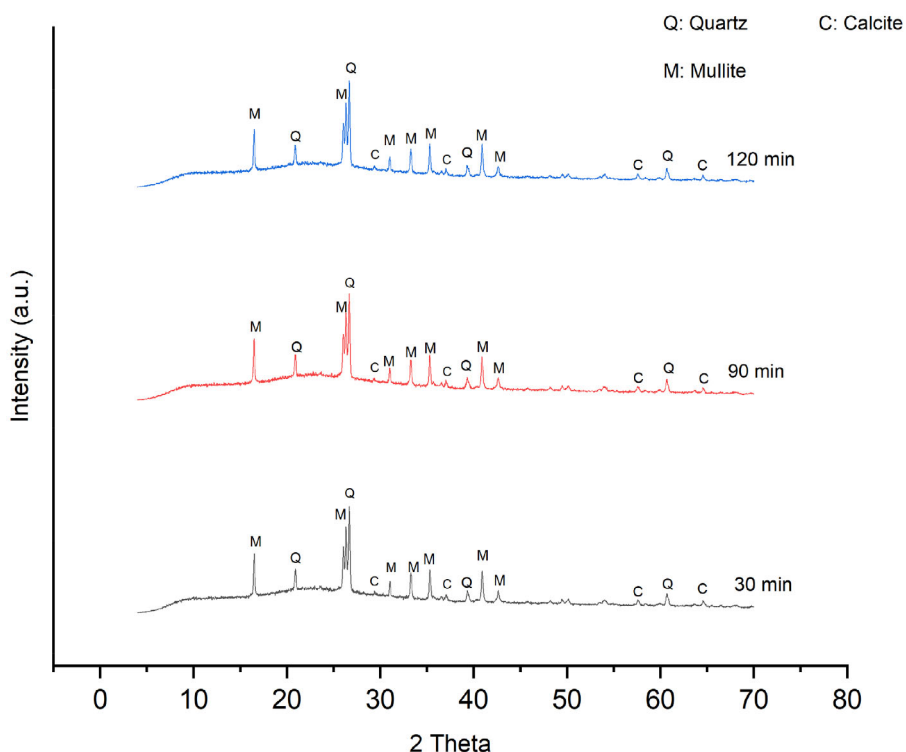


FIGURE 6 X-ray diffraction spectra for carbonated solid residues after direct carbonation with pure water (70°C , 0.5 g mL^{-1} , 100 rpm).

therefore there is an advantage to using AMD as an additional calcium source to enhance carbonation.

The lower S L^{-1} ratio of 0.2 g mL^{-1} used for carbonation using AMD as a reaction medium had the potential to contribute higher calcium through the dilution effect, while the higher stirring speed of 400 rpm promoted the mass transfer and dissolution of metal ions in solution to enhance the carbonation performance.^{24,50} Previous studies using CFA and hydrated lime, Jo et al.⁴⁰ and Lee et al.³⁴ obtained a higher CaCO_3 content of 16.1 wt% and 33.1 wt% while using fly ash material containing 15 wt% CaO and hydrated lime CaO >95%, respectively. The use of hydrated lime and the calcium present in the AMD in the latter study possibly contributed to the higher CaCO_3 content.

A recent mineralization study conducted using AMD achieved a relatively higher CO_2 sequestration capacity of $6.992 \times 10^{-4} (\text{g-CO}_2 \text{ g}^{-1}\text{-solution})$ due to the use of an external reaction accelerator in the form of an ultrasound cavitation device.³¹ A previous study exploring a different reaction medium, namely seawater, also achieved a high carbonation efficiency of 94.8%.⁵¹ It has also been previously indicated that a higher CO_2 uptake capacity of $0.016 \text{ g CO}_2/\text{g fly ash}$ using low calcium fly ash can be achieved when using a higher concentration of CO_2 .¹¹

The CO_2 uptake for the present work was based on the CaCO_3 content in the carbonated fly ash from direct carbonation with AMD. From the maximum CaCO_3 content of 6.68 wt%, the CO_2 uptake was $29.4 \text{ g-CO}_2 \text{ kg}^{-1}$ fly ash. The CO_2 uptake capacity was directly proportional to the pressure drop because the maximum CO_2 uptake capacity was obtained for the highest pressure drop of 1.86 MPa at 4 MPa after 120 min of carbonation. The results obtained in the present study were similar to those achieved by Dananjayan et al.⁴⁶ who obtained a CO_2 uptake of $26.3 \text{ g-CO}_2/\text{kg-fly ash}$ despite the slightly higher CaO content of 6.74 wt% used in their study. This can be explained by the additional calcium provided by AMD in this work. Montes-Hernandez et al.⁵² also achieved a similar CO_2 uptake of $26 \text{ g-CO}_2/\text{kg fly ash}$ with 4.1% of CaO content used in their work.

A previous study showed a higher CO_2 uptake while using fly ash containing a higher CaO content (32.4 wt%).⁵³ Muriithi et al.⁴⁷ also achieved a higher maximum storage capacity of $62.35 \text{ g-CO}_2 \text{ kg}^{-1}$ fly ash due to the higher CaO (wt%) in their fly ash, using Ca-rich brine solution as a reaction medium. The reactor carbonation efficiency was also estimated in this study. This was estimated to be $5.6 \text{ kg-CO}_2 \text{ m}^{-3} \text{ h}^{-1}$.

Similar to the results presented before using water for carbonation, the trend in CaCO_3 content found in the carbonated fly ash using AMD as the reaction solvent was related to the calcium concentration after carbonation, as observed in Figure 8.

Figure 8 shows that there was a high increase in the calcium concentration in the initial 30 min and a high concentration of calcium after carbonation with AMD. The highest calcium concentration was around 845 mg L^{-1} , indicating that more calcium could be precipitated to CaCO_3 using AMD, hence the higher maximum CaCO_3 content of 6.68 wt% found in the carbonated fly ash. Again,

the relatively high concentration was due to the initial calcium concentration available in AMD, and possibly its acidity, including the change in process conditions for the AMD case. The decrease in the concentration of calcium after carbonation compared to the concentration obtained from the calcium extraction test (Figure 3) can be attributed to the precipitation of calcium to form CaCO_3 . The substantial increase in the calcium concentration initially observed in Figure 6 can be confirmed by the measured pH with time as shown in Figure 9. Figure 9 presents the pH results obtained after direct carbonation with AMD.

Based on the solution pH, the calcium extraction was responsible for the increase in the pH in the initial 30 min, as OH^- ions were released from fly ash, which resulted in a pH increase. However, after

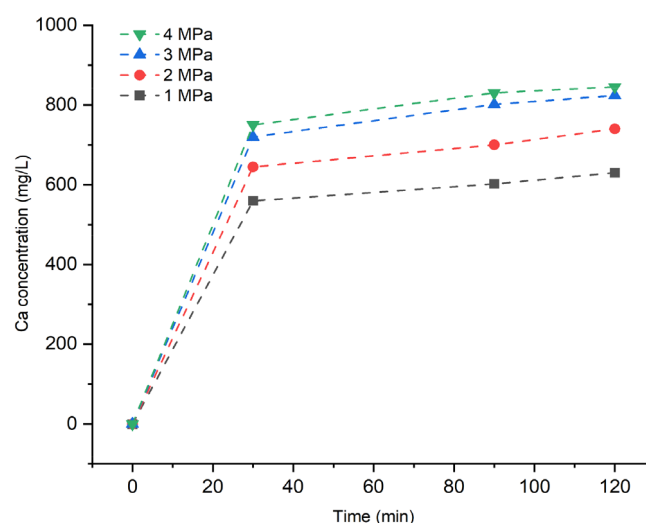


FIGURE 8 Calcium concentration after direct carbonation with acid mine drainage (AMD) (70°C , 0.2 g mL^{-1} , 400 rpm).

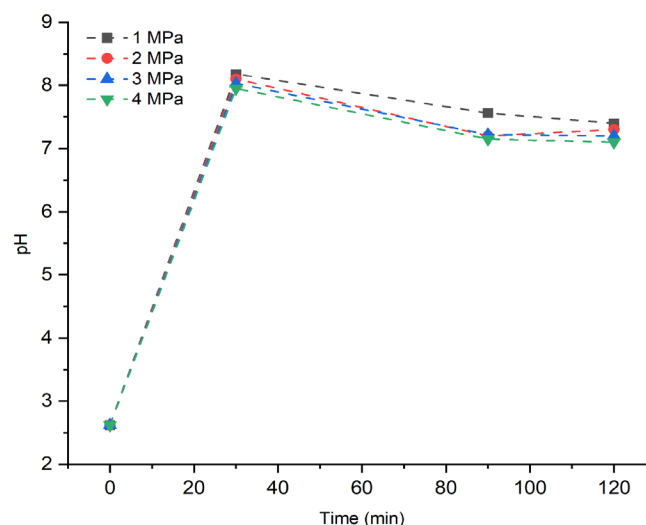


FIGURE 9 pH values over time after direct carbonation with acid mine drainage (AMD) (70°C , 0.2 g mL^{-1} , 400 rpm).

the initial period, more CO_2 dissolved in the alkaline solution, which was the preferred condition for CO_2 dissolution due to Le Chatelier's principle, thus causing the pH to decrease.

The concentrations of Fe, Mg, and Al in the raw AMD sample were 990, 330, and 244 mg L^{-1} and decreased to 6.5, 6.8, and 4.4 mg L^{-1} after carbonation, respectively. Hence, the percentage removal of most major elements was almost 100%. The precipitation of certain metals, therefore, including Fe^{2+} and Al^{2+} to form insoluble metal hydroxides from the AMD also caused an offset to the increase in solution pH. The decrease in pH was a result of CO_2 dissolution and the reaction of CO_2 with calcium, which resulted in the neutralization of the pH. In the equilibrium system, higher pressure resulted in a slightly lower pH of 7.1 at 4 MPa after 120 min due to the higher partial pressure of CO_2 , which possibly resulted in a slightly higher H^+ concentration. A similar trend in the pH was exhibited in a previous study conducted by Lee et al.³⁴

The results from Figure 10 show the XRD pattern for solid residues obtained after 30, 90, and 120 min of direct carbonation with AMD.

The XRD patterns showed the presence of major crystalline phases such as quartz and mullite contained in the raw fly ash. However, new phases in the form of gypsum (CaSO_4), hematite (Fe_2O_3) and ettringite ($\text{Ca}_6\text{Al}_2(\text{SO}_4)_3(\text{OH})_{12}26\text{H}_2\text{O}$), were also present. The presence of calcite again provided evidence for the conversion of calcium to CaCO_3 . The presence of gypsum (CaSO_4) suggests a drop in the original concentration of sulfate (SO_4^{2-}) from AMD after carbonation, which proves that calcium reacted with SO_4^{2-} , resulting in the formation of the new phase, namely gypsum.⁸

4.2.3 | Comparison between carbonation with water and AMD

The CaCO_3 content and calcium concentration after direct carbonation using AMD exhibited a similar trend to direct carbonation using pure water. However, a continued increase in the CaCO_3 content for carbonation with AMD was observed beyond the initial 30 min in all cases, due to higher calcium concentration available for CaCO_3 precipitation until a maximum CaCO_3 content of 6.68 wt% was reached. This was higher compared to the 4.33 wt% achieved for carbonation using pure water and was directly related to the higher access to calcium during carbonation with AMD. The additional calcium was from the initial concentration of 363 mg L^{-1} found in the AMD, which was 7.2 mg-Ca, considering the 200 mL volume of AMD used for the carbonation reaction.

Considering that the highest calcium concentrations after direct carbonation with water and AMD were 56 mg-Ca and 169 mg-Ca respectively, the difference between the concentrations means that AMD was responsible for the enhanced extraction of calcium during carbonation and the higher CaCO_3 content in the fly ash. Thus, the carbonation with the AMD solution proved beneficial to the carbonation performance of fly ash. It is worth noting that the change in process conditions for the AMD carbonation could have also contributed to the enhanced sequestration potential of fly ash.

The maximum CaCO_3 content found after 120 min at 4 MPa for carbonation with water and AMD was limited by competition between ionic species, as the presence of silica contributed to the formation of calcium silicate (CaSi) phases. The presence of gypsum

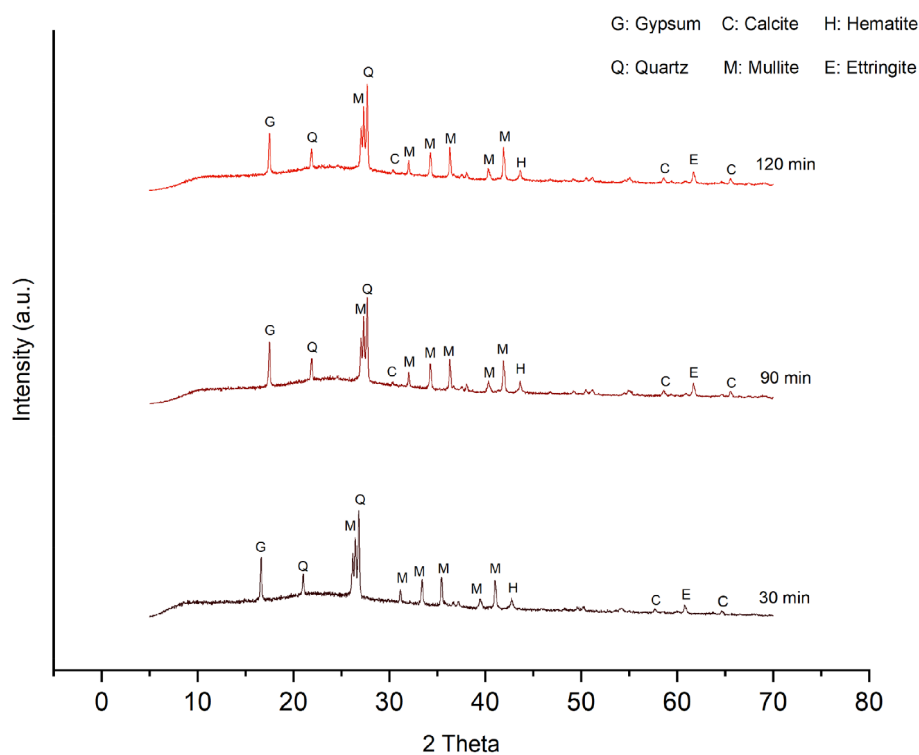


FIGURE 10 X-ray diffraction spectra for carbonated solid residues after direct carbonation with acid mine drainage (AMD) (70°C, 0.2 g mL⁻¹, 400 rpm).

(CaSO₄) during carbonation with AMD certainly suggested calcium was also responsible for the formation of other calcium-bearing phases besides CaCO₃.

4.3 | Energy balance: Power requirement

The carbonation process requires electrical energy for mechanical processes and heat energy. The electricity required by the current process was 18.9 kWh t⁻¹-CO₂. It was calculated from the difference between the total power requirement, which was 3987.2 kWh t⁻¹-CO₂, and the total heat requirement, which was 3968.3 kWh t⁻¹-CO₂. This equals 0.019 t-CO₂ emitted t⁻¹-CO₂ fixed, considering that the basic unit for coal-fired power generation in South Africa is 1.00 kg-CO₂ kWh⁻¹.⁵⁴ The CO₂ emission considering electricity is small enough, which indicates net CO₂ reduction can be achieved by the process, meaning the technology possesses significant potential in being applied at a commercial scale.

While the required amount of heat was 3968.3 kWh t⁻¹-CO₂, which accounts for almost the entire energy amount, this is not viable if waste heat cannot be utilized. Provided the temperature required for the process was only 70°C, waste heat generated from coal-fired power plants can be utilized. Generally, coal-fired power plants generate waste heat, therefore this fly ash mineral carbonation process must be operated on-site. Furthermore, the temperature in a cooling water circuit of a power station is at the level required by the current process, hence, the temperature for such a process can be maintained from cooling water.

The power and heat calculated here is proportional to the volume of the reactants. Therefore, the total power consumption can be directly scaled up to a reactor of any volume (pilot or commercial scale) for retrofitting. Future work must consider developing more comprehensive assessments of the energy requirement for MC processes.

5 | CONCLUSION

The benefit of utilizing AMD to provide additional Ca for enhanced carbonation performance was demonstrated in the present work. This was demonstrated by the higher CaCO₃ content in the carbonated fly ash of 6.68 wt% and the actual CO₂ uptake capacity of 29.4 g-CO₂ kg⁻¹ fly ash for AMD carbonation. The optimum conditions for this process were 120 min of carbonation at 4 MPa, using a S L⁻¹ ratio of 0.2 g mL⁻¹ and a stirring rate of 400 rpm. The study showed that 60% of the available calcium for such a process can be converted to CaCO₃ for the Ca-H₂O-CO₂ system. The electricity required by the process amounted to 18.9 kWh t⁻¹-CO₂, corresponding to 0.019 t-CO₂ emitted t⁻¹-CO₂ fixed in fly ash. The required amount of heat for the process was high, and the process is not viable if not operated on-site, producing CO₂ by means of the waste heat available. Furthermore, it is possible that the temperature for the mineral carbonation

process can be achieved by the water from a cooling water circuit of a power station.

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DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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