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Influencing Factors of the Mineral Carbonation Process of Iron Ore Mining Waste in Sequestering Atmospheric Carbon Dioxide

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Abstract: Mining waste may contain potential minerals that can act as essential feedstock for long-term carbon sequestration through a mineral carbonation process. This study attempts to identify the mineralogical and chemical composition of iron ore mining waste alongside the effects of particle size, temperature, and pH on carbonation efficiency. The samples were found to be alkaline in nature (pH of 6.9–7.5) and contained small-sized particles of clay and silt, thus indicating their suitability for mineral carbonation reactions. Samples were composed of important silicate minerals needed for the formation of carbonates such as wollastonite, anorthite, diopside, perovskite, johannsenite, and magnesium aluminum silicate, and the Fe-bearing mineral magnetite. The presence of Fe_2O_3 (39.6–62.9%) and CaO (7.2–15.2%) indicated the potential of the waste to sequester carbon dioxide because these oxides are important divalent cations for mineral carbonation. The use of small-sized mine-waste particles enables the enhancement of carbonation efficiency, i.e., particles of <38 μm showed a greater extent of Fe and Ca carbonation efficiency (between 1.6–6.7%) compared to particles of <63 μm (0.9–5.7%) and 75 μm (0.7–6.0%). Increasing the reaction temperature from 80 °C to 150–200 °C resulted in a higher Fe and Ca carbonation efficiency of some samples between 0.9–5.8% and 0.8–4.0%, respectively. The effect of increasing the pH from 8–12 was notably observed in Fe carbonation efficiency of between 0.7–5.9% (pH 12) compared to 0.6–3.3% (pH 8). Ca carbonation efficiency was moderately observed (0.7–5.5%) as with the increasing pH between 8–10. Therefore, it has been evidenced that mineralogical and chemical composition were of great importance for the mineral carbonation process, and that the effects of particle size, pH, and temperature of iron mining waste were influential in determining carbonation efficiency. Findings would be beneficial for sustaining the mining industry while taking into account the issue of waste production in tackling the global carbon emission concerns.

Keywords: carbon sequestration; carbon storage; iron ore; mineral carbonation; mining waste; sustainable production

1. Introduction

A sustainable approach in mitigating the impact of mining activities in relation to the emission of carbon dioxide (CO_2) needs to be implemented by countries throughout the world in response to the Sustainable Development Goals (SDG) set by the United Nations. This can be achieved through sustainable adaptation approaches as stipulated under the relevant SDG domains such as SDG 13 (climate action) and SDG 9 (industry, infrastructure, and innovation). In order to reduce the emissions of CO_2 into the atmosphere and to mitigate the environmental impact caused by mining activities such as from the waste produced, various mitigation technologies have been introduced as potential approaches to help tackle the problems.

Malaysia undertakes a variety of mining activities for materials such as iron, gold, tin, bauxite, copper, manganese, silver, and titanium (metallic minerals); gypsum ($CaSO_4 \cdot 2H_2O$), feldspar ($KAlSi_3O_8$-$NaAlSi_3O_8$-$CaAl_2Si_2O_8$), kaolin ($Al_2Si_2O_5(OH)_4$), limestone ($CaCO_3$), mica, sand and gravel, clays and silica sand (non-metallic minerals); and coal [1]. Regardless of the decline in production of the mined minerals, the mining sector remains one of the most important sectors in fulfilling demand and maintaining the Malaysian economy [2,3]. In order to sustain a constant supply of minerals to the global marketplace, Malaysia's government has continuously encouraged the exploration of minerals throughout the country. However, the excessive mining of minerals has not only provided Malaysia with economic growth but has also created some environmental problems mainly associated with the mining waste. The mining wastes can be defined as materials with no ore minerals, which are discarded from ore processing as they are lacking in economic importance and are valueless as compared to the ore, and are normally disposed in the form of stockpiles. This situation may bring harm to human health as well as potential environmental damage as the stockpiles that are exposed to weathering have a high potential to generate mine tailings which may contain high levels of toxic substances [4] For instance, sulfate and some trace elements commonly flow into tailing drainages thus initiating acid mine drainage problems which can be highly acidic [5,6]. As the acidic mine tailing flows into the water body, it will create further environmental problems.

On the other hand, global greenhouse gas emissions have risen drastically and have become one of the biggest worldwide concerns, as the effects are intense and uncontrollable [7,8]. Among the main sources of greenhouse gas emissions are nuclear power plants and the burning of fossil fuels and biomass, which are all energy- and industry- related concerns. Inevitably, the mining industry has also been associated with the emission of CO_2 [9]. As compared to the other greenhouse gases, CO_2 has become a serious concern as the percentage of this emission is the highest and CO_2 is more persistent in the atmosphere and has the potential to create massive environmental problems relating to global warming and climate change effects [10].

With regard to potential CO_2 mitigation approaches, various technologies have been adopted including those that use waste materials as feedstock for the process [9–11]. Some recent studies have been focusing on the use of mineral carbonation technology that incorporates the utilization of industrial waste materials [12–15]. In mineral carbonation technology, researchers have been emphasizing two types of carbonation processes which are direct and indirect mineral carbonation. The emphasis has been on the role of divalent cations such as Ca^{2+}, Mg^{2+}, and Fe^{2+} and as they are essential feedstocks for mineral carbonation processes [9,11]. Direct mineral carbonation will complete within one single reaction step, meanwhile indirect mineral carbonation processes require more than one activation step. Two major methods for mineral carbonation are direct gas–solid and aqueous mineral carbonation. Direct gas–solid is the most straightforward method for mineral carbonation. However, the conversion rate remained very slow despite the high temperatures and pressure used. Applying high pressure shows insignificant results, while applying heat treatment to feedstock can enhance the reaction rate but was deemed unworthwhile due to its high energy requirement [16,17]. Hence, several studies have suggested that gas–solid reaction rates are too slow for industrial-scale application [17]. Considering the quality of the results obtained, aqueous mineral carbonation is more preferable than a direct gas–solid method as it has higher reaction kinetics and better conditions for the carbonation process [12]. Aqueous mineral carbonation is the process of converting gaseous CO_2 into a final, stable, solid form, which usually involves two steps of reaction namely dissolution and precipitation. In direct aqueous mineral carbonation, sodium bicarbonate ($NaHCO_3$), sodium chloride ($NaCl$), distilled water, and minerals are utilized along with the addition of CO_2 via a diffusion process of surface and gas dispersion within an aqueous phase [13]. Using this method, solids are directly able to react with water and CO_2 [12]. In order to optimize the carbonation efficiency, variables such as pH, particle size, and temperature can be adjusted [18]. It has been evidenced that altering

these parameters by using smaller particle sizes of feedstock, increasing the operating temperature, and selecting appropriate pH levels for the dissolution and precipitation processes will make it more favorable to carbonation reaction [18–20]. The products that are expected to be produced from the direct mineral carbonation of iron ore mining waste are carbonate minerals such as Fe_2O_3, $CaCO_3$, and $MgCO_3$ [21], depending on the chemical and mineralogical contents of the minerals.

On the other hand, indirect mineral carbonation involves two crucial parts for the carbonation process to proceed, which are extraction and precipitation. Both of the steps occur separately and can be controlled in two different stages in the indirect mineral carbonation process [22]. Additives and solvents such as acids, bases, and ammonium salts were used where cations were leached from their respected minerals and reacted with carbon dioxide resulting in carbonate precipitation. A pH-swing method that combines the use of these reagents was developed in Japan [23] and later patented by Yogo et al. [24] to further optimize the carbonation process [25]. The use of various acid solutions and ammonium salts has been investigated for improving Ca and Mg ion dissolution, followed by the precipitation process in a multi-step method of carbonation [19,26,27]. The effects of varying experimental conditions have been studied in previous research. Among others, Azdarpour et al. [28] successfully obtained efficient Fe and Ca carbonation of red gypsum using less than 45 μm particle size with a constant carbon dioxide pressure of 20 bar, a temperature of 200 °C, and an ammonia solution concentration of 1M. Park and Fan [29] were experimenting on different solvents at ambient pressure and high temperature in serpentine dissolution of an Mg- and Fe-rich solution, prior to iron oxide and magnesium carbonate formation. In spite of using acids, bases, and salt solutions as solvents, a cheaper alternative was investigated by Han et al. [30] who proposed using water as the solvent for fly ash dissolution. Low reaction conditions were applied (i.e., ambient pressure and temperature) to reduce energy usage. Overall, it can be seen from most of the studies adopting direct or indirect carbonation that lower operating conditions can be applied with relatively lower energy requirements. This shows that the method was promising but the expensive chemicals used may be the limiting factor if industrial application is to be considered in the future.

There are still limited studies that integrate the mineralogical and chemical composition of iron ore mining waste and its potential for carbon sequestration. Furthermore, the optimum conditions for the occurrence of mineral carbonation cannot be assumed or determined with a certain fixed value, as it is found to be greatly dependent on the experimental conditions and material properties [31]. Waste materials from iron ore mining that are typically discarded from ore processing and dumped at the mining site may contain potential feedstock for mineral carbonation. Therefore, it is crucial for a laboratory-scale carbonation experiment to be conducted, so that this can be scaled-up in a pilot test and tested further in the industry. Thus, this study aims to provide information on the mineralogical and chemical composition of iron ore mining waste in order to identify the potential of the waste materials to undergo mineral carbonation processes, and to investigate the effects of varying parameters such as particle size, pH, and temperature on the carbonation efficiency. This would be useful for identifying suitable feedstock for mineral carbonation using waste material, while sustaining the mining industry by considering the issue of waste production in tackling the global carbon emission concerns.

2. Materials and Methods

2.1. Mine-Waste Sampling

Iron ore mining-waste samples were collected from an iron ore mining site operated by Jalur Galian Sdn. Bhd. located in the district of Jerantut, Pahang, with coordinates of N 434844.601, E 476902.551, as shown in Figure 1. The samples were collected from a series of tailing ponds and a dumping area within the mine occupying an area of 0.8 ha, depth 5.0 m and 3.9 ha, depth 7.0 m, respectively. The whole operation of the mine includes iron ore exploration in the main open pit mine and further processing for iron ore extraction

in the treatment facility. The collected samples were of mining-waste materials, which were produced during mining operations and ore processing in the form of waste rocks and sediments. The collected samples were ascertained from within the iron mine, i.e., the tailing ponds (tailing pond 1, tailing pond 2, and tailing pond 3), mine tailings, and waste dump. Triplicate samples were collected from each location and composite samples were obtained. Samples were kept in zip-locked bags, labelled accordingly, and brought to the laboratory for preparation and sample analysis. A total of 17.6 kg of sediments were collected from the tailing ponds and a total of 20.0 kg of waste rock samples were recovered from the waste dump. Samples were subjected to mineral and chemical analyses, prior to the carbonation experiment.

Sampling points:
1. Waste dump   2. Tailing pond 1   3. Tailing pond 2   4. Tailing pond 3   5. Mine tailings
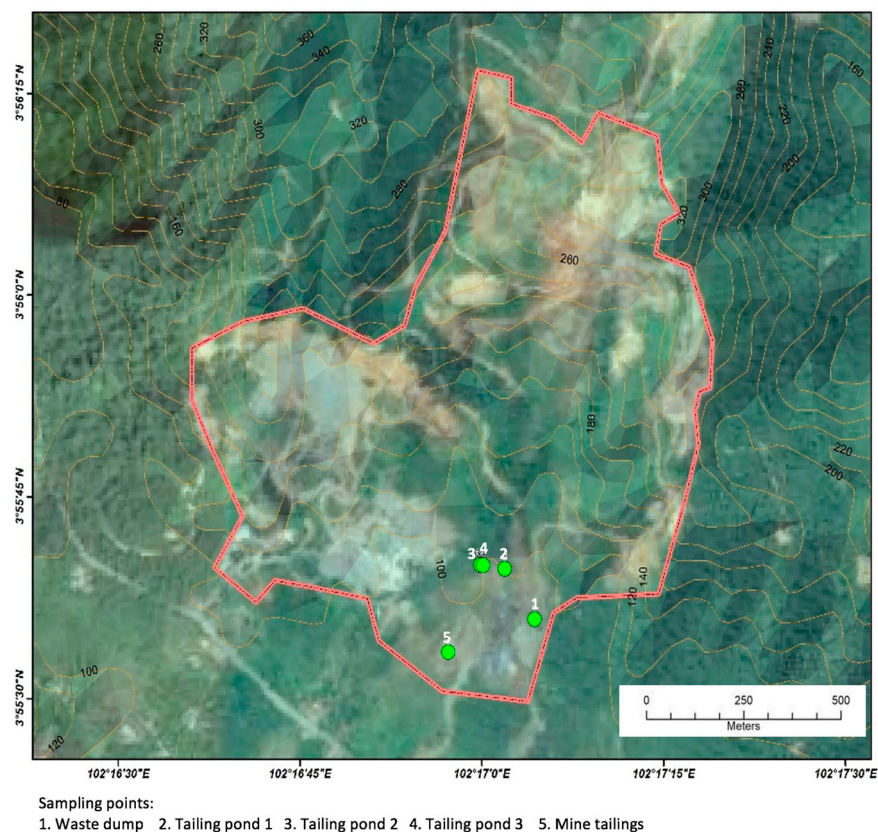
Figure 1. Location of the selected mine-waste sampling points.

2.2. pH and Particle Size Distribution Analysis

The pH of sediments and grounded waste rock samples were determined using a soil testing method adapted from the British standard (BS) 1377 [32]. Samples and distilled water were mixed with a ratio of 1:2.5, where 30 g of raw sample and 75 mL of distilled water were used. The pH of the solution was measured after 24 h using a pH meter. Particle size analysis was performed using dry sieving followed by wet sieving, i.e., the pipette method. The pipette method is considered to be more accurate than dry sieve analysis for sediment-type samples [33]. In dry sieving, particles were sieved until fractions of <2 mm were obtained, before undergoing pipette analysis to determine smaller-sized particle fractions. The percentages of sand, silt, and clay in the sediment samples were determined using the pipette method as described in Teh and Talib [34]. This method involves several steps in which the distribution of the sand (50 μm–2 mm), silt (2 μm–50 μm), and clay (less than 2 μm) fractions in the raw iron ore waste sample were obtained. After the initial steps of removing organic substance, the raw waste sample was pipetted at specific times and depths of the suspension in a 1000 mL measuring cylinder. The pipetted suspension was then condensed and oven-dried, and finally weighed to determine the pipetted fraction-

mass ratio. This analysis was done before the actual carbonation experiments were carried out as an early indication that the samples are mostly clays, which are potentially useful in mineral carbonation. A higher percentage of smaller particle size fractions (i.e., silt and clay) indicates that the waste has potential in sequestering carbon dioxide due to the larger surface area exposed for the occurrence of reactions [19]. While the presence of a large amount of large-sized particles (sand) indicated that the samples needed to undergo a pre-preparation process to produce a smaller particle size in order to optimize the mineral carbonation efficiency.

2.3. Mineralogical and Chemical Analysis

The collected samples were analyzed through X-ray diffraction (XRD) and scanning electron microscopy (SEM) with energy dispersive X-ray (EDX) analysis to determine the mineralogical and chemical composition. The XRD analysis was used to identify the major and minor mineral phases of the samples. An XRD instrument (model Bruker-AXS D8 Advanced, USA), counting for 0.2 s per step of scattering angle ranging between 5 and 80°, at a rate of 1° per min (0.02° step size) and with a detection limit of 1–2%, was used. Data from the XRD analysis were refined using Rietveld program Topas 4.2 (Bruker AXS). The Rietveld refinement method presents the widths and geometrical shapes of the diffraction peaks to produce a diffraction pattern for each mineral present in the sample. Through SEM analysis, the morphological structures of the mining waste were observed. The SEM instrument (model Hitachi SU3500) was used with a detection limit of 0.1%. EDX spectrum with peaks corresponding to each element in the sample were identified and data are presented in the form of major oxide percentage.

2.4. Mineral Carbonation Experiment

A carbonation experiment was undertaken to evaluate several operating variables that can influence the mineral carbonation process. The experiment was conducted using a customized 250 mL closed stainless steel reactor (Figure 2). For this experiment, the measured variables were particle size, temperature, and pH, while constant parameters were pressure; which was kept constant at 1 bar (ambient pressure), and stirrer rate; which was kept constant at 300 rpm (Table 1). For mineral carbonation to take place, CO_2 gas was supplied to the reactor during the experiment through a carbon dioxide supply tube. The reaction was allowed to take place for about one hour for each experiment. In the experiment, a sample with a weight of 10 g was mixed with 2.67 g of 0.64M $NaHCO_3$ and 2.92 g of 1M NaCl, which were dissolved with 50 mL of distilled water each. The mixtures were then mixed and poured into a Teflon tube to be placed into the reactor. Four different pH levels were applied for all the samples: the actual pH of the sample and pH 8, 10, and 12 (adjusted pH). In order to increase the pH of the sample, an NaOH solution of 1M was added into the solution. For the effect of particle size distribution, three different particle sizes were utilized which were <38, <63, and <75 μm. Meanwhile, for temperature as the adjusted variable, three different temperatures were used which were at 80 °C, 150 °C, and 200 °C. Yan et al. [20] have successfully achieved the highest carbonation efficiency using an operating temperature and pressure of 150 °C and 40 bar, and Azdarpour et al. [19] have found that carbonation conversion efficiency dropped once the temperature reached 200 °C in their study, whilst a temperature of 80 °C was included in this mineral carbonation experiment to investigate whether a lower operating temperature can achieve satisfying results comparable with the higher energy conditions. At the end of experiment, the carbonation product was drained, filtered, and dried for further analysis using a thermo gravimetric analyzer. The variability of the experimental parameters was based on the experience from our trial experiment [33]. The variables and constant parameters applied throughout the carbonation experiment were designed to identify the conditions appropriate for the carbonation process to occur more effectively, as measured by the carbonation efficiency.
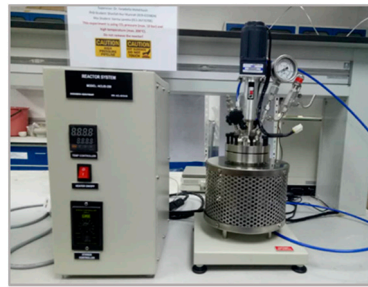
Figure 2. 250 mL stainless steel reactor used for the mineral carbonation experiment.

Table 1. Operating variables for the mineral carbonation experiment of iron mine waste.

	Type of Variables								
	pH				Particle Size (μm)			Temperature (°C)		
Operating variables	Actual pH	8	10	12	<38	<63	<75	80	150	200
Constant parameters	Particle size = 63 μm Temperature = 80 °C				pH = 8.5 Temperature = 80 °C			pH = 8.5 Particle size = 63 μm		

2.5. Thermogravimetric Analysis

For identifying the carbonation efficiency of the mining-waste samples, thermogravimetric analysis was performed. The purpose of this analysis was to determine the percentage weight loss of the samples after undergoing mineral carbonation. The concept of this analysis was to introduce the samples to high temperature conditions ranging from 0 °C to 950 °C. Loss of sample weight within the range of 200–450 °C and from 600–850 °C indicate the presence of Fe_2CO_3 and $CaCO_3$, respectively [33]. The carbonation efficiency was calculated using the equations below.

Fe/Ca mass in Fe_2CO_3/$CaCO_3$:

$$\% \text{ weight loss} \times \frac{\text{molecular weight of Fe or Ca}}{\text{molecular weight of CO}_2} \times \text{mass of solid residue} \tag{1}$$

Carbonation efficiency (%):

$$\frac{\text{Fe/Ca mass in FeCO}_3/\text{CaCO}_3}{\text{Fe/Ca total mass in feeding material}} \times 100 \tag{2}$$

2.6. Statistical Analysis

Statistical analysis was performed using the SPSS statistical software package. One-way ANOVA, an independent sample t-test, and a Tukey post-hoc test were applied to analyze if there is any significant difference between each parameter and between the carbonation efficiency of Fe and Ca in the samples.

3. Results and Discussion

3.1. pH and Particle Size Distribution

The pHs of samples showed that they were in alkaline state, with the pH ranging from 6.9 to 7.4 (Figure 3). Specifically, the pHs were 6.9 (tailing pond 2), 7.2 (tailing pond 1), 7.2 (tailing pond 3), 7.4 (mine tailings), and 7.5 (waste dump). It has been known that pH holds a very important role in affecting the rate of carbonation reaction. Thus, the actual pHs of the samples were determined prior to the carbonation experiment first. The pH was a set variable in the carbonation experiment that followed in order to observe the effect of pH variation on the carbonation reaction. Certain pH conditions can help increase the carbonation efficiency of the samples, e.g., alkaline conditions above pH 8 [35,36].
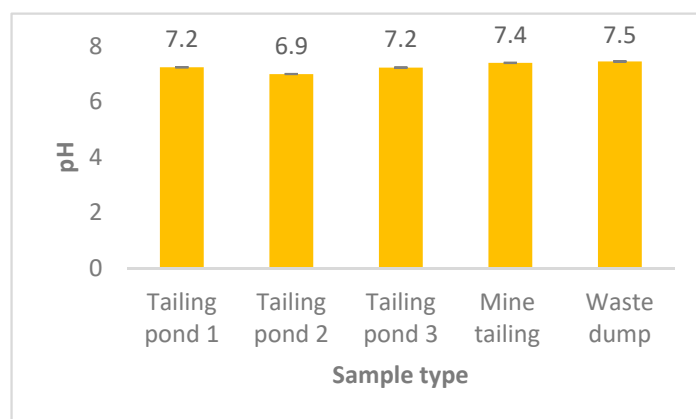
Figure 3. pH of mine-waste samples.

Particle size distribution analysis revealed that the texture of iron ore mining waste was in the form of sand for tailing pond 1, clay loam for tailing pond 2, sandy loam for tailing pond 3, and loamy sand for mine tailings (Table 2). A sample from the waste dump was classified as waste rocks and was not subjected to particle size distribution analysis using the pipette method. The smaller the particle size, the more efficient the carbonation process as greater surface sites are available for the carbonates to form, hence this has been regarded as one of the important mechanisms in determining mineral carbonation efficiency. It has been known that small-sized particles (e.g., clay) are essential in accelerating carbonation reactions [37]. Samples with a higher amount of clay were from tailing pond 2 with a clay content of 30.36%, followed by tailing pond 3 (11.00%), mine tailings (6.80%), and tailing pond 1 (0.23%). Silt content was found in a greater proportion in tailing pond 2 (43.98%), followed by tailing pond 3 (22.27%), mine tailings (11.37%), and tailing pond 1 (2.97%). Similar to clay, silt components are also important for mineral carbonation due to the presence of the small-sized particles. On the other hand, the sample with the most sand content was tailing pond 1 (96.80%), followed by mine tailings, tailing pond 3, and tailing pond 2, with a sand percentage of 81.84 %, 66.74 %, and 25.66 %, respectively. Literature suggested that smaller particle sizes (i.e., 38 μm and 40 μm) were found to be the best option for increasing the carbonation efficiency [17,19]. As the samples were found to contain considerable amounts of particle size fractions that can potentially affect the carbonation process, particles of different sizes were included in the subsequent experiments in order to compare the rate of the carbonation process.

Table 2. Particle size distribution of the samples.

Sample	Particle Size Distribution (%)			Soil Texture Class
	Clay	Silt	Sand	(USDA)
	(<2 μm)	(2–50 μm)	(>50 μm)	
Tailing pond 1	0.23	2.97	96.80	Sand
Tailing pond 2	30.36	43.98	25.66	Clay loam
Tailing pond 3	11.00	22.27	66.74	Sandy loam
Mine tailing	6.80	11.37	81.84	Loamy sand

3.2. Mineralogical and Chemical Composition of Mine Waste

The mineralogical composition of the samples was obtained from the XRD analysis. The XRD diffractograms of samples are shown in Figure 4. The mineral phase majorly found in all of the samples was quartz (SiO_2). Most importantly, the samples must contain potential minerals that are required for mineral carbonation such as Ca-, Fe-, and Mg-bearing minerals. Silicate minerals potentially useful for mineral carbonation were found in the form of Ca- and Mg-silicates such as anorthite, $CaAl_2Si_2O_8$; wollastonite, $CaSiO_3$;

diopside, $(Ca(Mg,Al)(Si, Al)_2O_6)$; johannsenite, $Ca_4Mn_4Si_8O_{24}$; and magnesium aluminum silicate, $MgO \cdot Al_2O_3 \cdot SiO_2$ (Table 3). In the case of iron-bearing minerals, iron oxides such as magnetite, Fe_3O_4, can be beneficially utilized for mineral carbonation.

(a)

(b)

(c)

(d)

(e)
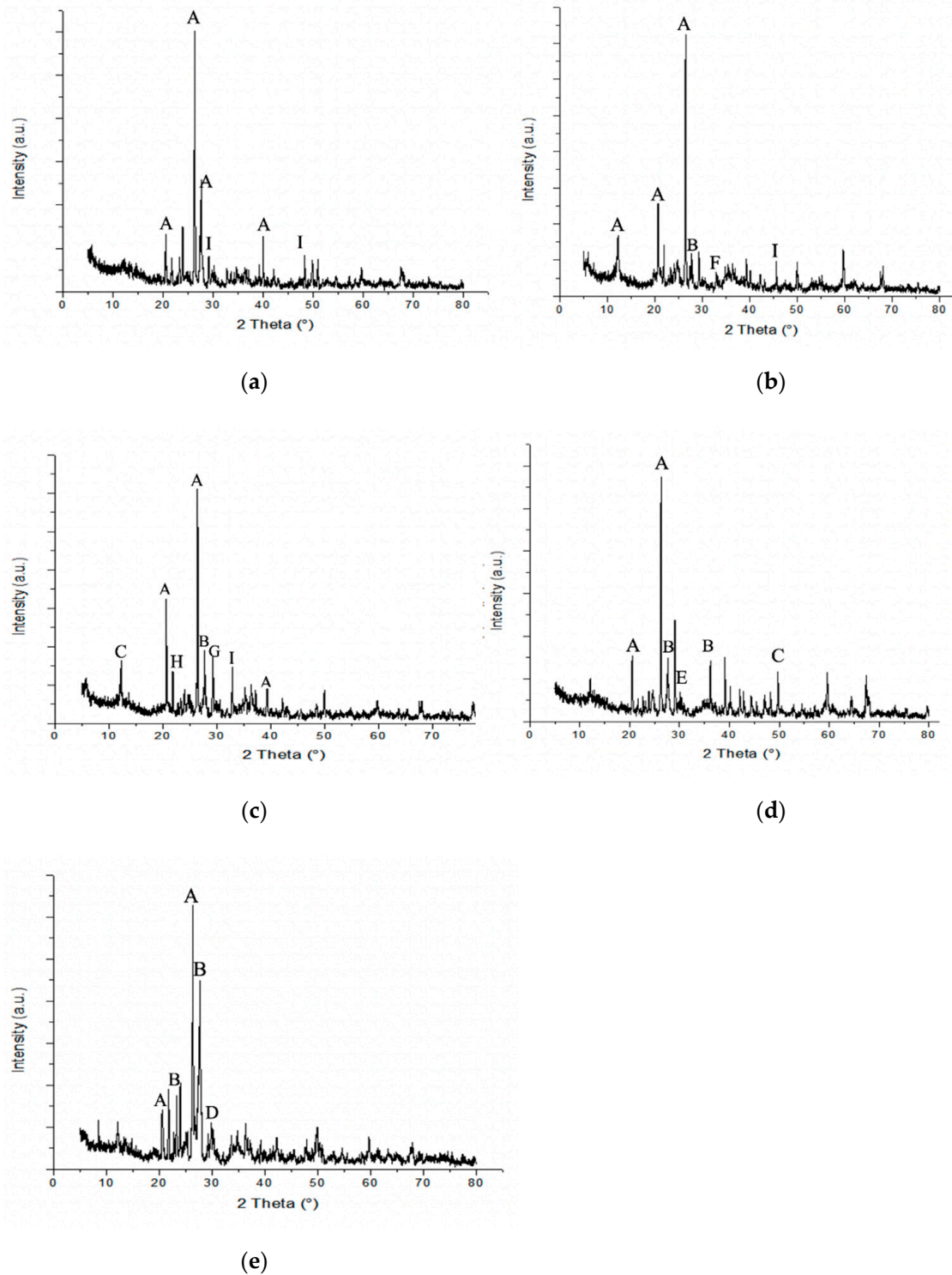
Figure 4. XRD diffractograms of samples from five different sampling sites: (**a**) tailing pond 1; (**b**) tailing pond 2; (**c**) tailing pond 3; (**d**) mine tailings; and (**e**) waste dump. The labelled alphabet indicates the presence of significant minerals in the mining wastes: quartz (A), anorthite (B), titanium silicon (C), wollastonite (D), diopside (E), perovskite (F), johannsenite (G), magnesium aluminum silicate (H), and magnetite (I).

Table 3. Summary of the mineral composition of the mine-waste samples. '$\sqrt{}$' label indicates the presence of significant minerals in the sample.

No.	Minerals	Sample				
		Tailing Pond 1	Tailing Pond 2	Tailing Pond 3	Mine Tailing	Waste Dump
1.	Quartz, SiO_2	$\sqrt{}$	$\sqrt{}$	$\sqrt{}$	$\sqrt{}$	$\sqrt{}$
2.	Anorthite, $CaAl_2Si_2O_8$		$\sqrt{}$	$\sqrt{}$	$\sqrt{}$	$\sqrt{}$
3.	Titanium Silicon, $TiSi_2$			$\sqrt{}$	$\sqrt{}$	
4.	Wollastonite, $CaSiO_3$					$\sqrt{}$
5.	Diopside, $(Ca(Mg,Al)(Si, Al)_2O_6)$				$\sqrt{}$	
6.	Perovskite, $Ca(TiO_3)$		$\sqrt{}$			
7.	Johannsenite, $Ca_4Mn_4Si_8O_{24}$			$\sqrt{}$		
8.	Magnesium Aluminum Silicate, $MgO \cdot Al_2O_3 \cdot SiO_2$			$\sqrt{}$		
9.	Magnetite, Fe_3O_4	$\sqrt{}$		$\sqrt{}$		

Minerals such as wollastonite and magnetite are the most remarkable minerals that are commonly used in mineral carbonation industries, and these minerals were found in the waste dump, tailing pond 1, and tailing pond 3. However, in the iron ore mining-waste samples, Mg-bearing minerals such as diopside and magnesium aluminum silicate were only minorly present compared to Ca- and Fe-bearing minerals (Table 3). Other than quartz, the major minerals found in tailing pond 1 were magnetite, while for tailing pond 2 anorthite and perovskite were present. On the other hand, for tailing pond 3, the major minerals present were anorthite, titanium silicon, johannsenite, magnesium aluminum silicate, and magnetite. The mineral composition of mine tailings comprised of anorthite, titanium silicon, and diopside. While the major minerals found in the waste dump were anorthite and wollastonite. As observed from the XRD results, all of the samples contained the Ca-, Fe-, and Mg-bearing minerals that are needed for the mineral carbonation process. Generally, the more dominant minerals found in the sediment samples were quartz followed by anorthite. The presence of anorthite in the samples was depicted by their flake-like anorthite crystal shapes as identified from the SEM micrographs analysis (Figure 5). Clearly, the presence of Ca- and Mg-silicate minerals is essential for mineral carbonation and can be useful as a supplementary cementitious material [33].

From the EDX analysis, the chemical composition of the iron ore mining waste has been identified in percentage of major oxides such as SiO_2, Fe_2O_3, CaO, MgO, K_2O, TiO_2, and SO_3 (Table 4). As mentioned earlier, in order to perform mineral carbonation experiments using the waste materials, samples must contain particular oxides such as CaO, MgO, or Fe_2O_3, which serve as the essential feedstocks for the process. As anticipated, the most abundant oxide in the samples was Fe_2O_3 (mainly of magnetite) which was dominating in all of the samples with a percentage of 62.95% (tailing pond 2), 61.68% (tailing pond 1), 61.00% (tailing pond 3), 57.79% (mine tailings), and 39.58% (waste dump). Silica, SiO_2 was found in the range of 15.93–37.97%. CaO was present at 7.19–15.24%; the highest was found in the mine tailings. The proportion of CaO in the samples was attributable to the presence of minerals such as anorthite, wollastonite, diopside, perovskite, and johannsenite as identified from the XRD analysis. However, a lower amount of MgO was noticeable of between 1.32–6.75%, which was associated with the minerals diopside and magnesium aluminum silicate. Therefore, the findings suggest that the sediments of the tailing ponds and mine tailings, and the waste rocks of the waste dump of iron ore mining, have the potential to undergo mineral carbonation as suitable feedstock for the process due to their mineralogical and chemical composition.

(**a**)

(**b**)

(**c**)

(**d**)

(**e**)
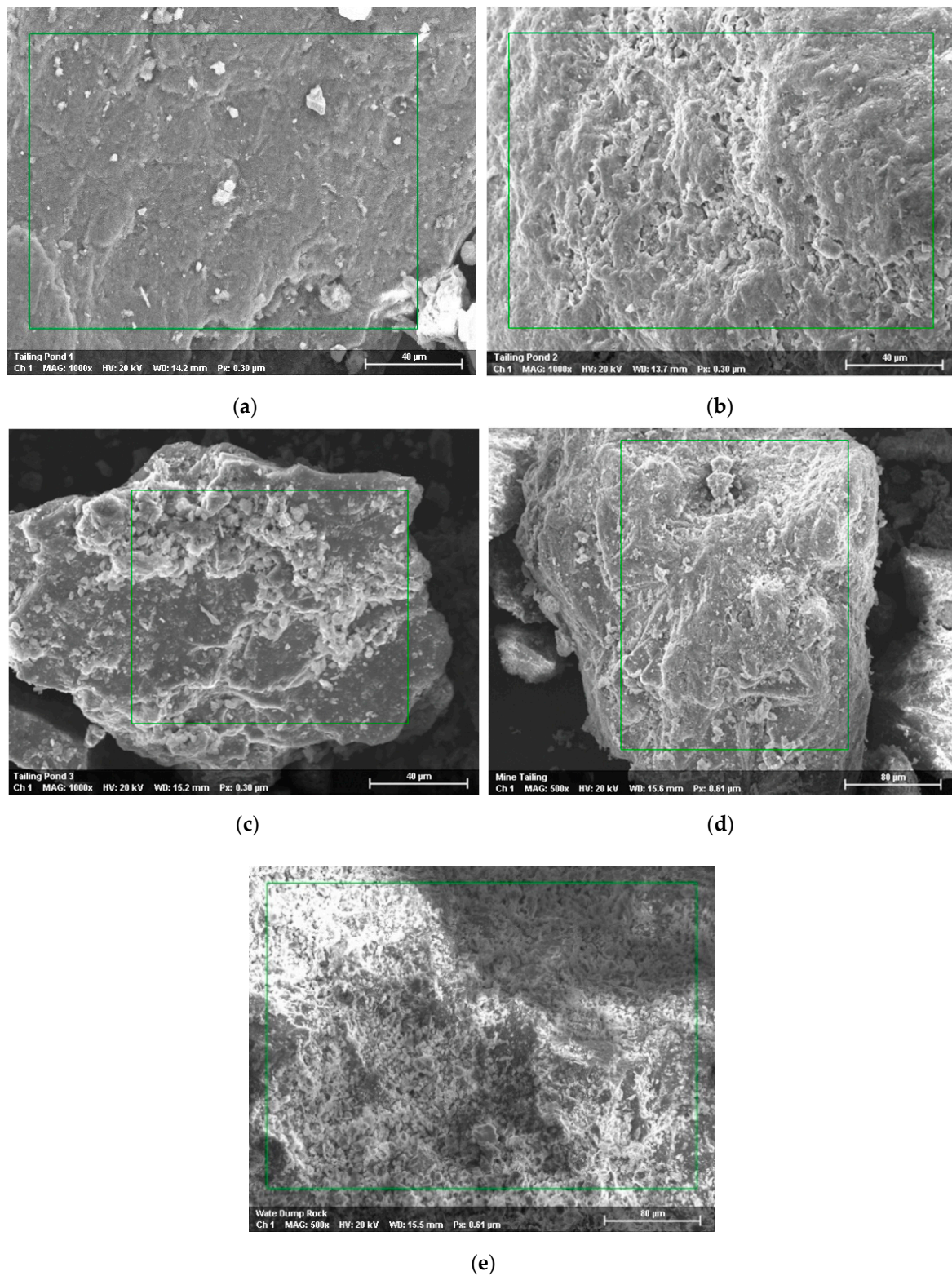
Figure 5. Scanning electron micrographs of samples (**a**) tailing pond 1; (**b**) tailing pond 2; (**c**) tailing pond 3; (**d**) mine tailing; and (**e**) waste dump.

3.3. Factors Influencing the Mineral Carbonation Process of Iron Mining Waste

3.3.1. Effect of Particle Size

When examining the effect of the experimental variables, carbonation efficiency was calculated for Fe and Ca ions which are the more important cations responsible for mineral carbonation in the iron mine-waste samples, as identified earlier. The effect of particle size on carbonation efficiency was investigated by applying variable particle sizes of 38 μm, 63 μm, and 75 μm with a constant pH of 8.5 and temperature of 80 °C. Results are presented as Ca and Fe carbonation efficiencies as these are the important divalent cations in the samples that govern mineral carbonation. However, Ca-bearing minerals have been the more dominant factor for carbonation as opposed to Fe-bearing minerals. As observed in Figure 4, reduction in particle size has a direct effect on the Ca carbonation efficiency of the samples where the overall efficiency decreased with the increase of particle size. This is because a reduction in particle size, i.e., the smaller/finer particles, enhances the release of ions contained in the samples [37,38]. Grinding and crushing processes will destroy the crystalline structure thus improving the reaction rate as the surface area for the carbonation reaction to occur during the dissolution process increases when particle size is reduced [19,20,22].

Table 4. Chemical composition of the mine-waste samples given in (% wt.) of major oxides.

| Oxide Elements	Percentage of Oxides (%)				
	Tailing Pond 1	Tailing Pond 2	Tailing Pond 3	Mine Tailing	Waste Dump
SiO_2	15.93	21.31	20.47	14.05	34.97
Fe_2O_3 *	61.68	62.95	61.05	57.79	39.58
CaO *	9.57	7.19	11.52	15.24	10.16
MgO *	3.50	2.72	1.32	6.75	2.10
K_2O	0.97	1.47	1.49	1.06	5.31
TiO_2	0.86	1.45	0.97	0.77	1.79
SO_3	3.95	1.31	1.09	2.96	1.43

Loss on ignition (LOI) for sample is 3.5%, 3.3%, 3.0%, 1.8%, and 4.1% for tailing pond 1, tailing pond 2, tailing pond 3, mine tailing, and waste dump, respectively. * Important divalent cations for mineral carbonation.

This reduction pattern of carbonation efficiency, with regard to the effect of particle size, can be more clearly observed for the Ca carbonation efficiency compared to the Fe carbonation efficiency (Figure 6a,b). However, the Fe carbonation efficiencies were comparably higher than the Ca carbonation efficiencies, i.e., ranging between 0.71–6.66% and 0.71–3.88%, respectively. This had been anticipated because these samples are dominated by iron oxide minerals as opposed to Ca- and/or Mg-silicate minerals as noticed earlier. At a particle size of <38 μm and <63 μm, the Ca carbonation efficiencies of the tailing pond samples were noticeably higher than samples from the mine tailings and the waste dump (Figure 6a). On the other hand, for Fe carbonation efficiency, it can be seen that the carbonation efficiency was relatively high in tailing pond 2 compared to other samples. This can be reflected by the particle size distribution of the sample that is categorized as having a high amount of clay and silt (clay loam). Furthermore, the chemical composition analysis also confirmed that this sample contains a higher amount of iron oxides compared to other samples, thus resulting in the highest Fe carbonation efficiency. Regardless of Fe or Ca carbonation efficiency, and comparing between particle size fractions, generally high carbonation efficiencies were observed at particle size of <38 μm. This supports the hypothesis that small-sized particles may enhance the mineral carbonation reactions.

3.3.2. Effect of Temperature

The effect of temperature on carbonation efficiency was investigated by applying variable reaction temperatures of 80 °C, 150 °C, and 200 °C, with constant particle size of 63 μm and a pH of 8.5. As with the effect of particle size, Fe carbonation efficiencies were noticeably higher than the Ca carbonation efficiencies (Figure 7a,b). The Fe carbonation

efficiencies were ranging between 0.89–5.82% and 0.84–3.99%, respectively. However, the effects of varying the reaction temperatures were not directly seen. Despite this, for Ca carbonation efficiency, the effect of increasing the reaction temperature can be observed for the tailing pond samples (tailing pond 1, tailing pond 2, and tailing pond 3), i.e., higher carbonation efficiency occurs when the temperature was increased from 80 °C to 200 °C. For these tailing pond samples, the highest efficiencies were observed when the temperature was increased to between 150–200 °C. In contrast, the mine tailings and waste dump samples showed decreasing carbonation efficiency when the temperature was increased. It was also shown that the Fe carbonation efficiencies were noticeably higher for tailing pond 2 samples compared to other samples (Figure 7a). The effect of increasing temperature on Fe carbonation efficiency was observed for tailing pond 2 and tailing pond 3 samples when the temperature was increased from 80 °C to 200 °C. It can be seen that with a sufficient amount of iron oxides, the Fe carbonation efficiencies of these samples were increased to their highest rate at temperature of between 150–200 °C. Other samples showed a decreasing pattern of Fe carbonation efficiency as the temperature increased. This may happen as the increase in reaction temperature assists in the extraction of Fe and Ca from the samples as the reaction kinetic increases. As stated by Azdarpour et al. [19], there is always an optimum temperature in a direct aqueous carbonation which results in maximum carbonation efficiency, and this is influenced by the mineralogical and chemical composition of the samples. Despite the low carbonation efficiencies obtained, carbonate product can be successfully precipitated at an ambient pressure as applied in this experiment, although it is generally formed at elevated temperature [39]. As noted earlier, the carbonation efficiency of tailing pond 2 samples remains the highest due to the mineralogical and chemical composition of the sample.

(a)

(b)
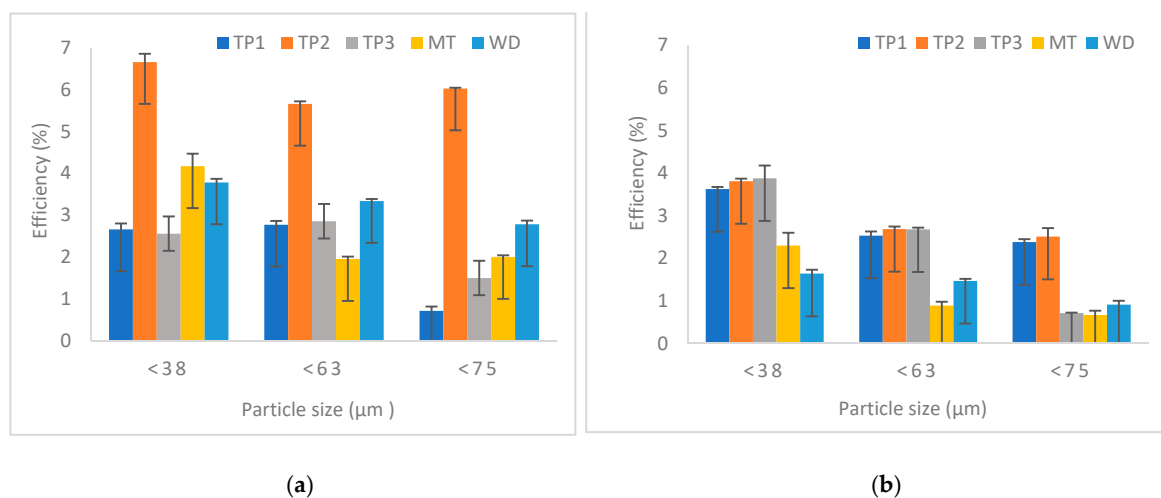
Figure 6. (**a**) Fe; (**b**) Ca carbonation efficiency in relation to particle size. Standard deviations are shown in error bars.

3.3.3. Effect of pH

The effect of pH on carbonation efficiency was investigated by applying variable reaction pHs, ranging from the actual pH of the samples to pH 8, 10, and 12, with a constant particle size of 63 μm and temperature of 80 °C. Figure 8a,b shows the Fe and Ca carbonation efficiencies of the iron ore mining waste, respectively. From the results of pH analysis, the iron ore mining wastes were considered as alkaline-based materials. Different types of samples may require different pH conditions to have an optimum reaction rate. As stated by Hassan et al. [38], the most favorable reaction pH for this type of carbonation of a sample would be in alkaline pH ranging between 8 to 12. As observed from Figure 8a, the effect of increasing reaction pH can be seen on the Fe carbonation efficiency. The Fe carbonation efficiencies were markedly increased as pH was increased from around pH 7 to pH 12 in samples of tailing pond 1, tailing pond 2, and tailing pond 3. Meanwhile, the

Fe carbonation efficiencies of the mine tailings were also increased when pH was increased up to pH 10. Similarly, for Ca carbonation efficiency (Figure 8b), the increasing pattern can be seen for the mine tailing samples as pH increases to pH 10. On the other hand, the Ca carbonation efficiency of the other samples showed a decreasing trend as pH was increased from 10 to 12. Generally, the Ca carbonation moderately occurred at a pH of between 8 to 10. Therefore, it can be seen that under this alkaline pH range studied, the carbonation reaction does occur variably. This is because mafic, ultramafic, and sedimentary rocks prefer a more alkaline condition for a better carbonation reaction, as increases in pH promote a higher dissolution rate of carbonates. This is particularly pronounced for Fe-bearing minerals because Fe precipitation greatly occurs in the range of circum-neutral state to alkaline range. In the presence of iron oxides, the synthesis for CO_2 capture involves the reaction between iron oxides and CO_2 to produce iron carbonates, as given in the reaction below.
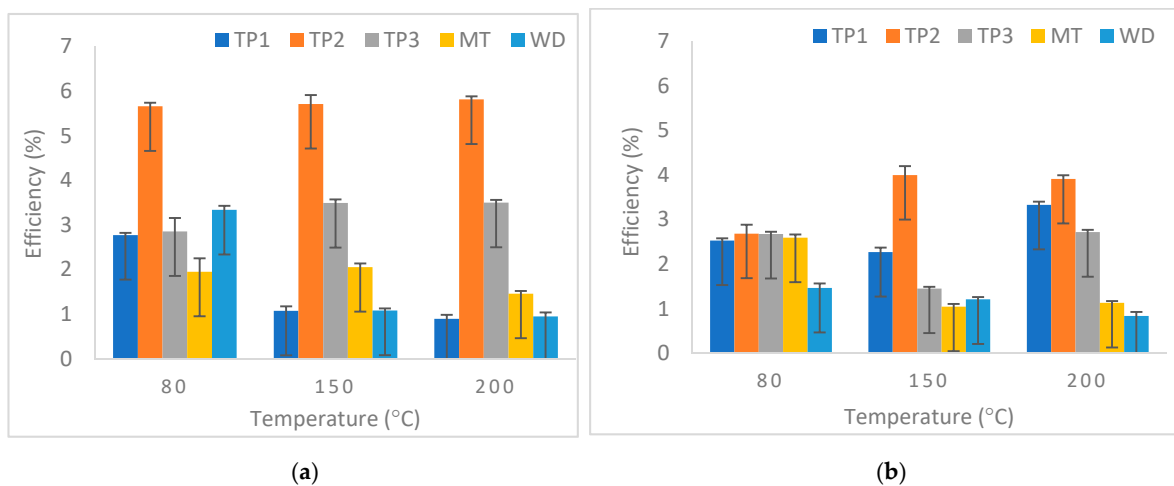
$$Fe_2O_3(s) + Fe(s) + 3CO_2(g) \rightarrow 3FeCO_3(s) \tag{3}$$

Figure 7. (**a**) Fe; (**b**) Ca carbonation efficiency in relation to temperature. Standard deviations are shown in error bars.
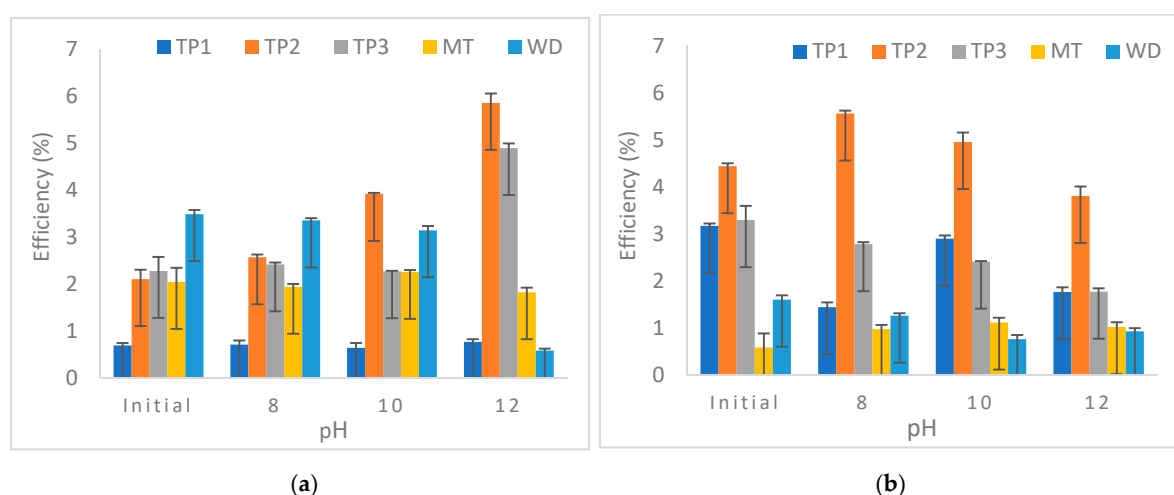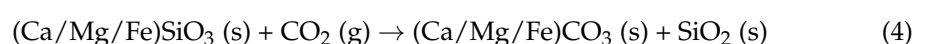

Figure 8. (**a**) Fe; (**b**) Ca carbonation efficiency in relation to pH conditions. Standard deviations are shown in error bars.

Silicate minerals that contain Ca-, Mg-, and Fe-bearing minerals with the presence of CO_2 will form carbonate minerals as shown in the following reaction [40,41]:

$$(Ca/Mg/Fe)SiO_3 (s) + CO_2 (g) \rightarrow (Ca/Mg/Fe)CO_3 (s) + SiO_2 (s) \tag{4}$$

In order to determine if there is any significant difference between each variable, a one-way ANOVA statistical test was used. Post-hoc analysis was conducted if there were any significant differences in the selected variables. The statistical significance of carbonation efficiency as influenced by different variations of particle size, pH, and temperature was evaluated. Results of the one-way ANOVA at a 95 % significance level showed that there is a statistically significant difference in the carbonation efficiency between different particle sizes as determined by the one-way ANOVA: $F_{(2, 42)} = 5.474$, $p = 0.008$. A Tukey post-hoc test revealed that the carbonation efficiency using a particle size of <38 μm (3.83 ± 1.46) was statistically significantly higher ($p < 0.05$) than a particle size of <75 μm (2.20 ± 1.34). However, there was no statistically significant difference at the $p > 0.05$ in the mean carbonation efficiency with different temperatures: $F_{(2, 42)} = 1.112$, $p = 0.339$. Similarly, the ANOVA indicated that there was no statistically significant difference at the $p > 0.05$ in the mean carbonation efficiency with different pH: $F_{(2, 42)} = 1.153$, $p = 0.326$. Despite statistically no significant differences in the variation of temperature and pH used in this study, these variables do have an influence on the mineral carbonation. On the other hand, there is a statistically significant difference between the samples in affecting the carbonation efficiency. There is a statistically significant difference in the carbonation efficiency between samples by one-way ANOVA: $F_{(2, 40)} = 3.354$, $p = 0.019$. A Tukey post-hoc test revealed that the mean carbonation efficiency for tailing pond 2 (4.11 ± 0.27) was significantly higher (p-value <0.05) than tailing pond 1 (2.60 ± 0.24), tailing pond 3 (2.41 ± 0.32), mine tailings (2.24 ± 0.76), and the waste dump (1.18 ± 0.11). The carbonation efficiency of the tailing pond 2 sample was mostly the highest among the other samples, as reflected by its high amount of iron oxides and the presence of a Ca-silicate mineral mainly composed of anorthite, alongside the fine particle size distribution of clay and silt. Overall, the Fe and Ca carbonation efficiencies found in this study were comparable to some previous studies on the mineral carbonation of a red gypsum, e.g., 1.25–12.50% at a particle size of <45 μm and temperature of up to 400 °C [19,40]. In fact, the various types of feedstocks may behave differently based on their characteristics (i.e., chemical, morphology, and elemental composition), therefore the preferable condition for the mineral carbonation to occur cannot be assumed or determined with a certain fixed value as it is found to be greatly dependent on the experimental conditions and material properties [31,41].

4. Conclusions

This study has explored the potential of iron ore mining waste to be used as suitable feedstock for the mineral carbonation process. Mineralogical and chemical component analysis were performed to evaluate the potential of the samples to undergo a mineral carbonation process. It has been found that the iron ore mining waste can be potentially utilized for mineral carbonation due to the alkaline nature of the pH (i.e., pH 6.9 to 7.5). Mineralogical components of the samples helped in the formation of carbonates due to the presence of Ca- and Mg-silicate minerals such as anorthite, wollastonite, diopside, and johannsenite, alongside Fe oxide such as magnetite. The chemical composition of the iron ore mining waste confirms the presence of a high amount of Fe_2O_3 (ranging between 39.6–62.9%), and some amount of CaO (7.2–15.2%) and MgO (1.3–8.8%) which are important divalent cations that can influence the carbonation efficiency. In order to evaluate the process efficiency, operating variables such as particle size, reaction temperature, and pH of the samples were made variable in the mineral carbonation experiment. Small-sized particles of <38 μm are important to enhance the efficiency of the process, resulting in the range of 1.6–6.7% (Fe and Ca) carbonation efficiency compared to particles of <63 μm and 75 μm (efficiency between 0.7–6.0%). In terms of temperature variation, a higher Fe and Ca carbonation efficiency (0.9–5.8% and 0.8–4.0%, respectively) was observed as temperature was increased from 80 °C to 150–200 °C. The increase in the Fe carbonation efficiency of between 0.7–5.9% was notably observed as pH increased from 8–12; Fe carbonation efficiency was 0.6–3.3% at pH 8. However, the Ca carbonation moderately occurred (0.7–5.5%) at pH of between 8 to 10. Clearly, the mineralogical and chemical composition of the

mine waste played an important role for mineral carbonation and the effects of pH, particle size, and temperature were of great importance in influencing the process.

The results of the carbonation efficiency found in this study are comparable to some other previous works investigating the same variables. Despite this, other variables that are not considered in this work could be incorporated into future experiment to enhance process efficiency. Findings have demonstrated that mine-waste materials could be as useful as other industrial by-products in sequestering atmospheric carbon dioxide. While this study has been focusing on the use of iron mine waste, various other types of mining waste can be investigated based on their mineral and chemical composition, thereby providing some useful insight into waste minimization issues in the mining industry.

Generally, the findings would be useful for mineral carbonation industries in finding potential feedstock by using waste materials to mitigate global carbon dioxide emissions. From an environmental perspective the utilization of waste material from the mining industry would benefit not only the environment but also the industry as it helps tackle the issue of waste production while minimizing the carbon footprint of their operation. In addition, the produce created from mineral carbonation processes can also be commercialized into various forms of carbonated products, which is another interesting aspect of the technology. Therefore, it has been demonstrated that the application of the mineral carbonation process using mine waste can be regarded as an alternative solution for sustainable carbonation technology.

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