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Potential investigation of concrete fines as an alternative material: A novel neutralizer for acid mine drainage treatment

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ABSTRACT

The concomitant generation of concrete fines as a byproduct during aggregate recycling is problematic in entire concrete recycling system. The properties of concrete fines mostly composed of hydrated cement limits the availability in construction use. Effective utilization of concrete fines must be explored to improve the negative environmental impact from the cement and concrete industries. Developing concrete fines as alternative material is one of promising options. This study investigated the potential of concrete fines as novel neutralizer for acid mine drainage (AMD) to explore the effective use of concrete fines. The neutralization performance, including identification of main substance that provide alkalinity, removal mechanism of As, Fe, and comparison with conventional neutralizers were confirmed by discussing the effect of dosage and particle size on AMD neutralization. The sedimentation performance was determined, and the neutralized sludge that was derived from concrete fines showed good settling properties: compactness, and dewatering ability. Moreover, CO_2 emissions in AMD neutralization were considered, and CO_2 emission reduction by $Ca(OH)_2$ and $CaCO_3$ substitution by concrete fines was assessed. This first fundamental investigation of concrete fines utilization in AMD treatment determined the potential for neutralization and established the prime consideration of CO_2 emissions.

1. Introduction

Recycling of waste cement and waste concrete has been investigated to reduce negative environmental impacts from the cement and concrete industries (Tam et al., 2018; Wang and Zhang, 2018). Concrete wastes can be recycled physically as roadbed (Thai et al., 2021), pavement (Zhao et al., 2020), and construction material (such as recycled aggregate) (Oh et al., 2021; Shi et al., 2016). Such waste can be recycled and reused chemically (Ho et al., 2021a) in soil stabilization (Abraham et al., 2018; Kluge et al., 2018), water treatment (Hongo et al., 2014; Iizuka et al., 2019, 2012; Sasaki et al., 2014, 2012), gas treatment (Iizuka et al., 2011; Wu et al., 2009, 2008), and in CO_2 sequestration and utilization (Ho et al., 2021b, 2020, 2019).

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The recycling of waste concrete to produce concrete is a good approach to preserve natural resources and prevent negative environmental impacts from cement and concrete industries (Akhtar and Sarmah, 2018; Bui et al., 2018). The recycled aggregates, including large particles (recycled stone; 5–40 mm), and small particles (recycled sand; <5 mm) are used as roadbed material and newmade concrete. However, concomitantly generated fine powders (i.e., concrete fines; <200 μm) mostly composed of hydrated cement are generated as byproduct which is regarded as problematic in entire concrete recycling system. Since concrete fines are composed mainly of hydrated cement with a small particle size, the water absorption ability is higher than recycled aggregates, resulting in the reduction of strength (Zhao et al., 2015). Besides, concrete drying shrinkage and freeze-thawing occurs in the hydrated cement section, indicating the concrete fines, mostly hydrated cement, are difficult to be used and required further treatment. Hence, the effective utilization of byproduct from aggregate recycling must be explored to improve the negative environmental impact from the cement and concrete industries. Thus far, research on byproduct recycling and use has been limited and effective methods for use have not yet been established. The potential for concrete fines use should be explored to increase the feasibility and benefit of aggregate recycling. One of possibilities is to utilize as an alternative material in environmental pollution issue.

In this study, the use of concrete fines from aggregate recycling as an alternative neutralizer was applied for treatment of acid mine drainage (AMD). AMD is the main pollution source in the Japanese mining industry and in many countries globally. AMD neutralization and toxic element immobilization are important issues (Moodley et al., 2018). Conventional processes that require large amounts of neutralizing agent (such as limestone or hydrated lime) are expensive and consume resources. Alternative alkaline sources for neutralization include fly ash (Gitari et al., 2006, 2018, 2013, 2010; Madzivire et al., 2019, 2014, 2011, 2010), steel slag (Zheng et al., 2020), paper mill waste (Pérez-López et al., 2010), cement kiln dust (Mackie and Walsh, 2015), recycled aggregate (Mahedi et al., 2020), concrete sludge (Iizuka et al., 2022), and pervious concrete (Shabalala et al., 2017). Concrete fines (a byproduct of recycled aggregate production) consist mainly of hydrated cement and are a possible candidate for AMD neutralization because of their alkali characteristics. The main concern in AMD treatment is solution neutralization and toxic element immobilization. Concrete fines addition allows for an increase in pH at least above 8, and preferably above 10 because most toxic elements are removed (Bortnikova et al., 2020; Masindi et al., 2018).

In this first fundamental investigation of concrete fines utilization in AMD treatment, synthetic concrete fines were tested with simulated AMD. The neutralization reaction mechanisms were examined by discussing dosage and influence of particle size. A comparison of the neutralization performance with conventional neutralizers (such as $CaCO_3$ and $Ca(OH)_2$) is provided. The basic sedimentation performance was investigated. Considerations related to CO_2 emissions were studied. In general, this work aims to enhance the value of concrete fines that were generated from aggregate recycling by exploring their use in AMD treatment as a neutralizer.

2. Experimental

2.1. Materials

Materials that were used in this study were prepared to simulate the concrete fines that are generated from aggregate recycling. Fines were synthesized by mixing ordinary Portland cement powder (401 J, Japan Cement Association, Japan) with distilled water in a mass ratio of 100:55. The mixture was placed in a mold (180 mm × 220 mm × 40 mm) at room temperature for 28 days. The solidified concrete fines were crushed by a crushing mill (M20 universal mill, IKA, Germany) and sieved to the desired particle size. Concrete fines (<53 μm) were composed mainly of Ca (35.54 mass%), Si (8.64 mass%), Al (1.88 mass%), Fe (1.4 mass%), Mg (0.43 mass%), and S (0.41 mass%). The $CaCO_3$ and $CaSO_4$ contents were 3.25 mass% and 1.72 mass%, respectively. The primary mineral phases in concrete fines are portlandite ($Ca(OH)_2$), calcite ($CaCO_3$), larnite (Ca_2SiO_4) and ettringite ($3CaO{\cdot}Al_2O_3{\cdot}3CaSO_4{\cdot}32H_2O$). Conventional neutralizers were used to compare the performance with the concrete fines. Reagent-grade $Ca(OH)_2$ (96%, FUJIFILM Wako Pure Chemical Corporation, Japan) and $CaCO_3$ (99.95%, FUJIFILM Wako Pure Chemical Corporation, Japan) were used. The average particle sizes of the $Ca(OH)_2$ and $CaCO_3$ were 12.9 and 31.3 μm, respectively. Because $CaSO_4$ does not react in the neutralization reaction, the ideal neutralization potential of the neutralizers is defined as the total Ca minus Ca existing as $CaSO_4$. The theoretical effective Ca in concrete fines, $Ca(OH)_2$ and $CaCO_3$ was 35 mass%, 52.6 mass%, and 40 mass%, respectively. The AMD solution was prepared by using iron(III) sulfate n-hydrate (99.5%, FUJIFILM Wako Pure Chemical Corporation, Japan) and arsenic acid solution (60%, Wako Pure Chemical Corporation, Japan) to simulate AMD that is generated at Japanese mine sites. The simulated solution pH was 2 with Fe^{3+} and As concentrations of 300 and 5 mg/L, respectively.

2.2. Analytical methods

The elemental composition was analyzed by inductively coupled plasma optical emission spectrometry (ICP-OES; Spectro Arcos, Spectro Analytical Instruments, Germany). A solid raw sample of concrete fines was analyzed as follows. Concrete fines (100 mg) were dissolved in 10 mL of an acidic mixture (25 vol% HCl (35%, FUJIFILM Wako Pure Chemical Corporation, Japan) + 60 vol% HNO_3 (60%, FUJIFILM Wako Pure Chemical Corporation, Japan) + 15 vol% HF (30%, FUJIFILM Wako Pure Chemical Corporation, Japan)) and 15 mL of H_3BO_3 (saturated solution) at 200 °C for 2 h. The $CaCO_3$ content in the raw sample of concrete fines was determined by thermal gravimetric analysis (TG, Thermoplus Evo TG 8120, Rigaku,

Tokyo, Japan). The pH was measured by a pH meter (D73, HORIBA Co., Kyoto, Japan). The particle size was determined from the particle size distribution analyzer (Microtrac MT3000 II, MicrotracBEL Corp, Japan). The crystalline phase was analyzed by X-ray diffractometry (XRD; D2 Phaser, Brucker AXS, Germany) with a scan rate of 2.22°/min and ICDD-PDF database. Filters (0.45 μm, ADVANTEC, Japan) were used in vacuum filtration to isolate the solid component from the neutralized sludge. The neutralized residues were dried in air at room temperature (21 ± 3 °C) for more than 18 h.

2.3. Experiments

Batch experiments were carried out by mixing different quantities of concrete fines to determine the effect of dosage on AMD neutralization. Concrete fines (0.4, 0.8, and 1.2 g) and 200 mL AMD solution were used at stirring rate of 200 rpm and room temperature (21 ± 3 °C). The sampling times were 0, 1, 2, 3, 5, 10, 20, 30, 60, and 120 min. To evaluate the effect of particle size on the neutralization performance, the particle size of concrete fines was sieved to: <53, 53–106, 106–212, 212–1000, and 1000–1180 μm. AMD solution (200 mL) was mixed with 0.8 g concrete fines of different particle sizes at 200 rpm for sampling times of 0, 1, 2, 3, 5, 10, 20, 30, 60, and 120 min. To compare the neutralization performance, 0.8 g neutralizer was mixed with 200 mL AMD solution at 200 rpm and aliquots of the mixture were removed for analysis after 0, 1, 2, 3, 5, 10, 20, 30, 60 and 120 min. It was anticipated that the particle size of the neutralizer might affect the neutralization and sedimentation performance. To minimize this effect, concrete fines (D_{50}: 17.29 μm) having particle sizes similar to those of conventional neutralizers were prepared to compare the performance of $CaCO_3$ (D_{50}: 31.3 μm) and that of $Ca(OH)_2$ (D_{50}: 12.9 μm). To determine the sedimentation performance, 1 L AMD solution was mixed with desired neutralizers (i.e., $Ca(OH)_2$, $CaCO_3$ and concrete fines) in a solid–liquid ratio of 4 g/L and with a reaction time of 120 min. A 1 L graduated cylinder having a circular cross-section and a diameter of 62 mm along with a height of 422 mm was used to measure the sludge volume. Sedimentation tests with mixed solutions were conducted at a reaction time from 0 to 90 min with a discussion of the sludge height and sludge properties. The masses of entire sludge and wet solid fractions were weighed before and after vacuum filtration through 0.45 μm filter paper. The mass of dry solids in the sludge was determined after drying at 105 °C for more than 18 h. The water content of the sludge was obtained from the difference in the masses of the wet and dry solids in the sludge. The mass/content percentages (i.e., wet solid mass percentage, dry solid mass percentage and water content percentage) were calculated based on the solid mass/water contents and the entire sludge masses.

3. Results and discussions

3.1. Neutralization performance

3.1.1. Effect of dosage

The effect of the dosage of concrete fines (<53 μm) on AMD neutralization was studied. The pH increased rapidly and reached equilibrium at ~11.5 at 120 min for 4 and 6 g/L as shown in Fig. 1a. At 2 g/L, the pH increased to 4.67 at 5 min and to 8.64 at 120 min. The greater dosage of concrete fines provided a higher alkali content as $Ca(OH)_2$, $CaCO_3$, and 3CaO·2SiO2·4H2O, which resulted in a higher pH. As and Fe were the main targets for removal. Table S1 shows that the Fe concentration was ~0.31 mg/L at 1 min and less than 0.1 after 5 min at 2 g/L. In all cases, As was removed within 1 min and Fe was removed within 5 min, which indicates that the concrete fines could eliminate these toxic elements. Possible reactions of AMD neutralization by concrete fines can be explained by Eqs. (1)–(8). Eqs. (1)–(4) show the neutralization reactions between the primary alkaline species (i.e., $Ca(OH)_2$, $CaCO_3$, 3CaO ·2SiO2 ·4H2O and 3CaO ·Al_2O_3·3$CaSO_4$·32H_2O) and sulfuric acid. Eqs. (5)–(8) indicate that Fe^{3+} is removed with an increase in pH by concrete fines addition and FeOOH precipitation (Jönsson et al., 2006; Santomartino and Webb, 2007). The As removal mechanism is considered based on coprecipitation with Fe. As coprecipitation with Fe was determined from previous studies (Tokoro, 2015; Tokoro et al., 2020, 2010). The initial As/Fe molar ratio in the treated solution in this study was less than 0.25, which indicates that As removal occurred by surface complexation on FeOOH during coprecipitation (Takaya et al., 2021). The Ca concentration increased with an increase in concrete fines dosage as shown in Fig. 1b, which indicates the greater amount of alkali dissolution, especially of $Ca(OH)_2$. This result is consistent with the pH variation with reaction time and can be explained by Eqs. (1)–(4). The XRD patterns of concrete fines and the concrete fines-neutralized residue (6 g/L) are shown as Fig. 2. No peaks of portlandite and ettringite existed in the concrete fines-neutralized residue pattern. This result indicates that portlandite and ettringite were consumed as alkali suppliers. In particular, the intense portlandite peak initially found in concrete fines completely disappeared, confirming that this material was the primary contributor to neutralization. The sulfur concentration was ~400–450 mg/L, which is constant with time and no gypsum peak was visible in the XRD pattern. Therefore, gypsum may not be precipitated in the neutralized residue. In general, AMD neutralization by $Ca(OH)_2$ and ettringite occurs according to Eqs. ((1), (4), (5) and (8)).

Possible reactions of AMD neutralization with concrete fines generated from aggregate recycling

$$Ca(OH)_2 + H_2SO_4 \rightarrow Ca^{2+} + SO_4{}^{2-} + 2H_2O \quad (1)$$

$$CaCO_3 + H_2SO_4 \rightarrow Ca^{2+} + SO_4{}^{2-} + H_2O + CO_2\uparrow \quad (2)$$

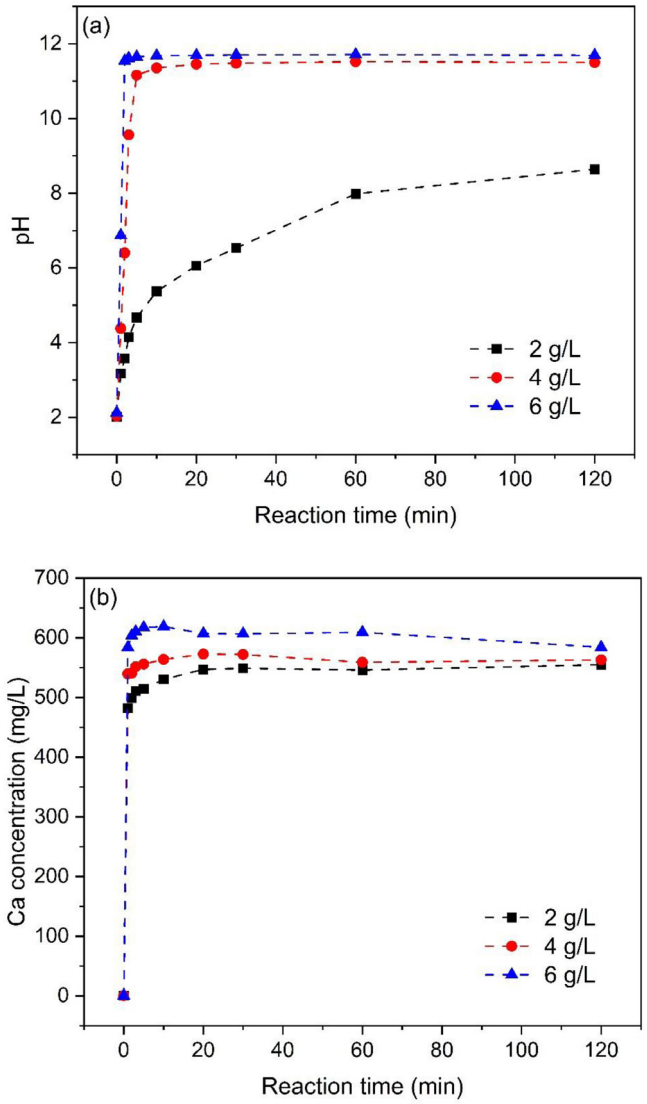

Fig. 1. Changes in (a) pH and (b) Ca concentration with time for different dosages. (Conditions: 200 mL AMD solution, <53 μm concrete fines, 200 rpm stirring rate and 21 ± 3 °C.)

$$3CaO{\cdot}2SiO_2{\cdot}4H_2O + 3H_2SO_4 \rightarrow 3Ca^{2+} + 3SO_4{}^{2-} + 2SiO_2 + 7H_2O \quad (3)$$

$$3CaO{\cdot}Al_2O_3{\cdot}3CaSO_4{\cdot}32H_2O + 3H_2SO_4 \rightarrow 6Ca^{2+} + 6SO_4{}^{2-} + Al_2O_3 + 35H_2O \quad (4)$$

$$2Fe^{3+} + 3Ca(OH)_2 \rightarrow 2FeOOH + 3Ca^{2+} + 2H_2O \quad (5)$$

$$2Fe^{3+} + 3CaCO_3 + H_2O \rightarrow 2FeOOH + 3Ca^{2+} + 3CO_2\uparrow \quad (6)$$

$$2Fe^{3+} + 3CaO{\cdot}2SiO_2{\cdot}4H_2O \rightarrow 2FeOOH + 3Ca^{2+} + 2SiO_2 + 3H_2O \quad (7)$$

$$2Fe^{3+} + 3CaO{\cdot}Al_2O_3{\cdot}3CaSO_4{\cdot}32H_2O \rightarrow 2FeOOH + 3Ca^{2+} + Al_2O_3 + 3Ca^{2+} + 3SO_4{}^{2-} + 31H_2O \quad (8)$$

3.1.2. Effect of particle size

The effect of particle size on AMD neutralization was investigated. The solution pH increased more rapidly for a small particle size as shown in Fig. 3a. The pH plateaued, and was similar in all cases except for 1000–1180 μm, where the pH reached 5.32 at 120 min. The specific surface areas of the smaller particles were larger, which provided a greater availability for alkali dissolution. Table 1 shows the As and Fe concentrations for different concrete fines sizes. The removal rate was affected by particle size. For a particle size smaller than 106 μm, As and Fe were removed within 1 min. A larger concrete fines particle size yielded a slower removal rate, even at 1000–1180 μm, As and Fe could be removed at 120 min.

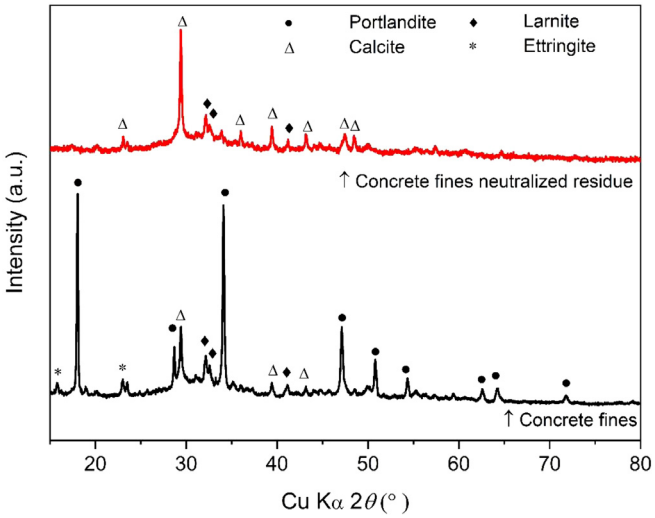

Fig. 2. XRD patterns of concrete fines and concrete fines-neutralized residue; portlandite ($Ca(OH)_2$; PDF 00-044-1481), calcite ($CaCO_3$; PDF 01-083-1762), larnite (Ca_2SiO_4; PDF 00-049-1673) and ettringite ($3CaO \cdot Al_2O_3 \cdot 3CaSO_4 \cdot 32H_2O$; PDF 00-013-0350).

Table 1
As and Fe concentration for different particle sizes of concrete fines.

Particle size (μm)	As						Fe					
	1 min	10 min	20 min	30 min	60 min	120 min	1 min	10 min	20 min	30 min	60 min	120 min
<53	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
53–106	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
106–212	0.54	<0.01	<0.01	<0.01	<0.01	<0.01	35.21	<0.1	<0.1	<0.1	<0.1	<0.1
212–1000	4.73	0.32	<0.01	<0.01	<0.01	<0.01	289.2	64.7	<0.1	<0.1	<0.1	<0.1
1000–1180	5.11	4.88	2.43	1.21	0.41	<0.01	296.7	285.3	228.5	163.4	42.4	<0.1

The change in Ca concentration with time agrees with the explanation of pH variation with time. Small concrete fines yielded a faster increase in Ca concentration. The change in Si concentration with time for different sizes of concrete fines was measured. The Si concentration increased and then decreased with time. A larger particle size yielded a slower decrease in Si concentration. The increase in Si concentration was caused calcium silicate hydrate dissolution in concrete fines according to Eqs. (3) and (7). The Si concentration started to decrease at pH around 3. The same behavior resulted in previous studies (Ni et al., 2014), and may be ascribed to silicate minerals formation. Colloidal silicate precipitation reduces the Si concentration in solution. Besides, the addition of silicate could improve the removal of elements (Meng et al., 2000; Roberts et al., 2004). The use of silicates in concrete may enhance the removal performance, which is a novel concept for concrete use. At 53–106 and 106–212 μm, the Si concentration increased, decreased, and increased again, possibly because of the high dissolution rate of concrete fines and precipitation of colloidal hydroxide, and a slight redissolution of colloidal hydroxide, which is dependent on Si concentration and solution pH. The Si concentration plateaued in all cases except for 1000–1180 μm. The crystalline phases of the concrete fines and concrete fines-neutralized residues were analyzed. The crystalline phases in the particle size ranges <53, 212–1000, and 1000–1180 μm provide evidence of the difference in neutralization performance as shown in Fig. 4. Portlandite peaks existed in the neutralized residues with larger particle sizes, which indicates that the alkali dissolution was slow for the larger particle size. This result is consistent with the previous results in Fig. 3. The slow alkali dissolution resulted in a slow increase in pH and Ca solution concentration and a slow removal of As and Fe.

A higher reaction rate and pH were achieved with a smaller particle sizes. These results are consistent with previous literature in which AMD was neutralized by recycled aggregates (Mahedi et al., 2020). Furthermore, unlike recycled aggregate, concrete fines from aggregate recycling tends to be less than 200 μm and are almost free of aggregate contamination (Ho et al., 2021b), which indicates that a good neutralization performance is expected.

3.1.3. Comparison with conventional neutralizers

Conventional neutralizers (such as $Ca(OH)_2$ and $CaCO_3$) are used extensively in AMD treatment. $Ca(OH)_2$ is one of the most effective and common neutralizers that can neutralize AMD within a relatively short reaction time. $CaCO_3$ is usually used as a first-stage neutralizer when treating AMD to remove metals that precipitate at a relatively low pH in two-stage neutralization. Because the use of concrete fines from aggregate recycling could reduce cost, avoid natural exploitation,

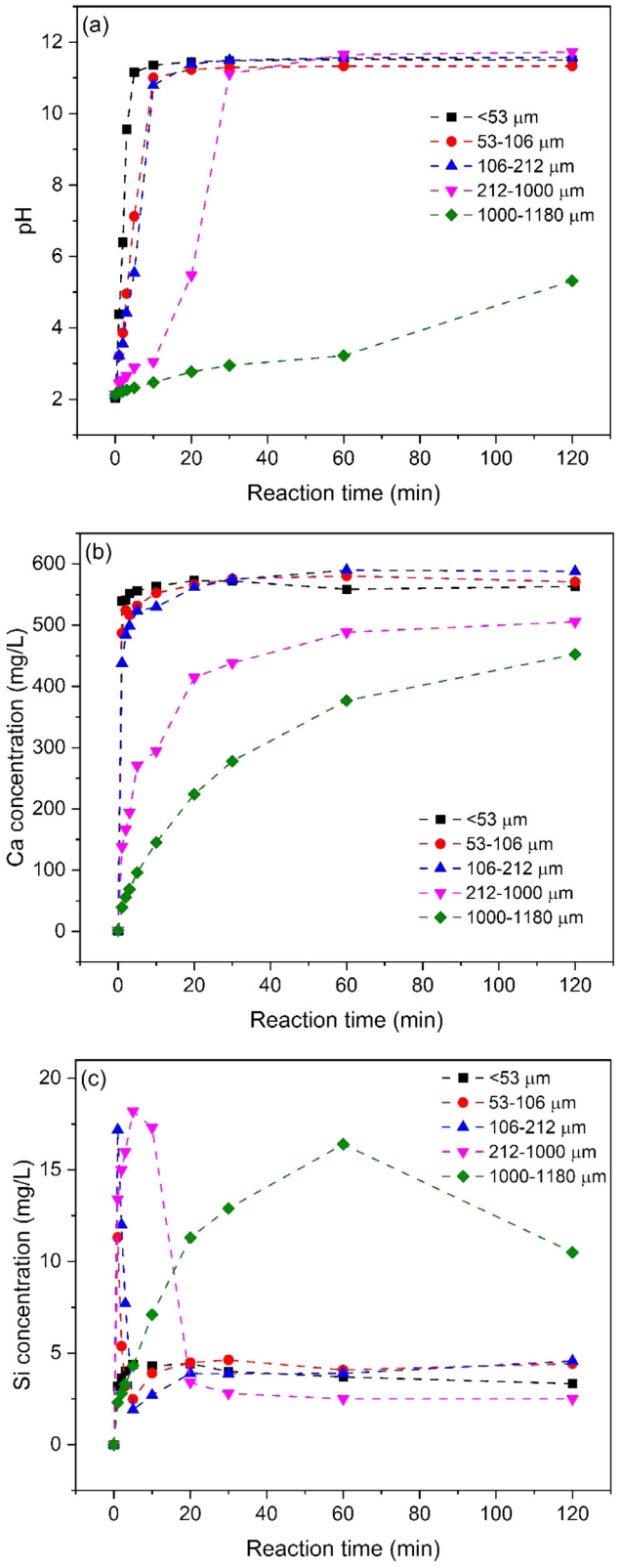

Fig. 3. Changes in (a) pH, (b) Ca concentration, (c) Si concentration with time for different concrete fines sizes. (Conditions: 200 mL AMD solution, 0.8 g concrete fines, 200 rpm stirring rate, 21 ± 3 °C.)

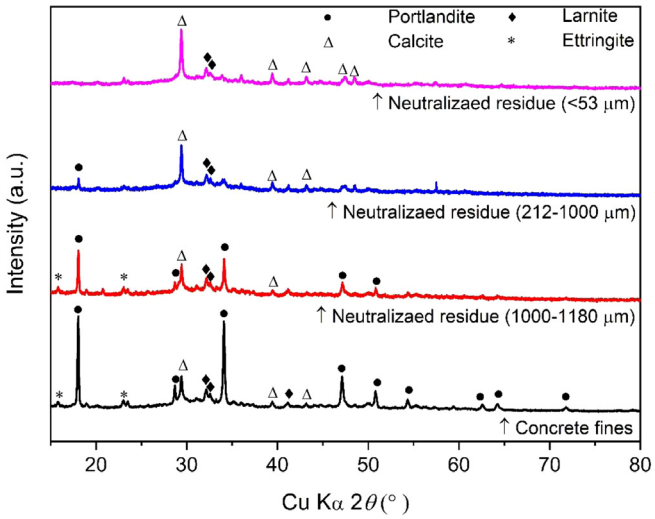

Fig. 4. XRD patterns of concrete fines and concrete fines-neutralized residue with different particle sizes; portlandite ($Ca(OH)_2$; PDF 00-044-1481), calcite ($CaCO_3$; PDF 01-083-1762), larnite (Ca_2SiO_4; PDF 00-049-1673) and ettringite ($3CaO \cdot Al_2O_3 \cdot 3CaSO_4 \cdot 32H_2O$; PDF 00-013-0350).

Table 2
As and Fe concentrations with different neutralizers.

Neutralizers	As			Fe		
	1 min	10 min	20 min	1 min	10 min	20 min
Concrete fines	<0.01	<0.01	<0.01	<0.1	<0.1	<0.1
$Ca(OH)_2$	<0.01	<0.01	<0.01	<0.1	<0.1	<0.1
$CaCO_3$	<0.01	<0.01	<0.01	0.19	0.11	<0.1

and achieve waste utilization, the neutralization performance of concrete fines were compared with $Ca(OH)_2$ and $CaCO_3$. AMD neutralizations with $Ca(OH)_2$ and $CaCO_3$ are described as follows:

AMD neutralization with $Ca(OH)_2$

$$Ca(OH)_2 + H_2SO_4 \rightarrow Ca^{2+} + SO_4{}^{2-} + 2H_2O \quad (1)$$

$$2Fe^{3+} + 3Ca(OH)_2 \rightarrow 2FeOOH + 3Ca^{2+} + 2H_2O \quad (5)$$

AMD neutralization with $CaCO_3$

$$CaCO_3 + H_2SO_4 \rightarrow Ca^{2+} + SO_4{}^{2-} + H_2O + CO_2\uparrow \quad (2)$$

$$2Fe^{3+} + 3CaCO_3 + H_2O \rightarrow 2FeOOH + 3Ca^{2+} + 3CO_2\uparrow \quad (6)$$

Fig. 5a shows the change in pH with time for different neutralizers. $Ca(OH)_2$ provided the greatest alkalinity at the same dosage, whereas $CaCO_3$ provided the least. Concrete fines are located in the middle because they comprise a mixture of $Ca(OH)_2$ and $CaCO_3$ with other amorphous calcium silicate hydrates. Table 2 shows the removal performance of As and Fe by the three neutralizers. For the $Ca(OH)_2$ and concrete fines, As and Fe were removed within 1 min. However, for the $CaCO_3$, Fe was removed within 20 min, which shows that the $CaCO_3$ reaction rate was the slowest. Fig. 5b shows the variation in Ca concentration with time for different neutralizers. $Ca(OH)_2$ yielded the highest Ca concentration because of its highest content of Ca (40 g Ca/mol/74 g $Ca(OH)_2$/mol $\times$ 100 = 54 mass%) and the greatest reactivity for neutralization. The Ca content in $CaCO_3$ (40 g Ca/mol/100 g $CaCO_3$/mol $\times$ 100 = 40 mass%) was slightly larger than in the concrete fines (35.5 mass%), which resulted in a slightly higher Ca concentration for $CaCO_3$ at the plateau. Fig. 6 shows that the portlandite peak in the $Ca(OH)_2$ case was reduced and disappeared, the peak of calcite in $CaCO_3$ remained, and only the calcite peak in the concrete fines remained. This result is consistent with a previous explanation of neutralizer reactivity.

3.2. Sedimentation performance

The sedimentation performance of AMD neutralization sludge that was generated by concrete fines without flocculant addition was compared with that by $Ca(OH)_2$ and $CaCO_3$. It should be noted that each reagents were prepared in

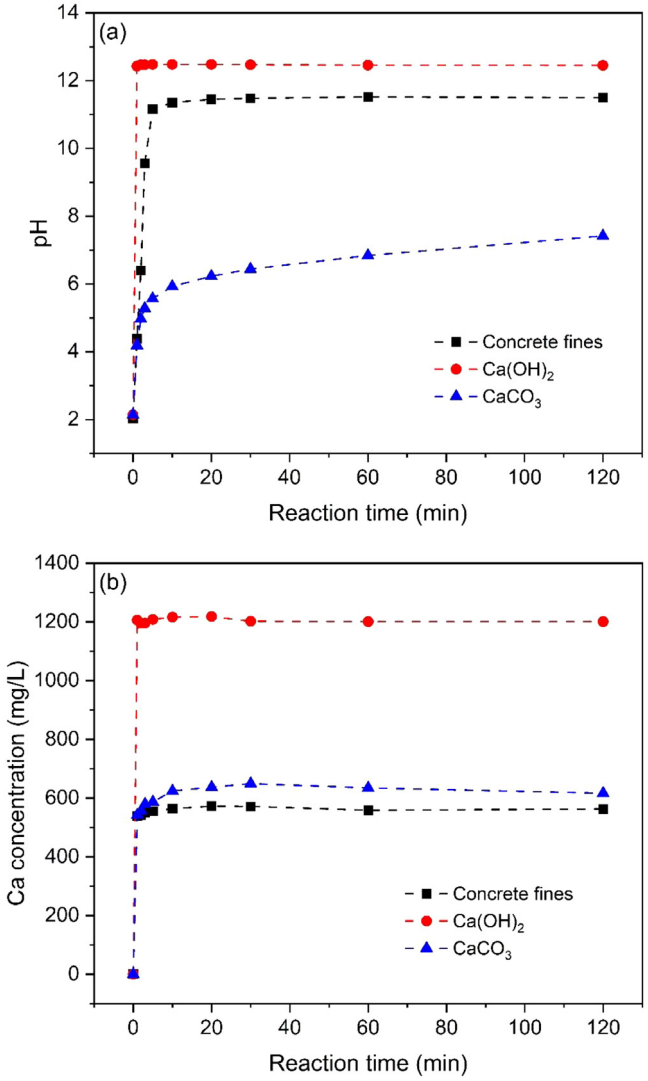

Fig. 5. Changes in (a) pH and (b) Ca concentration with time for different neutralizers. (Conditions: 200 mL AMD solution, 0.8 g neutralizers, 200 rpm stirring rate, 21 ± 3 °C.)

similar particle sizes to avoid the effect of particle sizes on sedimentation performance. The change in sludge height with sedimentation time is shown in Figure S1. The initial sedimentation rates were calculated from the slope of the initial section of the curves (Mackie and Walsh, 2015; Yan et al., 2013). The initial sludge sedimentation rates by $CaCO_3$ (0.06 cm/s) and concrete fines (0.05 cm/s) were higher than that by $Ca(OH)_2$ (0.004 cm/s). Other studies have reported the sedimentation rates of byproducts from the cement and concrete industries (such as cement kiln dust) and of $Ca(OH)_2$ (Mackie and Walsh, 2015). In addition, the sedimentation rate of cement kiln dust has been found to be faster than that of $Ca(OH)_2$ as well. Figure S1 shows that the sedimentation of $Ca(OH)_2$-neutralized sludge was incomplete at 90 min, whereas that of the $CaCO_3$-neutralized sludge and concrete fines-neutralized sludge was complete within 10 min. Figure S1 also indicates that the sludge volume of the concrete fines-neutralized sludge at 90 min was ∼10%, between $CaCO_3$-neutralized sludge (∼1.5%) and $Ca(OH)_2$-neutralized sludge (∼30%) because of the concrete fines composition. It is difficult to directly compare sludge properties because these are affected by the type of neutralizer that is employed and also by several other factors, including dosage amount of neutralizer, the elemental composition of the AMD, the addition of polymeric flocculants, the type of sedimentation container and the method used for the AMD treatment. Some prior studies described the volume of neutralized sludge derived by using alternative materials as neutralizers. Mackie and Walsh (2015) determined a sludge volume of approximately 40% based on trials using cement kiln dust and a polymeric flocculant at a sedimentation time of 60 min in high density sludge treatment at recycle ratio of 20:1. Tosun (2018) reported a sludge volume of approximately 42% using fly ash as neutralizer at a dosage of 10 kg/ton-AMD and

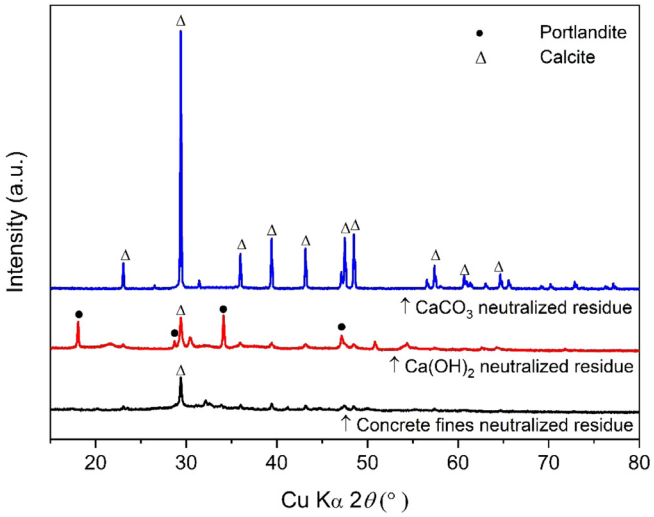

Fig. 6. XRD patterns of concrete fines-neutralized residue, $Ca(OH)_2$-neutralized residue, and $CaCO_3$-neutralized residue; portlandite ($Ca(OH)_2$; PDF 00-044-1481), calcite ($CaCO_3$; PDF 01-083-1762).

Table 3
Sludge properties with different neutralizers.

Neutralizers	Concrete fines	$Ca(OH)_2$	$CaCO_3$
Sludge mass (g)	85.6	290.9	28.1
Wet solid mass (g)	9.16	32.5	4.87
Wet solid mass percentage (mass%)	10.7	11.2	18
Dry solid mass (g)	3.41	2.56	2.72
Dry solid mass percentage (mass%)	3.98	0.88	10.1
Water content (g)	5.76	29.9	1.97
Water content percentage (mass%)	6.72	10.3	7.01

sedimentation time of 500 min. The sludge properties were analyzed as shown in Table 3. The $CaCO_3$-neutralized sludge was lightest, followed by the concrete fines-neutralized sludge, and the $Ca(OH)_2$-neutralized sludge. Based on the sludge volume and mass, the concrete fines-neutralized sludge had the highest density, which indicates that concrete fines-neutralized sludge had a good compactness. The lower water content of the AMD neutralization sludge is advantageous for dewatering. The water content was 6.72 mass%, 10.3 mass%, and 9.48 mass% in the concrete fines-, $Ca(OH)_2$-, and $CaCO_3$-neutralized sludge, respectively. Therefore, the use of concrete fines favors practical dewatering in AMD neutralization, which is advantageous in terms of capital and maintenance costs (Mackie and Walsh, 2015).

Although $Ca(OH)_2$ showed an efficient neutralization performance, its poor sedimentation performance may impede the AMD treatment. In contrast, $CaCO_3$ had a relatively low reactivity and lower alkalinity, but the sedimentation performance was good. Concrete fines generation from aggregate recycling showed a good neutralization and sedimentation performance and the fines are a promising candidate for AMD treatment.

Prior research has shown that one of the most promising applications for neutralized AMD sludge is as mine backfill material, which offers a so-called cradle-to-cradle solution (Vadapalli et al., 2012). Specifically, this material could be employed to provide support for pillars and walls in mines, to passively neutralize AMD and to reduce the ongoing generation of AMD by excluding air and water (Gitari et al., 2008; Madzivire et al., 2019). Neutralization sludge obtained from alternative materials such as fly ash has also been shown to be useful as mine backfill material (Gitari et al., 2008; Madzivire et al., 2019; Vadapalli et al., 2007). Like fly ash, concrete fines are alkaline and have pozzolanic properties and so concrete fines-neutralized sludge is expected to have applications as a material for mine backfill and passive AMD treatment. Other possible applications of concrete fines-neutralized sludge are used to produce geopolymer (Kumar Vadapalli et al., 2008), zeolitic (Somerset et al., 2005), ceramic materials (Liu et al., 2019a,b), and environmental friendly materials (Daradjat and Moersidik, 2021; Wang et al., 2013). The application of concrete fines-neutralized sludge is important and of interest and requires more investigation.

3.3. CO_2 emissions-related considerations

Climate change has become an increasingly pressing issue. Therefore, CO_2 emissions during AMD neutralization must be considered. CO_2 emissions from the perspective of neutralizers is discussed below. Replacement of $CaCO_3$ with concrete

fines could reduce Scope 1 emissions (such as direct CO_2 emissions from $CaCO_3$ for AMD neutralization) as shown by Eq. (2). The $CaCO_3$ content (3.25 mass%) in the concrete fines was determined by TG. According to Eqs. (2) and (9), for every gram of neutralizer, $CaCO_3$ releases 0.43 g-CO_2 theoretically more than when concrete fines are used.

$$M_{CO_2\text{-}CaCO_3} = (M_{CaCO_3} \times P - M_{concrete} \times W_{CaCO_3}/100) \times (m_{CO_2}/m_{CaCO_3}) \tag{9}$$

where $M_{CO_2\text{-}CaCO_3}$ (g) is the CO_2 avoidance amount by replacing $CaCO_3$ with concrete fines, M_{CaCO_3} (g) and $M_{concrete}$ (g) are the replaced amount of the $CaCO_3$ and the used amount of concrete fines, P is the purity fraction of the $CaCO_3$, W_{CaCO_3} (mass%) is the mass percentage of $CaCO_3$ in the concrete fines, m_{CO_2} and m_{CaCO_3} are molecular masses of CO_2 and $CaCO_3$, respectively.

Substitution of the use of $Ca(OH)_2$ by concrete fines could avoid Scope 3 emissions (such as indirect CO_2 emissions from $Ca(OH)_2$ production from $CaCO_3$). The production of 1 metric ton of $Ca(OH)_2$ emits 1.19 to 1.50 metric tons of CO_2 (Ochoa George et al., 2010). Considering the $CaCO_3$ content in the concrete fines and CO_2 emissions from $Ca(OH)_2$ production, substitution of each gram of $Ca(OH)_2$ by equivalent amount of concrete fines in AMD neutralization could theoretically avoid 1.18 to 1.49 g-CO_2 according to Eq. (10).

$$M_{CO_2\text{-}Ca(OH)_2} = Q \times M_{Ca(OH)_2} - (M_{concrete} \times W_{CaCO_3}/100) \times (m_{CO_2}/m_{CaCO_3}) \tag{10}$$

where $M_{CO_2\text{-}Ca(OH)_2}$ (g) is the CO_2 avoidance amount by replacing $Ca(OH)_2$ with concrete fines, Q (g-CO_2/g-$Ca(OH)_2$) represents the unit emissions of CO_2 for $Ca(OH)_2$ production (1.19 – 1.50), $M_{Ca(OH)_2}$ (g) is the replaced amount of $Ca(OH)_2$.

In addition to the effects of material substitutions, the CO_2 emissions associated with the transportation of concrete fines (because of fuel consumption by vehicles) must also be taken into account. Concrete fines are generated during aggregate recycling by concrete recycling facilities. The majority of the CO_2 emissions resulting from transportation are generated during the shipping of the waste concrete to the concrete recycling plant and the transportation of the concrete fines from the facility to the AMD treatment plant. The amount of CO_2 emissions (both direct and indirect), T (t-CO_2), can be calculated as

$$T = a \times b, \tag{11}$$

where a and b are the quantity of fossil fuel consumed during transportation (in metric tonnes) and the default emission factor of the fossil fuel that is used (typically diesel) (t-CO_2/t). To allow accurate calculations, the reductions in CO_2 emissions associated with the use of concrete fines in place of conventional neutralizers (i.e., $Ca(OH)_2$ and $CaCO_3$) should be considered. To do so, we can use the equations

$$a = a_1 + a_2, \tag{12}$$

where a_1 and a_2 are the quantities of fossil fuel consumed by the transportation of the waste concrete to the concrete recycling plant and of the concrete fines to the AMD treatment plant, and

$$c = c_1 + c_2, \tag{13}$$

where c_1 and c_2 are the quantities of fossil fuel consumed by the transportation of the raw materials to the chemical production plant for the production of conventional neutralizers and of the conventional neutralizers to the AMD treatment plant. The difference in CO_2 emissions from transportation when using concrete fines and when using conventional neutralizers, T_c, is therefore

$$T_c = (a - c) \times b. \tag{14}$$

These equations allow an evaluation of the complete CO_2 emissions from the material preparation to the AMD neutralization stage. In future, it will be necessary to clearly assign responsibility for CO_2 emissions to each individual stage or facility (such as the concrete production, recycling and AMD treatment plants). The ongoing development of inexpensive, stable supplies of renewable energy can be anticipated, and so the generation of transportation-related CO_2 emissions from fossil fuel consumption will be reduced. The use of concrete fines can therefore be regarded as potentially sustainable and environmentally friendly. Hence, the development of alternative neutralizers from byproducts or wastes is promising and should be further investigated.

4. Conclusions

The effective utilization of concrete fines for AMD treatment were investigated. The concrete fines from aggregate recycling were effectively used as novel neutralizer for AMD treatment. The neutralization performance, including identification of main substance that provide alkalinity, removal mechanism of As, Fe, and comparison with conventional neutralizers were confirmed. The sedimentation performance was determined and compared with conventional neutralizers. The CO_2 emissions-related consideration with use of concrete fines as alternative neutralizer was conducted. Based on the experimental results, the following conclusions can be drawn:

- A higher dosage and smaller particle size of concrete fines yielded a better neutralization performance. Alkali consumption, especially portlandite, was the main contributor in neutralization.

- The neutralization performance was comparable with conventional neutralizers.
- The sedimentation performance was determined. Sludge that was neutralized by concrete fines had good settling properties and a good compactness and dewatering ability.
- The substitution of $Ca(OH)_2$ and $CaCO_3$ with concrete fines could reduce CO_2 emissions in AMD neutralization. CO_2 emissions-related considerations for the utilization of concrete fines in AMD neutralization was established.

CRediT authorship contribution statement

Hsing-Jung Ho: Conceptualization, Formal analysis, Investigation, Data curation, Writing – original draft, Visualization. **Atsushi Iizuka:** Conceptualization, Methodology, Writing – review & editing, Resources, Supervision. **Viswanath Ravi Kumar Vadapalli:** Writing – review & editing. **Henk Coetzee:** Writing – review & editing. **Leslie Petrik:** Writing – review & editing. **Jochen Petersen:** Writing – review & editing. **Tunde Ojumu:** Resources, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary material related to this article can be found online at https://doi.org/10.1016/j.eti.2022.102985.

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