

## Reducing the environmental impact of soil cement stabilization using fine powder generated during crushing waste concrete with carbon dioxide fixed

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## ABSTRACT

Soil stabilization requires a significant amount of cement, which results in a considerable amount of CO<sub>2</sub> emitted during the process. In this study, to reduce the environmental impact of soil cement stabilization, a new soil stabilizer, which comprises of cement and carbonated recycled concrete fines (RCFs) is introduced. RCFs contain cement hydrate, which reacts with CO2 and transforms into calcium carbonate; hence, they can be used as a carbon capture and utilization (CCU) material. In this study, RCFs are carbonated in dry and wet conditions to be formed into a CCU material that can fix 10% of CO<sub>2</sub> by mass by the abovementioned two carbonation methods. In addition, test specimens mixed with sand, clay, cement, and CO<sub>2</sub>-fixed RCFs simulating soil-cement columns are prepared to investigate their physicochemical properties. The compressive strength of specimens composed of carbonated RCFs is 1.15 to 1.46 times higher than those without RCFs. This is attributable to the difference in the composition of hydrate in the soil-cement columns. Specimens with carbonated RCFs contain ettringite, which is a hydrate that enhances the strength, whereas it is not detected in specimens without RCFs; however, calcium hydroxide, which does not contribute to the strength, is detected. The dissolution of hexavalent chromium in all the specimens is less than 0.01 mg/L, which is lower than the environmental standard of Japan (0.05 mg/L). We conclude that carbonated RCFs is applicable as an environmentally friendly soil stabilizer.

Keywords: recycled concrete fines, carbon capture and utilization, soil stabilization, soil-cement column

#### **1** INTRODUCTION

Recently, reduction in CO<sub>2</sub> emissions has been required as a countermeasure against global warming, and in the field of geotechnics, decarbonization is additionally required because a significant amount of cement is consumed during soil stabilization, which emits a considerable amount of CO<sub>2</sub>. Some studies have been conducted to reduce environmental impact of soil stabilization, such as adding ground-granulated blast-furnace slag, which is a by-product of steel production that serves as a binder (Kawamura and Kasai, 1999), and performing soil stabilization using ladle furnace slag (Manso, 2013).

One of the countermeasures to reduce the environmental impact of soil–cement stabilization is to use recycled concrete fines (RCFs). A significant amount of RCFs is generated during the producing of recycled fine and coarse aggregates from demolished concrete, and these RCFs contain cement hydrates such as calcium hydroxide and dicalcium silicate (Jiang et al., 2022). Currently, RCFs are not widely applied, and various studies pertaining to the utilization of RCFs have been conducted. Adding RCFs to cement mortar to partially replace cement contributes have been reported to increase strength and reduce initial cracking (Wang et al., 2022). Meanwhile, the use of RCFs as a carbon capture and utilization (CCU) material is being considered because RCFs can absorb  $CO_2$  by soaking powder in water containing a high concentration of  $CO_2$  (Ho et al., 2021; Shen et al., 2021).

In this study, a soil stabilizer composed of cement and carbonated and  $CO_2$ -fixed RCFs is introduced to further reduce the environmental impact of soil stabilization. The RCFs are carbonated using two methods, and the amount of  $CO_2$  fixed in the RCFs is investigated via thermogravimetry differential

thermal analysis (TG/DTA). To clarify the changes in the amounts and types of hydrates in the RCFs before and after carbonation, X-ray diffraction (XRD) analyses for uncarbonated, wet carbonated, and dry carbonated RCFs are conducted. To confirm the change in strength properties due to the addition of uncarbonated and carbonated RCFs, unconfined compressive tests, XRD analyses, and mercury intrusion porosimetry tests are conducted. Additionally, hexavalent chromium dissolution tests are conducted. Previous studies indicate an increase in hexavalent Cr dissolution from cement mortar after carbonation due to the decomposition of hydrates that fixed hexavalent Cr (Kuroda and Koshiishi, 2010).

## 2 RECYCLED CONCRETE FINES

## 2.1 RCFs material

The RCFs used in this study were composed of demolished concrete from office building in Tokyo. Table 1 shows the compressive test results of concrete core obtained from the slab of each floor. The compressive strengths of the samples were considered to be varied due to differences in the design strength of the concrete. The average compressive strength of the cores was 27.4 N/mm<sup>2</sup>. This concrete was blasted under dry conditions, which resulted in the formation of recycled fine and coarse aggregates, and the RCFs generated as a by-product during this recycling process was used in this study.

Sampling floor	First basement	First floor	Second floor	Third floor	Fourth Floor	Fifth floor	Sixth floor	Seventh Floor
Compressive strength	38.6	24.6	16.4	30.0 average	27.8 27.4 st	24.5 andard de	24.5 viation : 6	33.1 5 7
[N/mm <sup>2</sup> ]				average : 27.4 standard deviation : 6.7				

**Table 1.** Compressive strength of concrete core obtained from slab of each floor

## 2.2 Carbonation methods

In this study, dry and wet RCFs carbonation methods are used. Figure 1 shows the dry and wet carbonation system used in this study. In the dry carbonation method, naturally dried RCFs were exposed for 9 days in a chamber with a  $CO_2$  concentration of 10%, temperature of 20 °C, and relative humidity of 60%; furthermore, they were stirred and weighed every 3 days. When the mass change of the RCFs stabilized, the carbonation of RCFs was regarded as complete. In the wet carbonation method, a chamber containing a slurry of naturally dried RCFs and water mixed at a ratio of 1:2 by mass was prepared, and pure  $CO_2$  gas was blown into the slurry at 20 L/min. When the pH of the slurry reached 6.8, carbonation was regarded as complete. After wet carbonation was completed, the RCFs and water were separated using filter and dried naturally to obtain carbonated RCFs.



Figure 1. Dry carbonation system (left). Wet carbonation system (right)

## 2.3 Physicochemical properties of RCFs

Figure 2 shows the particle size distribution of the RCFs carbonated under dry and wet conditions obtained via laser diffraction analysis. The median diameters of the two types carbonated RCFs were both 33.4  $\mu$ m, and the particle size distributions were almost identical. Table 2 shows the densities of the RCFs before and after carbonation obtained using the gas replacement method. The density of the RCFs carbonated under wet conditions was slightly higher than that of uncarbonated RCFs.



Figure 2. Particle size distribution of RCFs

Types of RCFs	Uncarbonated RCFs	Dry carbonated RCFs	Wet carbonated RCFs				
Density [Mg/m <sup>3</sup> ]	2.46	2.45	2.57				

Figure 3 shows the XRD patterns of the uncarbonated, dry carbonated, and wet carbonated RCFs. Regarding the uncarbonated RCFs, the peaks of cement hydrates such as calcium hydroxide and ettringite appeared; however, these peaks disappeared after carbonation. This indicates that cement hydrates reacted with  $CO_2$  to form calcium carbonate, whereas and after carbonation, cement hydrates did not appear in the RCFs.



Figure 3. XRD patterns of RCFs

## 2.4 Mass of CO<sub>2</sub> fixed in RCFs

The mass of CO<sub>2</sub> fixed in the RCFs was investigated via TG/DTA. The relationship between the change in the sample weight loss and temperature can be determined via TG/DTA. In this study, weight loss occurred at temperature from 550 °C to 950 °C owing the decomposition of calcium carbonated (CaCO<sub>3</sub>), and the percent weight loss in this temperature range was regarded as the mass percentage of fixed CO<sub>2</sub> (Chang & Chen, 2006; Wu et al., 2022). Figure 4 shows the TG curves of uncarbonated, dry carbonated, and wet carbonated RCFs. Their weight loss percentages in this temperature range were 8.15%, 11.12% and 10.93%, respectively. RCFs had already fixed 8% CO<sub>2</sub> by mass before the carbonation process, and after carbonation, the mass of CO<sub>2</sub> fixed in RCFs increased by 3%; hence using carbonated RCFs, approximately 10% of CO<sub>2</sub> by mass can be fixed, including CO<sub>2</sub> fixed prior to carbonation. Uncarbonated RCFs had already absorbed CO<sub>2</sub> because it reacted with atmospheric CO<sub>2</sub> in the months following production, and limestone aggregates can be used in demolished concrete.



Figure 4. TG curves of RCFs

## 3 SOIL-CEMENT COLUMN

#### 3.1 Test specimens

To investigate the applicability of carbonated RCFs for soil stabilization, test specimens with simulated soil-cement columns were prepared. The type of cement used in this study is Portland blast-furnace slag cement type B (based on the Japanese Industrial Standard), the mass of cement per unit volume (1 m<sup>3</sup>) of soil was set to 300 kg, and the water-cement ratio was set to 1.0 for all specimens. The sandy soil used in the experiment was composed of silica sand and kaolin clay at a mass ratio of 9:1 with a water content of 18%. All specimens were cylindrical with a radius of 50 mm and a height of 100 mm.

Table 3 shows mix proportion and mass of each specimen. The effects of the type and amount of carbonated RCFs added to the soil–cement column on the physicochemical properties was investigated. In addition, to investigate the effect of reduced water–powder ratio due to powder addition on the strength of the soil–cement column, specimens with sandstone powder, which featured low reactivity and a particle size distribution similar to that of the RCFs, were prepared. Uncarbonated and carbonated RCFs, as well as sandstone powder were added at 30%, 60% and 90% of the cement mass. Mass of specimen was measured on specimens which had been cured for 4 weeks by balance with 0.01 g sensitivity.

Case	Mass of cement [kg/m <sup>3</sup> ]	Mass of water [kg/m <sup>3</sup> ]	Types of added powder	Mass of powder [kg/m³]	Mass ratio of powder to cement	Mass of specimen [g]
1	300	300	-	-	-	378.34
2	300	300	Uncarbonated RCFs	90	0.3	385.43
3	300	300	Uncarbonated RCFs	180	0.6	387.18
4	300	300	Uncarbonated RCFs	270	0.9	388.08
5	300	300	Dry carbonated RCFs	90	0.3	382.54
6	300	300	Dry carbonated RCFs	180	0.6	383.75
7	300	300	Dry carbonated RCFs	270	0.9	383.59
8	300	300	Wet carbonated RCFs	90	0.3	382.11
9	300	300	Wet carbonated RCFs	180	0.6	382.75
10	300	300	Wet carbonated RCFs	270	0.9	384.86
11	300	300	Sandstone powder	90	0.3	381.76
12	300	300	Sandstone powder	180	0.6	385.97
13	300	300	Sandstone powder	270	0.9	388.33

**Table 3.** Mix proportion and weight of each specimen

## 3.2 Unconfined compression tests

Figure 5 shows the compressive strength increase ratio compared with Case 1 for each specimen with a curing period of 4 weeks. The compressive strength of the specimens for Case 1, which did not contain any powder, was 5.41 N/mm<sup>2</sup>. The compressive strengths of the specimens containing sandstone

powder were 1.05 to 1.13 times higher than those without powder. In the specimens containing uncarbonated RCFs, the compressive strengths were 1.24 to 1.39 times higher than those without powder. The compressive strengths of the specimens containing dry carbonated RCFs were 1.15 to 1.52 times higher, whereas those containing wet carbonated RCFs were 1.16 to 1.46 times higher than those without powder.



Figure 5. Compressive strength increase ratio compared with Case 1

Based on these results, a greater increase in compressive strength was observed in the specimens containing uncarbonated and carbonated RCFs than in those with sandstone powder, the effectiveness of adding RCFs in increasing the strength of the soil–cement column was confirmed, and an increase in the RCFs ratio increased the strength of the soil–cement column. Focusing on the strength variation with the RCFs type, the difference in strength was insignificant up to a mass ratio of powders to cement of 0.6, and at ratio of 0.9, the specimens containing dry carbonated RCFs indicated the highest strength. These strength variations appeared to be due to changes in the hydration reaction depending on the RCFs type, which resulted in different in the types and amounts of hydrates produced.

## 3.3 Mercury intrusion porosimetry test

To investigate the change in pore distribution of the specimens with added RCFs and sandstone powder, a mercury intrusion porosimetry test was performed on each specimen cured for 4 weeks. Figure 6 shows the cumulative intruded volume and log differential intrusion of each specimen. The cumulative intruded volume, which represents the total pore volume in the specimen, was the highest in the specimens for Case 1, which did not contain any powder, followed by those with sandstone powder and dry carbonated RCFs, wet carbonated RCFs, and uncarbonated RCFs. Meanwhile, studies show that the pores of concrete with a diameter of 0.05 to 0.5 µm decreased owing to hydration (Kwak et al., 2002; Fujikura & Oshita, 2011). The log differential intrusion, which represents the pore size distribution, showed that specimens with RCFs had fewer pores with diameters in this range than specimens without powder. These tests suggest that the addition of RCFs resulted in the formation of more hydrates, which reduced the volume of pores and increased the strength.



Figure 6. Cumulative intruded volume (left). Log differential intrusion (right)

#### 3.4 XRD analysis of soil-cement column

An XRD analysis of soil–cement column specimens were conducted to determine the types of hydrate in each soil–cement specimen. Figure 7 shows the XRD patterns of the specimens with 60% added powder after 4 weeks of curing. Calcium hydroxide, monosulfate, and hemicarbonate dominated in the specimen which did not contain any powder, whereas ettringite, hemicarbonate and monocarbonate dominated in the specimens with uncarbonated and carbonated RCFs, and calcium hydroxide was not observed in the specimens with carbonated RCFs. Based on XRD analysis of RCFs (Figure 3), the carbonated RCFs did not indicate peaks of these hydrates prior to the hydration reaction; hence, it was assumed that other hydrates were generated by the hydration reaction in the soil–cement column.



Figure 7. XRD patterns of soil-cement specimens

Studies shows that ettringite  $(3CaO \cdot Al_2O_3 \cdot 3CaSO_4 \cdot 32H_2O)$  contributes to the strength development of concrete (Gu et al., 1997). In addition, previous study indicates that adding CaCO<sub>3</sub> to cement increases the amount of ettringite produced (Lee et al., 1998). Uncarbonated and carbonated RCFs contained CaCO<sub>3</sub>. Figure 8 shows the mechanism of increase of ettringite. CaCO<sub>3</sub> reacts with monosulfate  $(3CaO \cdot Al_2O_3 \cdot CaSO_4 \cdot 12H_2O)$  to form monocarbonate  $(3CaO \cdot Al_2O_3 \cdot CaCO_3 \cdot 11H_2O)$ , and CaSO<sub>4</sub> (Eq. (1)), and CaSO<sub>4</sub> reacted with monosulfate to form ettringite (Eq. (2)).

 $3CaO \cdot Al_2O_3 \cdot CaSO_4 \cdot 12H_2O + CaCO_3 \rightarrow 3CaO \cdot Al_2O_3 \cdot CaCO_3 \cdot 11H_2O + CaSO_4 + H_2O$ (1)  $3CaO \cdot Al_2O_3 \cdot CaSO_4 \cdot 12H_2O + 2CaSO_4 + 20H_2O \rightarrow 3CaO \cdot Al_2O_3 \cdot 3CaSO_4 \cdot 32H_2O$ (2)



Figure 7. Mechanism of increase of ettringite

The improvement in the strength of the soil–cement column with uncarbonated and carbonated RCFs is attributable to the formation of ettringite. By contrast, calcium hydroxide, which does not contribute to the strength, was detected in the specimens without RCFs.

#### 3.5 Hexavalent chromium dissolution tests

As the RCFs were generated from demolished concrete, the dissolution of heavy metal was a concern. In this study, elution tests for hexavalent Cr, which is high water solubility and toxic, based on the Japan Ministry of Environment Notification No.46 were conducted. Elution tests were conducted on the soil–cement specimens with dry carbonated RCFs after they were cured for 1 week. Table 4 shows the test result; for all cases, the dissolution of hexavalent Cr was less than 0.01 mg/L, which is lower than the environmental standard of Japan (0.05 mg/L).

#### Table 4. Dissolution of hexavalent Cr

	Case5	Case6	Case7	
Dissolution of hexavalent Cr [mg/L]	١	Not detected (less th	nan 0.01)	

#### 4 CONCLUSION

In this study, the physicochemical properties of RCFs generated during the crushing of waste concrete with CO<sub>2</sub> fixed and the applicability of soil–cement stabilization with carbonated RCFs were investigated. The density of RCFs slightly increased after wet carbonation, whereas it did not change after dry carbonation; meanwhile, the particle size distribution did not change regardless of the carbonation condition. The RCFs absorbed CO<sub>2</sub> and fixed 10% of CO<sub>2</sub> by mass. By adding carbonated RCFs, the compressive strength of the soil–cement column was 1.2 to 1.5 times higher than that of a typical soil–cement column with carbonated RCFs indicated a lower pore volume than those without RCFs and a hydrate that enhanced the strength was detected in the specimen. In addition, hexavalent Cr dissolution recorded was lower than the Japanese standard. Hence, we conclude that fine powder generated during the recycling waste concrete with CO<sub>2</sub> fixed can be used as an environmentally friendly soil stabilizer.

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