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Application of Accelerated Carbonation Reaction for Low Alkalinity of Recycled Aggregate

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Abstract

As Construction and Demolition (C&D) debris increases every year, systems have been adopted to compel the use of recycled aggregate made from C&D debris, and the use of recycled aggregate in the construction field has increased. But environmental problems linked to the alkalinity of recycled aggregate have occurred, and a study on approaches to lower the alkalinity of recycled aggregate is needed. It was certified by this study that a large amount of recycled aggregate could be carbonated in the C&D debris midterm-treatment field. As a result, the density and the water absorption of recycled aggregate after carbonation reaction was improved, and pH of recycled aggregate was lowered from over 11 to 9.4. X-ray Diffraction (XRD), Scanning Electron Microscope (SEM), Energy Dispersive Spectroscopy (EDS), and Thermogravimetry/Differential Thermal Analysis (TG/DTA) methods also indicated the carbonation of recycled aggregate.

Keywords : recycled aggregate, alkalinity, accelerated carbonation

1. Introduction

1.1 Necessity and Background

The total amount of construction wastes generated in Korea in 2008 reached 64,400,000 tons and the recycled aggregates produced from this reached about 45,990,000 tons per year[1]. Such an increase in the utilization of recycled aggregate is attributable to the active promotion of incentives for aggregate use and the development of technology to improve the quality of recycled aggregates produced from construction wastes to ensure the effective use of resources and the preservation of the national environment according to the Act on the Promotion of Construction Waste Recycling in 2003. The use of recycled aggregate has increased continuously with the improvement of recycled aggregate quality and the support of such a system.

On the other hand, it was pointed out that use of recycled aggregate for the preservation of the national environment might cause environmental problems, following press coverage reporting that mass deaths of migratory birds were caused by a strong alkali leachate generated from use of recycled aggregates at the horizontal drains installed at the reclamation work site of Sihwa in late October of 2008. Subsequently, the standard on recycled aggregate quality was revised to install drain or collecting well or to use preprocessed recycled aggregate for the reduction of alkali in regions where the spill of alkali water was

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suspected if recycled aggregate would be used[2].

Recycled aggregate has cement paste or mortar attached on its surface or contains them in powder in large quantities. This cement consists of calcium -silicate-aluminate compound series, and since it creates high alkali calcium hydroxide [$Ca(OH)_2$] by reaction of hydration, the mechanism[3] showing that the pH of the solution eluted from the recycled aggregate shows strong alkali of 11 ~ 12 is generally known. Accordingly, alkalinity can be reduced if $Ca(OH)_2$ is reduced. Alkalinity can be reduced by converting $Ca(OH)_2$ to calcium carbonate($CaCO_3$) using the carbonization reaction of concrete[4].

Figure 1. Recycled aggregate shape[5]

Since the technology of absorbing and converting carbon dioxide (CO_2) to recycled aggregate accords with CO_2 reduction policy and global issues, the ripple effects are expected to be tremendous if such technology is developed.

1.2 Purpose

This study is about the technology to reduce the chemical alkalinity of recycled aggregate using an accelerated carbonation reaction produced through CO_2 gas. This study aims to recognize the carbonation effects of a recycled aggregate acceleration carbonation device by installing recycled aggregate acceleration carbonation devices at recycled aggregate production sites, and then comparing and analyzing the physical and chemical features of recycled aggregate before and after the carbonation reaction.

2. Experiment Plan

2.1 Overview of Alkali Reduction Process of Recycled Aggregate

The carbonation process to reduce the alkalinity of recycled aggregate applied in this study can be largely divided into a process in the rotating mixer type chamber (reactor) that induces the pH reaction of the recycled aggregate and a process in the CO_2 circulation equipment configured to supply, store and use CO_2 in circulation.

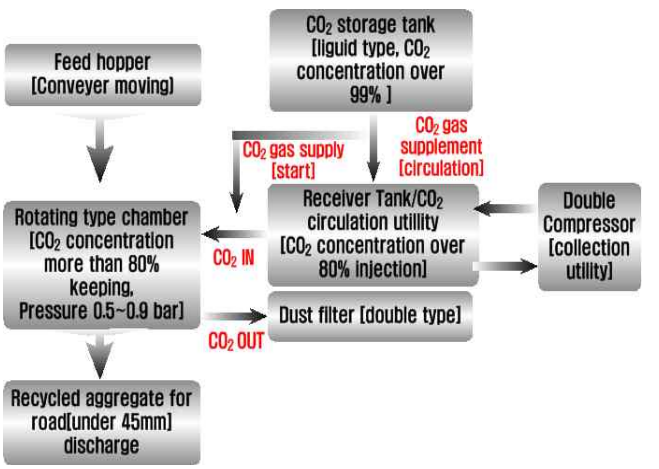


Figure 2. Recycled aggregate carbonation process

Reaction equipment includes CO_2 sensor and pressure gauge that measure CO_2 density and pressure, as well as switchgear that seals to keep the pressure of reactor. Circulation equipment can be divided into air compressor, dust collector, and receiver tank. CO_2 storage and supply equipment consists of 4.9 t capacity liquefied gas storage equipment.

The process is as follows: put recycled aggregate

in the rotating mixer type chamber → seal the rotating mixer type chamber → discharge air in the rotating mixer type chamber(-0.5 bar) → CO_2 injection · carbonation reaction (5 min, keep CO_2 density over 80 %, repeat pressure of 0.5~0.9 bar, rotate 15 rpm) → CO_2 gas recovery(move and store circulation device) → aggregate discharge process.

This device sets the 1 time carbonation time to up to 5 minutes for continuous carbonation reaction process of recycled aggregate. 1 time processing capacity was set to 3.4 t for maximum processing of 250 t per day.

Figure 3. Recycled aggregate pH reducing facility

2.2. Materials and Experiment Methods

Test materials are the recycled aggregates that were produced from construction wastes through a 2nd crushing for the construction of a road less than 40mm thick by an intermediate treatment company of construction wastes located at Icheon, Gyeonggi-do.

To analyze the effects of the acceleration carbonation reaction on the pH reduction of recycled aggregate, tests were performed on the recycled aggregate as indicated in Table 1, before and after the carbonation process for alkali reduction processing of Section 2.1.

Density and absorption rate of recycled aggregate were tested on the aggregate with bigger particle size than 5 mm according to KS F 2503 Test Method of Density and Absorption Rate of Coarse Aggregate[6] and with smaller particle size than 5mm according to KS F 2504 Test Method of Density and Absorption Rate of Fine Aggregate[7], since there is no separate test method for road construction.

A pH measurement method for recycled aggregate was presented at Test Method of Waste Pollution Process[8] and Test Method of Soil Pollution Process[9], as announced by the Ministry of Environment. However, according to the foreign study[10], in which pH result differs since the method of pulverizing and measuring specimen differs according to use conditions of the actual granulated recycled aggregate, this study collected 200 g of recycled aggregates with particle size less than 20 mm that have high pH among the recycled aggregate actually used; set the Solid in beaker: Liquid ratio to 1: 2.5, elution time to 10 minutes, and measured the pH of gushout water using pH meter.

For fine analysis, to check the carbonation reaction according to change of chemical components before and after carbonation reaction of recycled aggregate, XRD, SEM/ EDS, and TG/DTA were analyzed targeting carbonation processing recycled aggregate showing the highest pH after carbonation reaction.

Table 1. Test items for recycled aggregate

test items		Test methods	Test number of times
Density (g/㎤)	Under 5㎜	KS F 2504	5
	Over 5㎜	KS F 2503	
Water absorbtion (%)	Under 5㎜	KS F 2504	5
	Over 5㎜	KS F 2503	
pH (hydrogen ion concentration)		pH test of Recycled aggregate less than 20mm	5
Micro analysis		XRD analysis	1
		SEM/EDS analysis	1
		TG/DT analysis	1

3. Experiment Results and Analysis

3.1 Density and Absorption Rate

Applicable standards on absorption rate, density and quality standard of recycled aggregate for road subbase have not yet been prepared. To check the density and absorption rate, a test was conducted in accordance with KS F 2503 Test Method of Density and Absorption Rate of Coarse Aggregate and KS F 2504 Test Method of Density and Absorption Rate of Fine Aggregate.

As a result, it was found that density of recycled aggregate before carbonation reaction was 2.02~2.04 g/㎤ for particles less than 5mm, and 2.28~2.30 g/㎤ for over 5mm. Density of recycled aggregate after carbonation reaction was 2.09~2.12 g/㎤ for particles less than 5mm, a 2.9~4.7 % increase in density compared to those before carbonation reaction; and 2.35~2.38 g/㎤ for over 5mm, a 3.0~3.8 % increase in density.

On the other hand, absorption rate of recycled aggregate before carbonation reaction was 11.84~12.10 % for particles less than 5mm, and 6.17~6.66 % for those over 5mm. After carbonation reaction, it was 5.28~5.69 %, which is a 7.6~11.8 % decrease from those before carbonation reaction for aggregate less than 5mm, and 10.67~11.04 %, which is a 7.8~17.9 % decrease from those before carbonation reaction, confirming that the carbonation reaction has a quality improvement effect on the recycled aggregate.

This is because recycled aggregate adhered mortar gets densified, similarly to what has been shown in previous study results[11,12] that calcium carbonate created by carbonation reaction fills the pores of mortar and raises the watertightness.

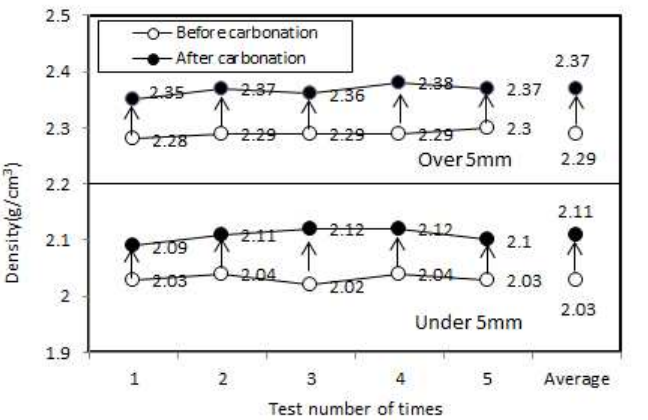


Figure 4. Density change of recycled aggregate before and after carbonation

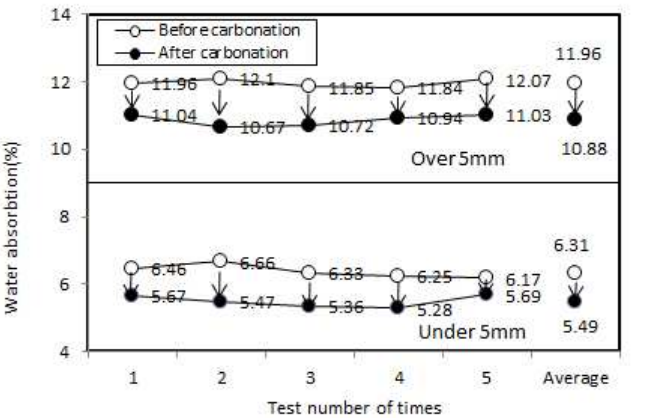


Figure 5. Water absorbtion change of recycled aggregate before and after carbonation

3.2 Recycled Aggregate pH

Cement element adhered to or mixed into recycled aggregate in the powder type shows strong alkalinity. Since it may create alkali leachate and affect the surrounding environment if it reacts with water, it is regulated in the recycled aggregate quality standard that generation of alkali leachate should be considered in the use of recycled aggregate.

In the carbonation reaction test of this recycled aggregate, it was found that pH of recycled aggregate before reaction was 11.6~11.8, strong alkalinity, while that of recycled aggregate after carbonation reaction was 9.3~9.5, showing that it had pH reduction effect of 18~21 %.

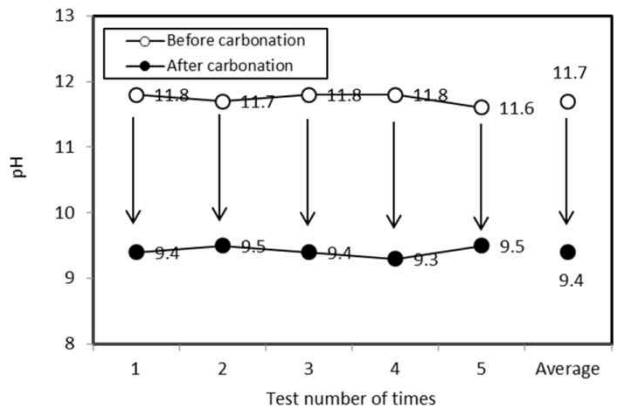


Figure 6. pH change of recycled aggregate before and after carbonation

3.3 XRD Analysis

XRD analysis is the method used to analyze the element of specimen according to X-ray diffraction. Reaction of $Ca(OH)_2$ and degree of $CaCO_3$ creation can be recognized.

Figure 7 shows the results of analysis for recycled aggregate adhered mortar before carbonation reaction. It can be confirmed that peaks of $Ca(OH)_2$ such as 17.9°, 34.1° and 47.1° form the main peak. In addition, peak of $CaCO_3$ can be observed in recycled aggregate adhered mortar before carbonation reaction, which means that most concretes are over 20 years, carbonation has been in progress from the surface and $CaCO_3$ is extracted since such parts are included.

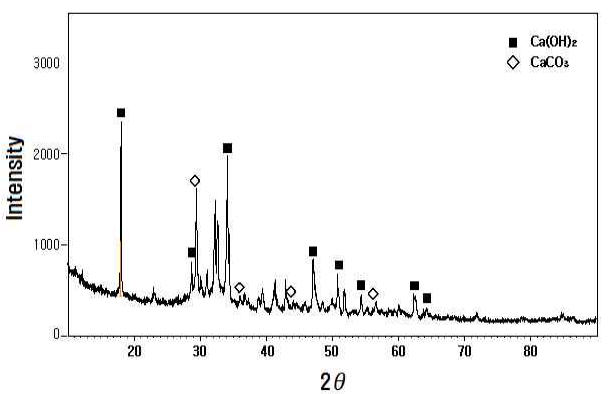


Figure 7. Recycled aggregate XRD peak before carbonation

Figure 8 is the result of analysis on recycled aggregate adhered mortar after carbonation reaction. It is clear that peaks of $CaCO_3$ are found at 29.6°, 33.8°, 43.5° and that $Ca(OH)_2$ also exists in part. Accordingly, an increase in $CaCO_3$ and a decrease in $Ca(OH)_2$ is evident as a result of the carbonation processing.

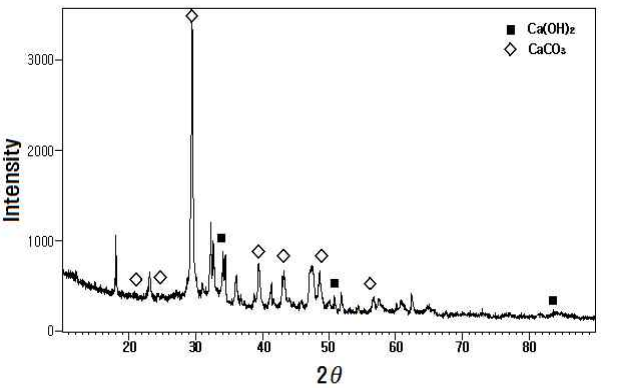


Figure 8. Recycled aggregate XRD peak after carbonation

3.4 SEM/EDS Analysis

SEM analysis and EDS were conducted for the adhered mortar before and after carbonation reaction.

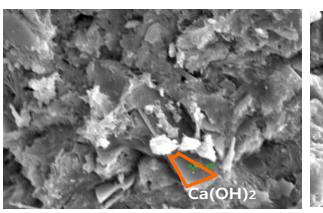


Figure 9. $Ca(OH)_2$ before carbonation

Figure 10. $CaCO_3$ after carbonation

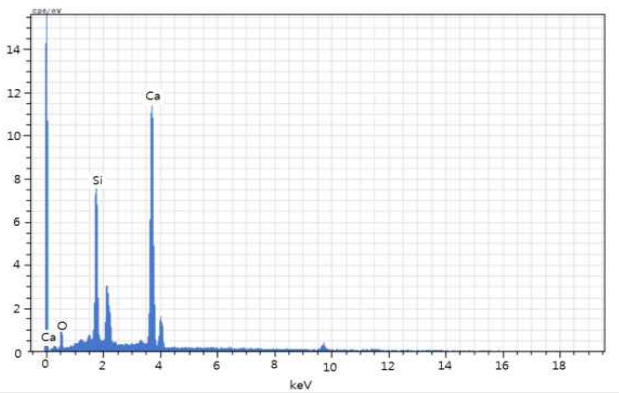
According to Gallucci[13] and De Silva[14], the shape of calcium hydroxide exists in the form of a hexagonal plate. Calcium carbonate has a round and circular pipe shape.

Figure 9 shows the SEM photo of flat-type hexagonal substances of recycled aggregate mortar. The existence of calcium hydroxide hydrate in bulk can be confirmed. Figure 10 shows the SEM photo of recycled aggregate adhered mortar after

carbonation reaction. As shown in the photo, it shows the typical round pipe shape of calcium carbonate.

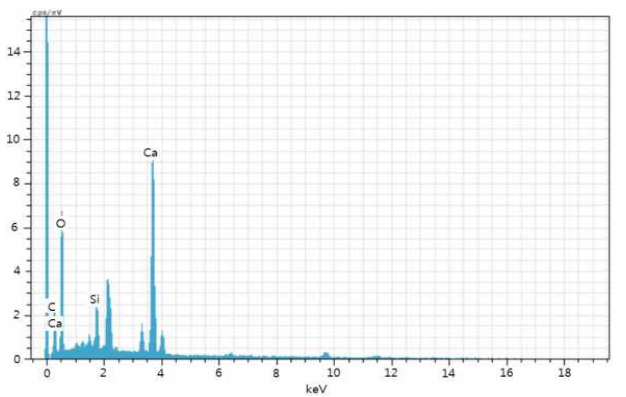
Figure 11 is the result of EDS analysis for recycled aggregate adhered mortar before carbonation reaction. In theory, $Ca(OH)_2$ consists of 54 % Ca, 43 % O, and 3 % H. The analysis showed that Ca content is 63 % and O content is about 15 % so it is determined to be $Ca(OH)_2$, not carbonated.

Figure 12 is the result of EDS analysis for recycled aggregate adhered mortar after carbonation reaction. In theory, $CaCO_3$ consists of 40 % Ca, 48 % O and 12 % C. The analysis of the substance estimated to be calcium carbonate showed O 48 %, Ca 38 %, C 6 %, the unique properties of calcium carbonate.



Ingredient	O	Ca	C	K	mSi
wt, %	15.25	63.31		1.47	18.72

Figure 11. EDS analysis result before carbonation



Ingredient	O	Ca	C	K	Si
wt, %	47.31	38.04	6.46	3.96	2.81

Figure 12. EDS analysis result after carbonation

3.5 TG/DTA Analysis

According to a previous study [15], $Ca(OH)_2$ shows a heat absorption reaction in the temperature range of 300~500 ℃, and the thermal weight of $CaCO_3$ reduces as the evaporation by heat occurs in the temperature of 500~80 ℃.

As shown in Figure 13, for recycled aggregate adhered mortar before carbonation reaction, heat absorption reaction by calcium hydroxide occurs at around 490 ℃, so it is confirmed that calcium hydroxide exists in large quantities in mortar. It is also shown that a tiny quantity of calcium carbonate is dissolved and CO_2 evaporated at about 700 ℃.

Recycled aggregate adhered mortar after carbonation in Figure 14 showed no specific reaction in the temperature range of 450~500 ℃, so $Ca(OH)_2$ was not detected. It was found that there was a weight decrease of 4.2 % after 700 ℃, so calcium carbonate was extracted.

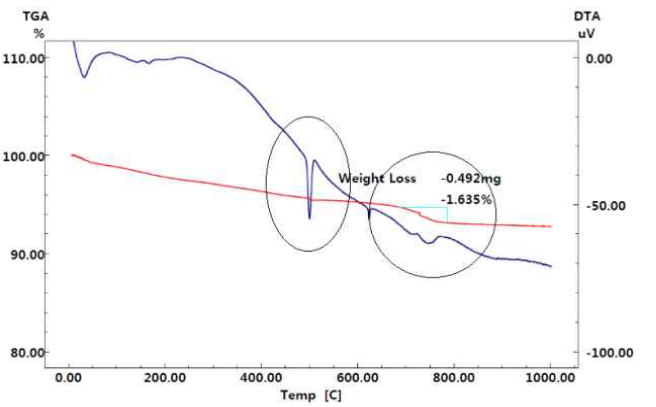


Figure 13. TG/DTA result before carbonation

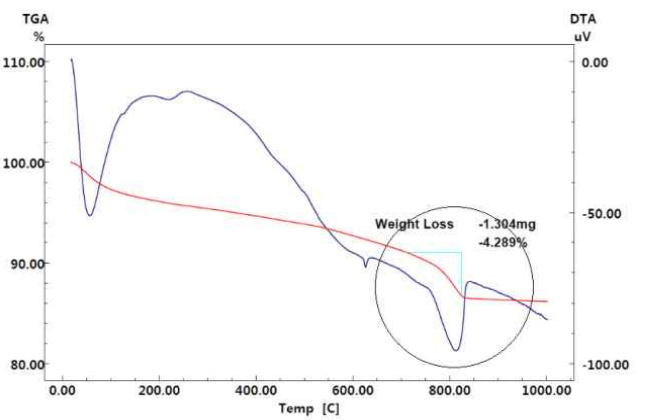


Figure 14. TG/DTA result after carbonation

4. Conclusion

By utilizing the acceleration carbonation reaction for pH reduction of recycled aggregate, the following conclusions were obtained.

1) It was confirmed that recycled aggregate could be neutralized using neutralization equipment (1 time reaction amount of 3.4 t, reaction time of 5 minutes, CO_2 density over 80 %) installed to reduce the strong alkalinity of recycled aggregate produced in large quantities by an intermediate treatment company.
2) Recycled aggregate could improve density, absorption reduction and quality by reducing the pore through the creation of calcium carbonate by carbonation reaction with adhered mortar, the cause of quality decline.
3) By measuring pH using the method to measure pH of recycled aggregate not pulverized in consideration of use conditions, it was shown that the average pH 11.7 of recycled aggregate can be reduced to pH 9.4 through accelerated carbonation reaction.
4) By analyzing XRD, SEM, EDS, TG/DTA to check the carbonation reaction of recycled aggregate, it was confirmed that the carbonation reaction of recycled aggregate was surely executed.

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