

# Study on the Effect of Carbonation of Various Low Quality Recycled Fine Aggregates for Modification and Properties of Mortar

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**Abstract.** The use of recycled aggregate is desirable to realize a carbon-neutral and sustainable society. In addition, the widespread use of concrete using large amounts of recycled aggregate that can absorb CO<sub>2</sub> will contribute to reducing the environmental impacts. The authors have proposed modification of recycled aggregate by accelerated carbonation technology, however it have not conducted many studies on the type of fine aggregate, which is considered to greatly affect the properties of concrete. Therefore, in this study, accelerated carbonation was performed using low-quality recycled aggregate with various properties to confirm its modification effect. As a result, it was confirmed that the finer the particle size, such as fine grains in the aggregate, the higher the adhered paste ratio, and thus the greater the CO<sub>2</sub> absorption. In addition, the physical properties of the mortar using this carbonated aggregate were measured, and it was proven that the tendency of mortar improvement was higher when fine aggregate with a high modification rate was used. In addition, in the results of trying various modification methods, it became clear that the key point is to modify the fine particle in the aggregate.

**Keywords:** Recycled aggregate, Accelerated carbonation technology, Modification of recycled technology.

## 1 Introduction

Low-quality recycled aggregate concrete should be widely used to build a resource-recycling society. However, low-quality recycled aggregates have limited application in concrete due to the low quality of the aggregate itself. In particular, there are very few applications of low-quality recycled fine aggregate. On the other hand, technologies to absorb and fix CO<sub>2</sub> in concrete and its materials are being developed with a view to achieving carbon neutrality in 2050<sup>1)2)</sup>. Recycled aggregates can utilise the CO<sub>2</sub> absorbed and fixes during the service life of concrete structures before demolition, and

the expose of new concrete fracture surfaces by crushing during demolition can absorb  $\text{CO}_2$ . Recycled aggregates can make a significant contribution to carbon recycling as one of the CCU (Carbon dioxide Capture and Utilisation) materials. Low-quality recycled fine aggregates are considered suitable for CCU materials because they have a large potential to absorb and fix  $\text{CO}_2$  due to the large proportion of mortar and cement paste (adhered mortar and adhered paste) attached to and mixed with the aggregate. Against the above background, for the future spread of recycled aggregate concrete technology is needed to improve the quality of the recycled aggregate itself (modification) as well as to convert low-quality recycled aggregate into CCU material. Therefore, one of the authors suggested a modification technique for recycled aggregate by accelerated carbonation<sup>3)</sup>. In the future, it is necessary to investigate methods for effectively modifying low-quality recycled fine aggregate by carbonation treatment. In this study, accelerated carbonation was carried out using various low-quality recycled aggregates and the influence of aggregate properties on the modification effect was investigated. Secondly, the effect of different periods and methods of carbonation treatment on the modification effect of low-quality recycled aggregate and the relationship between the strength and air permeability improvement rate of hardened mortar using these low-quality recycled fine aggregates were investigated.

## **2 Outline of the experiment**

### **2.1 Carbonation treatment methods.**

The carbonation method used in this experiment is accelerated carbonation. The carbonation treatment was carried out in an accelerated carbonation chamber at temperature of  $20^\circ\text{C}$ , a relative humidity of 60% and a  $\text{CO}_2$  concentration of 5%. During accelerated carbonation, the aggregate was stirred once a day to ensure that  $\text{CO}_2$  was distributed throughout the aggregate. accelerated carbonation periods of 7 days (acc 7D), 14days (acc 14D), 28 days (acc 28D) and 56 days (acc 56D) were carried out to determine the effect of the period of accelerated carbonation on the modification effect of the recycled fine aggregate. Additionally, to further enhance the modification effect of accelerated carbonation, recycled fine aggregate impregnated with calcium hydroxide solution (CH) and CH impregnated with accelerated carbonation for 7 days (CH 7D) were also conducted. This is because the calcium hydroxide solution penetrates the voids in the attached paste, and the impregnated calcium hydroxide, in addition to the calcium hydroxide present in the attached paste, is carbonated, which is thought to enhance the modification effect.

## 2.2 Recycled fine aggregate.

Table-1 shows the physical properties of the recycled fine aggregate used in this study. The raw concrete was all demolished concrete. The source of the demolished concrete is confirmed to be the building structures. This leads to assumption that the cement type of the original concrete of the recycled aggregate is Ordinary Portland Cement (OPC). The original concrete aggregate for **a** to **d** was confirmed to be crushed aggregate from visual inspection of the original concrete. For **e**, it was confirmed that limestone aggregate may have been used as part of the fine aggregate, and for **f**, it was confirmed that limestone coarse aggregate was used. The quality categories of the recycled fine aggregate used were those according with the standard of Annex A of JIS A 5023 (Recycled aggregate concrete L) and those whose water absorption deviated from that standard. The specific surface area and attached paste ratio are not specified in JIS but were measured because they are indices related to CO<sub>2</sub> absorption and fixation. The specific surface area was calculated from the total surface area per unit volume of recycled fine aggregate; the surface area was determined from sieve tests. The attached paste ratio was obtained from hydrochloric acid dissolution. However, as limestone aggregate may have been used for **e** and **f**, these were estimated from the relationship between the percentage of attached paste and water absorption in **a-d**. The specific surface area was the largest for **c** and the attached paste ratio was the largest for **d**.

**Table 1.** Physical properties of the recycled fine aggregate

No.	Drying-density (%)	Water absorption rate (%)	Fine particle fraction (%)	Coarse-grained rate (FM)	Specific surface area (cm <sup>2</sup> /g)	Attached paste ratio (%)	Lime stone
<b>a</b>	2.04	10.42	3.30	3.64	57.2	33.7	-
<b>b</b>	1.94	13.44	<b>12.2</b>	3.37	83.2	36.7	-
<b>c</b>	2.00	12.01	8.6	3.11	<b>104.1</b>	34.4	-
<b>d</b>	1.94	13.24	5.58	3.20	77.5	<b>39.4</b>	-
<b>e</b>	2.04	10.54	7.10	3.43	70.6	33.3	○
<b>f</b>	1.94	13.03	6.90	3.45	72.0	37.2	○

## 3 Modification of recycled aggregates

### 3.1 Effects of carbonation methods on the modification effect of recycled fine aggregate.

The effect of the carbonation method on the modification effect of the recycled fine aggregate was checked using the **d** aggregates listed in Table-1. Figure-1 shows the density and water absorption of the recycled aggregate before and after carbonation.

The modification effect tended to increase with increasing carbonation duration. Comparing the percentage improvement in dry density and water absorption between acc 7D and acc 56D, the modification effect tended to increase as the duration of carbonation treatment increased. However, no difference was found between the modification effects of acc 28D and 56D. The improvement in dry density and water absorption of CH 7D was 5.1% and results showing a greater modification effect than acc 7D. This is due to the carbonation of the impregnated calcium hydroxide, which is considered to have a greater modification effect than acc 7D. These results indicate that the modification effect can be increased by extending the carbonation period of the recycled fine aggregate. In addition, the results also show that the modification effect can be further improved by CH impregnation.

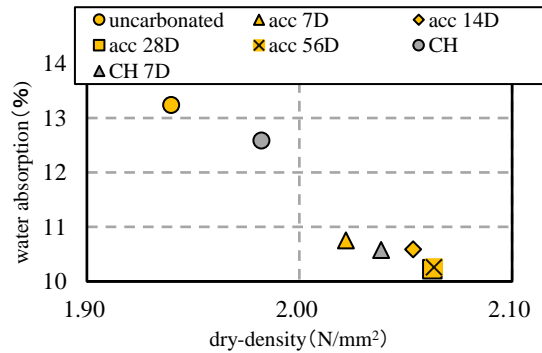


Figure-1. Density and water absorption of the recycled aggregate

### 3.2 Effects of modification of recycled aggregate by accelerated carbonation.

As the accelerated carbonation period was set at 7days in 3.1, this chapter considered the effects of modification of recycled aggregate by 7days of accelerated carbonation. Figure-2 shows the relationship between density and water absorption of recycled fine aggregate before and after accelerated carbonation. By accelerated carbonation, density and water absorption were improved in all aggregates. The highest modification effect, **c**, improved the dry density and water absorption about twice as much as the lowest modification effect, **e**. Comparing these aggregate properties, there is no significant difference in the attached paste ratio, but considering that **c** is the aggregate with the highest specific surface area of all the aggregates used and that the specific surface area of **e** is not so large, the effect of the aggregate particle size can be considered. Therefore, Figure-3 shows the attached paste ratio for each aggregate particle size. The attached paste ratio increased with decreasing aggregate particle size. These results suggest that if there is no significant difference in the amount of paste attached to the aggregate, the aggregate with a smaller grainsize has a larger amount of paste attached to and a larger specific surface area, which makes the modification effect by accelerated carbonation.

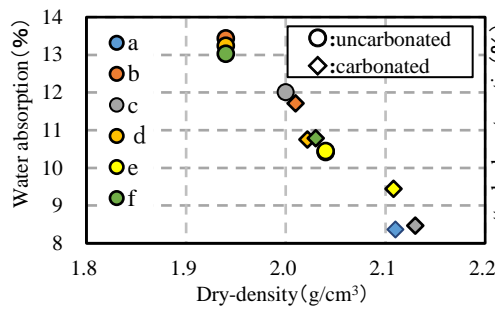


Figure-2. Relationship between density and water absorption

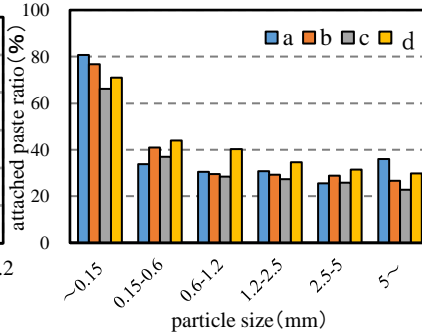


Figure-3. Attached paste ratio

### 3.3 *CO<sub>2</sub> absorption of recycled fine aggregate by carbonation treatment.*

It was found that the modification effect differs depending on the particle size of the recycled fine aggregate and the carbonation treatment method. The CO<sub>2</sub> absorption of **d** aggregate was measured, as the CO<sub>2</sub> absorption was related to these differences in the modification effect. CO<sub>2</sub> absorption was determined by differential thermogravimetric analysis (TG-DTA). Samples were taken by randomly, weighing 500 g. This was used as a representative sample. The samples were all milled to less than 150  $\mu$ m using a grinding machine and used for the measurements. However, only CH 7D was finely milled in quantities of less than 500 g and used as a sample. Throughout the measurement, nitrogen gas flowed in at a rate of 150-200 mL/min. The temperature range for the mass loss in the calcination reaction was assumed to be 550-850 °C. The ratio of the mass loss to the sample mass was subtracted before and after carbonation to give the CO<sub>2</sub> absorption. Figure-4 shows the CO<sub>2</sub> absorption of recycled fine aggregate. Comparing different carbonation periods, a longer carbonation period resulted in an increase in CO<sub>2</sub> absorption. The CH 7D resulted in greater CO<sub>2</sub> uptake than the acc 7D. This is thought to be due to the carbonation of the impregnated calcium hydroxide as well as the results of aggregate modification. Based on these results, the modification effect of recycled fine aggregate is related to the amount of CO<sub>2</sub> absorption.

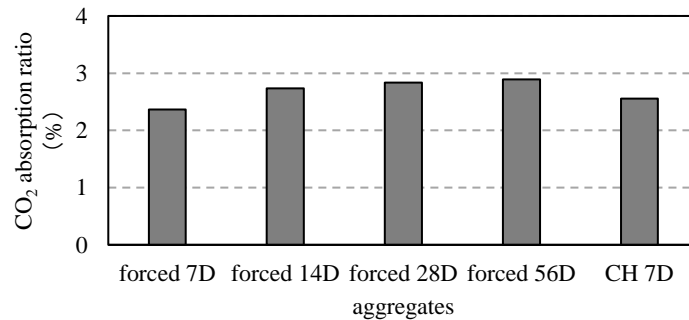


Figure-4. CO<sub>2</sub> absorption of recycled fine aggregate [**d** agg.]

## 4 Investigations in mortar with recycled fine aggregate.

### 4.1 Outline of the experiment.

In the preparation of mortars with low-quality recycled fine aggregate, the mix proportion with a cement to fine aggregate ratio of 1:3 mortar with 50% W/C and OPC was used. Compressive strength test and air permeability test were conducted. For strength tests, mortar bar specimens of  $40 \times 40 \times 160$  mm were prepared and tested in accordance with JIS R 5201 after 28 days standard curing. For the permeability tests, cylindrical specimens of  $\phi 100 \times 25$  mm. were prepared, and standard curing was carried out for 28 days. After curing, the specimens were placed in a drying chamber at  $40^\circ\text{C}$  and 30% relative humidity until their mass became constant, and then air was permeated through them at a pressure of 0.1 MPa using a permeability tester. The permeability was measured by the water displacement method and the permeability was calculated.

### 4.2 Effect of accelerated carbonation on improving the quality of mortar.

Mortar specimens were prepared using low-quality recycled fine aggregate before and after accelerated carbonation as shown in Table-1. Figure-5 shows the compressive strength and the permeability of mortar specimens before and after accelerated carbonation. Accelerated carbonation improved compressive strength for all types. The improvement rate for each aggregate type was the highest for c with 67.2%. On the other hand, the smallest improvement rate, e, was 5.8%, which was similar to the trend for aggregate modification. Accelerated carbonation improved the permeability for all types. These results suggest that the improvement in strength and permeability of the mortar specimens was related to the  $\text{CO}_2$  absorption, as the trend was close to that of the aggregate modification.

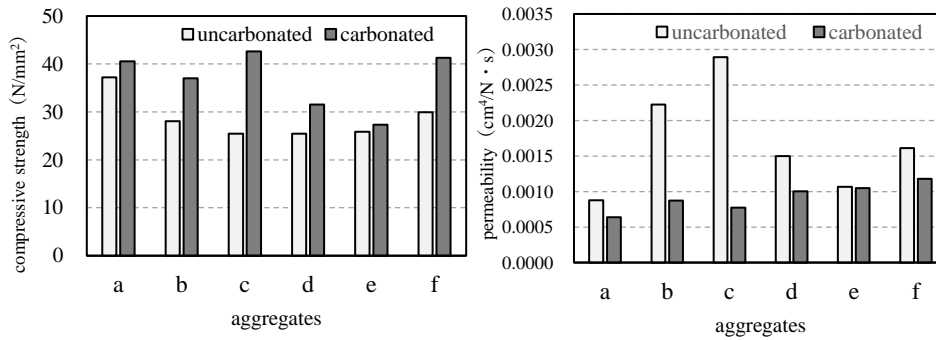


Figure-5. Compressive strength and the permeability of mortar specimens

#### 4.3 Effect of accessibility to CO<sub>2</sub> absorption and fixation on the strength and air permeability of mortar.

According to the results of this study, the CO<sub>2</sub> absorption of the recycled fine aggregate and the associated aggregate modification are responsible for the improvement of the quality of the mortar specimens. Therefore, the aggregate particle size was adjusted to change the ease of CO<sub>2</sub> absorption and fixation using c aggregate and compared the improvement in strength and permeability of the mortar. Aggregate particle sizes were sorted into five categories with the particle sizes adjusted to achieve a mass of 20% of each. Table-2 shows the amount of paste in each particle size range. In the grain size-adjusted samples (1) to (5), the ease of CO<sub>2</sub> absorption and fixation was varied by partially accelerated carbonation of the respective particle size ranges, thereby changing the amount of attached paste and surface area. Figure-6 shows the compressive strength and permeability of the mortar specimens. Compressive strength was increased for all types compared to the uncarbonated specimens. For permeability compared to the uncarbonated specimens, the permeability tended to improve for all types. The results for both compressive strength and air permeability were particularly improved for the specimens in (1). The improvement trend of the permeability was greater when carbonating the smaller particle size range. These are related to the ability to absorb and fix CO<sub>2</sub>, which may have influenced the improvement in compressive strength and permeability. In conclusion, it was found that carbonation treatment effectively improves the quality of the mortar by modifying the aggregate in areas where the amount of attached paste and the specific surface area are large.

Table-2. The amount of paste in each particle size range

No.	(1)	(2)	(3)	(4)	(5)
Particle size range(mm)	0~0.15	0.15~0.6	0.6~1.2	1.2~2.5	2.5~5.0
Amount of paste(kg/m <sup>3</sup> )	185	104	80	76	72

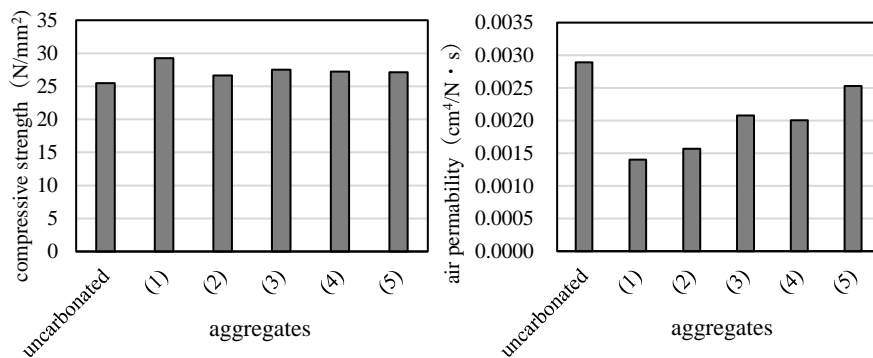


Figure-6. Compressive strength(left) and air permeability(right)

## Conclusion

1. Aggregates with smaller particle sizes have a larger specific surface area of adhered paste, which facilitates the absorption and fixation of CO<sub>2</sub>, and thus the modification effect of accelerated carbonation is greater when there is no significant difference in the amount of paste adhered to the aggregate.
2. The modification effect of recycled coarse aggregate can be improved by extending the period of accelerated carbonation of the aggregate and by CH impregnation.
3. The strength of the mortar can be improved by modifying the physical properties of the recycled fine aggregate through CO<sub>2</sub> absorption by the carbonation treatment, which tends to improve the strength of the hardened body.

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