Experimental Study on CO_2 Curing for Enhancement of Recycled Aggregate Properties

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ABSTRACT

In this paper, the results of an experimental program on studying the use of a carbonation process to enhance the properties of recycled aggregates are presented. Hardened concretes prepared by using different water-to-cement ratios in the laboratory, were crushed to produce recycled aggregates with different particle sizes. Before and after the laboratory carbonation process, the physical properties of the recycled aggregate, including water absorption and density were determined. The extent of CO_2 curing of the recycled aggregate was quantified by assessing the carbonation percentage of the aggregates. Carbonation resulted in reduction in water absorption values and increase in density and these showed that the properties of recycled aggregate were improved. The factors influencing the CO_2 curing process, including curing time, particle size and moisture contents of recycled aggregate, were investigated.

Keywords: recycled aggregate; CO_2 curing; carbonation; mechanical properties

1. INTRODUCTION

To achieve sustainable development in the construction industry, there is a need to recycle and reuse construction waste derived from demolition of old concrete structures as secondary aggregates for producing new building materials. However, due to the adhered old cement mortar, the quality of recycled concrete aggregate (RCA) has been widely reported to be of inferior to that of natural aggregates (Liu, Xiao & Sun 2011, Poon, Chan 2007). In consideration of the constituent of the old cement mortar adhering to the surface of RCA, improving the low quality of RCA through accelerated carbonation is possible to some extent because the calcium hydroxide, which is one of the main cement hydration products in the old cement mortar adhering to the surface of RCA, can react with carbon dioxide accompanied by an increase in solid volume(Johannesson, Utgenannt 2001), which is formulated by the following reaction:

$$Ca\,(OH)_2 + CO_2 = CaCO_3 + H_2O \qquad\qquad\qquad (1)$$

The other hydration products, such as calcium silicate gel, may also be carbonated and decalcified to generate the $CaCO_3$ (Yousuf et al. 1993). During the carbonation process, the precipitation of calcite ($CaCO_3$) in pore space can decrease the porosity of the matrix, thus strengthen the weak surface and lower the water absorption of the porous cement mortar (Lange, Hills & Poole 1995)(Walton et al. 1997).

This study presents an innovative pretreatment method to enhance the properties of RA by using a CO_2 curing process. Different from the conventional pretreatment methods for recycled aggregates, which normally try to remove the attached cement mortar, the CO_2 curing pretreatment aims to densify the old cement mortar and then strengthen the RA through a well known carbonation reaction between CO_2 and cement hydration products.

2. CO_2 CURING EXPERIMENT

2.1 Experimental program and measurement

In this study, four concrete mixes with different water to cement ratios and mix proportions were prepared in the laboratory and used as parent concretes for producing the recycled aggregates (RA). The mixes proportioning are shown in Table 1. After 28-days of water curing, the compressive strength of the parent concretes was determined (shown in Table 1). After 90-days of water curing, all the parent concrete samples were crushed manually and sieved to

produce the recycled aggregates with particle sizes ranging from 5 mm to 20 mm. In this paper, each kind of recycled aggregate was notated with RA followed by the strength grades of the parent concretes.

Table 1 Details of parent concrete

	Water, Kg/m^3	Cement (ASTM Type I) , Kg/m^3	Sand(<5mm), Kg/m^3	Crushed granite (10-20mm), Kg/m^3	28d Compressive strength, MPa
C30	205	300	697	1143	30.2
C45	180	350	706	1158	48.3
C60	185	425	696	1092	61.6
C80	165	485	685	1094	74.0

Before carrying out the CO_2 curing, the apparent density and water absorption of each kind of RA were determined according to BS EN 1097-6, the results of which are tabulated in Table 2. Additionally, the porosity values of the RA (p) were determined as a ratio of pore volume (V_p) to the bulk volume of the RA (V):

$$p = V_p / V \tag{2}$$

The pore volume of RA (V_p) was determined from the difference between the saturated surface dry (M_{ssd}) and oven-dry (M_{od}) masses:

$$V_p = \frac{M_{ssd} - M_{od}}{\rho_w} \tag{3}$$
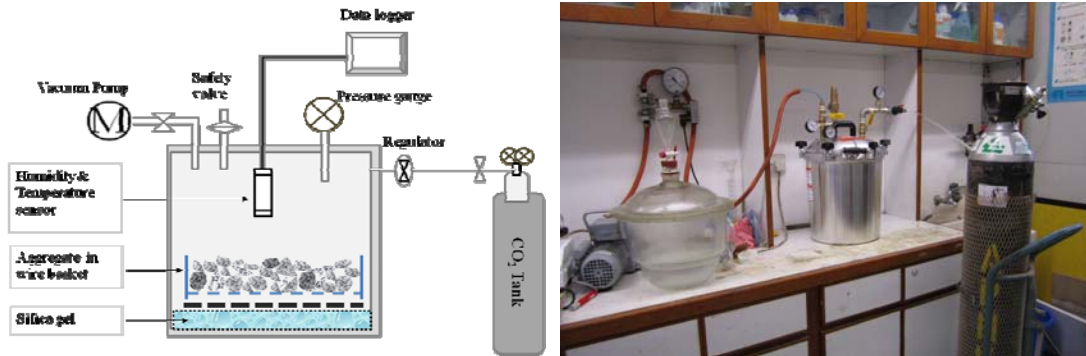
While the bulk volume of RA (V) was calculated as:

$$V = \frac{M_{ssd} - M_{sub}}{\rho_w} \tag{4}$$

Where, ρ_w means water density; M_{sub} means mass of saturated samples submerged in water.

Table 2 Physical properties of RA before CO_2 curing

	Apparent density, kg/m^3	Water absorption, %	Porosity, %
RA30	2639	6.58	15.66
RA45	2623	5.98	14.30
RA60	2620	5.95	14.23
RA80	2602	5.81	13.84

Figure 1. Experimental setup of CO_2 curing

The laboratory CO_2 curing setup is illustrated in Fig. 1. An air-tight cylindrical vessel was used as the curing chamber, which was vacuumed to -0.5 bar before the pure CO_2 gas injection. The CO_2 pressure in chamber was controlled by a regulator and kept at ＋0.1bar. Considering the adverse impact of high humidity on carbonation, anhydrous silica gel was put inside the chamber to remove the evaporated water from the specimens during the carbonation process.

After 24 hours of CO_2 curing, the apparent density, water absorption and porosity of carbonated RA were determined again, as the results are shown in Table 3. Compared to the results shown in Table 2, there were slight variations in apparent density before and after the CO_2 curing; while the water absorption and capillary porosity of RA derived from different strength grades of parent concretes were reduced. This was the result of carbonation products filling the pores as the carbonation of calcium hydroxide, which is one of the main reactions, is accompanied by an 11.8% increase in solid volume (Johannesson, Utgenannt 2001). The other cement hydration products can also react with CO_2 leading to solid phases precipitation in the pores.

Table 3 Physical properties of RA after CO_2 curing

	Apparent density		Water absorption		Porosity, %
	Mean, kg/m^3	Std Dev	Mean,%	Std Dev	
RA30	2619	3.45	5.25	0.42	12.68
RA45	2606	7.71	4.71	0.19	11.65
RA60	2670	3.77	4.61	0.33	11.18
RA80	2661	5.66	4.41	0.11	10.94

Obviously, the extent of the carbonation reaction and improvement in physical properties of RAs were interrelated. To quantify the carbonation degree of RA after the CO_2 curing process, carbonation percentage (ε), which was defined as the ratio of the experimental determined mass gain to the theoretical mass gain of RA by the carbonation reaction:

$$\varepsilon = \frac{\Delta M_e}{\Delta M_t} \times 100\% \tag{5}$$

Where, ΔM_e and ΔM_t represent respectively the experimental mass gain and theoretical mass gain of RA. ΔM_e was determined by weighing the RA samples before and after the CO_2 curing; while ΔM_t was the maximum theoretical uptake of CO_2 by the cement mortar adhered to RA, and was calculated by:

$$\Delta M_t = \frac{M_c \times CO_2\%_{max}}{(M_c + M_s + M_a + M_c \times 0.23)(1 + Q)} \tag{6}$$

Where, M_c, M_s and M_a are the proportions (by mass) of cement, sand and coarse aggregate in the parent concrete respectively; Q represents the moisture content of RA before carbonation; $CO_2\%_{max}$ represents the maximum theoretical CO_2 captured by Portland cement, which can be calculated based on the oxide contents of cement using the following formula (Steinour 1959):

$$CO_2\%_{max} = 0.785(CaO - 0.7SO_3) + 1.091MgO + 1.420Na_2O + 0.935K_2O \tag{7}$$

The theoretical mass gain ratio, which were determined as the mass ratio of mass gain to the mass of carbonated RA, for the different RAs after complete carbonation were calculated and are shown in Table 4.

Table 4 Theoretical mass gain ratio of RA after complete carbonation

Aggregate type	RA30	RA45	RA60	RA80
Moisture content, %	3.04	3.66	3.37	4.27
Mass gain ratio, %	6.92	7.82	9.40	10.53

2.2 Effect of water absorption values of RA on carbonation

After 1 hour of CO_2 curing, the experimental determined mass gain ratio of RA is shown in Fig. 2 (a), while Fig. 2 (b) indicates the carbonation percentage (ε). It should be noticed that RA with higher water absorption values attained a higher mass gain ratio and carbonation

percentage. This is because higher water absorption values of RA means the open porosity inside RA is higher, which can supply an easier access for CO_2 penetrating within the RA. However, the open porosity is only one of the critical factors influencing the carbonation percentage of RA; once most of open porosity inside RA is fulfilled with free water, CO_2 still cannot penetrate into RA easily. In that case, the CO_2 diffusion in saturated RA (concrete) limits the carbonation process of interior part of RA; this is also a reason why carbonation percentage of RA is not directly proportional to the water absorption of RA, as shown in Fig. 2(b).
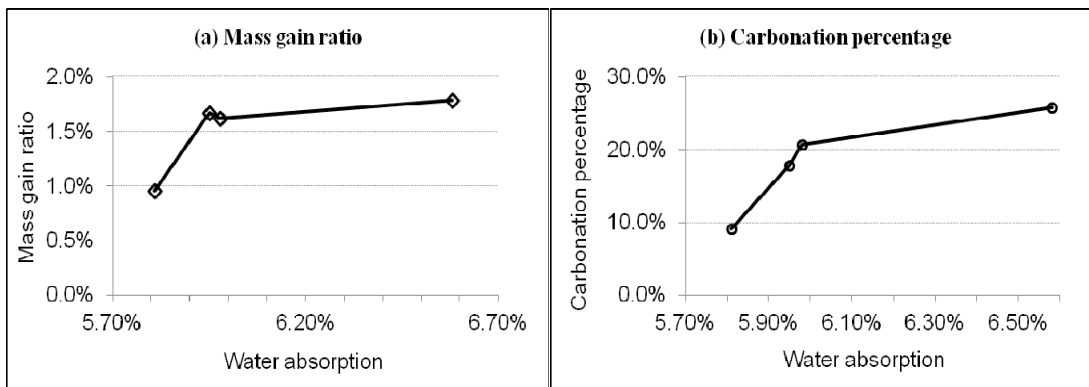
**Figure 2. Experimental mass gain % and carbonation percentage**

2.3 Effect of size of RA on carbonation

RA30 with particle size ranged from 5-10 mm, 10-14 mm and 14-20 mm were used to investigate the effect of particle size of RA on carbonation. After 1 hr of CO_2 curing, the mass gain and the carbonation percentage were determined and are shown in Figs. 3(a) and 3(b) respectively. Obviously, RA with a smaller particle size and a larger specific surface can take up more CO_2 during the CO_2 curing process. The carbonation percentage was up to 56% for RA with a particle size of 5-10 mm and was only 37% for the 14-20 mm RA. This indicates there was a great potential to capture CO_2 with smaller RAs.
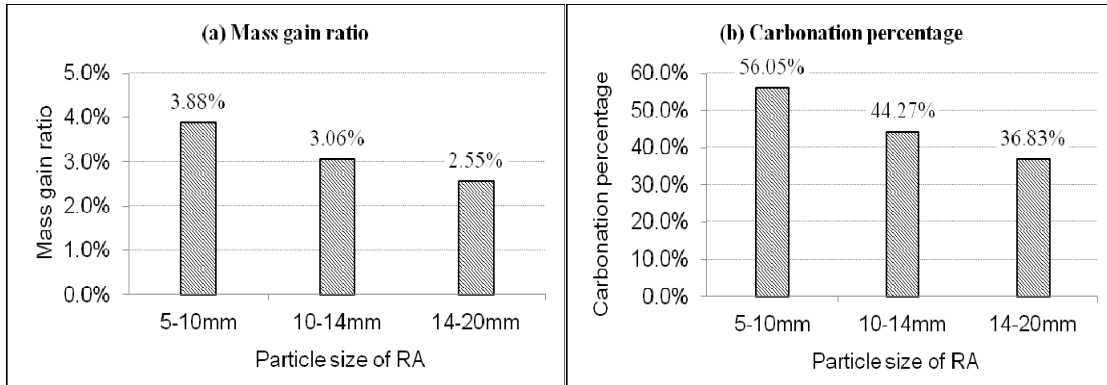
Figure 3. Mass gain ratio and carbonation percentage of RA30

## 2.4 Effect of CO_2 curing time on carbonation
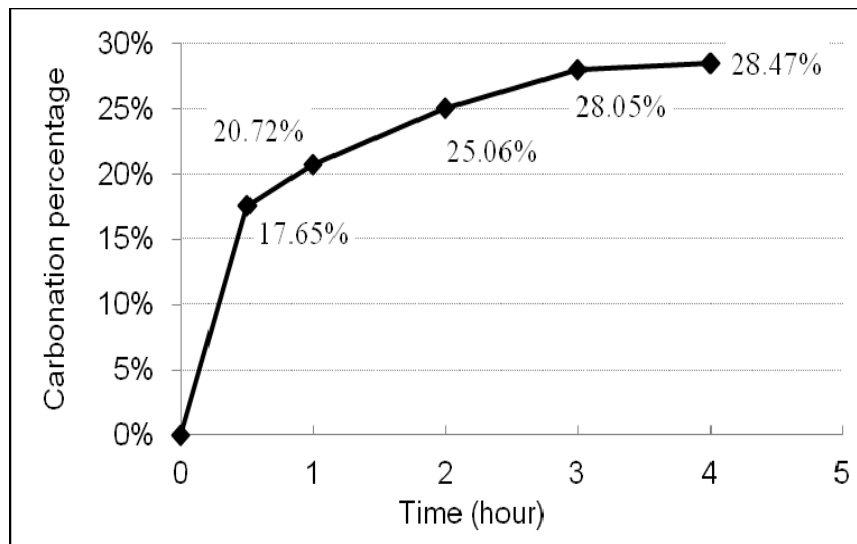
Figure 4. Carbonation percentage of RA versus carbonation curing time

In order to study the effect of CO_2 curing time on carbonation percentage, RA45 with particle size of 10-14 mm was cured respectively for 0.5, 1, 2, 3 and 4 hours. Figure 4 indicates the carbonation percentage of RA versus curing time. The increase in CO_2 curing time led to a higher carbonation percentage of RA. After 2 hours of CO_2 curing, RA45 attained a 25% carbonation percentage, which was increased only to 28.47% after 2 more hours of curing.

2.5 Effect of moisture content of RA on carbonation

RA60 with a particle size of 10-14 mm was used to study the effect of moisture content on mass gain and carbonation percentage of RA. Before the carbonation process, the aggregates were pretreated under different conditions for moisture content control as follows: (i) dried in an oven at 105°C for 2 hours, (ii) stockpiled RA without treatment, (ii) soaked in water for 2 hours. The moisture contents of the RAs after the above three pretreatment conditions were determined to be 0.08%, 3.37% and 5.03% respectively. Figs. 5(a) and 5(b) present the mass gain ratio and carbonation percentage of the RAs after 1 hour CO_2 curing. It is clear from the results that appropriate moisture content is critical for the carbonation process. The stockpiled RA with a moisture content of 3.37% attained the highest carbonation percentage, which was a magnitude higher than that for the other two batches.
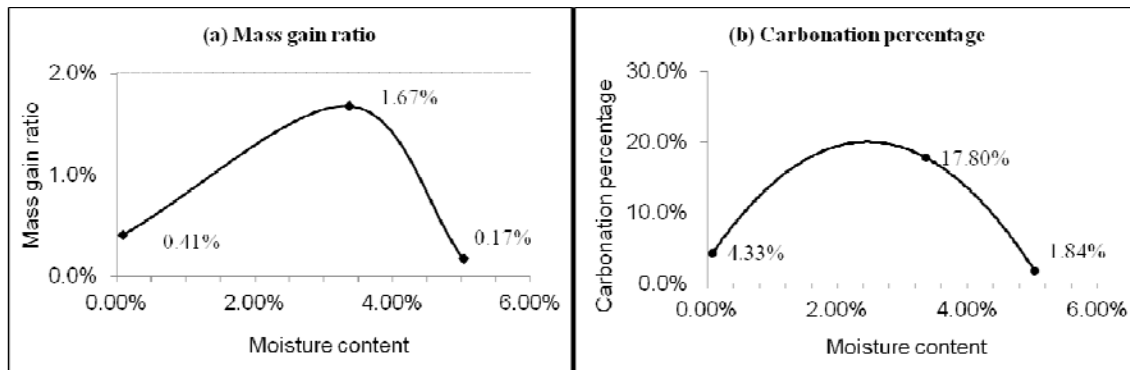
Figure 5. Mass gain ratio and carbonation percentage versus moisture content

3. SUMMARY AND CONCLUSIONS

Based on the positive effects of natural carbonation process on concrete properties, an innovative pretreating method: CO_2 curing process, was evaluated in this study for enhancing the properties of recycled aggregate (RA). The experimental results confirmed that the CO_2 curing process can densify the mortar adhered on RA. After the CO_2 curing process, there was a significant reduction in water absorption and porosity of the RA. Owing to the large specific surface area, RA with smaller particle sizes was more easily to be carbonated. The moisture content of RA significantly influenced the carbonation percentage since the dry matrix could not provide sufficient water for carbonation reactions and the pores on surface of wet matrix was filled with water blocking CO_2 penetration. Furthermore, the carbonation process

proceeded rapidly within the first 2 hours but slowed down sharply after that.

ACKNOWLEDGEMENT

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