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Use of a CO_2 curing step to improve the properties of concrete prepared with recycled aggregates

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ABSTRACT

The paper presents the results of an experimental study on properties of concrete prepared with recycled mortar aggregate (RMA) that has been modified by a CO_2 curing method. The experimental investigation was conducted in two parts. Firstly, the properties such as density, 10% fine value, and water absorption of CO_2 improved RMA were determined. Secondly, the fresh, hardened and durability properties including slump, compressive and tensile splitting strength, drying shrinkage and chloride penetrability of the concretes prepared with RMA and CO_2 cured aggregates (CI-RMA) were determined. It was found that the density, and 10% fine value of the CI-RMA was higher, and the water absorption of the CI-RMA was lower when compared to the untreated RMA. For the concrete, not only was there an improvement in the mechanical properties and resistance to chloride ion penetration for the concrete prepared with CI-RMA, but also the drying shrinkage was decreased.

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1. Introduction

Research work on recycled aggregate concrete had been well documented in the literatures [1–5]. Generally, the mechanical properties, such as compressive and tensile splitting strengths of the concrete made with recycled aggregates, were found that the concrete strengths diminish by up to a 40% at 100% RA replacement ratio compared with the natural aggregate concrete [6], and the drying shrinkage increased by up to 60% [7]. Similar findings were reported in more recent studies [8–12]. Most of the adverse effects are attributed to the higher water absorption properties of the recycled aggregates (RA) due to the presence of old cement mortars in the RA, particularly in the finer fraction.

Recycled aggregate had higher water absorption and lower strength, which affects the crushing strength of the RA and the bond between the RA and the new cement paste. Therefore, techniques have been developed to minimize this adverse effect. These techniques aim to consolidate the adhering mortar layer and reduce the porosity of RA, thereby they would improve the interfacial bond between RA and new cement paste in the new concrete.

Shayan and Xu [13] reported the use of lime and silica fume to improve the surface properties of RA and the results clearly demonstrated that the improved RA can be used to produce 50 MPa structural concrete with durability properties similar to those of natural aggregate concrete. Tsujino et al. [14] conducted experiments on using two types of surface improving agent, an oil-type and a silane-type, to improve the properties of RA. Otsuki et al. [15] suggested that a double mixing method was able to enhance the compressive strength of the concrete prepared with coarse RA by improving the interfacial transition zone between the RA and the cement paste. Poon and Chan [16] found that the adverse effects (i.e., strength reduction) due to the use of fine recycled aggregates could be minimized by the deployment of the double mixing method, which can be easily implemented in pre-cast concrete production. Tam and Tam [17] reported that the additions of silica fume in a two-stage mixing approach could fill up the weak areas in the RA and thus developed a stronger interfacial layer around aggregates, and hence a higher strength of the concrete.

The authors previously used PVA to improve the properties of RA [18]. It was found that the 10% fines value of the PVA improved RA was higher, and the water absorption were lower when compared to the untreated RA. The results show that there was not only an improvement in the mechanical properties of the concrete made with the treated RA, but also the shrinkage of new concrete decreased while the resistance to chloride-ion penetration improved.

Carbonation is known to improve surface hardness, strength, and durability of cement-based products by pore refinement of the cement paste matrix. Carbonation can be helpful in non-reinforced cement-based products. However, for reinforced cement-based products, as the pH of carbonated cement paste

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reduces due to carbonation, reinforcing steel losses its passivity and becomes vulnerable to corrosion [19]. Carbonation in cement-based products can be defined as a reaction between the CO_2 dissolved in water and the cement hydration product $Ca(OH)_2$ in the pore water [20]. This reaction produces calcium carbonate ($CaCO_3$) and water. Calcium silicate hydrates and calcium aluminate hydrates also react with CO_2 in the presence of moisture to produce calcium carbonate and hydrates of silicates and aluminates and water [21–24]. It was estimated at full carbonation when all CaO reacts with carbon dioxide, CO_2 uptake by cement could reach to 50% by cement mass, i.e., one ton of cement could uptake half a ton of CO_2. Full carbonation could happen in weathering carbonation of concrete in more than 30 years when atmospheric CO_2 slowly but progressively reacts with hydration products in matured concrete. Nevertheless it is not a desired reaction since hydration products are decomposed and strength of concrete is reduced. On the other hand, early carbonation during concrete curing provides an ideal process window for CO_2 utilization.

Approximately one third of all CO_2 emissions are due to human activity from fossil fuels used for generating electricity, with each power plant capable of emitting several million tonnes of CO_2 annually. A variety of other industrial processes also emit large amounts of CO_2 from each plant, for example oil refineries, cement works, and iron and steel production. These emissions could be reduced substantially, without major changes to the basic process, by capturing and storing the CO_2. Other sources of emissions, such as transport and domestic buildings, cannot be tackled in the same way because of the large number of small sources of CO_2. There many ways in which CO_2 emissions can be reduced, such as increasing the efficiency of power plant or by switching from coal to natural gas. However, most scenarios suggest that these steps alone will not achieve the required reductions in CO_2 emissions.

Normally, there are three distinct processes for CO_2 capture and storage. First, capturing CO_2 from the gas streams emitted during electricity production, industrial processes or fuel processing; second, transporting the captured CO_2 by pipeline or in tankers; and third storing CO_2 underground in deep saline aquifers, depleted oil and gas reservoirs or unmineable coal seams. All three processed have been in use for decades, albeit not with the purpose of storing CO_2. Further development is needed, especially on the capture and storage CO_2.

Recycled concrete aggregate (RCA) contains a large amount $Ca(OH)_2$ due to the presence of old cement mortar. Using a carbon dioxide (CO_2) curing step would not only improve the RCA properties, but it also can capture CO_2 emitted from industrial processes.

The aim of this study was to improve the properties of RMA by using a CO_2 curing method. The paper reports the influence of the CO_2 cured recycled aggregates on the strength development and durability properties of new concrete. In this study, the experimental investigation was conducted in two parts. Firstly, the optimal condition of CO_2 curing for the recycled mortar aggregates (RMA) was determined. The RMA was cured in a 100% CO_2 chamber under a + 0.1 Bar pressure for 6, 12, 24, 48 and 72 h, and the properties of the aggregate were subsequently determined. Secondly, the fresh and hardened properties of the concrete prepared with the CI-RMA were determined. A comparison was made between the properties of the concrete prepared with untreated RMA or NA and CI-RMA.

2. Materials collected and their characterizations

2.1. Cement

The cement used in this study was an ASTM Type I Portland cement with a density of 3.15 g/cm^3 and specific surface area of 3960 cm^2/g which was supplied by Green Island Cement Company Limited in Hong Kong. The chemical compositions of the cement are given in Table 1.

Table 1
Chemical compositions of cement.

Materials	Composition (%)						
	LOI	SiO_2	Fe_2O_3	Al_2O_3	CaO	MgO	SO_3
Cement	2.97	19.61	3.32	7.33	63.15	2.54	2.13

2.2. Aggregates

Natural and recycled mortar aggregates were used as the coarse aggregate in the concrete mixtures. Crushed granite was used as the natural aggregate, and the recycled aggregates were crushed old cement mortar derived from two types old mortars prepared in the laboratory with 90 days compressive strength of 40 MPa and 65 MPa. The two types of mortars were named RMA1 and RMA2, respectively. Moreover, natural river sand with a fineness modulus of 2.11 was used as the fine aggregate in the concrete mixtures.

2.3. Co_2

A commercially available CO_2 gas source with a CO_2 concentration higher than 99% was used for curing the recycled mortar aggregate.

3. Experimental details

3.1. Preparation of recycled mortar aggregates (RMA)

The old cement mortar mixtures RMA1 and RMA2 were prepared with sand to cement ratios of 3.0 and 2.5, and water to cement ratios of 0.55 and 0.45, respectively. The mortar mixes were cast in 150 mm cubic steel moulds, compacted using a vibrating table, and covered with a plastic sheet and cured in the laboratory. The specimens were demolded after 24 h and further cured in a water curing tank at 27 ± 1 °C until the ages of 90 days were reached. At 90 days, all the specimens were taken out from the curing tank. After testing for the compressive strength, all the specimens were manually crushed by using a steel hammer. The size fractions between 20–10 mm and 10–5 mm of the crushed materials were used as 20 mm and 10 mm coarse RMA respectively.

3.2. CO_2 curing

An air tight steel-cylindrical vessel with a volume of about 33 L was used as the curing chamber. About 4 kg of crushed RMA was put into the chamber, and then it 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 0.1 Bar. Five different curing periods were used viz. 6, 12, 24, 48 and 72 h. Considering the adverse impact of high humidity on carbonation, a sufficient quantity of silica gel was put at the bottom of the chamber to absorb evaporated water from the aggregate. The CO_2 curing process is illustrated in Fig. 1. The above procedures were repeated until sufficient quantities of CI-RMA were obtained.

3.3. Concrete specimens casting and curing

The new concrete mixes were prepared with a water-to-cement ratio of 0.50 and the cement content was kept constant at 380 kg/

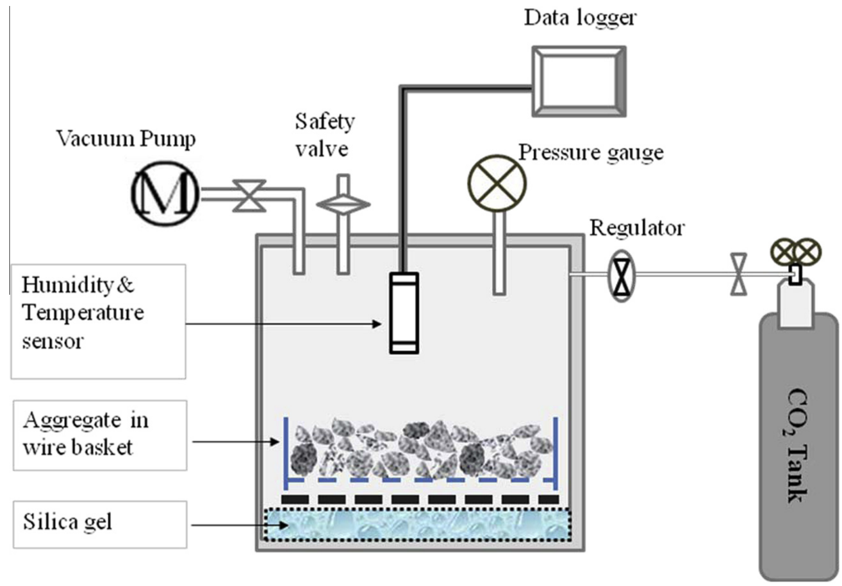


Fig. 1. Schematic of CO_2 curing experiment.

m^3. The proportions of the concrete mixes were designed using the absolute volume method. The mix proportions of the concrete mixtures with the aggregates at the saturated surface-dried (SSD) condition are shown in Table 4. The actual proportions at mixing were adjusted according to the moisture contents and water absorption values of the aggregates.

For each concrete mix, 100 mm cubes, 75 × 75 × 285 mm prisms and 200 × 100 mm dia. cylinders were cast. The cubes and the prisms were used to determine the compressive strength and the drying shrinkage of the concrete, respectively. The cylinders were used to evaluate the splitting tensile strength, static modulus of elasticity and the resistance to chloride-ion penetration of the concrete. All specimens were cast in steel moulds, compacted using a vibrating table, and covered with a plastic sheet and cured in the laboratory. The specimens were demolded 24 h and further cured in a water curing tank at 27 ± 1 °C until the ages of 3, 7, 28 and 90 days were reached.

3.4. Testing

3.4.1. Determination of properties of aggregates

The water absorption, density, and Ten percent Fines Value (TFV) of the natural aggregate, recycled mortar aggregate and CO_2 improved recycled mortar aggregate (CI-RMA) were tested according to BSEN 1097-3, BSEN 1097-5, BS 812-111, respectively.

3.4.2. Slump

The slump of the fresh concrete prepared was measured using the standard slump test apparatus. A portion of about 20 L (three times the quantity required for the slump test) of fresh concrete were taken for the slump test. Slump values were regularly measured at the intervals of 15 min, with the first value measured immediately after mixing and the last value measured at the 180th minute. Plastic films were used to cover the concrete mixtures during the intervals between tests.

3.4.3. Compressive and tensile splitting strength

The compressive and tensile splitting strengths of concrete were determined using a Denison compression machine with a loading capacity of 3000 kN. The loading rates for the compressive and splitting tensile tests were 200 kN/min and 57 kN/min in accordance with BSEN 12390-3 and BSEN 12390-6, respectively. The compressive and tensile splitting strength tests were carried out at the ages of 3, 7, 28 and 90 days.

3.4.4. Static modulus of elasticity

The static modulus of elasticity (E values) of the natural and recycled aggregate concrete were determined on the 100 × 200 mm cylindrical specimens according to ASTM C 469-65 at the ages of 28 and 90 days.

3.4.5. Drying shrinkage and weight loss test

The drying shrinkage and rate of weight loss was determined by measuring the specimen length and weight change in accordance with ASTM C596. The prismatic concrete specimens with sizes of 75 × 75 × 285 mm were used. Each specimen was fitted with a stainless steel stud at both ends. The specimens were removed from the moulds 24 h after casting and cured in water for another 48 h. At the age of 3 days, the specimens were removed from the water tank, wiped with a damp cloth, and the length and weight was immediately measured; this was regarded as the initial length and weight of the specimens. Then the specimens were placed in an environmental chamber at a controlled temperature of 23 ± 2 °C and a relative humidity of 50 ± 5%. The drying shrinkage and rate of weight loss of all the specimens was monitored at 1, 4, 7, 28, 56, 90 and 112 days. The accuracy in the length change measurement was ±0.0001 mm.

3.4.6. Determination of chloride-ion penetrability

The resistance to chloride ion penetrability of the concrete mixtures was determined in accordance with ASTM C1202 using a 50 mm thick × 100 diameter concrete disk that was mechanically cut from the 100 × 200 mm concrete cylinders. The resistance of the concrete against chloride ion penetration is represented by the total charge passed in coulombs during a test period of 6 h. The test was carried out on the concrete specimens at the ages of 28 and 90 days.

4. Results and discussion

4.1. Properties of CI-RMA

The test results of the properties of CI-RMA such as density, water absorption and 10% fine value of the CO_2-improved recycled

Table 2
Physical properties of aggregates.

Property	Particle size (mm)	Aggregate type				
		Natural granite	RMA1	RMA2	CI-RMA1	CI-RMA2
Density (Kg/m^3)	20	2.62	2326	2355	2345	2371
	10	2.62	2326	2355	2351	2379
Water absorption (%)	20	0.89	11.82	9.30	7.32	4.84
	10	0.87	12.25	10.81	7.57	4.95
10% Fine value (KN)	14	156	96	116	108	134

Table 3
Physical properties of CO_2-cured recycled mortar aggregate.

Aggregate Type	Curing time (h)	Physical properties				
		Density (Kg/m^3)		Water absorption (%)		10% fine value (KN)
		20 mm	10 mm	20 mm	10 mm	14 mm
RMA1	6	2330	2339	10.12	10.86	98
	12	2337	2346	8.03	8.77	101
	24	2345	2351	7.32	7.57	108
	48	2347	2352	7.13	7.42	109
	72	2349	2354	6.99	7.28	111
RMA2	6	2358	2360	8.12	8.99	120
	12	2363	2365	6.35	6.26	126
	24	2371	2379	4.84	4.95	134
	48	2373	2381	4.68	4.78	136
	72	2375	2383	4.54	4.66	138

mortar aggregates are shown in Tables 2 and 3 together with those for natural granite and RMA. The values are the averages of three measurements. It was found that the properties of RMA improved with CO_2 curing. The density and 10% fine value of RMA increased with an increase in CO_2 curing time. Moreover, the water absorption of RMA was significantly decreased. Table 3 shows that the properties of 48 and 72 h CO_2 treated RMA were only marginally better than that of the 24 h CO_2 treated RMA; and hence, it was considered that 24 h CO_2 treatment was optimum.

4.2. Initial slump and slump loss of concrete

Figs. 2 and 3 depicts the initial slump and the changes of concrete slump with time for the concrete mixtures, respectively. The reported slump values are also the averages of three measurements. It can be seen that, compared with the control, the initial slump of the concretes prepared with both RMA and CI-RMA increased. The mix prepared with RMA2 had the highest slump value of 165 mm. This was due to the higher initial free water content in the concrete mixture. This was in turn due to the higher water absorption value of the RMA (see Tables 3 and 4) which was used at the air dried condition with a moisture content at mixing much lower than the water absorption value. Additional amounts of water were added to maintain the mix proportions as listed in Table 2. It is also shown in Fig. 3 that the rate of slump loss was quicker for the RMA concretes within the first hour after mixing, but was slower afterwards. To reach zero-slump, the concrete mixture prepared without recycled aggregate took about 150 min, while the mixtures prepared with RMA and CI-RMA took over 3 h.

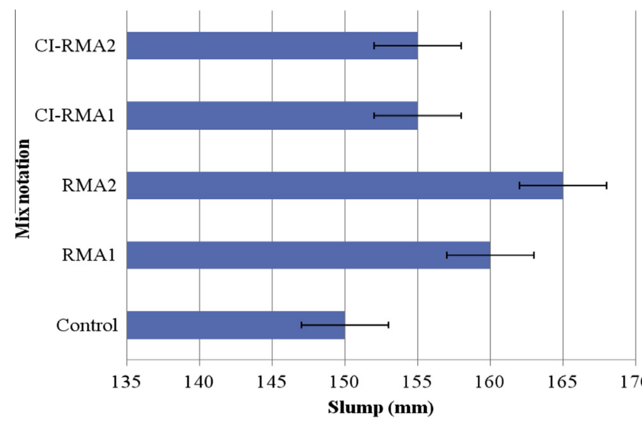


Fig. 2. Initial slump of concrete.

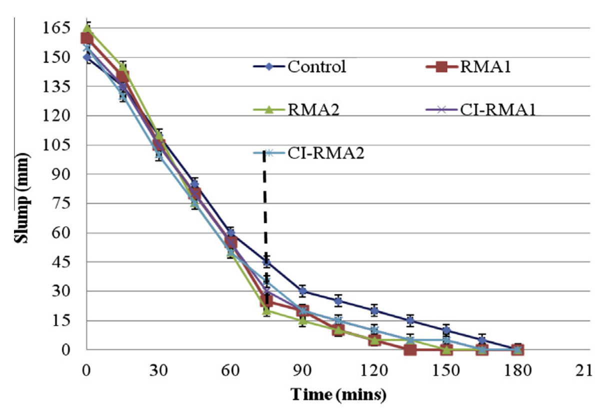


Fig. 3. Slump loss of concrete.

Table 4
Proportions of concrete mixtures.

Notation	Proportion (kg/m^3)							
	Cement	Water	W/C	Sand	10 mm granite	20 mm granite	10 mm RMA	20 mm RMA
Control	380	190	0.50	704	367	735	–	–
RMA1	380	190	0.50	704	–	–	367	664
RMA2	380	190	0.50	704	–	–	352	653
CI-RMA1	380	190	0.50	704	–	–	361	665
CI-RMA2	380	190	0.50	704	–	–	359	658

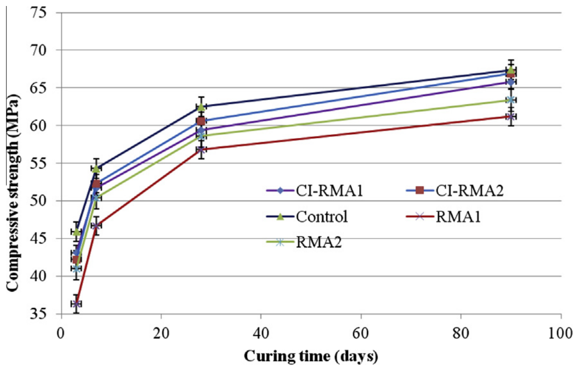


Fig. 4. Development of compressive strength of concrete.

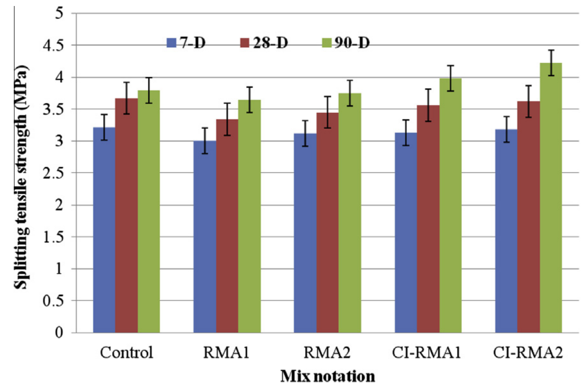


Fig. 5. Tensile splitting strength of concrete.

Table 5
Relative compressive strength of concrete.

W/C	Age (days)	Control	RMA1	RMA2	CI-RMA1	CI-RMA2
0.50	3	1	0.79	0.89	0.94	0.92
	7	1	0.86	0.93	0.95	0.96
	28	1	0.91	0.94	0.95	0.99
	90	1	0.91	0.94	0.97	0.99

Table 6
Relative tensile splitting strength of concrete.

W/C	Age (days)	Control	RMA1	RMA2	CI-RMA1	CI-RMA2
0.50	7	1	0.93	0.97	0.98	0.99
	28	1	0.91	0.94	0.97	0.99
	90	1	0.96	0.99	1.05	1.11

4.3. Compressive and tensile splitting strength

The development of the compressive strength of the concrete mixtures up to 90 days is presented in Fig. 4. Table 5 gives the relative compressive strength (expressed as percentages of that of the control mixtures (100% natural aggregate)) of the concrete mixtures. It is seen that at all test ages, the compressive strength of concrete mixtures prepared with untreated RMA was lower than that of control concrete. Although both the CI-RMA1 and CI-RMA2 improved the compressive strength of the concrete mixtures, the compressive strength of the concrete mixtures was still lower than that of the control at the all tested ages. At 28 days the compressive strength of concrete prepared with RMA1, RMA2, CI-RMA1, and CI-RMA2 was 10%, 8%, 3% and 1% lower than that of the control concrete. The concrete mixtures prepared with CI-RMA2 had higher compressive strength than that of the mixtures with CI-RMA1. At 90 days, the compressive strength of concrete made with CI-RMA2 was only 1% lower than that of control concrete. This may be attributed to the CO_2 curing regime improved the physical and mechanical properties of the recycled mortar aggregates. It can be seen from Table 2 that the water absorption of 20 mm CI-RMA1 and CI-RMA2 was 38.1% and 47.9% lower than that of RMA1 and RMA2, respectively. Moreover, the 10% five value of CI-RMA1 and CI-RMA2 was 12.5% and 15.5% higher than that of RMA1 and RMA2, respectively.

Table 6 provides the relative tensile splitting strength (expressed as percentages of that of the control) of the concrete mixtures and Fig. 5 shows the development of the tensile splitting strength with age. It can be seen that before 28 days the tensile splitting strength of the concrete mixtures with RMA and CI-RMA was lower than that of the control concrete. However, at 90 days, the tensile splitting strength of the concrete mixtures prepared with CI-RMA1 and CI-RMA2 were 5% and 10% higher than that of the control concrete. This is consistent with the results of the authors' previous studies [18,25].

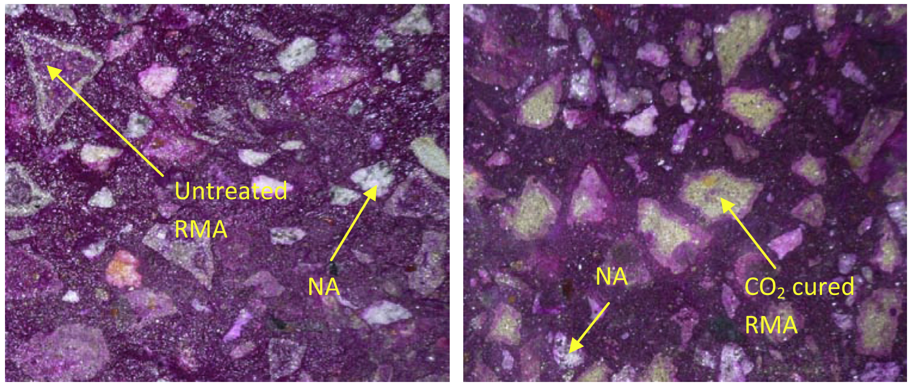
Carbonation is known to improve surface hardness, strength, and durability of cement-based products by pore refinement of the cement paste matrix. Fig. 6 shows the comparison of results from phenolphthalein spraying on split concrete at 28 days. It can be seen from Fig. 6A that the whole surface of split concrete with untreated RMA was red except natural granite. However, the color of split CO_2 improved recycled mortar aggregate CI-RMA did not change (See Fig. 6B). It is mean that the RMA has been completely carbonated in the concrete. Therefore, the strength and durability of the concrete prepared with CI-RMA was improved by CO_2 curing regime.

4.4. Static modulus of elasticity

The results of static modulus of elasticity (*E*-values) of the concrete mixtures are shown in Fig. 7 and Table 7 provides the relative *E*-values (expressed as percentages of that of the control) of the concrete mixtures. It is seen that the *E*-values of concrete mixtures with RMA1 and RMA2 were 15% and 12% lower than that of the corresponding control concrete, respectively. Although both the CI-RMA1 and CI-RMA2 improved the *E*-values of the concrete mixtures, the *E*-values of the concrete mixtures were still lower than that of the control at the all tested ages. Moreover, the concrete mixtures prepared with CI-RMA2 had higher *E*-values than that of the mixtures with CI-RMA1. At 90 days, the *E*-values of the concrete mixtures prepared with CI-RMA1 and CI-RMA2 were 6% and 2% lower than that of the control concrete.

4.5. Drying shrinkage and rate of weight loss

The development of drying shrinkage and rate of weight loss of the concrete mixtures are shown in Figs. 8 and 9, respectively. Each presented value is the average of three measurements. It can be seen that at all test ages, the drying shrinkage and the rate of weight loss of the concrete mixtures prepared with RMA and CI-RMA were higher than that of the control mixture. However, at 112 days, the drying shrinkage of the concrete mixtures made with CI-RMA1 and CI-RMA2 was approximately 10% and 15% lower than that of the concrete prepared with RMA1 and RMA2, respectively. This is due to the rate of weight loss of the concrete prepared with CI-RMA1 and CI-RMA2 was approximately 24% and 26% lower than that of the concrete prepared with RMA1 and RMA2, respectively. This may be explained by the fact that CI-RMA had a lower water absorption capacity than that of RMA (see Table 3).



(A) **(B)**

Fig. 6. Comparison of results from phenolphthalein spraying on split concrete at 28 days (A) with untreated RMA; (B) with CI-RMA.

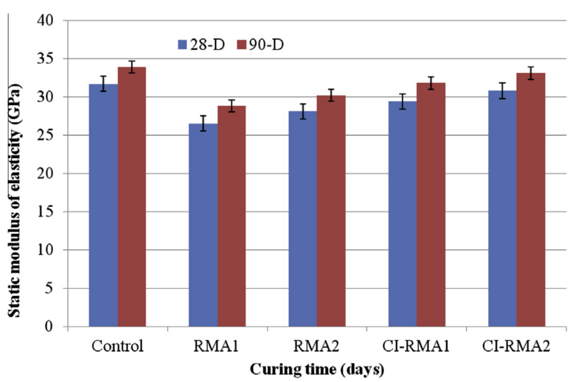


Fig. 7. Static modulus of elasticity of concrete.

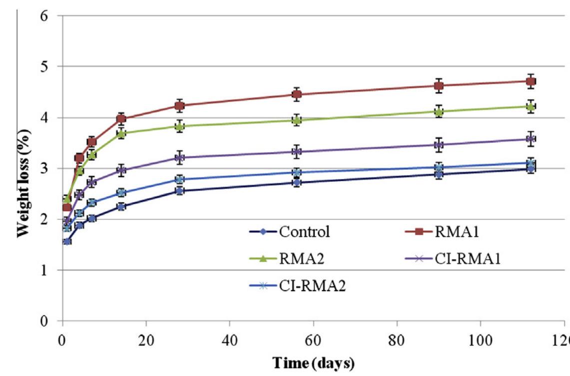


Fig. 9. Weight loss of concrete.

Table 7
Relative static modulus of elasticity of concrete.

W/C	Age (days)	Control	RMA1	RMA2	CI-RMA1	CI-RMA2
0.50	28	1	0.84	0.87	0.93	0.97
	90	1	0.85	0.89	0.94	0.98

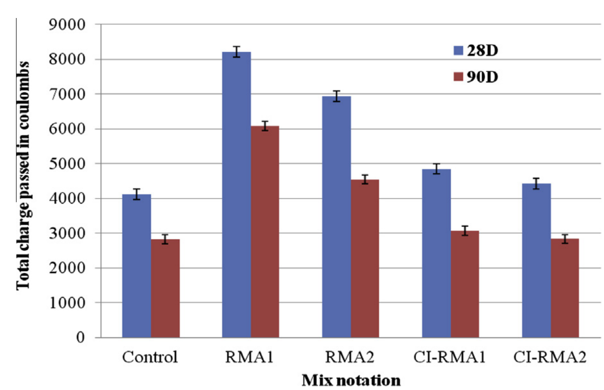


Fig. 10. Total charge passed (in coulombs) of concrete.

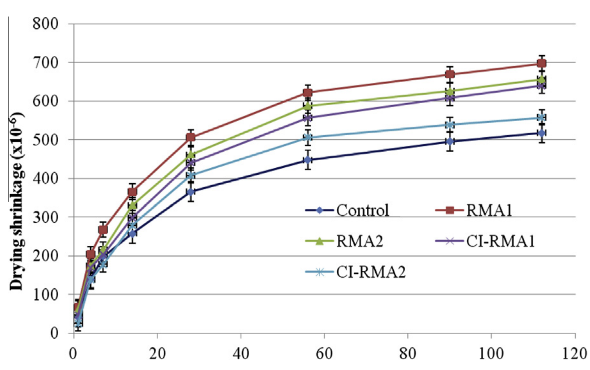


Fig. 8. Drying shrinkage of concrete.

4.6. Chloride-ion penetrability

Fig. 10 shows the test results of resistance to chloride-ion penetration at 28 and 90 days. The results show that when compared with the RMA concrete mixture, the resistance to chloride-ion penetration was significantly increased with the incorporation of the CI-RMA in the concrete mixes. At 28 days, the concretes made with CI-RMA1 and CI-RMA2 had approximately 41% and 46% higher resistance to chloride penetration than that of the concrete made with RMA1 and RMA2, respectively, and the values are similar to that of the control concrete. This may be attributed to the CO_2 curing regime improved the water absorption of RMA. Table 2 shows the water absorption of CI-RMA1 and CI-RMA2 was 38.1% and 47.9% lower than that of RMA1 and RMA2, respectively.

5. Conclusion

Based on the test results and discussion above, the following conclusions can be drawn:

- The compressive strength of the concrete made with RMA and CI-RMA was still lower than that of the control concrete at 28 days. However, at 90 days, the compressive strength of the concrete made with CI-RMA2 was similar to the control concrete.
- Before 28 days, the tensile splitting strength of concrete mixtures with RMA and CI-RMA were lower than that of the control concrete. However, at 90 days, the tensile splitting strength of the concrete mixtures prepared with RMA and CI-RCA were slightly higher than that of the control concrete.
- The drying shrinkage of the concrete mixtures made with CI-RMA1 and CI-RMA2 was approximately 10% and 15% lower than that of the concrete with RMA1 and RMA2, respectively.
- At 28 days, the concretes made with CI-RMA1 and CI-RMA2 had approximately 41% and 46% higher resistance to chloride penetration than that of the concrete made with RMA1 and RMA2, respectively.
- The results are encouraging and show that the CO_2 cured RMA can be used to produce structural concrete with durability properties that are similar to natural aggregate concrete.

Acknowledgements

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