Carbon Storage through Concrete Block Carbonation Curing

Hilal El-Hassan and Yixin Shao

Abstract—The effect of initial curing on carbonation curing of lightweight concrete masonry units (CMU) was examined. Initial curing was performed from 4 to 18 hours at a relative humidity of 50% and temperature of 25°C. Based on cement content, four-hour carbonation curing allowed concretes to uptake 22% to 24% CO₂ with initial curing and 8.5% without initial curing, while prolonged 4-day carbonation recorded an uptake of 35%. Carbonation curing can replace steam curing in CMU production to accelerate hydration and recycle cement kiln CO₂ in a beneficial manner.

Index Terms—Concrete masonry unit, curing, carbonation, carbon uptake.

I. INTRODUCTION

Concrete masonry units (CMU) have been widely used in building construction as load bearing and non-load-bearing walls. The North American market for concrete blocks and bricks is projected to increase to 4.3 billion units per year by 2014 [1].

In comparison to cast-in-place concrete, masonry block structures not only exhibit superior performance due to precast quality, but also represent a low environmental impact construction system. With reference to 1 m^2 of solid concrete wall, a masonry wall using 200-mm CMU requires 48% less material due to internal cavities, leading to 65% less cement, and 65% less CO₂ emission.

CMUs produced in North America are typically steam cured. While steam curing accelerates strength gain and shortens the production cycles, the process itself is energy intensive. It is estimated, for one cubic meter of concrete in block form, atmospheric pressure steam curing consumes 0.59 Gigajoules per m^3 of concrete and autoclave curing consumes 0.71 Gigajoules per m^3 of concrete [2].

The alternative curing method for CMU production is carbonation curing which uses high purity carbon dioxide (99.5% of CO₂) or low purity flue gas (14% of CO₂) for accelerated hydration and durability improvement.

However, carbonation curing has never been adopted in large-scale production. This was possibly because flue gas carbonation was not effective in hydration acceleration and pure gas carbonation was expensive. The latter situation may change in the near future. Large quantities of high purity, low cost carbon dioxide could soon be available as regulations requiring reductions in CO_2 emissions are developed. In this case, pure gas carbonation can simultaneously accelerate the strength, stabilize the dimension, and enhance the durability. By reducing the hydroxyl ion and precipitating CaCO₃ on the surface layer, carbonation curing could improve the concrete resistance to sulfate attack, freeze-thaw cycling, and acid attack [3]. Since carbonation is a CO_2 uptake process [4], recovered cement kiln CO_2 can be recycled into concrete products to make contribution to carbon emission reduction.

The purpose of this research is to develop a carbonation curing process that can be used to replace steam curing for CMU production. High purity CO_2 (99.5%) will be used to simulate the recovered cement kiln flue gas carbon dioxide. The goal is to shorten the carbonation duration to 2 to 4 hours with the help of initial curing ranging from 0 to 18 hours. The effect of initial curing on degree of carbonation reaction is evaluated to promote maximum possible carbon uptake in CMU. The carbon uptake is estimated using mass gain and thermal analysis.

II. EXPERIMENTAL INVESTIGATIONS

A. Sample Preparation

The carbonation curing parameters will be studied using rectangular slab concrete samples of 38 mm thickness to simulate the typical web or face shell of a hollow concrete masonry unit (CMU). The optimized process parameters are then applied to carbonation curing of 200-mm CMU. The samples are prepared according to the commercial CMU mix design with lightweight expanded slag aggregates. The as-received aggregates are nearly saturated with a water content of 5%. Table I summarizes the mixture proportion of both slab and CMU samples. By mass, it includes a water-to-cement ratio of 0.4, an aggregate-to-cement ratio of 6.23, and cement content of 0.13. Each rectangular slab sample weighs approximately 680 grams with a density of 1850 kg/m³ and each CMU block weighs 15 kg with a density of 1839 kg/m³. For rectangular slabs, raw materials were mixed in a pan mixer, cast into a $127\times76\times38~\text{mm}$ mold and compact formed using a vibrating hammer to simulate the industry production of CMU.

Because of the dry mix, concrete was demolded right after casting for initial curing. For 200-mm CMU blocks, materials were mixed in a mechanical drum mixer and compact formed by a manual block machine. They were typical 200-mm CMU blocks with the thickness of the web or face shell ranging from 25 to 33 mm. The density was

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calculated based on ASTM C140 [5].

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	Slab (g)) CMU (g) Mass (kg/m ³)		Percent (%)			
Cement	88	1967	241	13			
Water	35	787	96	5			
Expanded Slag	554	12251	1502	82			
Sample	677	15004	1839	100			

TABLE I: MIXTURE PROPORTIONS

B. Curing Procedures

Curing procedures of different batches using rectangular slab samples are summarized in Table II. For comparison, Batches 1-4 are steam cured and Batches 5-15 are carbonation cured. One set of slab batches were also prepared for normal hydration in sealed plastic bag to serve as control. Steam curing took place in a steam cooker for a period of 4 hours with maximum temperature of 80°C and relative humidity of 95%. Initial curing of 0, 4, 6, and 8 hours at 22°C and relative humidity of 80% was applied prior to steam. Carbonation curing setup is shown in Fig. 1. Initial curing was performed on fresh concrete by 0, 4, 6, 8, and 18 hours respectively in an environmental chamber at a relative humidity of 50% and a temperature of 25°C. The purpose of initial curing was to reduce the free water on the surface and allow diffusion of carbon dioxide. Initial curing of 0 hour was actually immediate carbonation of fresh concrete and served as reference. Initial curing of 18 hours

was to simulate an overnight curing and was likely the longest preset that can be accepted by commercial production. Concrete slab samples after initial curing were placed in a sealed chamber in Fig. 1, which was then vacuumed to about 0.7 bars and filled with carbon dioxide gas to a pressure of 1 bar. The chamber was placed on a digital balance to obtain the mass curve of concrete during carbonation. The carbonation duration varied from 2 to 4 hours. A period of 96 hours was also investigated to study the effect of extreme exposure time.



Fig. 1. Schematic of carbonation setup

TABLE II. CURING I ROCEDURES										
		Initial curing			Steam curing		Carbonation curing		Subsequent Hydration	
Batch #	Condition	RH	Т	t	RH	Т	t	t	Water	t
		(%)	(°C)	(hours)	(%)	(°C)	(hours)	(hours)	Spray (g)	(days)
1	0a + 4s	-	-	0	95±5	75±5	4	-	-	28
2	4a + 4s	80±5	22±1	4	95±5	75±5	4	-	-	28
3	6a + 4s	80±5	22±1	6	95±5	75±5	4	-	-	28
4	8a + 4s	80±5	22±1	8	95±5	75±5	4	-	-	28
5	0a + 4c	-	-	0	-	-	-	4	-	28
6	$0a + 4c^{w}$	-	-	0	-	-	-	4	1±0.2	28
7	4a + 4c	50±1	25±0.2	4	-	-	-	4	-	28
8	$4a + 4c^{w}$	50±1	25±0.2	4	-	-	-	4	17±2	28
9	6a + 4c	50±1	25±0.2	6	-	-	-	4	-	28
10	8a + 4c	50±1	25±0.2	8	-	-	-	4	-	28
11	18a + 4c	50±1	25±0.2	18	-	-	-	4	-	28
12	$18a + 4c^{w}$	50±1	25±0.2	4	-	-	-	4	29±2	28
13	18a + 2c	50±1	25±0.2	18	-	-	-	2	-	28
14	18a + 96c	50±1	25±0.2	18	-	-	-	96	-	28

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TABLE II:	CURING	PROCEDURES

Note: a - Initial air curing; s - Steam curing; c - Carbonation; RH - Relative humidity; T - Temperature; t - Time; W - Water sprayed after carbonation

The effect of initial curing and carbonation curing were evaluated based on water loss and carbon uptake. To compensate for the water loss during initial curing and carbonation curing, water spray was applied to Batches 6, 8 and 12 to restore the original water content and examine its effect on subsequent hydration after carbonation. The temperature, relative humidity, pressure, samples' initial and final mass, and mass of water condensed on the wall of the chamber were recorded. The best combination of initial curing and carbonation curing from slab tests was selected for 200-mm CMU production. Control concrete as reference to carbonation underwent same initial curing of 0, 4, 6, 8, and 18 hours in an environmental chamber at a 50% RH and a 25° C for each hydration control batch.

C. Carbon Uptake Estimation

In order to measure the degree of carbonation, three methods were utilized for the estimation of carbon uptake: mass gain, mass curve, and thermal decomposition analysis.

Mass gain method estimates CO_2 uptake in concrete by comparing mass of samples before and after carbonation (1). Carbonation-induced water loss was collected by absorbent paper and added to the final mass. By treating the system as a closed system, it was imperative to include the evaporated water, which was initially inside the samples prior to carbonation.

CO_2 uptake (%) = (Final mass + Mass of water loss - initial mass) / (Mass of cement) (1)

Mass curve method was executed by placing the carbonation setup on a digital balance, which was zeroed after vacuuming the chamber. A mass curve was recorded as mass versus time until the end of the process at which time CO_2 was released and the residual mass, M, was measured. The system was calibrated by repeating the tests using CO_2 -insensitive styrofoam samples of the same volume to obtain second residual mass, m. The difference between M and m represented the CO_2 uptake by concrete (2). The data obtained from mass gain and mass curve are independent from any carbon content existing before carbonation.

$$CO_2$$
 uptake (%) = (M - m)/(Mass of cement) (2)

Thermal decomposition analysis was also performed to estimate the amount of carbonates in concrete. Instead of using a classical thermogravimetry device, which allows only a few micrograms of powder, a furnace of maximum temperature of 1100°C was employed to test large concrete samples with mass range of 35-70 for each. Separation of paste from concrete was thus avoided. Concrete samples were heated up to 105°C, 550°C, and 1000°C to quantitatively measure the evaporable water, bound water in hydration products, and carbon dioxide in carbonates respectively [6]. The mass loss between 550°C and 1000°C represents carbon uptake by carbonation (3):

$$CO_2$$
 uptake (%) = (Mass at 550°C - Mass at 1000°C) /

(Mass of Cement) (3)

III. EXPERIMENTAL RESULTS AND DISCUSSION

A. Effect of Initial Curing on Internal Water Content

Initial curing in a controlled environment (25°C) and 50% RH) was introduced in the carbonation process to justify the water content in concrete. To explore the possibility of application of carbonation curing to CMU production, the entire curing process cannot exceed 24 hours in comparison to current steam curing practice. Therefore, the process window of initial curing is limited to 0, 4, 6, 8, and 18 hours at 25°C and 50% RH. The water loss curve due to initial curing up to 14 days is plotted in Fig. 2.

Percent water loss is a ratio of mass loss during initial curing in a specified time over total initial water mass. The total initial water mass is the sum of the mixing water (water-to-cement ratio of 0.4) and the water in wet expanded slag aggregates (5% of the total slag mass). The mass loss during initial curing was recorded by a digital balance over a period of 14 days at 25°C and 50% RH.

It was apparent that water loss was proportional to the duration of initial curing within 24 hours. The most significant loss occurred in first 4 hours, reaching 32%. Water loss due to 6 and 8 hours initial curing was basically

no different from 4 hours. However, 18 hours curing increased the water loss to 51%, which was very close to that by 24 hours. Eighteen hours seemed to be the maximum possible time for initial curing and could be executed through an overnight shift. At 14 days, the water loss reached 81%.

B. Effect of Initial Curing on Carbonation Reaction

Degree of carbonation is characterized by carbon uptake. Three methods are used to quantify CO_2 uptake in carbonated concrete. Fig. 3 shows water loss collected from carbonation process and the carbon uptake by mass gain method (1). Two groups of data are presented in Fig. 3. First group includes the first 5 batches with constant carbonation time of 4 hours and varied initial curing of 0, 4, 6, 8 and 18 hours to study the effect of initial curing.

The second group involves the last three batches with constant initial curing of 18 hours and varied carbonation duration of 2, 4 and 96 hours to investigate the effect of carbonation time. Percent water loss due to exothermic carbonation curing is defined as a ratio of water collected in chamber after carbonation over total initial water mass.



Fig. 3. Water loss due to carbonation and carbon uptake

In first group of 4-hour carbonation, immediate carbonation with no initial curing resulted in a carbon uptake of 7.5% with a carbonation water loss of 2.2%. It was indicative of a low degree of reaction. After 4, 6, and 8-hour initial air curing, water loss due to initial curing was of close value of 32-33%. Nevertheless, their carbon uptake was different at 21.3, 22.8, and 23.5% with carbonation water loss at 8.2, 9.6, and 10.2%, respectively. Obviously, initial curing reduces free water, making room for gas diffusion and calcium carbonate precipitation. Initial curing of 18 hours removed 51% free water and promoted carbon

uptake to 24.2% with a carbonation water loss of 5.9%. Prolonged initial curing is not directly beneficial to reaction efficiency.

In the second group, initial curing was fixed at 18 hours. When 2-hour carbonation was compared with 4-hour carbonation, longer carbonation evaporated more free water and promoted higher carbon uptake. In prolonged carbonation, carbon uptake by 96 hours was increased by 44% in comparison to that by 4 hours. However, water evaporation was reduced possibly due to the re-absorption of water during the 4-day process. A carbon uptake of 34.9% in 4-day carbonation represented a degree of carbonation of nearly 70%, if full carbonation is considered as 100% [7]. Obviously, longer carbonation time could promote carbon uptake and enhance the carbon storage capacity of concrete.

To verify the carbon uptake by mass gain method (1), mass curves were obtained. Five batches were compared in Fig. 4 with initial curing of 0, 4, 6, 8 and 18 hours. During the 4-hour carbonation process, 60-70% of the reaction occurred in the first hour and 80-90% in the second hour. The third method to quantify the calcium carbonates in concrete is thermal decomposition analysis. As seen in Table III, mass loss was categorized into three components: evaporable water (up to 105°C), bound water (between 105 and 550°C) and carbon dioxide (between 550 and 1000°C). Higher bound water indicates more hydration products. It is noted that zero initial curing has the lowest uptake but highest bound water. Other samples, as 18a + 4c, have higher uptake, which is associated with high carbonates, but low bound water content. It can be concluded that the hydration products and carbonation products cannot coexist in high quantities in one sample, or in other words, hydration products can be consumed in the carbonation reaction to produce more carbonates.



However, once the water spray technique is applied to batch with 18-hour initial curing followed by 4-hour carbonation curing, both hydration and carbonation products appear to be high. Apparently, water compensation after initial curing followed by carbonation curing is effective and beneficial for subsequent hydration and overall performance. Table III also compares bound water content between carbonated and steamed concretes. After the same initial curing, the amount of hydration products is of close value in carbonated and steamed concretes, suggesting carbonation can technically replace steam to accelerate hydration. Carbon dioxide content in the last

column of Table III is calculated based on total concrete mass used in thermal analysis. If the value is divided by 13%, the cement content, carbon uptake is converted to cement based and comparable to the other two methods.

TABLE III: THERMAL DECOMPOSITION ANALYSIS AFTER 28 DAYS

	Sample	Evaporated		Combined		CO ₂ ^c	
Sample	Mass	Water ^a		Water ^b			
	(g)	M (g)	% ^d	M (g)	% ^d	M (g)	% ^d
0a + 4c	47.25	0.90	1.90	1.66	3.51	0.55	1.16
0a + 4s	48.26	0.72	1.49	1.86	3.85	0.03	0.06
0a	38.84	1.10	2.83	1.55	4.29	0.01	0.03
4a + 4c	76.45	1.09	1.43	2.27	2.97	2.16	2.83
4a + 4s	35.58	0.52	1.46	1.03	2.89	0.03	0.08
4a	45.91	0.91	1.98	1.24	2.70	0.02	0.04
6a + 4c	44.33	0.49	1.11	0.90	2.03	1.32	2.98
6a + 4s	38.76	0.73	1.88	0.85	2.19	0.03	0.08
6a	34.92	0.98	2.81	1.07	3.06	0.02	0.06
8a + 4c	47.26	0.42	1.04	1.40	2.96	1.44	3.05
8a + 4s	55.4	0.66	1.19	1.50	2.71	0.05	0.09
8a	47.28	1.16	2.45	1.59	3.36	0.03	0.06
18a + 4c	65.15	0.67	0.83	1.17	1.80	2.05	3.15
$18a + 4c^{e}$	71.14	0.70	1.97	1.37	3.65	1.14	3.20
18a + 2c	50.69	0.47	0.93	0.86	1.70	1.43	2.82
18a + 96c	40.57	0.46	1.13	0.98	2.42	1.86	4.58
18a	43.61	0.50	1.15	1.32	3.03	0.04	0.09

NOTE: M - Mass; a - Difference in mass between room temperature and 105°C; b - Difference in mass between 105°C and 550°C; c - Difference in mass between 550°C and 1000°C; d - Percentage of initial sample mass; e - Sprayed after carbonation

C. Carbonation Curing of 200-mm CMU

Process parameters are selected for 200-mm CMU production. To maximize carbon uptake within a 24-hour timeframe, it appears 18-hour initial curing followed by 4-hour carbonation can reach a carbon uptake of 24% based on cement. Its carbonation degree is about 48%.

Standard 200-mm CMU block was made in the laboratory and carbonated in a setup similar to that shown in Fig. 1. The fresh blocks underwent an initial curing of 18 hours at 50% RH and 25°C.

Water loss due to initial curing was about 50%, very close to that observed in slab tests. The blocks were then carbonated at 0.1 MPa gas pressure for 4 hours. Water loss due to carbonation was about 5%. Carbon uptake by three block samples, at the age of 1 day, is presented in Table IV. The three methods were used in carbon estimation. In comparison, the three blocks demonstrated an uptake in the range of 23-25%. These results agreed with that of the slab samples, whose uptake was approximately 24%. CMU can be produced by carbonation process to replace steam and achieve superior carbon uptake capacity.

D. Carbon Storage in Blocks and Bricks

Reaction of cement with carbon dioxide at early age is a CO_2 sequestration process. If one 200-mm block with a mass of 15 kg contains 13% cement could have a CO_2 uptake of 24% based on cement, one block has the capacity to store 0.47 kg of CO_2 in a thermodynamically stable calcium carbonate form. Assuming every block or brick has the same carbon storage capacity, the projected annual production of 4.3 billion units in US market can thus consume 2 million tons of CO_2 per year.

The capacity for carbon capture and storage (CCS) in geologic formation is approximately 1 million tonnes per year per site [8]. CO_2 utilization in concrete blocks and bricks production is equivalent to carbon sequestration in two geologic formation sites. The cement annual production in the United States is about 100 million tonnes with CO_2 emission of 80 million tonnes. If all block and brick plants adopt carbonation curing with the same carbon uptake rate, CO_2 utilization in their production lines alone could reduce carbon emission by 2.5% for cement industry.

	Carbon Uptake (%)				
CMU #	Mass Mass		Thermal		
	Gain	Curve	analysis		
1	23.8	24	23.3		
2	23.1	23.6	23.9		
3	23.6	23.8	23.5		

TABLE V: ENERGY CONSUMPTION AND COST ESTIMATE

Process	Р	V	Р	Cost
FIOCESS	(kWh)	(m ³)	(kWh/m ³)	(\$/m ³)
Carbonation (18a+4c)	9	0.058	154.58	9.27
Carbonation	40.5	0.058	695.61	41.74
(4a+4c)				
Steam Curing [2]	-	1	164	9.84

Note: P - Power; t - time; V - Volume.

E. The Network Operation and Cost Estimate

To implement CO₂ utilization at the vicinity of carbon sources, a network needs to be established. It shall include carbon capture, compression, transport, storage, and utilization. The network will be operational based on the assumption that large quantities of high purity and low cost carbon dioxide will be produced as regulations requiring reductions in CO₂ emissions are developed and will be otherwise transported to remote area for underground geologic storage. For CO₂ utilization in block production, the pressurized high purity CO_2 is instead transported to block plants. The cost estimate was made by comparing carbonation curing with steam curing in Table V. It is assumed that the cost estimate starts when the pressurized gas arrives at block plants. The static carbonation process does not require additional energy except the initial air curing at 50% RH and ambient temperature. For laboratory setting used in this preliminary study, the environmental chamber used consumes 2.25 kW, but can fit up to 160 slab samples with a total concrete volume of 0.058 m^3 . Accordingly, per cubic meter of concrete, initial curing by 4-hour and 18-hour consumes 155 and 696 kWh and cost \$9.27 and \$41.74 respectively, assuming 1 kWh of electricity costs \$0.06. Energy consumption in atmospheric steam curing is well studied. It is approximately 164 kWh for cubic meter concrete [2] and costs \$9.84. With reference to steam, the cost by 4-hour initial curing is comparable but the cost by 18-hour initial curing is too high. In order to reduce the cost without compromising the superior performance, the initial curing duration must be modified while maintaining more than 50% water loss. Technically, an environment for initial curing at 50% RH and ambient

temperature can be easily created on production site with no need of an environmental chamber. There is room to reduce the energy consumption in carbonation by using different initial curing and eliminating vacuum. While the industrial steam curing has been optimized in the last 30 years for mass production, the cost of the network operation for carbonation should also be analyzed and optimized to make the system economically feasible and sustainable.

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