

Article

Research on Influences of Ultrasonic Vibration Agitation Stirring on Carbonation Resistance of Cement-Based Materials after Absorption of CO₂

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Abstract: To disclose influences of ultrasonic vibration agitation on the carbonation resistance of cement-based materials after absorption of CO₂, the variation laws in internal carbonization zone were explored by the testing carbonization depth and carbonization range (pH variation range) of cement mortar after CO₂ absorption at different ages. Results demonstrated that when CO₂ absorption volumes of the cement mortar before carbonization were 0.44%, 0.88%, 1.32%, 1.76%, and 2.20% (28 d), the carbonization depth under ultrasonic vibration decreased by 5.5%, 12.3%, 21.7%, 20.7%, and 26.7% compared to those under mechanical stirring, respectively. When the ultimate CO₂ absorption volume increased to 2.2% of cement mass, the extended degree of cement mortar was 103.23 mm, which decreased by 5.4% compared to that before CO₂ absorption. pH variation values of the carbonization range under ultrasonic vibration presented a rising trend with the increase of CO₂ absorption volume of cement mortar before carbonation. This indicated that, with the increase of CO₂ absorption volume of cement mortar before carbonation increases under ultrasonic vibration, the carbonization process of the hardened body of cement mortar might be decelerated to some extent. Additionally, changes in internal composition and physical images of cement-based materials after absorption of CO₂ were analyzed through microtest means like SEM and XRD. A carbonation resistance model was constructed, thus enabling disclosure of the variation mechanism of carbonation resistance of cement-based materials after absorption of CO₂ under mechanical stirring and ultrasonic vibration. Results demonstrated that the higher CO₂ absorption volume of fresh slurry generated more “nano-level” CaCO₃ crystal nucleus. Accordingly, it could improve the porous structure of the cement mortar, decrease the quantity of capillary tubes significantly, improve the compaction degree of cement-based materials effectively, and lower the diffusion rate of CO₂ in the cement paste base, thus improving the carbonation resistance. Research conclusions have important significance to decrease CO₂ emissions and improve carbonation resistance of concrete.

Keywords: fresh cement mortar; ultrasonic vibration agitation; CO₂ absorption; carbonation performances; pH value



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1. Introduction

With rapid economic development, fossil fuels are being consumed at a high rate, leading to a rapid increase in CO₂ concentration in the atmosphere. Rising surface and seawater temperatures are a direct consequence of this, accelerating the thawing of polar caps, alpine glaciers, and the rising sea level [1–3]. It is reported that rises in sea level may bring catastrophic influences and disasters to human life, with many islands and beaches expected to disappear, rises in groundwater levels, and damage to water supply pipeline systems, harbor facilities, and buildings. In particular, people in coastal regions will face considerable challenges in coastal regions [4–6]. Furthermore, global warming is threatening the natural ecosystem balance and human survival. Hence, greenhouse gas

emission reduction has become one of the hot topics in energy policy and environmental administration [7–9].

At present, sealing and solidification of CO₂ are the most common methods to decrease carbon emissions. Mineral carbonization and marine sequestration are the most common methods. However, the natural carbonization of minerals takes a long time since it is very slow. To accelerate the reaction, it is necessary to enhance minerals in advance, which may incur excessive energy consumption. Therefore, sealing potentials of mineral carbonization are not so optimistic. Besides, marine sequestration may increase CO₂ concentration on seawater surface, thus changing chemical characteristics of marine ecosystem and causing various problems like ocean acidification. Hence, concrete was applied to absorb, recycle, and solidify CO₂. This has important significance to realize sustainable development of the concrete industry, relieve greenhouse gas emission, and solve global warming. Therefore, it is urgent to explore a new method for CO₂ sealing and solidification. As everyone knows, cement in concrete may produce a lot of Ca(OH)₂ during hydration, which accounts to a high proportion (about 20–30%) in cement. Most of Ca(OH)₂ exists in cement stone pores as crystals. Ca(OH)₂ can react with CO₂ in solution to produce CaCO₃. This is also another important reason that concrete has such considerable CO₂ absorption volume. Compared with mineral carbonization and marine sequestration, CO₂ absorption based on concrete has stronger operability and more promising application prospects.

China's strategies for CO₂ emission reduction are significantly different from those of other countries. While the CCUS (CO₂ capture, use and storage) strategy is adopted in foreign countries, China adopts the CCUS strategy whereby CO₂ is collected and used as a resource [10–13]. So far, research on the influence of ultrasonic vibration agitation stirring on CO₂ absorption by fresh paste has achieved some fruits, with some studies reporting that the ultrasonic “cavitation effect” can improve the CO₂ absorption rate and the total absorbed amount by the cement mortar.

Interestingly, the ultrasonic cavitation effect is stronger when the ultrasonic frequency is lower. An ultrasonic frequency of 20 kHz has been found to maximise the CO₂ absorption rate and ultimate absorption amount. When the ultrasonic frequency is 20 KHz, the water cement ratio is 0.7, and the mixing rate is 280 ± 5 r/min; the amount of CO₂ absorbed by the cement paste accounts for 3.17% of the cement mass, while under mechanical mixing, the amount of CO₂ absorbed by the cement paste accounts for 2.64% of the cement mass with the same water–cement ratio and mixing rate. In addition, some studies have shown that ultrasonic vibration mixing can significantly improve the uniform distribution of cement particles in freshly mixed pastes, thereby effectively improving the compressive strength of cement-based materials, while the impact on flexural strength is not significant. Ultrasonic vibration mixing molding can effectively reduce the porosity and pore size of carbonated cement paste, making the pore distribution of cement paste more uniform and greatly improving the compactness, thereby better improving and strengthening the internal pore structure of cement and improving the durability of cement-based materials [14,15].

Nevertheless, the carbonation of concrete is a complicated physiochemical process. Ca(OH)₂ is one of the main hydration product of cement, the content of which can be easily determined given a fixed mixing ratio of cement [16–19]. However, during the stirring of concrete, absorbed CO₂ gases can react with, and thus consume, some of the Ca(OH)₂ present in the cement to produce CaCO₃. This will inevitably cause a portion of Ca(OH)₂ to be consumed in the concrete. Will the consumption of Ca(OH)₂ affect the internal carbonation resistance of the concrete? This is very important for reinforced concrete because carbonation will reduce the alkalinity of the concrete, which will weaken the protective effect of the concrete on the reinforcement. Hence, it is very necessary to study the influences of cement-based materials after CO₂ absorption on the carbonation resistance of concrete.

2. Test Study

2.1. Raw Materials

The cement used in this study was P·O 42.5 cement produced by the Xuzhou Zhonglian Cement Group. The mean grain size, density, standard-consistency water requirement, fineness (0.08 mm square-hole sieve), and specific surface area were 14.813% X_{av} (μm), 3.14 g/cm³, 28.1%, 1.02%, and 3300 cm²/g, respectively. The specific chemical composition and mineral composition are listed in Tables 1 and 2. The CO₂ was high-purity ($\geq 99.5\%$) CO₂ produced by a special gas plant in Xuzhou. Tap water was also used.

Table 1. Chemical composition of the P·O 42.5 cement.

Chemical Composition	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	f-CaO	Loss
Content (%)	22.1	5.34	3.44	65.33	2.11	0.39	0.13

Table 2. Mineral composition of the P·O 42.5 cement.

Composition	C ₃ S	C ₂ S	C ₃ A	C ₄ AF
Content (%)	54.04	22.84	8.39	10.42

2.2. CO₂ Absorption Device

The ultrasonic vibration agitation stirring device developed by the research team is used as a stirring device for CO₂ absorption under ultrasonic vibration agitation. It is composed of a transducer (chosen according to the desired amplitudes and vibration frequency of the vibration system), an ultrasonic power unit, an amplitude-change pole, a motor, stirring blades, a flowmeter, sealing gaskets, and other parts (Figure 1).

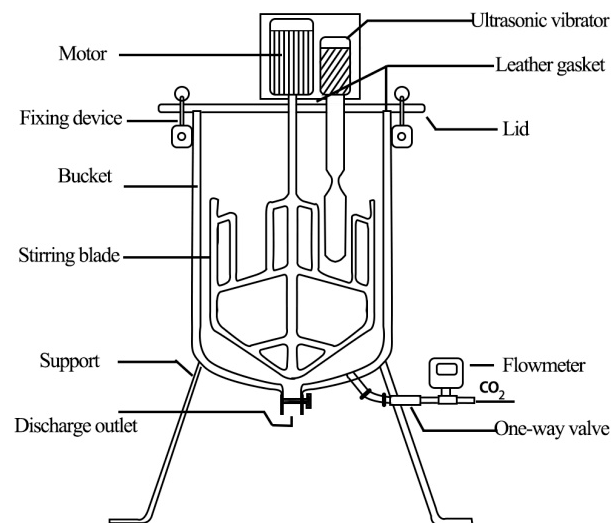


Figure 1. Schematic diagrams of ultrasonic agitation devices (reprinted with permission from Ref. [13]).

2.3. Specimen Grouping

Specimens were grouped into Q and R. Group Q refers to specimens with different CO₂ AA under ultrasonic vibration agitation. Group R refers to specimens with different CO₂ AA under mechanical agitation.

CO₂ absorption volume (G) of cement mortar is expressed as the percentage of the absorbed CO₂ mass of cement mortar in cement mass. The relationship between G and the CO₂ flow (Q) which was read by the CO₂ flowmeter was:

$$G = \frac{Q}{V_0} \times \frac{M_0}{m} \times 100\% \quad (1)$$

where Q refers to the supplied CO₂ flow (mL). V_0 represents the molar volume of CO₂ and it is about 22.4×10^3 mL under standard conditions. M_0 represents the molar mass of CO₂ and it is about 44 g/mol. m refers to cement mass (g).

After repeated tests (G increased from a low volume to the ultimate volume successively), the CO₂ absorption volume parameter increased orderly at the rate of 0.44%, which valued 0.44%, 0.88%, 1.32%, 1.76%, and 2.20%, respectively. This series of coefficients not only have laws, but also are more intuitive.

CO₂ AA in Q1, Q2, Q3, Q4, Q5, Q6 of Group Q as well as R1, R2, R3, R4, R5, R6 of Group R are 0%, 0.44%, 0.88%, 1.32%, 1.76%, and 2.20% of cement mass, respectively. (Note: The number of each ages (3 days, 7 days, 28 days) of specimens for all test were 3).

2.4. Specimen Preparation

(1) Preparation of cement mortar specimens after CO₂ absorption under mechanical agitation

A total of 3 kg cement and 1.5 kg water were added into the mixing tank of the CO₂ absorption device to prepare cement slurry with a water-to-cement ratio (W/C) of 0.5. The stirring rate was set at 210 ± 5 r/min. After the slurry was stirred well, the CO₂ flow valve was opened, and the mixture vibrated and stirred at the same time. The CO₂ AA increased at 0.44% of the cement mass. Then, 2025 g of carbonised cement paste was released from the mixing tank. The released carbonised cement paste was divided into 3 parts equally (675 g each), to which 1350 g of standard sands (GB/T17671-1999) were added. Rectangular solid mortar specimens (40 mm × 40 mm × 160 mm) were prepared. All specimens were demolded after curing for 24 h and then cured to the regulated age under standard conditions of 20 ± 2 °C and humidity >95%. Samples were then used for relevant tests.

(2) Preparation of cement mortar specimens after CO₂ absorption under ultrasonic vibration agitation stirring

The ultrasonic vibration agitation stirring device was initiated and the ultrasonic vibration agitation frequency was set at 20 kHz. The mortar specimens of the paste after CO₂ absorption under ultrasonic vibration agitation stirring were prepared by the same process as that of the mechanical agitation device.

2.5. Concrete Carbonization Test Procedure

(1) Preprocessing of specimens and accelerated carbonation test

After 28 days of standard curing, specimens were dried for 48 h under a constant temperature of 60 °C. Then, specimens were cooled for 48 h in a natural environment. Next, concrete specimens were put in the accelerated carbonation box and the accelerated carbonation test was carried out according to the Standards for Long-term Performance and Durability Test Methods of Ordinary Concretes (GB/T 50082-2009). Test parameters included a CO₂ mass fraction of 20%, a carbonation temperature of 20 ± 2 °C and a humidity level of $70 \pm 5\%$. The carbonation ages were set for 3, 7, and 28 d, respectively [20]. Take out the carbonized 3D concrete specimen and break it on a press. Cut a block with a thickness of about 50 mm from a section, clean the powder at the section position, immediately spray 1% phenolphthalein indicator, and measure the depth of carbonization. Then place the three-dimensional carbonized test block in the carbonization boxes 7 d and 28 d using the same method for carbonization testing.

(2) Calculation of carbonation depth

The concrete carbonation specimens at the age of 3, 7, and 28 d were laid out evenly to measure the distance from the specimen surface to the color development point. Test points were set every 5 mm and each test's data were recorded. Next, the mean value was calculated. The average carbonation depth of concrete was calculated according to the following equation [21]:

$$\overline{d_t} = \frac{\sum_{i=1}^n d_i}{n} \quad (2)$$

where d_i refers to the average carbonation depth, d_i is the carbonation depth at any point on the carbonation surface, and n denotes the total number of measuring points.

3. Test Contents and Methods

3.1. Carbonization Performances Test

CO₂ AA (i.e., the percentage of CO₂ absorption mass by cement paste in the cement mass) in Group Q and Group R were 0%, 0.44%, 0.88%, 1.32%, 1.76%, and 2.20%. The fresh cement pastes were carbonised for 3, 7, and 28 d, and the carbonation depths of cement mortar with different CO₂ AA under ultrasonic vibration agitation and mechanical were tested. The carbonation depths at different ages under ultrasonic vibration agitation stirring and mechanical agitation were compared and the effects of ultrasonic vibration agitation stirring on the carbonation resistance of the hardened body of carbonised cement mortar were investigated.

3.2. pH Test

The 28 d cement mortar specimens with different CO₂ AA under mechanical agitation and ultrasonic vibration agitation stirring were broken up and the chromaticity was measured using the phenolphthalein indicator to obtain the specific depth of the complete carbonation zone. Subsequently, specimens were collected through drilling from the concrete cross-section surface. To increase the accuracy of the pH variation value, drilling points were set every 2 mm up until the center of each specimen. Each specimen was collected and put in a reagent bottle with a ground stopper quickly and sealed up. Each specimen was ground into powders and poured into a 25 mL beaker, where they were mixed with distilled water in a solid-to-water ratio of 3:1 to form a paste. The pH values of each mixture were recorded every 5 min. By comparing with mechanical agitation, the effects of ultrasonic vibration agitation on the carbonation process of the pH variation zone of cement mortar could be investigated.

3.3. XRD Test

To avoid poor test effects for low CO₂ AA and few reaction products of corresponding cement specimens, the specimen with 2.20% of CO₂ AA was chosen for XRD analysis of carbonised cement paste. In this experiment, specimens were divided into three groups, including specimens without CO₂ AA, specimens with 2.20% of CO₂ AA under mechanical agitation, and specimens with 2.20% of CO₂ AA under ultrasonic vibration agitation stirring. These three groups of molded specimens were cured for 28 d and then ground into powder samples. Powder samples were polished by a polisher until a diameter of about 10 µm. The powders were pinched and rotated between two fingers until there was a feeling of particles. All prepared samples were put in bags and sealed up before tests. The XRD test device used the D8 ADVANCE type XRD analyser from the Modern Analysis and Computation Center of China University of Mining and Technology. Diffraction spectra of testing samples were recorded and phase analysis was carried out [22]. The working parameters of the XRD analyser include a Cu target K α ray, a tube voltage of 40 kV, and a tube current of 30 mA.

3.4. SEM Test

The 28 d Q0, Q2, Q4 and Q6 samples were broken and their CO₂ AA were measured to be 0%, 0.88%, 1.76%, and 2.20%, respectively. Each specimen was prepared into a 1 mm thick sample and then immersed in absolute ethyl alcohol for 48 h to terminate the hydration of cement. Specimens were dried for 24 h in a constant-temperature 65 °C blast oven and then put in a carbon coater for surface spraying [23–25].

The microstructures of the prepared 28 d testing specimens were observed using an SEM under a magnification of 10,000 $\times$. Paste specimens before and after CO₂ absorption were observed before carbonation. The microstructural changes after carbonation for 28 d and internal causes were analysed.

4. Test Results and Analysis

4.1. Carbonation Performances Analysis

(1) Mechanical agitation

(1) Carbonation age of 3 d

The carbonation of cement mortar after CO₂ absorption under mechanical agitation at 3 d is shown in Figure 2. Clearly, the area of color developing zone in specimens decreases with the increase of CO₂ AA. The average carbonation depth under different CO₂ AA was calculated according to test results in Figures 2–4. The results are listed in Table 3. It can be seen from Table 3 that with an increase in CO₂ AA, the carbonation depths in the hardened body of fresh cement mortar at 3 d are 3.1, 2.9, 3.3, 3.5, 3.5, and 3.6 mm, respectively. The carbonation depth of cement mortar with 0.44% of CO₂ absorption is 6.9% lower than that of cement mortar without CO₂ absorption. However, the carbonation depth increases by 13.8%, 6.1%, 0%, and 2.9% with the continuous increase of CO₂ AA in the later stages.

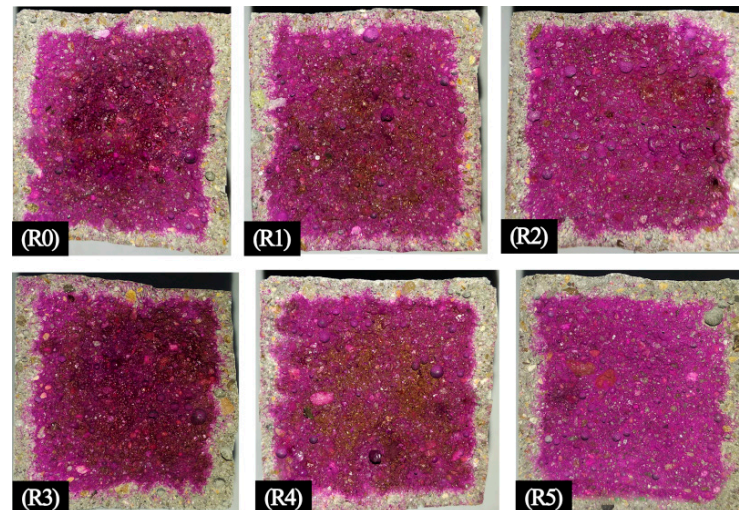


Figure 2. Image of carbonation depth under mechanical agitation (3 d).

(2) Carbonation age at 7 d

The carbonation of cement mortar after CO₂ absorption under mechanical agitation at 7 d is shown in Figure 3. Clearly, the area of color developing zone in specimens decreases compared to that at 3 d, but it changes slightly macroscopically with the increase of CO₂ AA. It can be seen from Table 3 that the carbonation depth of cement mortar with 0.44% of CO₂ absorption at 7 d is 2.3% lower than that of cement mortar without CO₂ absorption. When the CO₂ AA increases to 0.88% and 1.32%, the carbonation depths are increased by 4.7% and 2.2%, respectively. As the CO₂ AA continues to increase to 1.76%, the carbonation depth declines by 2.2%. When the CO₂ AA reaches 2.20%, the carbonation depth remains the same. In summary, the carbonation changes of hardened paste at 7 d are not stable due to influences by CO₂ AA.

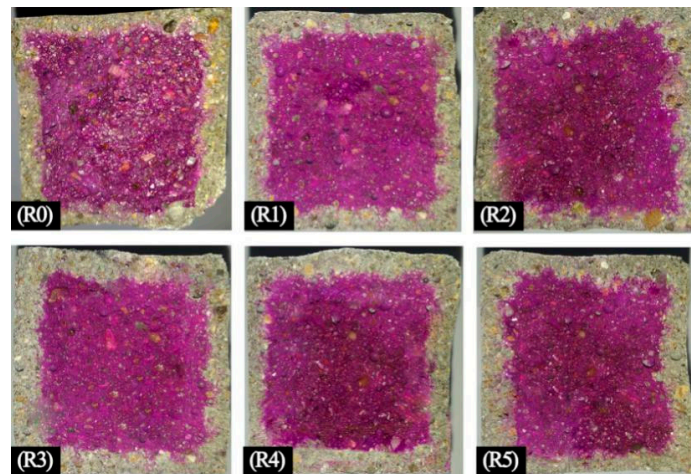


Figure 3. Image of carbonation depth under mechanical agitation (7 d).

(3) Carbonation age at 28 d

The carbonation of cement mortar after CO₂ absorption under mechanical agitation at 28 d is shown in Figure 4. Clearly, the area of color-developing zone in specimens decreases compared to that at 7 d and it continues to decrease significantly with the increase of CO₂ AA. At 28 d, carbonation depths of the hardened body of fresh cement mortar are 5.6, 5.5, and 5.7 mm as well as 6.0, 5.8, and 6.0 mm with the increase of CO₂ AA. When the CO₂ AA is 0.44%, the carbonation depth is 1.8% lower compared to that before CO₂ absorption. Subsequently, CO₂ AAs are increased by 1.8%, 7.1%, 3.6%, and 7.1%, respectively.

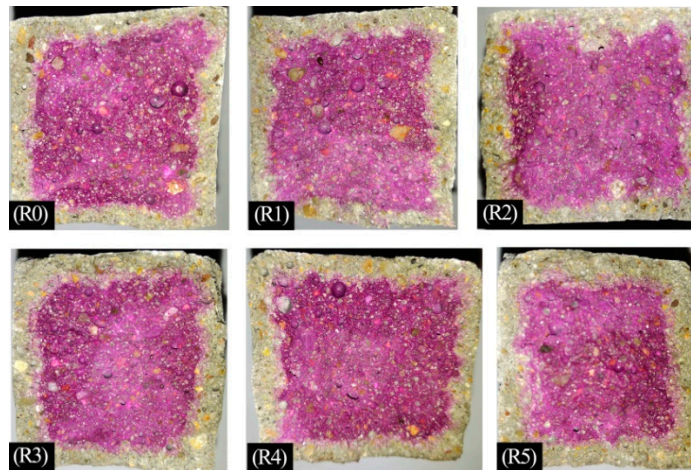


Figure 4. Image of carbonation depth under mechanical agitation (28 d).

Table 3. Absorb different amounts of CO₂ and carbonize depth at different ages (mechanical agitation).

Specimen No.	R0	R1	R2	R3	R4	R5
CO ₂ AA (%)	0	0.44	0.88	1.32	1.76	2.20
3 d/mm	3.1 ± 0.2	2.9 ± 0.1	3.3 ± 0.2	3.5 ± 0.1	3.5 ± 0.2	3.6 ± 0.2
7 d/mm	4.4 ± 0.3	4.3 ± 0.4	4.5 ± 0.3	4.6 ± 0.3	4.5 ± 0.2	4.5 ± 0.4
28 d/mm	5.6 ± 0.3	5.5 ± 0.3	5.7 ± 0.4	6.0 ± 0.4	5.8 ± 0.4	6.0 ± 0.5

In view of increasing and decreasing percentages of carbonation depths at different ages, the carbonation depth of hardened paste generally increases with the increase of CO₂ AA. It changes slightly at 7 d, but it increases to some extent with increasing curing age and CO₂ AA. This reveals that the hardened body of cement mortar with CO₂ absorption

under mechanical agitation can influence the carbonation resistance of concrete to some extent. However, such influences are not obvious.

(2) Ultrasonic vibration agitation

(1) Carbonation age at 3 d

The carbonation of cement mortar after CO₂ absorption under ultrasonic vibration agitation at 3 d is shown in Figure 5. The average carbonation depth under different CO₂ AA was calculated according to the test results in Figures 5–7. The results are listed in Table 4. It can be seen from Figure 5 that, with the increase of CO₂ AA, the area of color-developing zones increases to some extent. It can be calculated from Table 4 that, for each increase in CO₂ AA, the carbonation depth at 3 d decreases by 7.7%, 8.3%, 4.5%, 9.5%, 5%, and 0%, respectively.

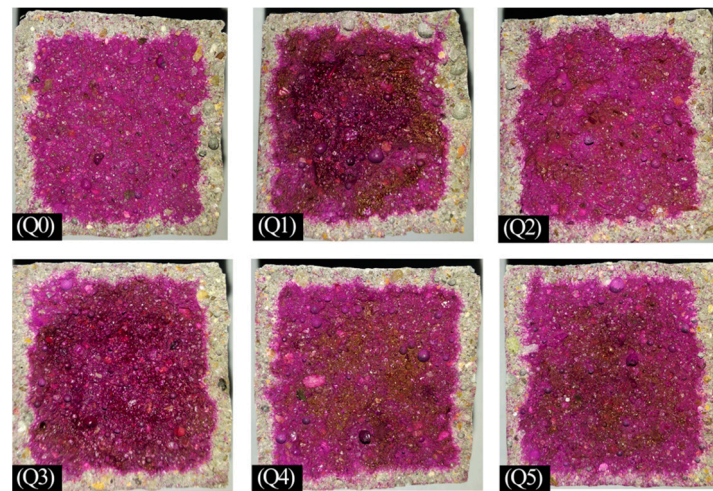


Figure 5. Image of carbonation depth under ultrasonic vibration agitation (3 d).

(2) Carbonation age at 7 d

The carbonation of cement mortar after CO₂ absorption under ultrasonic vibration agitation at 7 d is shown in Figure 6. The area of color-developing zones is smaller compared to that at 3 d. With the increase of CO₂ AA, the area of color-developing zones still increases to some extent. It can be calculated from Table 4 that with the increase of CO₂ AA, the carbonation depth at 7 d decreases by 5%, 5.2%, 5.6%, 2.9%, 6.1%, and 6.5%, respectively.

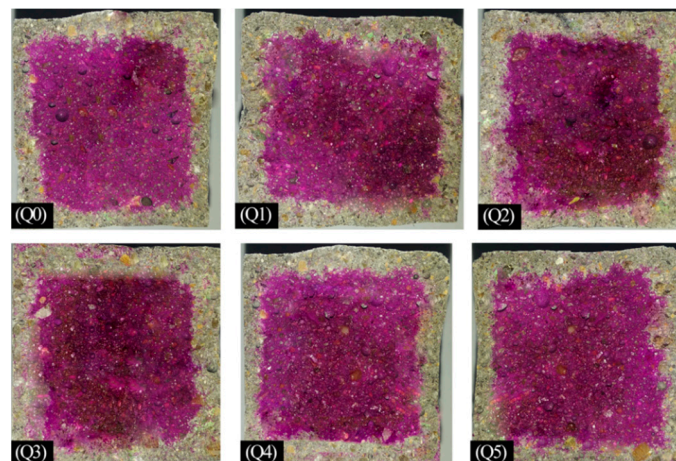


Figure 6. Image of carbonation depth under ultrasonic vibration agitation (7 d).

(3) Carbonation age at 28 d

The carbonation of cement mortar after CO₂ absorption under ultrasonic vibration agitation at 28 d is shown in Figure 7. The area of color developing zones further decreases compared to that at 7 d. Similar to the observations at 3 d and 7 d, the area of color-developing zones still increases to some extent with the increase of CO₂ AA. It can be calculated from Table 4 that with the increase of CO₂ AA, the carbonation depth at 28 d decreases by 1.9%, 4%, 6.4%, 2.2%, 4.5%, and 4.8%, respectively.

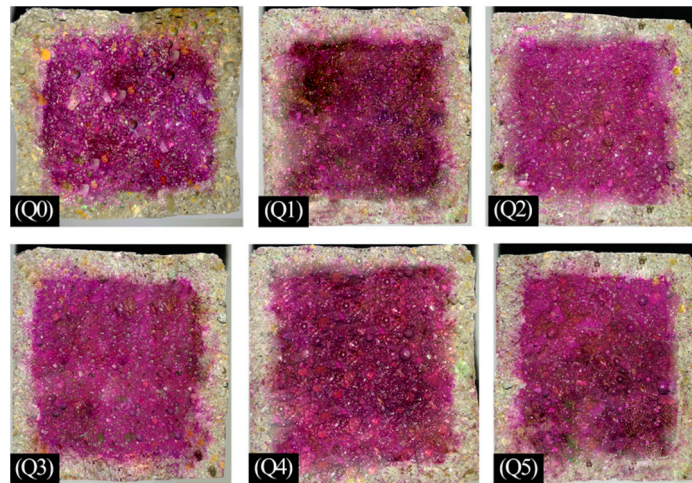


Figure 7. Image of carbonation depth under ultrasonic vibration agitation (28 d).

Table 4. Absorb different amounts of CO₂ and carbonize depth at different ages (ultrasonic vibration agitation).

No.	Q0	Q1	Q2	Q3	Q4	Q5
CO ₂ AA/%	0	0.44	0.88	1.32	1.76	2.20
3 d/mm	2.8 ± 0.1	2.6 ± 0.2	2.4 ± 0.1	2.3 ± 0.2	2.1 ± 0.1	2.0 ± 0.1
7 d/mm	4.2 ± 0.3	4.0 ± 0.2	3.8 ± 0.3	3.6 ± 0.2	3.5 ± 0.3	3.3 ± 0.2
28 d/mm	5.3 ± 0.4	5.2 ± 0.3	5.0 ± 0.4	4.7 ± 0.4	4.6 ± 0.3	4.4 ± 0.4

(3) Comparative analysis of two stirring methods

To disclose the effects of ultrasonic vibration agitation on carbonation depth clearly, the carbonation depths of specimens with different CO₂ AA s at different ages under mechanical agitation and ultrasonic vibration agitation stirring are compared and analysed. The results are shown in Figure 8. Obviously, the carbonation depths of cement mortar specimens at 3 d, 7 d, and 28 d under mechanical agitation increase only slightly with the increase of CO₂ AA. The carbonation depth of cement mortar under ultrasonic vibration agitation declines quite considerably with the increase of CO₂ AA. This reveals that the carbonation depth of hardened paste at different ages is negatively related to the CO₂ AA under ultrasonic vibration agitation.

In a word, when CO₂ absorption volumes of the cement mortar before carbonization were 0.44%, 0.88%, 1.32%, 1.76%, and 2.20% (28 d), the carbonization depth under ultrasonic vibration decreased by 5.5%, 12.3%, 21.7%, 20.7%, and 26.7% compared to those under mechanical stirring, respectively. When the ultimate CO₂ absorption volume increased to 2.2% of cement mass, the extended degree of cement mortar was 103.23mm, which decreased by 5.4% compared to that before CO₂ absorption. Additionally, the carbonation resistance of cement mortar molded under ultrasonic vibration agitation is improved. The reasons are introduced as follows. Since ultrasonic vibration agitation increases the collision probability of cement particles effectively, the hydration rate of cement particles, the CO₂ absorption rate, and the neutralization rate of Ca(OH)₂ hydration product are increased

significantly, thus enabling the construction and development of a more uniform and compacted microstructure of cement paste [14].

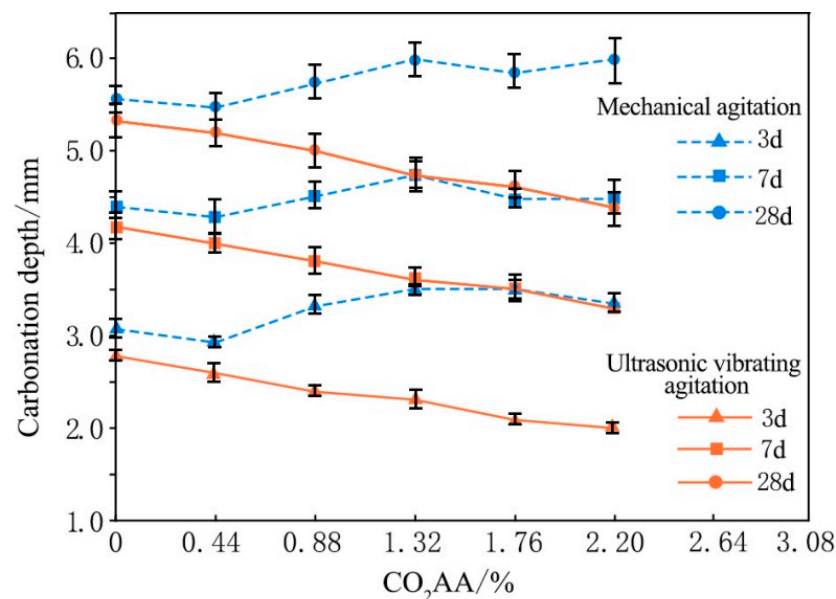


Figure 8. Carbonization depth curves of different CO₂ absorption stages.

4.2. pH Changing Zone

Since the carbonation depth measurement method based on the phenolphthalein indicator cannot distinguish changes in pH value within incomplete carbonation zones accurately [26], the carbonation process of the hardened paste is represented more clearly and intuitively through the length of the pH changing section. This has very important significance to elaborate the carbonation process of hardened paste accurately [27,28]. The pH distribution patterns of hardened pastes with different CO₂ AA under mechanical agitation and ultrasonic vibration agitation stirring are introduced as follows. The test results of pH values at different depths are compared. Therefore, the pH distribution and variation laws of the carbonation zone of the hardened paste under ultrasonic vibration agitation with the increase of CO₂ AA are disclosed more clearly.

(1) Mechanical agitation

The test results and distribution patterns of the pH value of the carbonation zone of the hardened paste after CO₂ absorption under mechanical agitation (28 d) are shown in Table 5 and Figure 9.

Table 5. pH value at 28 d in carbonized area of cement mortar with different CO₂ absorption (mechanical agitation).

Carbonation Depth/mm	0	2	4	6	8	10	12	14	16	18	20
CO ₂ 0%	8.5 ± 0.5	8.5 ± 0.6	8.5 ± 0.5	9.5 ± 0.7	11.7 ± 0.8	12.0 ± 1.1	13.0 ± 1.2	13.2 ± 1.1	13.1 ± 1.2	12.9 ± 1.0	13.0 ± 1.1
CO ₂ 0.44%	8.5 ± 0.6	8.5 ± 0.7	8.5 ± 0.6	9.6 ± 0.8	11.0 ± 0.9	12.2 ± 1.1	13.1 ± 1.0	13.0 ± 1.2	13.0 ± 1.1	13.1 ± 1.2	13.1 ± 1.0
CO ₂ 0.88%	8.5 ± 0.6	8.5 ± 0.7	8.5 ± 0.8	9.3 ± 0.6	10.5 ± 0.9	11.6 ± 1.0	13.1 ± 1.3	12.9 ± 1.3	13.2 ± 1.0	12.9 ± 1.1	13.1 ± 1.1
CO ₂ 1.32%	8.5 ± 0.6	8.5 ± 0.7	8.5 ± 0.6	8.5 ± 0.8	9.7 ± 0.9	11.1 ± 0.9	13.0 ± 1.2	13.1 ± 1.2	12.9 ± 1.1	13.0 ± 1.2	13.1 ± 1.2
CO ₂ 1.76%	8.5 ± 0.5	8.5 ± 0.7	8.5 ± 0.8	9.0 ± 0.2	10.2 ± 1.0	10.9 ± 1.1	12.9 ± 1.2	13.0 ± 1.1	13.0 ± 1.2	13.2 ± 1.2	13.1 ± 1.3
CO ₂ 2.20%	8.5 ± 0.6	8.5 ± 0.7	8.5 ± 0.6	8.5 ± 0.6	9.5 ± 1.0	10.8 ± 0.9	12.8 ± 1.1	13.1 ± 1.1	13.0 ± 1.0	13.1 ± 1.1	13.2 ± 1.0

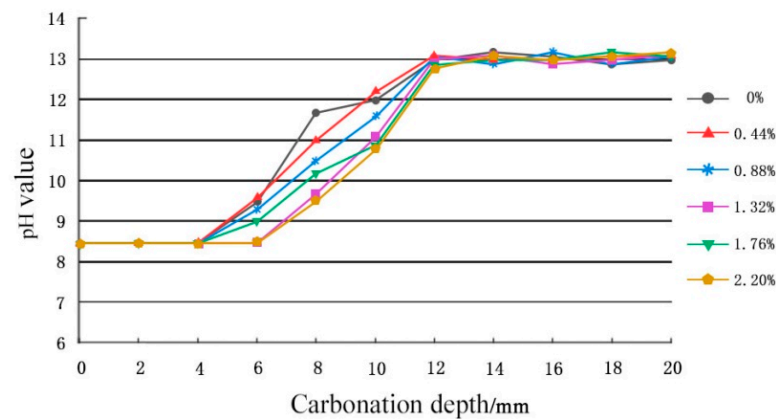


Figure 9. pH distribution of CO₂-absorbing cement mortar for 28 days (mechanical agitation).

The pH values at carbonation depths of 2, 4, 6, 8, 10, 12, 14, 16, 18, and 20 mm when the CO₂ AA is 0%, 0.44%, 0.88%, 1.32%, 1.64%, and 2.20% were tested.

According to the tested pH values at different carbonation depths, zones with great variation in pH value are mainly concentrated within the carbonation depth range of 6 to 12 mm. It can be seen from Figure 9 that the pH of the carbonation zone decreases with the increase of CO₂ AA by the cement mortar.

(2) Ultrasonic vibration agitation stirring

The test results and distribution patterns of the pH value of the carbonation zone of the hardened paste after CO₂ absorption under ultrasonic vibration agitation stirring (28 d) are shown in Table 6 and Figure 10.

Table 6. pH value at 28 d in carbonized area of cement mortar with different CO₂ absorption (ultrasonic vibration agitation).

Carbonation Depth/mm	0	2	4	6	8	10	12	14	16	18	20
CO ₂ 0%	8.5 ± 0.2	8.5 ± 0.2	8.5 ± 0.2	8.6 ± 0.2	9.8 ± 0.2	11.2 ± 0.2	13.1 ± 0.2	13.0 ± 0.2	13.1 ± 0.2	12.9 ± 0.2	13.1 ± 0.2
CO ₂ 0.44%	8.5 ± 0.2	8.5 ± 0.2	8.5 ± 0.2	8.7 ± 0.2	10.0 ± 0.2	11.5 ± 0.2	13.3 ± 0.2	13.1 ± 0.2	13.1 ± 0.2	13.2 ± 0.2	13.0 ± 0.2
CO ₂ 0.88%	8.5 ± 0.2	8.5 ± 0.2	8.5 ± 0.2	8.7 ± 0.2	10.4 ± 0.2	12.0 ± 0.2	13.1 ± 0.2	12.9 ± 0.2	13.2 ± 0.2	13.1 ± 0.2	13.1 ± 0.2
CO ₂ 1.32%	8.5 ± 0.2	8.5 ± 0.2	8.5 ± 0.2	8.8 ± 0.2	10.9 ± 0.2	12.3 ± 0.2	13.0 ± 0.2	13.2 ± 0.2	13.0 ± 0.2	13.3 ± 0.2	13.2 ± 0.2
CO ₂ 1.76%	8.5 ± 0.2	8.5 ± 0.2	8.5 ± 0.2	9.0 ± 0.2	11.2 ± 0.2	12.7 ± 0.2	13.1 ± 0.2	13.2 ± 0.2	13.1 ± 0.2	13.2 ± 0.2	13.1 ± 0.2
CO ₂ 2.20%	8.5 ± 0.2	8.5 ± 0.2	8.5 ± 0.2	9.0 ± 0.2	11.5 ± 0.2	13.1 ± 0.2	13.2 ± 0.2	13.0 ± 0.2	13.2 ± 0.2	13.1 ± 0.2	13.2 ± 0.2

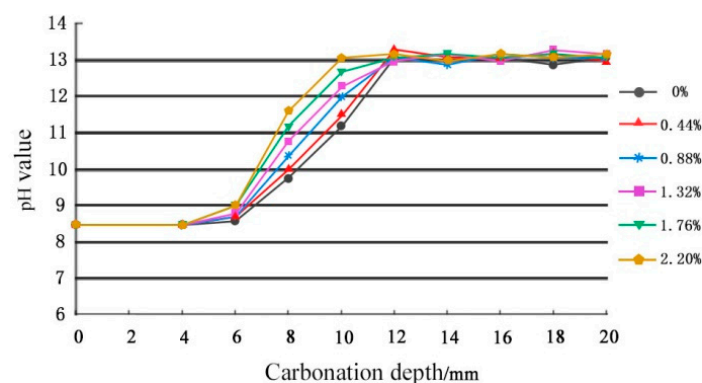


Figure 10. pH distribution of CO₂-absorbing cement mortar for 28 days (ultrasonic vibration agitation).

It can be seen from Figure 10 that zones with large pH variations are concentrated within the carbonation depth range of 6 to 12 mm, which is similar to that under mechanical agitation. Moreover, the pH value increases gradually with the increase in carbonation depth and increases with the increase in CO₂ AA by the cement mortar.

(3) Comparative analysis between two stirring methods

In the early carbonation stage, the pH value of specimens at carbonation depths of 0~4 mm is 8.5 under both mechanical agitation and ultrasonic vibration agitation stirring. This zone is called the complete carbonation zone. The pH value of specimens at carbonation depths of 4~12 mm increases to about 13.0 (Figures 9 and 10). When the carbonation depth increases to 12 mm, the pH of cement mortar remains constant at about 13.0, reaching a stable state. It indicates that the Ph value of concrete increases with the increase in carbonation depth under both mixing methods, and the alkalinity increases.

In addition, with the increase in the amount of CO₂ absorbed by the freshly mixed cement paste under the two mixing methods, the pH value of the hardened cement mortar in the carbonation zone changes in reverse order. Under mechanical agitation, the pH value of the carbonation zone generally decreases with the increase in CO₂ AA by the fresh cement mortar. However, the pH value of the carbonation zone under ultrasonic vibration agitation stirring is relatively stable and almost presents a rising trend.

The major reasons for this observation are introduced as follows. Before molding of specimens, there is no variation law of pH at the same carbonation depth with the increase of CO₂ AA under mechanical agitation, due to the insufficient distribution uniformity in the paste. Instead, the pH value declines gradually. Influenced by the ultrasonic “cavitation effect”, the internal structural distribution is more uniform and more compacted under ultrasonic vibration agitation stirring. With the increase of CO₂ AA, the porosity of specimens decreases continuously. When the specimens are further carbonized with CO₂ in air, the invasion speed of CO₂ into the specimens decreases with the decrease of internal porosity. As a result, the pH value presents a rising trend. This proves that under ultrasonic vibration agitation stirring, the carbonation slows down after the cement-based materials absorb more CO₂, which is conducive to improving the carbonation performance of cement-based materials.

4.3. XRD Analysis of Hardened Paste

XRD spectra of cement mortar absorbing 2.20% CO₂ and the cement mortar without CO₂ absorption under two stirring methods are shown in Figure 11. The XRD spectra and phase analysis results of cement paste without CO₂ absorption under mechanical agitation, cement paste with 2.20% of CO₂ absorption under mechanical agitation, and cement paste with 2.20% of CO₂ absorption under ultrasonic vibration agitation stirring are shown in Figure 11a–c, respectively.

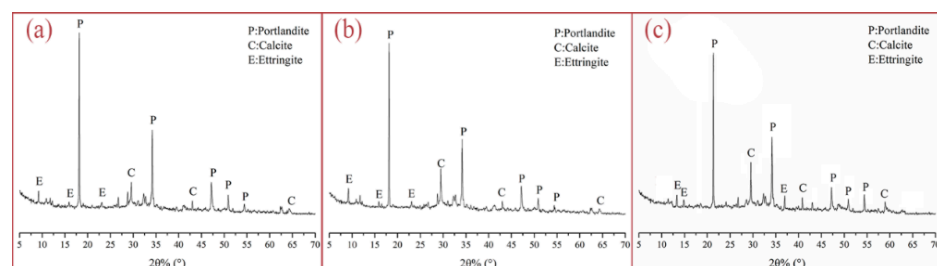


Figure 11. Diffraction results of cement slurry sample. (a) Unabsorbed CO₂ cement paste; (b) 2.20% CO₂ (mechanical agitation); (c) 2.20% CO₂ (ultrasonic vibration agitation).

(1) Pure cement paste

It can be seen from Figure 11a that there is a diffraction peak of Ca(OH)₂ (Portlandite) in the XRD spectra of pure cement paste, indicating that this specimen contains abundant Ca(OH)₂ crystals in hydration products at 28 d. The diffraction peaks of CaCO₃ (Calcite) and 3CaO·Al₂O₃·3CaSO₄·32H₂O (Ettringite) are relatively low [29,30].

(2) Cement paste after CO₂ absorption under mechanical agitation

After 2.20% CO₂ is supplied to the fresh paste under mechanical agitation (Figure 11b), the diffraction peak of Ca(OH)₂ in the diffraction spectra is lower than that of the pure cement mortar. However, the reduction amplitude is very small, indicating that Ca(OH)₂ crystals decrease only to some extent. This reveals that the supplied CO₂ into the fresh cement paste reacts with and partly consumes some of the Ca(OH)₂ in the cement, thus reducing the diffraction peak of Ca(OH)₂ (Portlandite) to some extent. Furthermore, the diffraction peak of CaCO₃ (Calcite) of pure cement paste increases compared to that on the XRD spectra of the cement paste after CO₂ absorption. This is because some of the CaCO₃ is produced from the reaction of the supplied CO₂ and substances in the cement paste during stirring.

(3) Cement paste after CO₂ absorption under ultrasonic vibration agitation stirring

After 2.20% CO₂ is supplied to the fresh paste under ultrasonic vibration agitation stirring (Figure 11c), the diffraction peak of Ca(OH)₂ declines only slightly compared to that under mechanical diffraction. This is mainly because the crystal number of Ca(OH)₂ in cement paste after CO₂ absorption under ultrasonic vibration agitation changes and Ca(OH)₂ can come into contact to react with CO₂ better. Hence, Ca(OH)₂ is consumed and CaCO₃ crystals are produced.

Additionally, the cement paste also may react with CO₂ in the air in the curing tank. Since ultrasonic vibration agitation increases the compaction of curing specimens, the CO₂ AA from air into the paste decreases accordingly, thus resulting in small influences on Ca(OH)₂ during the curing process [31–33].

It can be seen from Figure 11 that the diffraction peak of Ca(OH)₂ under ultrasonic vibration decreased to some extent compared to that under mechanical stirring (Portlandite). This proves that CO₂ absorption by the cement mortar under ultrasonic vibration had small influences on Ca(OH)₂. Additionally, peaks of the CaCO₃ crystal might be increased to some extent. To sum up, cement mortar after CO₂ absorption under ultrasonic vibration has small influences on (Portlandite) Ca(OH)₂, and CaCO₃ (Calcite) might increase to some extent.

4.4. Microstructure Analysis of Hardened Paste

The post-carbonation SEM images (amplification: 10,000×) of hardened pastes with different CO₂ AA under ultrasonic vibration agitation are shown in Figure 12. Pure cement paste before carbonation is shown in Figure 12a. After carbonation, flocculating constituents, calcium carbonate crystals, and ettringite whiskers can be observed on the gel surface. The flocculating constituent is relatively loose. It can be seen from Figure 12b that the flocculating constituent in the cement paste with 0.88% absorbed CO₂ before carbonation increases after carbonation. Compared with the pure cement paste, there are fewer pores and tighter flocculating constituents.

The flocculating constituent of cement paste which has absorbed 1.76% CO₂ before carbonation (Figure 12c) continues to increase after carbonation, and its compaction also increases to some extent. For the cement paste with a CO₂ AA of 2.20% before carbonation (Figure 12d), particulate flocculating constituents are distributed in high density, thus further decreasing the pore size and increasing compaction. Based on the above analysis, gels increase after carbonation of the cement paste with CO₂ absorption under ultrasonic vibration agitation. Such increasing amplitude is positively related to the CO₂ AA before carbonisation, thus blocking some capillaries and decreasing the pores of flocculating constituents.

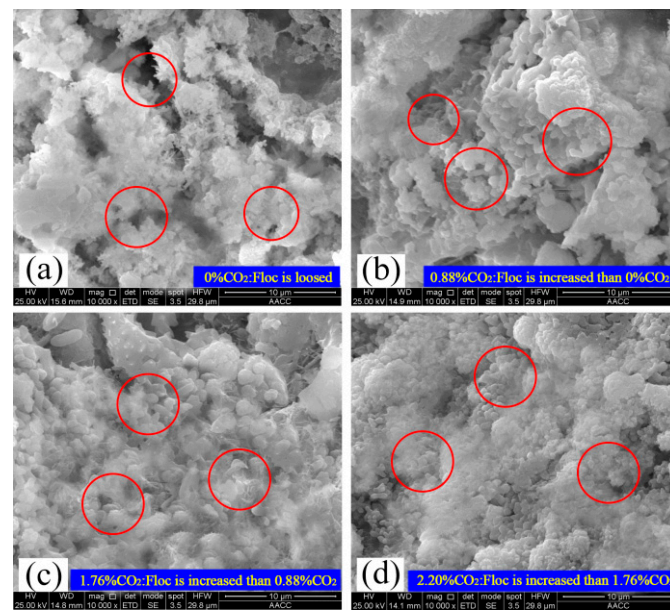


Figure 12. SEM image of carbonized cement by ultrasonic vibration agitation: (a) 0%; (b) 0.88%; (c) 1.76%; (d) 2.20%.

5. Concrete Carbonation Mechanism Analysis

5.1. Carbonation Mechanism Analysis of Ordinary Concretes

For the convenience of analysis of the carbonation mechanism of cement paste after CO_2 absorption, the carbonation mechanism of ordinary concrete is analysed first in this section. The internal mechanism before concrete carbonation is shown in Figure 13a. After cement particles react with water, C-S-H gel is produced and solid-phase crystals $\text{Ca}(\text{OH})_2$ are produced through hydration [34,35]. The internal mechanism after concrete carbonation is shown in Figure 13b. CO_2 in air reacts with $\text{Ca}(\text{OH})_2$ in the cement gel after it penetrates the pores and capillaries of the concrete, producing CaCO_3 which may precipitate due to its low solubility. To assure constant concentrations of Ca^{2+} and OH^- in the pore solution, $\text{Ca}(\text{OH})_2$ in the original solid phase is dissolved to maintain a balanced Ca^{2+} content.

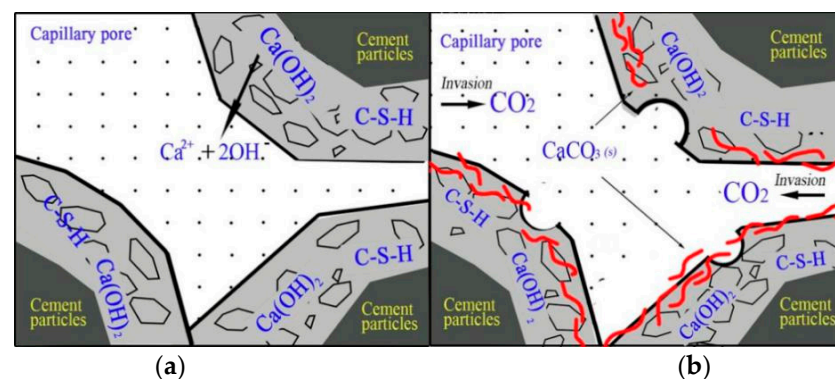


Figure 13. Carbonation mechanism of ordinary concrete: (a) Non carbonized; (b) After carbonization.

Furthermore, the product layer may restrict the reaction speed between CO_2 and alkaline components in concrete particles since CaCO_3 precipitates on capillary pore walls, but it may not influence the ultimate carbonation process of concrete. The continuous diffusion of CO_2 will enable it to continually react with $\text{Ca}(\text{OH})_2$, thus decreasing the $\text{Ca}(\text{OH})_2$ content and decreasing the alkalinity of the concrete. As such, the carbonation resistance of concrete is weakened.

5.2. Carbonation Microstructure Analysis

(1) Molding under mechanical agitation

In the early hydration stage, cement and water react to produce C-S-H gels. When the cement begins to mold, water in the hardened paste evaporates continuously, finally forming multiple pores with different sizes and shapes in the hardened body (Figure 14a). For the hardened body under mechanical agitation, the fresh paste has absorbed CO_2 before molding and reacts with $\text{Ca}(\text{OH})_2$ in the paste to produce CaCO_3 , thus increasing the hydration product in the cement. When the paste is hardened, the internal pore area decreases relatively, thus decreasing the CO_2 AA. Furthermore, there are small gel pores in the C-S-H gel through which some CO_2 may enter into hardened cement paste (Figure 14b).

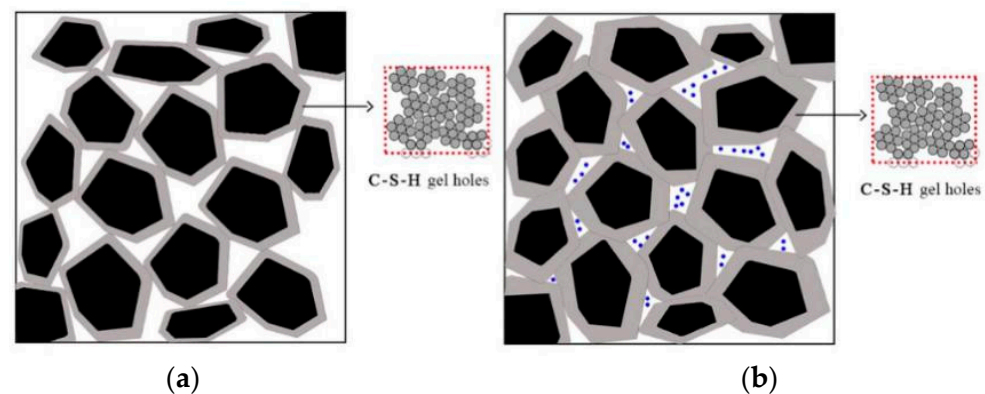


Figure 14. Mechanism of carbonation by mechanical agitation molding. (a) Non-invaded CO_2 ; (b) Invasion of CO_2 .

(2) Molding under ultrasonic vibration agitation

The internal structure of fresh cement before molding under ultrasonic vibration agitation is shown in Figure 15a. The internal structure is similar to that in Figure 14a and it is not introduced here again. For the hardened body formed under ultrasonic vibration agitation, the fresh paste absorbs CO_2 before molding and reacts with $\text{Ca}(\text{OH})_2$ in the paste to produce CaCO_3 . Under ultrasonic vibration agitation, the produced CaCO_3 crystals are separated into “nano-level” CaCO_3 nuclei that fill in spaces among the cement particles effectively. Due to the adsorption of nuclei, hydration products are adhered onto nuclei, such that internal spaces of the hardened paste are filled with nucleus-flocculating constituents. Hence, it is very difficult for CO_2 to enter the hardened body of the fresh paste. Moreover, there are small gel pores in C-S-H gels, through which some CO_2 may enter the hardened cement (Figure 15b).

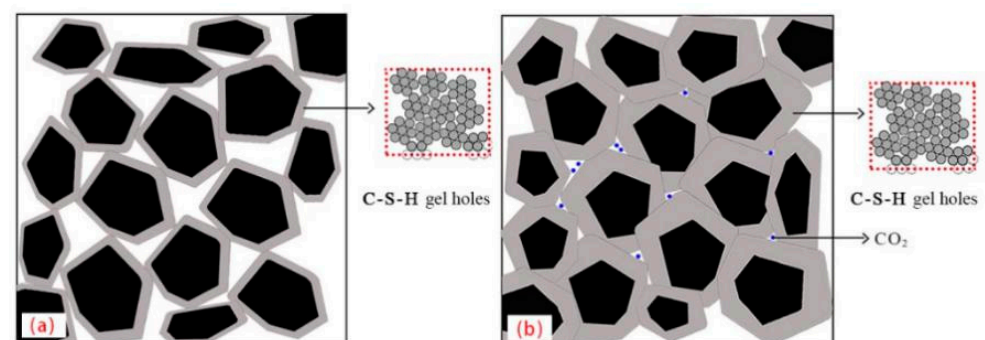


Figure 15. Carbonation mechanism of ultrasonic vibration agitation forming. (a) Non-invaded CO_2 ; (b) Invasion of CO_2 .

- (3) Microstructure analysis of CaCO_3 nuclei before molding of cement mortar with CO_2 absorption under ultrasonic vibration agitation

Due to the assistance provided by the ultrasonic “cavitation effect”, cement particle surfaces that are hydrated in the cement paste are broken continuously. As a result, gels that are produced on the surface peel off quickly, thus creating new cement particles. Since CaCO_3 flocculating constituents are segmented effectively under an ultrasonic wave, the abundant “nano-level” crystals are distributed among the cement particles uniformly (Figure 16a). When the slurry absorbs 0.44% CO_2 , the “nano-level” CaCO_3 nuclei among cement particles increase. Moreover, a significant amount of hydration products adhere to each crystal nuclei (Figure 16b). When the CO_2 AA reaches 1.32%, the CaCO_3 crystals continue to increase, thus making flocculating constituents among the cement particles more compact (Figure 16c). It can be seen from Figure 16d that when the CO_2 AA is 2.20%, the CaCO_3 crystals cover all of the flocculating constituents, thus making them more compact.

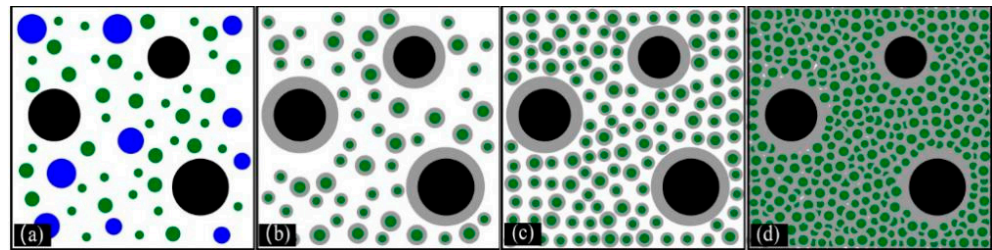


Figure 16. Microstructure of CaCO_3 crystal nucleus before formation of CO_2 -absorbing slurry. (a) No reaction occurred when the slurry absorbed CO_2 ; (b) Slurry absorbs 0.44% CO_2 at late hydration stage; (c) Slurry absorbs 1.32% CO_2 at late hydration stage; (d) Slurry absorbs 2.20% CO_2 at late hydration stage.

Based on the above mechanism analysis, the hardened body of fresh slurry formed under ultrasonic vibration had better carbonation resistance than the hardened body of slurry formed under mechanical stirring. Before molding, the higher CO_2 absorption volume generates more “nano-scale” CaCO_3 crystals which distribute uniformly in pores. This improves porous structure of cement mortar effectively. Since CaCO_3 has filling and activation effects, it can increase compaction degree of cement-based materials, thus decreasing the diffusion rate of CO_2 and improving the carbonation resistance of the cement-based materials.

6. Summary

- (1) Carbonation resistance of cement mortar species after CO_2 absorption under ultrasonic vibration is more obvious than that of species formed under mechanical stirring. Besides, carbonation resistance is positively related to CO_2 absorption volume before cement molding.
- (2) pH variation values of carbonization range under ultrasonic vibration increase with the increase in CO_2 absorption volume of cement mortar before carbonation, presenting a rising trend.
- (3) The diffraction peak of $\text{Ca}(\text{OH})_2$ of cement mortar after CO_2 absorption under ultrasonic vibration decreases to some extent compared to that under mechanical stirring, but such a reduction is not very obvious. This proves that CO_2 absorption by the cement mortar under ultrasonic vibration has small influences on $\text{Ca}(\text{OH})_2$.
- (4) After carbonization of CO_2 absorbed slurry under ultrasonic vibration, the quantity of capillary tubes decreases significantly with the increase of CO_2 absorption volume. As a result, porous structure is improved and compaction degree of cement-based material is increased, thus decreasing diffusion rate of CO_2 in the cement paste base. Finally, carbonation resistance of cement-based materials is improved effectively.

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