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Effect of Carbon Dioxide Curing on the Carbonation Durability of Concrete

Zijian Liu

College of Civil and Architectural Engineering, North China University of Science and Technology, Tangshan 063210, Hebei, China

Abstract

Concrete structures generate a large amount of carbon dioxide(CO_2) during the production and construction of raw materials, exacerbating environmental problems such as greenhouse effect. In order to achieve low-carbon and environmentally friendly construction projects, a large number of new technologies and products have been developed to reduce carbon emissions when using Portland cement concrete. Among them, using carbon dioxide to cure newly poured concrete is an effective method of carbon sequestration. However, further research is needed on its carbon sequestration efficiency and whether it will have adverse effects on the durability of concrete structures. This article summarizes the research status of using carbon dioxide to cure concrete, analyzes its carbon fixation efficiency, and based on a concrete carbonation durability life prediction model, analyzes the impact of carbon dioxide curing concrete on carbonation durability life.

Keywords

Carbon Dioxide; Durability; Concrete Curing.

1. Introduction

Currently, major industrialized countries around the world are facing energy shortages, environmental pollution, and climate change caused by massive carbon dioxide emissions. These problems can be alleviated to a certain extent by improving production processes to make industrial production more energy-efficient and environmentally friendly. Therefore, countries are actively formulating carbon reduction measures and setting carbon reduction targets to promote the improvement of production processes. The construction industry is an important industry that promotes social and economic development, improves the living environment of residents, and is also one of the main sources of carbon emissions. According to the China Carbon Accounting Database, China's total carbon emissions in 2023 are approximately 12.51 billion tons, accounting for 34% of the world's total carbon emissions. Among them, the total carbon emissions of the construction industry are about 6.48 billion tons, accounting for 51.8% of the country's total carbon emissions. The carbon emissions of the construction industry mainly come from the production of building materials (accounting for about 55.4% [1]). Concrete is the largest material in the construction industry, with a production volume of approximately 3.29 billion cubic meters in China in 2022. The carbon emissions generated during its production process account for approximately 28% of the total carbon emissions in the construction industry.

Therefore, in order to achieve the ultimate strategic goal of carbon neutrality, the construction industry is facing the pressure of transformation, and traditional high energy consuming and high carbon emitting operation methods are being replaced by energy-saving and low consumption innovative methods. When selecting materials, products containing a large amount of hidden carbon emissions have also been replaced by similar products with low

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hidden carbon emissions using new green production methods. In the path selection of achieving carbon neutrality, current methods mostly focus on carbon reduction techniques. In fact, the macro concept of carbon reduction includes two concepts: reducing carbon emissions and carbon sinks. In other words, besides focusing on reducing carbon emissions, developing the potential of carbon sinks and related technologies is also an indispensable main path to achieve current carbon reduction technologies. Carbon sink is a natural or artificial warehouse that absorbs and stores CO_2 in the atmosphere for a long time. Forests, oceans, soils, etc. are all good natural carbon sinks. Artificial carbon sink mainly refers to the capture, utilization, and storage of CO_2 in industrial processes through technological means.

In the construction industry, carbon sequestration technology is also actively being researched and developed. Among them, the technology of utilizing carbon dioxide mineralization has attracted much attention from researchers due to its rapid reaction kinetics of spontaneous absorption of CO_2. The reaction principle is to use CO_2 gas under certain pressure to undergo carbonization reaction with alkaline metal substances in the raw materials (such as calcium Ca, magnesium Mg, etc.) to form stable carbonates, thereby forming a carbon fixation effect.

2. Carbon Fixation Technology During the Curing Stage of Concrete

In the construction industry, this technology can be deeply coupled with the concrete curing process to create carbonation curing concrete technology. Concrete carbonation curing technology combines CO_2 sequestration with concrete material design. During the formation stage of fresh concrete, the concrete is placed in an appropriate temperature and pressure environment for CO2 curing. The mechanism is to use hydration products (such as Ca $(OH)_2$, C-S-H, etc.) [2] from the maintenance stage to react with CO_2. Ca $(OH)_2$ can react with CO_2 dissolved in pore solution to form $CaCO_3$ (Formula 1); C-S-H reacts with CO_2 to produce $CaCO_3$ and amorphous silica gel ($SiO_2 \bullet nH_2O$) (Equation 2) [3]. Based on the chemical reaction equation 2, tricalcium silicate and dicalcium silicate are converted into calcium carbonate and silica gel, and the volume increases. After CO_2 curing, calcium carbonate particles and silica gel fill the pores of concrete, making the concrete structure denser. These carbonized products can fill the pores of concrete, enhance the compactness of concrete while sequestering CO_2, and thus enhance the mechanical and durability properties of concrete [4]. Compared with standard curing (20 ± 5 ℃, relative humidity ≥ 95%), carbonation curing can achieve higher strength of concrete in a short period of time [3-5].

$$\boldsymbol{Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O} \quad (1)$$

$$\boldsymbol{xCaOySiO_2zH_2O + xCO_2 \rightarrow y(SiO_2tH_2O) + xCaCO_3 + (z - yt)H_2O} \quad (2)$$

The process of mineralized curing concrete can mix and utilize a large proportion of alkaline industrial solid waste as the core component of non in situ mineral mineralization reaction without leakage and pollution risk control. The CO2 mineralization curing concrete technology has the advantages of mild conditions and high product value. It can utilize solid waste on-site to synergistically absorb and capture CO_2, effectively solving the "source sink matching" problem in the implementation of concrete carbon fixation. It has become one of the emerging technologies for large-scale CO_2 and industrial solid waste comprehensive utilization. The carbon sequestration efficiency of concrete is usually evaluated by the amount of carbon sequestration, which is the mass ratio of CO_2 sequestered to cementitious materials [6].

As the carbonization reaction proceeds, $CaCO_3$ gradually precipitates and accumulates on the surface of the cementitious material. After carbonization, the microstructure of concrete

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becomes denser, and the total porosity and capillary porosity decrease [7]. At this point, the diffusion of CO_2 is inhibited, resulting in a decrease in carbon sequestration efficiency. Shi Caijun et al. [8] found that during the CO_2 curing process of concrete, the reaction between concrete and CO_2 mainly occurs in the first 15 minutes. The carbon sequestration efficiency and compressive strength of concrete increase with the increase of CO_2 pressure and carbonation time. However, in practical engineering, continuously increasing the curing time will cause a decrease in concrete performance. Research has shown that the optimal curing time is the time required for 30% to 40% moisture loss in concrete [9]. Excessive carbonation curing can lower the pH value of concrete, causing some hydration products to dissolve, thereby reducing the mechanical properties of concrete [10]. Therefore, some scholars such as Kashef Haghighi have attempted to increase CO_2 pressure to enhance carbon sequestration efficiency [2]. Existing research often uses high-purity CO_2 gas for experiments. When industrial gases such as natural gas processing, coal or biomass combustion are used as CO_2 sources, a certain amount of impurities are generated, which often require purification and pressurization. For example, sodium hydroxide can be used to remove acidic gases such as SO_2 and SO_3; The use of ethylene glycol based materials (such as triethylene glycol) can remove moisture [11]. Dwarakanath et al. [12] suggest that carbonation curing may not produce net CO_2 benefits. By estimating the carbon emissions during CO_2 capture, transportation, and utilization processes, as well as the energy consumption generated during concrete CO_2 storage. Out of the 99 published experimental data, 68 experiments showed an increase in carbon emissions. The author points out that the net CO_2 benefit of carbonation cured concrete is more likely to be negative. In addition, CO_2 curing can lower the pH of the concrete surface to 9.9, and subsequent hydration of cement makes it difficult to restore the pH value to 13. This increases the risk of steel corrosion and becomes a key factor limiting carbonation curing.

3. Carbonization Durability of Concrete

At the current level of technology, manufacturing and using concrete for engineering construction consumes a large amount of energy and generates significant carbon emissions. Therefore, the better the durability of concrete structures, the longer their service life, and the lower their carbon emissions per unit service time. So whether it has excellent durability is an important indicator to measure the quality of concrete structures, and the carbonation resistance of concrete is an important indicator to reflect its durability. When concrete is exposed to air, CO_2 in the air will come into contact with it, and the gaps in the concrete will become channels for CO_2 to gradually diffuse into the interior of the concrete. Over a long period of contact, CO_2 will undergo a series of complex physical and chemical reactions with alkaline hydrates in the concrete, which is the carbonation problem of concrete. As time goes by, the carbonation problem of concrete will continue to deepen. The calcium carbonate and other solid substances produced by the carbonation reaction will gradually block the voids of concrete, and the voids of concrete will also decrease with the deepening of the reaction, which to some extent hinders the diffusion of CO_2 and effectively improves the compactness of concrete! However, on the other hand, the carbonation reaction of concrete can lead to a reduction in the alkaline substances inside the concrete, resulting in a decrease in the pH value inside the concrete, and the concrete gradually transforms from its original alkaline state to a neutral environment; When the pH inside the concrete drops to a certain degree, it will cause damage to the passivation film on the surface of the steel bars. Without the protection of the passivation film, the steel bars will undergo corrosion reaction! Therefore, analyzing the carbonation reaction process of concrete and the corrosion reaction of steel bars is of great significance for studying the durability of concrete.

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Concrete carbonation is a long-term process, and passive carbonation may occur during its service life. Due to the increase in the initial mass ratio of CO2 in concrete cured with CO_2, its carbonation durability is inevitably weakened, and the degree of its impact is worrying. Passive carbonization reduces the alkalinity of concrete. When the pH value is below 10, the passivation film on the surface of the steel bar will be damaged, accelerating the corrosion of the steel bar. Corrosion of steel bars can cause volume expansion, leading to cracking or peeling of the concrete protective layer and accelerating further carbonation of the concrete. Affects the physical properties of concrete. The continuous carbonation process ultimately leads to concrete shrinkage, tensile stress, and may cause microcracks, reducing the flexural and compressive strength of concrete. Carbonization can also cause surface deformation of concrete, such as cracking and expansion. In addition, carbonation of concrete can lead to the loss of calcium ions in concrete, accelerate the aging of cement slurry, and thus affect the structural safety and service life of buildings.

The passive carbonation rate of concrete structures is mainly affected by the concentration of CO_2 in the environment, environmental temperature and humidity, and the concrete material. The most severe degree of carbonization occurs in high-temperature indoor enclosed environments with relative humidity of 50% to 70%, followed by high-temperature indoor ventilation environments, and the least severe is low-temperature indoor ventilation environments [13]. The reason is that when the relative humidity exceeds 70%, a water film will form on the surface of the concrete, making it difficult for carbon dioxide to diffuse into the concrete, resulting in mild carbonation; If the relative humidity is less than 50%, the carbonization reaction rate is slower; Under low temperature conditions, the diffusion coefficient and carbonation reaction rate of CO_2 in concrete are relatively slow, resulting in a relatively light degree of carbonation; Due to high temperature! The moderate humidity environment is conducive to the diffusion of CO2 and the occurrence of carbonization reactions, therefore the degree of carbonization is the most severe.

Yu Bo et al. comprehensively considered the effects of CO_2 diffusion rate and carbonation reaction rate on the carbonation depth of concrete, and quantitatively analyzed the influence of environmental factors such as temperature, relative humidity, and CO_2 concentration. A practical prediction model for analyzing the carbonation depth of concrete was established by modifying the theoretical model [14,15] by introducing environmental correction factors for temperature, relative humidity, and CO_2 concentration, as shown in Equation 3:

$$= k_j k_s k_e \sqrt{\frac{2D^0_{CO_2} C^0_{CO_2}}{m_0}} \sqrt{T} \tag{3}$$

In the formula, Xc is the effective protective layer thickness (m), and k_j is the position influence coefficient [13]; Ks is the coefficient of influence of working stress [16]; K_e is the product of environmental correction coefficients, and for the environmental conditions considered in reference [16], the value range of k_e is 0.020 to 0.140, D^0_{CO2} is the diffusion coefficient of CO_2 in concrete under standard environmental conditions (m2/s); C^0_{CO2} is the molar concentration of CO_2 under standard environmental conditions (mol/m^3); T is the carbonation time, and m0 is the amount of CO_2 absorbed per unit volume of concrete during complete carbonation (mol/m^3) [17]. For ordinary Portland cement concrete, the calculation formula for m0 is shown in Equation 4:

$$m_0 = (1 - a)8.22B \tag{4}$$

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In the formula, B is the amount of cementitious material per unit volume of concrete (kg/m^3), and a is the amount of mixed material in ordinary Portland cement (a=10%). After conversion, m_0 is equivalent to 32.6% of carbon sequestration. According to the practical prediction model for concrete carbonation analysis (Equation 3), the calculation formula for the service life of concrete structures under carbonation environment can be established:

$$\boldsymbol{T} = \frac{m_0}{2D^0_{CO_2}C^0_{CO_2}}\left(\frac{X_c}{k_j k_s k_e}\right)^2 \tag{5}$$

According to equation 5, the carbonation durability life of concrete structural components is linearly related to m_0. According to reference [18], the carbon sequestration of CO_2 cured concrete is 8% -10%, while according to reference [2], by adjusting the concrete mix ratio, the highest carbon sequestration of CO_2 cured concrete can reach 16.5% -23% Due to the fact that the carbon fixation process of CO_2 cured concrete is similar to the passive carbon fixation process during the service life of concrete structures, CO_2 infiltrates from the surface of the concrete to the interior, and the concrete protective layer on the outer side of the steel reinforcement is the area with a larger amount of carbon fixation. Therefore, the amount of carbon fixation during the maintenance stage will have a direct impact on the carbonation durability of concrete structures, which is equivalent to accelerating the shortening of their carbonation durability life. Therefore, when passive carbonation is complete, the amount of CO_2 absorbed per unit volume of concrete m0 'should be deducted, that is:

$$m'_{0max} = 32.6\% - 8\% = 24.6\%$$

$$m'_{0min} = 32.6\% - 23\% = 9.6\% \tag{6}$$

By replacing m_0 in equation 5 with m_0 ', the formula for calculating the service life of concrete structures cured with CO_2 in a carbonated environment can be obtained:

$$T_{max} = \frac{m'_{0msx}}{2D^0_{CO_2}C^0_{CO_2}}\left(\frac{X_c}{k_j k_s k_e}\right)^2$$

$$T_{min} = \frac{m'_{0min}}{2D^0_{CO_2}C^0_{CO_2}}\left(\frac{X_c}{k_j k_s k_e}\right)^2 \tag{7}$$

Comparing equations 5 and 7, the carbonation durability life of concrete structures with a carbon sequestration rate of 8% during maintenance is reduced by 24.5% , and the carbonation durability life of concrete structures with a carbon sequestration rate of 23% during maintenance is reduced by 70.5%, compared to ordinary concrete structural components, and the impact is very significant. According to the model prediction results in reference [13], it can be seen that in indoor environments with specific temperature, humidity, and CO_2 concentration, the carbonation durability life of ordinary concrete structures is close to the design service life of general building structures (50 years). If ordinary concrete structures are replaced with CO_2 cured concrete structures, their carbonation durability life may be lower than the design service life of general building structures. This is unacceptable.

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4. Conclusion

(1) Using carbon dioxide to cure concrete structures can promote carbonation of concrete, which helps to improve the storage rate and efficiency of carbon dioxide. However, at the current level of technology, the carbon sequestration area is concentrated on the protective layer outside the steel bars, which cannot fully utilize the internal concrete. During the simultaneous addition of carbon dioxide, carbon emissions are generated during the capture, compression, and transportation of carbon dioxide. Therefore, further research is needed to determine whether carbon reduction can ultimately be achieved.

(2) Carbon dioxide curing of concrete can lead to an increase in the initial carbon content on the surface of the concrete. Due to the high carbon fixation efficiency, which is generally 8% to 10% and can reach up to 23%, it significantly affects the carbonation durability life of concrete structural components (with a maximum reduction of 70.5%). Therefore, for environments with fast carbonization rates, carbon dioxide should not be used to cure concrete structures.

(3) If this technology can be improved to fully utilize the carbon fixation inside concrete, the impact on the carbonation durability life of concrete structural components will also be reduced.

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