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The Impact of Concrete Carbon Fixation Technology on the Durability of Concrete Structure

Zijian Liu

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

Abstract

Researchers in the engineering field have developed a large number of new technologies and products to fix carbon dioxide at various stages of concrete structure construction to neutralize the large amount of carbon dioxide emitted by the cement manufacturing industry during the production process. This includes injecting carbon dioxide into freshly mixed concrete, using carbon dioxide gas to cure the concrete, and using carbon dioxide treatment to strengthen recycled aggregates. However, further research is needed to determine whether these concrete carbon sequestration technologies will have adverse effects on the durability of concrete structures. This article summarizes the current research status of various concrete carbon fixation technologies and analyzes the impact of their carbon fixation efficiency on carbonation durability life. It is pointed out that although concrete carbon sequestration technology currently has many positive effects on the durability of concrete structures, this is based on the premise of low carbon sequestration efficiency. With the improvement of carbon sequestration efficiency, its impact on the durability of concrete structures is negative and will increase with the increase of carbon sequestration efficiency.

Keywords

Concrete carbon fixation, durability, concrete structures.

1. Introduction

Concrete, as the current mainstream building material, occupies a dominant position in various engineering constructions and is an important force in promoting the progress of human civilization. However, as an essential component of concrete, cement consumes a large amount of energy and raw materials during the production process, and emits a large amount of carbon dioxide (CO_2). The global cement industry emits approximately 1.35 billion tons of carbon dioxide annually, accounting for 5% to 7% of the total carbon dioxide generated by human activities. The huge energy consumption and carbon dioxide emissions in the cement industry have made it the core of the global warming debate. According to data released by the International Energy Agency, global carbon dioxide emissions are expected to reach 37.4 billion tons by 2023. Carbon dioxide and other greenhouse gases have caused a series of serious environmental problems, such as global warming and glacier melting. How to reduce carbon dioxide emissions and mitigate the impact of greenhouse effect has become a challenge that the world needs to face and solve together.

By utilizing the carbonation reaction between alkali metal ions (such as Ca^{2+}/Mg^{2+}) in natural minerals or solid waste and carbon dioxide, atmospheric carbon dioxide is converted into stable inorganic carbonates, thereby achieving carbon dioxide sequestration. This method is called carbon sequestration technology and is an important carbon dioxide reduction technology. At the same time, the hydration products of concrete ($Ca(OH)_2$, C-S-H, etc.) can react with carbon dioxide and have enormous potential for carbon sequestration. At different stages

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of the life cycle of concrete (mixing, curing, use, and secondary utilization), concrete can achieve permanent storage of carbon dioxide through carbonation reaction [1]. Among them, carbonation during the mixing, curing, and secondary utilization stages belongs to active carbonation [2], which involves injecting carbon dioxide into fresh concrete or carbonizing and curing concrete, recycled aggregates, and recycled powders. The carbonation during the use stage belongs to passive carbonation, which means that concrete spontaneously reacts with carbon dioxide in the atmosphere without human intervention. However, the carbonation rate of passive carbonation is influenced by external environment and fluctuates greatly. Therefore, research on carbon fixation in concrete mainly focuses on active carbonation.

However, the impact of concrete carbon sequestration technology on the durability of concrete structures cannot be ignored, and it is also a complex issue with significant duality: there are both positive and enhanced durability aspects, as well as potential negative risks that require vigilance.. The impact of carbon sequestration technology on the durability of concrete varies greatly at different stages.

2. Carbon Fixation Technology During the Concrete Mixing Stage

Injecting carbon dioxide during the concrete mixing stage is a direct carbon sequestration method that can significantly improve the carbonation rate of concrete [3]. The carbon fixation mechanism is to inject carbon dioxide into the concrete in the form of gas or dry ice during mixing. During the stirring process, carbon dioxide fully contacts and reacts with the pore solution and anhydrous calcium silicate ($CaOSiO_2$), generating calcium carbonate ($CaCO_3$) and hydrated calcium silicate (C-S-H) [4] (Equations 1 and 2).

$$\boldsymbol{3CaOSiO_2 + (3-x)CO_2 + yH_2O \rightarrow xCaO \cdot SiO_2 \cdot yH_2O + (3-x)CaCO_2} \tag{1}$$

$$\boldsymbol{2CaOSiO_2 + (2-x)CO_2 + yH_2O \rightarrow xCaO \cdot SiO_2 \cdot yH_2O + (2-x)CaCO_2} \tag{2}$$

At the same time, CO_2 reacts with the main hydration products in the cement slurry, calcium hydroxide ($Ca(OH)_2$), as well as unhydrated tricalcium silicate (C_3S) and dicalcium silicate (C_2S), to generate nano-sized or micrometer sized calcium carbonate ($CaCO_3$) and silica gel ($SiO_2 \bullet n H_2O$). The generated calcium carbonate can provide additional nucleation sites for concrete, thereby accelerating its carbonation process [5]. The generated $CaCO_3$ crystals and silica gel can effectively fill the capillary pores and microcracks of concrete, reduce porosity, and make the concrete structure denser.

Injecting carbon dioxide during the mixing stage can not only directly offset some of the carbon emissions of concrete, but also improve its mechanical properties and durability. Through rational design, this technology can effectively regulate the setting time, hydration behavior, and pore structure of concrete. The carbon sequestration efficiency of concrete is usually evaluated based on the amount of carbon sequestration, which is the mass ratio of sequestered carbon dioxide to cementitious materials. Injecting carbon dioxide gas during the mixing stage can not only improve the carbon sequestration efficiency of concrete, but also enhance its compressive strength and flexural strength. The reason is that carbon dioxide promotes the hydration of cement, increases the volume of hydration products, and makes the microstructure denser.

There are still some shortcomings that need to be addressed when adding carbon dioxide as an admixture to concrete. Firstly, adding in the form of gas will not improve carbon sequestration efficiency, increase stirring time and energy consumption. Adding carbon dioxide in the form of dry ice can improve carbon sequestration efficiency, but dry ice is prone to volatilization

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during cement hydration, and the gas produced can easily form connected pores inside the concrete. These connected pores will reduce the mechanical properties and durability of concrete. As the amount of dry ice increases, the strength of concrete decreases accordingly.

It should be noted that carbonation can cause a decrease in the pH value of the pore solution in concrete. Due to the short duration and low carbon sequestration efficiency of the concrete mixing stage, the mass of solidified carbon dioxide is usually only 0.3% to 1.4% of the mass of the cementitious material. Therefore, the carbonization process at this stage usually does not increase the risk of steel corrosion and does not affect the durability and alkalinity of concrete [5, 6]. However, 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 carbon dioxide in concrete caused by pre mixed carbon dioxide, its carbonation durability will inevitably weaken, and the degree of its impact is worrying. Passive carbonization will reduce the alkalinity of concrete. When the pH value is below 10, the passivation film on the surface of the steel bar will be damaged, thereby 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, thereby accelerating further carbonation of the concrete. This will affect the physical properties of concrete. The continuous carbonation process will ultimately lead to concrete shrinkage, tensile stress, and possible microcracks, thereby 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 the concrete, accelerate the aging of cement paste, and thus affect the structural safety and service life of buildings.

3. Carbon Fixation Technology During the Curing Stage of Concrete

Concrete carbonation curing technology combines carbon dioxide sequestration with the molding and curing of concrete materials. During the forming stage of fresh concrete, place the concrete in a suitable temperature and pressure environment for carbon dioxide curing. The mechanism is to use hydration products from the maintenance stage (such as $Ca(OH)_2$, C-S-H, etc.) [7] to react with carbon dioxide. $Ca(OH)_2$ can react with dissolved carbon dioxide in pore solution to form $CaCO_3$ (equation 3); C-S-H reacts with carbon dioxide to produce $CaCO_3$ and amorphous silica gel ($SiO_2 \bullet nH_2O$) (Equation 4) [8]. According to chemical reaction equation 2, tricalcium silicate and dicalcium silicate are converted into calcium carbonate and silica gel, resulting in an increase in volume. After carbon dioxide curing, calcium carbonate particles and silica gel fill the pores of concrete, making the concrete structure more compact. These carbonized products can fill the pores of concrete, enhance the compactness of concrete while sequestering carbon dioxide, thereby improving the mechanical properties and durability of concrete [9]. Compared with standard curing (20±5℃, relative humidity≥95%), carbonation curing can increase the strength of concrete in a short period of time [10].

$$\boldsymbol{Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O} \tag{3}$$

$$\boldsymbol{xCaOySiO_2zH_2O + xCO_2 \rightarrow y(SiO_2tH_2O) + xCaCO_3 + (z - yt)H_2O} \tag{4}$$

As the carbonization reaction proceeds, $CaCO_3$ gradually precipitates and accumulates on the surface of cement-based materials. After carbonization, the microstructure of concrete becomes denser, with a decrease in total porosity and capillary porosity [11], forming a dense shell on the outer layer of concrete. However, while the dense outer layer improves the impermeability of concrete, it also suppresses the diffusion of CO_2, leading to a decrease in carbon sequestration efficiency. Shi Caijun et al. [12] found that during the CO_2 curing process

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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 can lead to a decrease in concrete performance.

The use of carbon dioxide to cure concrete structures can promote concrete carbonation and help improve the storage rate and efficiency of carbon dioxide. However, at the current level of technology, carbon sequestration areas are mainly concentrated in 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. The 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 fixation efficiency of carbon dioxide, typically ranging from 8% to 10% and up to 23%, it has a significant impact on the carbonation durability life of concrete structural components (with a maximum reduction of 70.5%). Therefore, in environments with fast carbonization rates, carbon dioxide should not be used to cure concrete structures. If this technology can be improved to fully utilize the carbon fixation effect inside concrete, the impact on the carbonation durability life of concrete structural components will also be reduced.

4. Carbon Fixation Mechanism of Recycled Aggregates

China has generated an astonishing amount of construction waste during its rapid urbanization process, of which about 45% is waste concrete. To deal with these discarded concrete, researchers have developed recycled concrete technology. Recycled concrete is a new type of environmentally friendly building material that uses recycled aggregates as coarse aggregates in new concrete, allowing construction waste to regain value and extending the service life of concrete in another way, reducing its carbon emissions per unit life.

Recycled aggregate refers to the aggregate obtained by crushing and screening waste concrete after demolishing buildings. Its surface is covered with a layer of hardened cement slurry and has multiple edges and complex interfaces. However, due to the fact that the recycled aggregate component is waste concrete, which not only contains natural coarse aggregates from the original concrete, but also old cement slurry and fine powder, its engineering performance is not as good as natural aggregates [13], especially its resistance to chloride ion penetration [14]. Therefore, the steel bars in recycled concrete are more prone to corrosion, which seriously affects the durability of concrete structures and limits the practical application of recycled concrete in engineering. Compared to natural aggregates, recycled aggregates have a higher porosity, resulting in a larger specific surface area and water absorption rate. Therefore, when using recycled aggregate to prepare concrete, its unit water consumption is higher than that of ordinary concrete. In addition, the compressive strength and elastic modulus of recycled aggregate concrete are only two-thirds of those of natural aggregate concrete [15], so it is necessary to strengthen the pretreatment of recycled aggregate to improve its performance.

Carbon dioxide solidification can improve the performance of recycled aggregates and achieve carbon sequestration [16], but the impact of this method on the durability of recycled concrete still needs further research. The mechanism of carbon dioxide solidification of recycled aggregates is that carbon dioxide reacts with $Ca(OH)_2$ and C-S-H in the mortar attached to the surface of recycled aggregates to carbonize, converting carbon dioxide into carbonate to sequester carbon dioxide [17,18]. During the accelerated carbonation process of recycled concrete aggregates, carbon dioxide undergoes a series of complex physical and chemical reactions. Firstly, carbon dioxide diffuses into the interior through the pores of recycled concrete aggregates and dissolves in the water within the pores to form carbonic acid. Carbonic

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acid easily neutralizes with dissolved calcium hydroxide to form calcium carbonate [19]. As the surface of calcium hydroxide gradually forms relatively dense calcium carbonate crystals during the reaction process, the reaction rate between carbon dioxide and calcium hydroxide will also gradually slow down.

The carbonation reaction of hydrated calcium silicate cement (C-S-H) refers to the reaction between carbon dioxide and calcium ions separated from the cement, resulting in the formation of amorphous silica gel and various forms of calcium carbonate crystals, as shown in equation (5) [20]. The type of C-S-H carbonization reaction mainly depends on the initial Ca/Si ratio in the gel. With the carbonization of C-S-H gel, the Ca/Si ratio gradually decreases, the decomposition rate of C-S-H accelerates, and the number of calcium carbonate crystals generated from C-S-H carbonization gradually decreases. After the C-S-H carbonization reaction, calcite, aragonite, and vanadium minerals are usually formed.

$$(CaO) \cdot x(SiO)(H_2O) + xCO_2 \rightarrow CaCO_3 + SiO_2 \cdot (H_2O)_t + (z-t)H_2O \tag{5}$$

At present, research on the durability of carbon dioxide enhanced recycled aggregate concrete mainly focuses on its resistance to chloride ion penetration. Research has found that when using 100% carbon dioxide instead of reinforced recycled concrete aggregates, the electrical conductivity, chloride ion permeability, and air permeability of recycled concrete decrease by 15.1%, 36.4%, and 42.4%, respectively [21]. Zheng [22] studied the optimal conditions for carbon dioxide enhanced treatment of recycled aggregates, as well as the effects of carbon dioxide enhanced treatment on the apparent density, water absorption, and crushing index of recycled coarse aggregates. The resistance to chloride ion penetration of recycled concrete prepared from recycled coarse aggregate reinforced for 24 and 48 hours is comparable to that of ordinary concrete. The reason for the decrease in chloride ion permeability may be that the amorphous silica gel and various forms of calcium carbonate crystals generated by the mineralization of carbon dioxide in the aggregate fill the pores, improve the pore structure of the aggregate, reduce the channels for chloride ion diffusion, and thus enhance the ability of concrete to resist chloride ion invasion.

But this is only one aspect of the durability of concrete structures. On the other hand, the carbonation of concrete is a double-edged sword. On the one hand, it plays a role in carbon sequestration. Carbon dioxide undergoes a series of complex physical and chemical reactions with alkaline hydrates in concrete. Over time, carbonation reactions produce calcium carbonate and other solid substances, gradually blocking the pores in concrete. The pores in concrete also decrease as the reaction progresses, which to some extent hinders the diffusion of carbon dioxide and effectively improves the compactness of concrete. However, on the other hand, the carbonation reaction of concrete can lead to a reduction of alkaline substances inside the concrete, resulting in a decrease in the pH value inside the concrete. The concrete will gradually transition from its original alkaline state to a neutral environment. When the pH value inside the concrete drops to a certain extent, it will damage the passivation film on the surface of the steel bars, and the steel bars will undergo corrosion reactions without the protection of the passivation film. Although carbon dioxide enhanced recycled aggregate concrete has not undergone comprehensive carbonation curing, the highly carbonized recycled aggregate inside has a relatively low pH value. When these neutral islands in concrete are exposed to carbon dioxide in the external environment, carbonation gradually deepens, forming connected neutral channels, which may weaken the carbonation durability of concrete. However, there is currently limited research in this field. Therefore, there is an urgent need to further study the effect of carbon dioxide enhanced recycled aggregates on the carbonation durability of concrete.

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5. Conclusion

(1) Injecting carbon dioxide during the mixing stage can promote the carbonation of concrete. When gas is injected, it can improve the compactness of concrete, enhance the impermeability and durability of concrete structures. However, the carbon sequestration efficiency is low, which is a failure from the perspective of carbon sequestration. If the injection amount of carbon dioxide is increased, it will cause a decrease in the compactness and strength of the concrete, a decrease in the internal pH value, and a deterioration in durability.

(2) The use of carbon dioxide to cure concrete structures can promote concrete carbonation and help improve the storage rate and efficiency of carbon dioxide. However, at the current level of technology, carbon sequestration areas are mainly concentrated in the protective layer outside the steel bars, which cannot fully utilize the internal concrete. Due to its high fixation efficiency, typically ranging from 8% to 10%, with a maximum of 23%, carbon dioxide has a significant impact on the carbonation durability of concrete structural components. Therefore, in environments with rapid passive carbonation rates, carbon dioxide should not be used to cure concrete structures.

(3) Carbon dioxide enhanced recycled aggregates can significantly improve the resistance of ordinary recycled concrete to chloride ion penetration, with higher density and better compactness. However, carbon dioxide enhanced recycled aggregates can lower the pH value inside concrete, and its negative impact on the carbonation durability of concrete still needs to be studied. At present, there is a lack of research on whether carbon dioxide enhanced recycled aggregates will affect other durability properties of concrete, and further research is needed.

(4) The impact of concrete carbon sequestration technology on concrete durability is a complex issue with significant duality: there are both positive and durability enhancing aspects, as well as potential negative risks that require caution. Overall, when applied under controlled conditions, its positive impact usually dominates. Concrete carbon fixation technology has a positive and dominant impact on the long-term durability of structures by significantly improving the compactness, mechanical strength, and impermeability of concrete. It creates a physical barrier that greatly delays all degradation processes caused by medium invasion. However, this is based on the current low carbon sequestration efficiency of concrete carbon sequestration technologies. With the improvement of carbon sequestration efficiency, its impact on the durability of concrete is negative. How to improve carbon sequestration efficiency without negatively affecting the durability of concrete structures is an urgent problem to be solved.

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