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Study on Carbon Fixation Efficiency of Concrete Carbon Fixation Technology

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Abstract

Concrete emits a large amount of carbon dioxide during the production process, exacerbating environmental problems such as greenhouse effect. However, at the same time, the hydration products of concrete ($Ca(OH)_2$, C-S-H, etc.) can react with CO_2 and have great potential for carbon sequestration. At different stages of the life cycle of concrete (mixing, curing, service, and secondary utilization), concrete can achieve permanent CO2 sequestration through carbonation reactions. Whether concrete carbon sequestration technology can achieve carbon neutrality, the amount of carbon sequestration is the most important parameter, which directly determines whether various concrete carbon sequestration technologies have research value. Different concrete carbon sequestration technologies have significant differences in their carbon sequestration mechanisms and technical routes, resulting in varying amounts of carbon sequestration. This article summarizes the current research status of carbon sequestration technology in different stages of concrete, analyzes and compares its carbon sequestration efficiency, and predicts its development potential and application prospects.

Keywords

Carbon Dioxide; Carbon Fixation Efficiency; Concrete.

1. Introduction

The large amount of carbon dioxide emissions generated by human activities has led to an increase in the concentration of carbon dioxide in the atmosphere, which in turn causes the greenhouse effect and has a significant impact on the Earth's environment. The cement manufacturing industry is one of the main sources of carbon dioxide (CO_2) emissions. In the process of cement production, a large amount of energy and raw materials are consumed, and a large amount of CO_2 is emitted. The global cement industry emits about 1.35 billion tons of CO_2 annually, accounting for 5% to 7% of the CO_2 generated by human activities. The huge energy consumption and CO_2 emissions of the cement industry have put it at the center of the global warming debate. According to data released by the International Energy Agency, global CO_2 emissions will reach 37.4 billion tons in 2023. Greenhouse gases such as CO_2 have caused a series of serious environmental problems such as global warming and glacier melting. How to reduce CO_2 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 CO_2, CO_2 in the atmosphere is converted into stable inorganic carbonates, thereby achieving CO_2 sequestration. This method is called carbon sequestration technology, which is a significant CO_2 emission reduction technology. Concrete, as the most important raw material in the field of civil engineering and construction, emits a large amount of carbon dioxide during the manufacturing process. However, at the same time, the hydration products of concrete (Ca (OH) $_2$, C-S-H, etc.) can react with CO_2 and have great potential for carbon sequestration. At different stages of the life cycle of concrete (mixing, curing, service, and secondary utilization), concrete can achieve permanent CO_2 sequestration through carbonation reactions [2]. Among them, carbonization during the mixing, curing, and secondary utilization stages is active carbonization [3], which refers to injecting CO_2 into fresh concrete or carbonizing and curing concrete, recycled aggregates, and recycled powders. The carbonization during the service stage is passive carbonization, which refers to the spontaneous carbonization of concrete with CO_2 in the atmosphere without human intervention. However, the carbonization rate of passive carbonization is influenced by the external environment and has significant fluctuations. Therefore, the research focus of concrete carbon fixation is mainly on active carbonization. In the study of carbon sequestration in concrete, the carbon sequestration efficiency of concrete is usually evaluated by the amount of carbon sequestration, which is the mass ratio of CO_2 sequestered in a unit volume of concrete to the concrete cementitious material [4]. Whether concrete carbon sequestration technology can achieve carbon neutrality, the amount of carbon sequestration is the most important parameter, which directly determines whether various concrete carbon sequestration technologies have research value. Different concrete carbon sequestration technologies have significant differences in their carbon sequestration mechanisms and technical routes, resulting in varying amounts of carbon sequestration.

2. Carbon Sequestration Technology during the Mixing Stage of Concrete

Although hardened concrete can sequester CO_2 through carbonation reactions during service, it can only come into contact with CO_2 through the surface of the concrete, resulting in a low carbonation rate. Injecting CO_2 during the concrete mixing stage allows the interior of the concrete to fully contact CO_2, which can significantly increase the carbonation rate of the concrete [5]. The mechanism is to inject CO_2 into fresh concrete in gaseous or dry ice form. During the mixing process, CO_2 reacts with the pore solution and anhydrous calcium silicate ($CaOSiO_2$) to produce calcium carbonate ($CaCO_3$) and hydrated calcium silicate (C-S-H) [6] (Equations 1 and 2). The generated CaCO3 can provide additional nucleation sites for concrete, thereby accelerating its carbonation [7].

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

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

Injecting CO_2 during the mixing stage has a significant impact on the hydration behavior of cement, as well as the pore structure and macroscopic properties of concrete. Through rational design, this technology can effectively regulate the setting time, hydration behavior, and pore structure of concrete. During the mixing stage, there are unhydrated and partially hydrated cement particles in the concrete. At this point, the injected CO_2 gas dissolves into the water and can react with the pore solution and hydration products, indicating that fresh concrete has the potential to absorb CO_2 [8]. A case study shows that it is feasible to reduce carbon emissions throughout the production process of concrete components by 4.6% by injecting carbon dioxide, which can reduce cement load [9].

However, due to certain limitations, the efficiency of injecting CO_2 for carbon sequestration during the current mixing stage is relatively low, typically only 0.3% to 1.4% [7]. This is because dry ice is prone to volatilization during the hydration process of cement, and the gas generated can easily form interconnected pores inside the concrete. These connected pores will reduce the mechanical and durability properties of concrete. As the amount of dry ice increases, the degree of decrease in concrete strength also increases. Therefore, a large amount of dry ice cannot be added during the mixing stage. If CO_2 is added in gaseous form, due to its low concentration and short duration of concrete mixing, the carbon sequestration efficiency cannot be improved.

Meanwhile, injecting CO_2 during the mixing stage involves energy consumption and new carbon emissions, and further evaluation is needed to determine whether it can reduce the carbon footprint. Monkman et al. [9, 10] questioned the carbon sequestration effect during the mixing stage, stating that carbon emissions are generated during CO_2 capture, compression, and transportation processes. For every cubic meter of carbon fixed concrete produced, the power consumption is 0.018 kW/h, which is equivalent to emitting 9.23 grams of CO_2. Therefore, the key issue facing carbon sequestration technology at this stage is whether it can further improve the carbon sequestration efficiency, fully tap into the carbon sequestration potential of concrete, and truly achieve net zero CO_2 emissions.

3. Carbon Fixation Technology during the Curing Stage of Concrete

The carbonation curing technology for concrete involves placing the concrete in an appropriate temperature and pressure environment for CO_2 curing during the formation stage of fresh concrete. The mechanism is to use hydration products (such as Ca $(OH)_2$, C-S-H, etc.) from the maintenance stage to react with CO_2 [11]. Ca $(OH)_2$ can react with CO_2 dissolved in the pore solution to form $CaCO_3$ (equation 3); C-S-H reacts with CO_2 to produce $CaCO_3$ and amorphous silica gel ($SiO_2 \bullet nH_2O$) (Equation 4) [6]. Based on the chemical reaction equation 4, 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 [12]. Compared with standard curing (20 ±5℃, relative humidity≥95%), carbonation curing can achieve higher strength of concrete in a short period of time [12-13].

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

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

During the carbonization curing process, as the carbonization reaction progresses, $CaCO_3$ gradually precipitates and accumulates on the surface of the cementitious material. After carbonization, the microstructure of concrete becomes denser, and the total porosity and capillary porosity decrease [14]. At this point, the diffusion of CO_2 is inhibited, resulting in a decrease in carbon sequestration efficiency. Shi Caijun et al. [15] 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, continuously increasing the curing time will cause a decline in the performance of concrete in various aspects. Chen found through research that the optimal curing time for CO_2 cured concrete is the time required for 30% to 40% moisture loss in the concrete [16]. At the same time, excessive carbonation curing will lower the pH value of concrete, causing some hydration products to dissolve, thereby reducing the mechanical properties of concrete [7]. Therefore, some scholars such as Kashef Haghighi have attempted to increase CO_2 pressure to enhance carbon sequestration efficiency [11]. According to reference [17], the carbon sequestration of CO_2 cured concrete is 8% • 10%, while according to reference [11], by adjusting the concrete mix ratio, the maximum carbon sequestration of CO_2 cured concrete can reach 16.5% -23%. However, due to the fact that CO_2 permeates from the surface of the concrete to the interior during the curing process, the main area of carbon sequestration is the surface of the concrete. If it is a reinforced concrete component, it is the protective layer on the outer side of the steel bars. The carbon sequestration potential of the concrete inside the steel bars is difficult to fully utilize. Because CO_2 curing can lower the pH of the concrete surface to 9.9, 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 [6]. Therefore, although carbonation curing has a high carbonation efficiency, current conventional methods cannot further increase its carbonation amount.

Dwarakanath et al. [18] 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. Therefore, the technology of carbonization maintenance needs further optimization to reduce energy consumption and net carbon emissions.

4. Carbon Fixation Technology during the Service Stage of Concrete

The carbonation mechanism during the service stage of concrete [19] is that cement clinker generates $Ca(OH)_2$ and C-S-H through hydration reaction. CO2 diffuses into the interior of concrete through pores in the external environment and reacts with hydration products to produce $CaCO_3$ and $SiO_2 \bullet nH_2O$. Compared with the mixing stage, curing stage, and secondary utilization stage, the carbonation reaction of concrete in the service stage is very slow. In addition, due to factors such as small exposure area, low concentration of CO_2 in the atmosphere, and high compactness of concrete, the diffusion process of CO_2 is very slow, resulting in carbonation reactions usually only occurring on the surface layer of concrete (1-15mm). Moreover, excessive carbonization can lead to a decrease in the pH value of concrete, increasing the risk of steel corrosion. Therefore, in order to further improve the carbon sequestration efficiency of concrete, it is necessary to focus on plain concrete that does not require steel reinforcement. Some scholars have attempted to improve carbon sequestration efficiency by increasing the exposed area. Zhang Yuan et al. [20] studied the carbon sequestration potential of foam concrete and found that the carbon sequestration efficiency of foam concrete in the whole life cycle is higher than that of ordinary concrete. He et al. [21] studied autoclaved aerated concrete (SAAC) and proposed a carbonation model for SAAC based on Fick's first law. The results indicate that the carbon sequestration efficiency of SAAC is much higher than that of ordinary concrete, and the carbonation model established confirms that SAAC has great potential in carbon sequestration. However, due to the porous nature of SAAC, excessive carbonization may cause degradation of its mechanical and durability properties.

5. Conclusion

(1) During the concrete mixing stage, carbon fixation is carried out. Due to the short mixing time and low concentration of gaseous CO_2, dry ice will evaporate and produce voids during the mixing process, resulting in a low carbon fixation efficiency of only 0.3% to 1.4%. However, this technological route still has great potential in utilizing the interior of concrete for carbon sequestration.

(2) Carbon fixation during the concrete curing stage is technically safe and easy to implement. At the same time, it has a high carbon sequestration efficiency, reaching 8% - 10%. By adjusting the concrete mix ratio, the carbon sequestration amount of CO_2 cured concrete can reach up to 16.5% -23%. It is currently a feasible concrete carbon sequestration technology path. However, due to the fact that this technology can only utilize the surface layer of concrete for carbon sequestration, its potential for carbon sequestration is limited. At the same time, excessive

curing can cause a decrease in the pH value of concrete, increase the risk of steel corrosion, and become a key factor limiting carbonation curing.

(3) In the carbon sequestration technology during the service stage of concrete, new concrete materials with large exposed areas have great development prospects. Although its application scenarios are limited, it has a very high carbon sequestration efficiency and great development prospects.

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