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Influence of CO_2-enhanced Recycled Coarse Aggregate on Durability of Concrete

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Abstract: Recycling discarded concrete into recycled aggregates is a very environmentally friendly construction waste disposal method. However, recycled aggregates have performance defects such as low strength and high permeability. The use of carbon dioxide (CO_2) mineralization to regenerate aggregates can effectively solve this problem, while also effectively sequestering carbon. However, the durability of concrete made using CO_2 mineralized recycled aggregates instead of natural aggregates still needs further research. This article summarizes and analyzes the current research on the durability of CO_2 enhanced recycled aggregate concrete, and finds that most of the current research focuses on its chloride ion permeability, while research on its carbonation durability is extremely rare. Therefore, this article analyzes the adverse factors of CO_2 mineralized recycled aggregate concrete in terms of carbonation durability and provides suggestions for further research.

Keywords: CO_2-enhanced; Durability; Recycled Coarse Aggregate.

1. Introduction

Concrete is an important industrial product that promotes social and economic development, improves the living environment, and is also one of the main sources of carbon emissions. The production of concrete in China in 2022 is about 3.29 billion cubic meters [1], and the carbon emissions generated during its production process account for about 28% of the total carbon emissions in the construction industry, which accounts for more than half of China's total carbon emissions. As an important component of the construction industry, the concrete industry is urgently seeking reasonable and effective measures to reduce carbon emissions, which is of great significance.

Carbon sequestration technology is a strategically significant CO_2 emission reduction technology, which is based on the carbonation reaction between alkali metal ions (such as Ca^{2+}/Mg^{2+}) in natural minerals or solid waste and CO_2, converting CO_2 into stable inorganic carbonates and achieving CO_2 sequestration. Carbon sequestration technologies include land storage, ocean storage, etc [2]. Among them, land storage is the process of capturing and compressing industrial CO_2 emissions and burying them deep underground to reduce CO_2 emissions. Although this technology has a high carbon sequestration efficiency, the risk of CO_2 leakage will continue to increase with the increase of storage time. Therefore, land storage requires long-term monitoring and maintenance, with high economic costs [3]. The ocean is the largest and most active carbon reservoir on Earth, with enormous potential for carbon sequestration. There are two main types of carbon dioxide ocean storage: one is ocean water storage, which refers to ocean storage carried out in seawater and on the seabed (such as underwater carbon dioxide lakes); Another type is offshore geological storage, which mainly utilizes various natural geological structures on the seabed to store carbon dioxide, including deep-sea saline layer storage, oil and gas reservoir storage, and deep-sea basalt layer storage [4,5]. Ocean closures may cause changes in the pH value of seawater in some areas or trigger unforeseeable environmental problems, thereby affecting marine ecology [6]. The shortcomings and high costs of the above-mentioned storage technologies have prompted the construction industry to seek new carbon sequestration models within the industry. As a major emitter of carbon in the construction industry, the hydration products of concrete ($Ca(OH)_2$, C-S-H, etc.) can react with CO_2, thus possessing the potential for carbon sequestration.

2. Carbon Fixation Mechanism of Recycled Aggregates

China is currently in a stage of rapid urbanization, with the size of cities constantly expanding. The number of new buildings built each year ranks first in the world, while the number of old buildings demolished is also enormous. China generates over 1 billion tons of construction waste annually, of which approximately 45% is waste concrete [7]. Recycled concrete is a new type of environmentally friendly building material that uses recycled aggregates as coarse aggregates for the new version of 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 building demolition, which is covered with a layer of hardened cement slurry with multiple edges and complex interfaces on its surface. 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 workability is inferior to natural aggregates [8,9], especially its resistance to chloride ion penetration is poor [10,11]. Therefore, steel bars in recycled concrete are more prone to corrosion [12], which seriously affects the durability of concrete structures and limits the practical application of recycled concrete in engineering [13]. Compared with natural aggregates, recycled aggregates have a higher porosity, resulting in a larger specific surface area and water absorption rate. The water absorption rate of natural aggregates is generally 1%~5%, while that of recycled aggregates can reach 3%~12% [14]. Therefore, when using recycled aggregates 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 modify the recycled aggregate.

Due to the performance defects of recycled aggregates, it is necessary to strengthen the pre-treatment of recycled aggregates to improve their performance. The reinforcement pretreatment of recycled aggregates mainly includes physical reinforcement methods, chemical reinforcement methods, and wet treatment reinforcement methods [16]. These methods can improve the quality of recycled aggregates to a certain extent by removing or reducing the old cement slurry and fine powder in the recycled aggregates, filling the pores between the old cement slurry and aggregates. However, some methods require complex and expensive equipment, high energy consumption, and significant secondary pollution, which goes against the original intention of using recycled aggregate concrete as an environmentally friendly material. CO_2 curing can improve the performance of recycled aggregates [17, 18] and also achieve carbon sequestration [19], but the impact of this method on the durability of recycled concrete needs further research.

The mechanism of CO_2 curing recycled aggregates is: through the carbonation reaction between CO_2 and Ca $(OH)_2$ and C-S-H in the mortar attached to the surface of recycled aggregates, CO_2 is converted into carbonate to sequester CO_2 [20, 21]. CO_2 undergoes a series of complex physical and chemical reactions during the process of accelerating carbonation of recycled concrete aggregates. Firstly, carbon dioxide diffuses into the interior through the pores of recycled concrete aggregates and dissolves in the water in the pores to form carbonic acid, as shown in equation (1). Carbonic acid easily neutralizes with dissolved calcium hydroxide to form calcium carbonate, as shown in equation (2) [22]. The state in which calcium carbonate precipitates mainly depends on which kinetics and thermodynamics dominate [23]. As the surface of calcium hydroxide gradually forms relatively dense calcium carbonate crystals during the reaction, the reaction rate between carbon dioxide and calcium hydroxide will also gradually slow down.

$$CO_2 + H_2O \rightarrow H_2CO_3 \quad (1)$$

$$Ca(OH)_2 + H_2CO_3 \rightarrow CaCO_3 + 2H_2O \quad (2)$$

The carbonation reaction of hydrated calcium silicate cement (C-S-H) is the reaction of carbon dioxide with calcium ions separated from the cement, resulting in the formation of amorphous silica gel and various forms of calcium carbonate crystals, as shown in formula (3) [24]. The type of C-S-H carbonation reaction mainly depends on the initial Ca/Si ratio in the gel. As the C-S-H gel carbonation reaction progresses, the Ca/Si ratio gradually decreases, the decomposition rate of C-S-H accelerates, and the number of calcium carbonate crystals generated by the C-S-H carbonation reaction gradually decreases. After the C-S-H carbonization reaction, calcite, aragonite, and vanadium minerals are usually formed.

$$(CaO) \cdot x(SiO)(H_2O) + xCO_2 \rightarrow CaCO_3 + SiO_2 \cdot (H_2O)_t + (z-t)H_2O \quad (3)$$

3. The Impact of CO2 Cured Recycled Aggregates on the Durability of Concrete

At present, research on the durability of CO_2 reinforced recycled aggregate concrete mainly focuses on its ability to resist chloride ion penetration. Research has found that when using recycled concrete aggregates reinforced with 100% CO_2 substitution, the conductivity, chloride ion permeability, and air permeability of recycled concrete can be reduced by 15.1%, 36.4%, and 42.4%, respectively [25]. Kou et al. [26] used two types of mortar, 40MPa and 65MPa, to crush and produce recycled aggregates, which were then subjected to CO_2 mineralization treatment. The recycled aggregate concrete prepared after mineralization treatment showed a 41% and 46% increase in chloride ion permeability at 28 days, respectively. Xuan et al. [27] found that when the replacement rate of mineralized recycled aggregate was 100%, the chloride diffusion coefficient of recycled aggregate concrete decreased by 36.4%. Zhang et al. [28] found that the chloride ion diffusion coefficient decreased by 11 times after mineralization of recycled gravel aggregate mortar, while the chloride ion diffusion coefficient decreased by 6 times after mineralization of recycled crushed stone aggregate mortar. Zheng [29] studied the optimal conditions for CO_2 enhanced treatment of recycled aggregates, as well as the effects of CO_2 enhanced treatment on the apparent density, water absorption, and crushing index of recycled coarse aggregates. ASTM C1202 was used to test the electrical flux of 27 specimens, and PASTAT4000 electrochemical workstation was used to test the corrosion behavior of steel bars at different ages. The effects of different replacement rates of recycled coarse aggregates and different CO_2 strengthening times on the chloride ion permeability of recycled aggregate concrete and the corrosion performance of steel bars in recycled aggregate concrete at different corrosion ages were studied. The results indicate that: after 12, 24, and 48 hours of strengthening treatment with recycled coarse aggregate, the resistance of recycled concrete to chloride ion penetration was improved to varying degrees. At a strengthening time of 24 hours, the electrical flux of the concrete significantly decreased, and its resistance to chloride ion penetration was significantly improved. When the replacement rate of recycled coarse aggregate treated with CO_2 is 50%, the electrical flux of concrete with a strengthening time of 12, 24, and 48 hours decreases by 14%, 26%, and 31%, respectively, after 28 days. When the replacement rate of recycled coarse aggregate is 100%, the electric flux decreases by 17%, 34%, and 38%, respectively. The resistance to chloride ion penetration of recycled concrete prepared by strengthening recycled coarse aggregate after 24 and 48 hours is equivalent 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 produced by CO_2 mineralization of the aggregate fill the pores, improve the spatial structure of the aggregate, reduce the channels for chloride ion diffusion, and enhance the ability of concrete to resist chloride invasion.

But this is only one aspect of the durability of concrete structures. On the other hand, 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, the carbonation reaction produces calcium carbonate and other solid substances that gradually block the voids in concrete. The voids in concrete also decrease with the deepening of the reaction, 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 will lead to a decrease in the alkaline substances inside the concrete, resulting in a decrease in the pH value inside the concrete, and the concrete will gradually transform 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 passive film on the surface of the steel bars, and the steel bars will undergo corrosion reaction without the protection of the passive film. Although CO_2 reinforced recycled aggregate concrete has not undergone overall carbonation curing, the highly carbonized recycled aggregate inside has a relatively low pH value. When these neutral islands inside the concrete are exposed to CO_2 in the external environment, carbonization gradually deepens and forms a connected neutral channel, which may weaken the carbonation durability of the concrete. However, research in this area is currently scarce. Therefore, further research is urgently needed to investigate the effect of CO_2 enhanced recycled aggregates on the carbonation durability of concrete.

4. Conclusion

(1) CO_2 enhanced recycled aggregates can not only recycle waste building materials, but also play a role in carbon sequestration. At the same time, it can improve the mechanical properties of recycled aggregates, making it a promising green environmental protection technology path.

(2) CO_2 enhanced recycled aggregates can significantly improve the resistance of ordinary recycled concrete to chloride ion penetration, with higher density and better compactness.

(3) CO_2 enhanced recycled aggregates will reduce the pH value inside concrete, and whether it will have a negative impact on the carbonation durability of concrete remains to be studied. At the same time, there is currently a lack of relevant research on whether CO_2 enhanced recycled aggregates will affect other durability properties of concrete, and further research is needed.

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