Developing carbon-neutral construction materials using wastes as carbon sink

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Abstract. Developing ecofriendly and low-carbon construction materials is essential to meet the ambitious target of global net zero emission by 2050. The concept of using wastes as carbon sink is taking advantage of alkaline solid wastes, which are rich in calcium or magnesium as a medium for absorbing carbon dioxide (CO₂), and allowing manufacturing thermodynamically stable carbonate minerals for construction applications. Today, the major practical and successful applications of CO₂ in the construction industry are through accelerated carbonation of granulated aggregates and precast concrete blocks manufactured with different proportions of calcium-rich waste materials. CO₂ is also used as an activator in speciality nonhydraulic carbonate-binders or directly injected (in a liquid form) into fresh ready-mix concrete to produce a construction material with higher strength and lower carbon footprint. In addition, accelerated carbonation has become a promising approach to improve the quality of crushed concrete waste, and can be reused in new concrete to attain a sustainable concrete life cycle. In this paper, the progress in academic research of using CO₂ in wastes and cement-based products as well as the key advantages and limitations related to the large scale application in the construction industry have been discussed in detail.

1. Introduction
The atmospheric level of carbon dioxide (CO₂) has increased up to 414.5 ppm, which has forced the public to take the global warming issue seriously [1]. In December 2020, a truly Global Coalition for Carbon Neutrality was established to achieve a global net zero emission by 2050 [2]. As the world’s largest carbon emitter, China has pledged a “30–60 Target” to speed up the reduction in carbon intensity by 60%–65% less than the 2005 level, as compared to the CO₂ emission pledged in Paris Agreement [3].

Notably, 7%–10% of the global CO₂ emissions are attributed to the cement industry, which is needed to produce essential construction material, such as concrete. Every year, approximately 2.8 billion m³ of concrete is generated worldwide [4]. Therefore, developing ecofriendly and low-carbon construction materials using CO₂ capture through carbonation, is highly recommended. Numerous studies have shown that CO₂ curing or accelerated carbonation can enhance the properties of cement concrete, artificial recycled aggregate, and waste concrete [5 - 8].

Calcium silicate phases of cement clinker (e.g., C₃S and C₅S) and calcium-based hydration products (e.g., Ca(OH)₂) can easily react with CO₂ in water to form calcium carbonate (CaCO₃) and amorphous silica gel [9–10]. Monkman et al. [11] have identified the potential of CO₂ to unlock a
performance benefit in the cast-in-place concrete without impacting its durability. This process involves adding CO$_2$ into fresh concrete as a part of the batching and mixing steps. Moreover, using construction wastes as a carbon sink is achievable as well. Through this method, alkaline solid wastes rich in calcium or magnesium, will be used as a medium for absorbing CO$_2$.

Overall, to reduce the carbon footprint and fulfill the 30–60 Target, having comprehensive and systematic research on increasing the CO$_2$ sequestration in the whole concrete life cycle, as well as generating innovative ideas to speed up this journey is important. An overview of our current research works is illustrated in Figure 1.

![Figure 1. An overview of current research works](image)

In this paper, we focus on upcycling alkaline solid wastes to react with CO$_2$ via several technical routes to create commercially viable products. Further, the progress of academic research will be mainly detailed in the three following areas:
1. Production of steel slag granulated aggregate via carbonation
2. Dry-mix pressing carbonation products
3. Carbonation of ready-mix concrete

2. Production of steel slag granulated aggregate via carbonation
Application of accelerated carbonation on alkaline solid wastes, such as steel slag (generated from steel-making industry), is reckoned as an effective approach. It accounts for approximately 25% of the total waste production in China’s iron and steel-making field with an annual production of 101 million tons [12]. Carbonation of steel slag with low hydraulic but high CO$_2$ reactivity, such as basic oxygen furnace slag (BOFS), facilitates the manufacture of artificial aggregates, replacing the less occurring natural ones. The workflow is simplified and demonstrated in Figure 2.
With different CO₂ conditions (CO₂ concentration and temperature), post and synchronized carbonations during granulation, can produce lightweight and high-strength aggregates. Physical, mechanical, mineralogical, and microstructural properties have been investigated, and the results show that post carbonation can significantly increase the strength (atmost by 220%) as compared with the reference sample [7][10]. Owing to carbonate precipitates, approximately 40% higher microhardness value has been recorded. Although synchronized carbonation cannot be used to obtain high-strength aggregates, it seems promising in producing lightweight aggregates. By integrating the two approaches, the produced aggregates are found to have lower bulk density, higher water absorption, and higher CO₂ uptake. Nevertheless, the extensive heat released during the CO₂ granulation vaporizing too much water and unfilled voids are the issues that need to be resolved.

3. Dry-mix pressing carbonated products
Carbonation of hardened cement pastes with a high CO₂ concentration requires a longer carbonation time. The reaction only occurs on the surface layer, a few millimeters thick, attributed to the cement densification [13]. Dry-mix pressing approach requires low water to cement ratio, 0.06–0.2; and high casting press, 10–30 MPa, and it allows an immediate demolding feature and instant carbonation, facilitating the CO₂ diffusion and carbonation reactions through the whole pressed compacts. The workflow is summarized in Figure 3.

The carbonation strength increased remarkably, and the residual compressive strength of the CO₂ cured dry-mixed pressed blocks is higher than that of the air-cured blocks. This is owing to a greater degree of cement hydration and formation of CaCO₃, which has improved the integrity of microstructure and reduced porosity. In addition, a positive influence on the high temperature of CO₂ cured samples has been noticed owing to the conversion of Ca(OH)₂ into thermally stable carbonate products, such as CaCO₃.

4. Carbonation of ready-mix concrete
Ready-mix concrete (RMC) is widely adopted to fast-track the construction. For reinforced concrete structures, carbonation is usually considered as a hazard as it can lead to corrosion of the steel reinforcement and then, failure of the structure. However, early-age carbonation curing creates
concretes with low chloride penetration owing to its dense surface concrete layer [14]. Thus, carbonation cured concretes are likely to be more corrosion resistant than ordinary concrete if carbonation is limited to the surface layer.

In fact, different early-age carbonation rates play different roles: for a lower rate, the subsequent hydration after carbonation can increase the pH to a level that can prevent steel corrosion [15]; for a higher rate, steel could be substituted for corrosion-resistant materials, such as a fiber-reinforced polymer. For RMC, CO₂ instantly becomes a mixture of gas and solid (snow) before dispensing into the concrete mixer. It is added shortly after water and before the entire mixing is finished [11]. The reaction between CO₂ and Ca²⁺ ions can improve the compressive strength of the concrete. Therefore, carbonation has a great potential in improving the performance of the concrete (dry-mix and ready-mix), even for the concrete wastes and aggregates. The workflow of current research is encapsulated in Figure 4.

![Figure 4. Flowchart of carbonation of dry-mix and wet-mix (ready-mix) concretes](image)

Based on this research, CO₂ uptake is mainly related to the availability of active minerals in the concrete and the water-to-solid ratio (w/s). When w/s exceeds 0.4, an obvious decrease in CO₂ uptake is obtained. This is primarily owing to liquid saturation, which hinders the diffusion of CO₂ reaction. In addition, the leachability of heavy metals is altered, attributed to the pH of concrete wastes, which is almost neutralized after carbonation. The calcite formation in the pore space also decreases the porosity and densifies the structure, which is beneficial in strength gain. Nonetheless, further detailed studies, such as setting time, durability, and drying shrinkage, are required to improve the feasibility of this technique in the construction industry.

5. Conclusions

Overall, the stated approaches are feasible in reducing CO₂ from the perspective of construction industry. Among all, the steel slag granulated aggregate can reduce CO₂ the most as the carbonation can occur during granulation of aggregate, followed by the dry-mix pressing of carbonated blocks, owing to pores for CO₂ absorption. For RMC, minimal CO₂ is used during the carbonation, resulting in the lowest efficiency. Nevertheless, all the reductions are usually small, and there are several limitations that need to be resolved, as described below:

1. Sourcing appropriate raw material limits the use of alkali-activated materials
2. Recycling has negligible effects on global warming potential
3. Production processes have very limited scope for optimization

Therefore, to successfully translate carbonation technology to commercial scale application in the future, improving the CO₂ sequestration rate and CO₂ amount into cement-based or alkaline solid wastes construction materials, is critical. This could contribute toward sustainable development, offering an excellent avenue for CO₂ capture and storage while enhancing the quality and safety of construction material for the end-users.
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