

A MINI REVIEW OF MINERAL CARBONATION OF SOLID MATERIALS

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ABSTRACT

Mineral carbonation of solid materials (MC-SM) has garnered significant attention for its potential to reduce carbon dioxide emissions. This study presents a thorough overview of research trends and hotspots in MC-SM through bibliometric analysis. Articles were sourced from the Google Scholar core database, covering the period from 2000 to 2020, and were analysed in detail. The findings indicate that MC-SM is an emerging research area, with rapidly changing key topics. China and Switzerland lead in publication numbers and average citations, respectively. Carbonation curing typically enhances the strength and durability of materials due to carbonate formation. In terms of carbon dioxide storage capacity, materials such as carbide slag, reactive magnesium oxide cement, and β -C2S are effective, while fly ash, mine tailings, and carbon mixes show limited activity. The slow kinetics and low carbonation rates currently hinder the large-scale industrial application of MC-SM.

Keywords: Solid materials, carbon dioxide, waste materials, mineral carbonation.

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INTRODUCTION

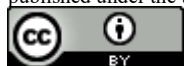
Since the Industrial Revolution, the amount of carbon dioxide in the atmosphere has increased, with annual worldwide emissions exceeding 30 billion tons and still rising. The pressing need to cut greenhouse gas emissions has rekindled interest in carbon sequestration materials based on cement. A promising method for storing carbon dioxide is provided by carbon capture, utilization, and storage technology, which includes mineral, geological, and ocean storage. It must contribute 20% of the emissions reductions required to meet the target of halving greenhouse gas emissions by 2050,

according to the International Energy Agency [1].

In ocean storage, captured carbon dioxide transported via ships or pipelines to the deep-sea floor for storage. However, this method is costly and can lead to ocean acidification. High concentrations of carbon dioxide in seawater can harm marine life, and dissolved carbon dioxide may eventually re-enter the environment. Additionally, ocean storage is still in the experimental phase.

Conventional geological storage involves injecting large volumes of supercritical

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carbon dioxide into porous and permeable geological formations. However, these formations are dynamic, which introduces risks such as carbon dioxide leakage and stratigraphic deformation. Furthermore, the reaction time between geological rock layers and carbon dioxide can take thousands of years due to low reactivity. In contrast, mineral storage of carbon dioxide is permanent, environmentally safe, and requires minimal monitoring. Consequently, mineral carbonation has attracted significant attention in recent years [2-4].

Mineral carbonation is defined as the reaction of raw materials containing metals such as calcium (Ca) and magnesium (Mg) with carbon dioxide (CO₂) to form insoluble carbonates. Based on our research group's literature survey, mineral carbonation raw materials can be categorized into two groups: natural ores and solid materials that contain Ca and Mg. The solid materials include systems like Ordinary Portland cement, magnesia cement, industrial solid waste, construction solid waste, and mining tailings. However, existing literature on mineral carbonation tends to focus on one specific area, leaving a comprehensive bibliometric review of carbon sequestration in solid materials lacking.

While research has addressed the mineral carbonation of natural minerals, steel slag, blast furnace slag, and cement-based materials, it has largely overlooked the carbonation of solid wastes, such as mine tailings, red mud, carbide slag, and recycled concrete powder. Additionally, the concept of carbon mixing—transporting carbon dioxide during concrete mixing—is often not discussed in reviews of mineral carbonation [5-7].

Reactive magnesium oxide cement has garnered increasing research interest due to its benefits over ordinary Portland cement,

such as a lower calcination temperature and a higher theoretical carbon dioxide sequestration rate. In contrast to reactive magnesium oxide, magnesium oxychloride cement, magnesium sulfate cement, and magnesium phosphate cement may exhibit different performance characteristics following carbonation. While several scholars have reviewed the carbonation of reactive magnesium oxide. This study offers several novel contributions: (1) It is the first comprehensive investigation that systematically broadens the research scope across various civil engineering disciplines; (2) A thorough bibliometric analysis of mineral carbonation in solid materials was performed, identifying suitable journals for sourcing important information in MC-SM research through a fuzzy comprehensive evaluation method. (3) The selection of materials with significant carbon sequestration potential was aided by the collection, normalization, and comparison of data on the carbon dioxide storage capacity of different types of solid materials [8].

A bibliometric study was carried out and relevant articles from 2000 to 2020 were gathered to give a more thorough grasp of developments in MC-SM. The most recent advancements in research and the difficulties encountered are highlighted in this report. The results show that most materials' early strength, durability, and mechanical qualities have been enhanced by carbonation. Progress has been made in carbon sequestration technology; however, new challenges have emerged, such as low carbonation efficiency, immature craftsmanship, and excessive energy consumption [9, 10]. To draw greater attention from researchers and address these issues, this review aims to help researchers comprehend the trends, advantages, and disadvantages of mineral carbonation, providing a scientific basis for informed

decision-making and evaluation in research.

METHODOLOGY

The bibliometric analysis method highlights the annual publications in MC-SM research from 2000 to 2020. Prior to 2008, the field experienced a limited number of yearly publications. However, from 2008 onwards, there has been a gradual and consistent increase in these publications. Notably the academic community launched an extensive investigation into mineral carbonation, leading to a significant rise in published works. This upward trend aligns with the growing focus on environmental issues and climate change, indicating that mineral carbonation is becoming an important and promising research area. The earliest proposal regarding the carbon dioxide fixation potential of calcium silicate concrete was made in 2006 [11]. MC-SM research is multidisciplinary, primarily encompassing the fields of Engineering, Materials Science, and Construction Building Technology. These domains fall under "Technology," one of the five main research areas. This suggests that MC-SM research is practice-oriented and directly addresses the need to mitigate the global greenhouse effect. Journal source visualization, an essential bibliometric method, provides the academic community with a systematic overview of knowledge landscapes and research directions. It allows for the intuitive identification of core journals, knowledge dissemination pathways, and centers of academic influence. Furthermore, analyzing journal sources helps readers efficiently access relevant information and determine the most suitable journals for their publications. The size of each node indicates the number of published articles, while the connecting lines represent mutual citations [12].

RESEARCH PROGRESS ON MINERAL CARBONATION OF SOLID MATERIALS

MC-SM is an effective method for reducing the greenhouse effect through the carbonation of various materials, including cementitious materials, industrial solid waste, construction waste, mine tailings,

carbon mixes, and more. However, it is important to note that this work does not cover the carbon sequestration of biochar, asphalt, plastic, and wood due to their distinct mechanisms differing from mineral carbonation.

Solid Waste of Construction

Based on particle size, recycled materials from crushed construction and demolition waste can be divided into three categories: recycled powder (RP, <0.15 mm), recycled fine aggregate (RFA, 0.15 mm–5.00 mm), and recycled coarse aggregate (RCA, 5.00–31.5 mm). While RP comprises recovered concrete powder (sometimes referred to as "recycled concrete fines" by certain scholars, but called recycled concrete powder in this review), recycled brick powder, and recycled glass powder, RCA and RFA are both categorized as aggregates. The widespread use of recycled aggregates (RA) and RP promotes sustainability and offers the possibility of mineral carbonation by lowering the need for natural resources [13–16].

The performance of recycled aggregate concrete (RAC) and RA (including RCA and RFA) has been the subject of extensive research over the last 20 years. The performance of RA is typically worse than that of natural aggregates because of the sticky old mortar and numerous interfacial transition zones. Additionally, natural aggregate concrete with the same water-to-cement ratio usually has stronger mechanical qualities, workability, and durability than RAC. Because of this, a number of RA modification methods have been put out recently, with the carbonation approach being acknowledged as a more sustainable and environmentally friendly choice than other modification technologies.

When producing recycled aggregates (RA), particles smaller than 150 μm are also generated, referred to as recycled concrete

powder (RCP). RCP consists of various components, including CaCO_3 , AFt, AFm, SiO_2 , CH, C–S–H, and unhydrated cement particles. However, RCP produced during RA generation has low reactivity, which limits its effectiveness as an inert filler when used in high quantities.

To enhance the performance of RCP, carbonation is a promising treatment. This process leads to the formation of CaCO_3 , which reduces porosity in RCP. Additionally, it promotes the creation of stable calcium aluminum hemicarboxate, calcium aluminum monocarbonate, and AFt, thereby improving the microstructure of the cement matrix.

Solid Waste of Industry

Industrial production generates substantial amounts of solid waste containing calcium and magnesium, which can be utilized for mineral carbonation. This includes materials such as steel slag, red mud, calcium carbide slag, coal gangue, fly ash, gypsum, and ground granulated blast furnace slag. Varied carbonation mechanisms arise from the varied compositions of these solid wastes.

Basic oxygen furnace slag, electric arc furnace slag, ladle furnace slag, argon oxygen decarburization slag, and induction furnace slag are the several varieties of steel slag. Direct and indirect carbonation are two of the carbonation techniques used for SS; direct carbonation is further subdivided into dry and moist carbonation.

Red mud, a highly alkaline industrial byproduct from alumina production, has seen limited utilization in construction, environmental management, mining, and agriculture, with only 2–3% effectively processed. Due to its mineral content, which includes calcium (Ca) and magnesium (Mg), red mud can undergo carbonation. This process reduces its alkalinity, providing a viable pretreatment method for its utilization [17].

Carbide slag (CS), a byproduct of calcium carbide hydrolysis during acetylene production, primarily consists of $\text{Ca}(\text{OH})_2$. The smelting and volatilization processes in calcium carbide production lead to lower levels of other components, such as CaO, SiO_2 , Al_2O_3 , and Fe_2O_3 .

Coal gangue, a mineral associated with coal, is generated during coal mining and washing, with approximately 0.15–0.2 tons produced for every ton of raw coal. Its chemical composition is diverse, containing elements like aluminum (Al), silicon (Si), and carbon (C). The main chemical components of coal gangue include SiO_2 , Al_2O_3 , Fe_2O_3 , CaO, MgO, TiO_2 , and K_2O , with the total content of SiO_2 and Al_2O_3 ranging from 60–90%.

Fly ash is commonly utilized in the production of solid materials, mine landfills, and agricultural applications, with a utilization rate of less than 60%. Recently, fly ash has garnered significant attention as a mineral carbonation material due to its large output. Carbonation enhances the stability of fly ash, thereby expanding its applications in civil engineering. The carbonation process can be classified into direct and indirect methods, with indirect carbonation being more costly and the presence of carbonates in fly ash reducing carbonation effectiveness.

Flue gas desulfurization gypsum, phosphogypsum, titanium gypsum, and red gypsum are some of the forms of byproduct gypsum that come from diverse industrial operations. $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ is the main component of byproduct gypsum, and its purity varies depending on how it is produced [18].

Cementitious Material

In a carbon dioxide cured Portland cement-based system, CO_2 reacts with calcium silicate and calcium silicate hydrate. The carbonation of adequately moistened C_2S and C_3S occurs within minutes to hours,

leading to the rapid formation of C–S–H, which contributes to early strength gain. The paste matrix consists of C₃S, C–S–H, C₂S, CaCO₃, and Ca(OH)₂. Aside from calcite, a gel-like amorphous phase of CSH with a lower CaO/SiO₂ ratio than typical C–S–H has been observed in CO₂-cured calcium silicates. This gel phase is sometimes referred to as decalcified C–S–H or polymerized silica gel, displaying morphological similarities to C–S–H produced from C₃S hydration [19-21].

In recent years, magnesium-based cements have gained significant attention. Common types include reactive magnesium oxide cement, magnesium oxychloride cement, magnesium phosphate cement, and magnesium sulfate cement.

Mine Tailings

Mine tailings represent the primary waste product of the mining industry, generating approximately 10–15 billion tons annually. The small particles found in mine tailings can reduce mineral crushing costs and exhibit potential for carbonation. However, the degree of carbonation in mine tailings remains low [22]. The carbonizable substances in mine tailings include magnesium/calcium oxides and magnesium/calcium silicates, such as (Mg, Fe)₂SiO₄, 3MgO·2SiO₂·2H₂O, Mg₃Si₂O₅(OH)₄, and CaSiO₃, which necessitate deposits of mafic/ultramafic (M/UM) rocks. The advantages of utilizing mine tailings for mineral carbonation are twofold, Mine tailings has large reserves that are widely distributed and readily accessible; and Its fine particle sizes help reduce crushing processing costs.

Currently, researchers have proposed various laboratory-scale strategies to address the main challenges of mineral carbonation in mine tailings. However, limited efforts have been made to apply these strategies on a pilot or large scale. It has been reported that processing and carbon

dioxide sequestration costs account for 85 % of the total expenses. In a pilot project by Mineral Carbonation International in Australia, serpentine was used as feedstock to capture carbon dioxide emissions from the Kooragang Island power plant. Despite this, the current carbonation yield of 35% is significantly below the levels needed for scaling up the process, even when factoring in potential sales of the MC products.

Carbon Mix Technology

Carbon mix technology involves transporting carbon dioxide into premixed concrete via mixer trucks, with a reaction process similar to that of Ordinary Portland Cement systems. In the carbon mix process, 963 grams of carbon dioxide are captured, resulting in a reduction of 17 kg of cement and an increase of 14 kg of sand, while also decreasing overall material transportation emissions by 124 grams of carbon dioxide [23-25].

Canadian company Carbon Cure has developed a comprehensive carbon mix system. This system connects a tank of liquid carbon dioxide to a gas control system and injector. The liquid carbon dioxide is precisely metered for injection into the truck, where it transforms into a mixture of carbon dioxide gas and solid carbon dioxide “snow.” The carbon dioxide is then delivered into the fresh concrete at a specific flow rate during a fixed injection interval, reacting with the hydrating cement during the initial mixing stage. The concrete is subsequently assessed and tested. Concrete treated with carbon mix demonstrates improved early strength, ranging from 3-17%. This treatment also results in reduced initial and final setting times, as well as an accelerated hydration rate and strength development. By utilizing carbon mix, the amount of cement required can be decreased. Although this reduction may initially weaken the concrete, it is possible to restore compressive strength performance after carbonation.

Life cycle assessment in Mineral Carbonation of Solid materials

Life Cycle Assessment is a valuable tool for environmental management and analysis, as it quantifies resource consumption and environmental emissions throughout the entire product lifecycle. Life cycle assessment has become a key focus in current research, with a notable increase in studies examining the life cycle assessment of MC-SM over the past decade.

In terms of cementitious materials, it has been suggested that carbon dioxide mineralization in concrete may reduce carbon dioxide emissions from the manufacturing of cement clinker by up to 15%, with theoretical reductions approaching 60%. It's common to oversimplify the benefits of hydrated cement carbonation in direct air capture. According to research cement reabsorbed over 13.8 billion metric tons of carbon dioxide worldwide. When taking into consideration the carbonation capability of cement, it was suggested that the net carbon dioxide emissions of ready-mixed concrete are roughly 70% lower than those of ordinary Portland cement. Prioritizing concrete mixtures with high supplemental cementitious material substitutions in carbon capture, utilization, and storage initiatives has been proposed [26].

In terms of industrial solid waste, the direct and indirect carbonation sequestration potential of alkaline industrial waste is estimated at 4.02 gigatons of carbon dioxide per year globally. It is reported that over 4 billion tons of industrial solid waste are generated annually in China alone, and if fully carbonated, this waste could capture more than 200 million tons of carbon dioxide as carbonates. Pan et al. found that the steel industry can address both waste management and CO₂ emissions by employing a high-gravity carbonation process, thereby promoting a circular economy. They prepared carbonated stainless steel-pressed

blocks and observed a 70% reduction in environmental impact compared to ordinary Portland cement-based blocks of equivalent compressive strength. In another study, compared the global warming potential of carbonated recycled cementitious materials-based mortar with that of ordinary Portland cement-based mortar and found that the former results in a about 75 % reduction in environmental impact. Additionally, using carbonated recycled aggregates instead of natural aggregates helps save environmental costs and reduce carbon dioxide emissions, providing significant environmental and economic benefits.

In Canada, nickel tailings can capture approximately 16% of the carbon dioxide emitted annually by planned mining operations. Specifically, 15 million tons of tailings produced each year generate 127.7 kilotons of carbon dioxide, with the potential to sequester 21 kilotons of carbon dioxide through passive mineral carbonation. Moreover, it has been found that mineral carbonation of platinum group element tailings could theoretically account for 43.6% of the 95% pure carbon dioxide produced annually by the synthetic fuels industry in South Africa. Additionally, mineral carbonation of serpentine minerals using flue gas can reduce carbon dioxide emissions by 215 kg for every 1 megawatt-hour (MWh) of electricity generated by a power plant.

Capacity of CO₂ Storage in Solid materials

Understanding the mineralization potential of different solid materials is improved by normalizing their carbon dioxide storage capacity. The CaO/MgO content and material shape have an impact on this capacity. Solid materials can be categorized as extremely active (>200 g/kg), active (100–200 g/kg), inactive (50–100 g/kg), and very inactive (<50 g/kg) based on their capacity to store carbon dioxide [27].

Because of their high carbonizable component content, carbide slag, reactive magnesium oxide cement (RMC), and β -C₂S are considered particularly active materials. With an average value of 517.8 g/kg, carbide slag has the highest CO₂ storage capacity due to its high Ca(OH)₂ content (85–95%). Significant levels of carbonizable MgO and C₂S are also present in RMC and β -C₂S, respectively.

In terms of solid waste, fly ash and mine tailings exhibit very low CO₂ storage capacity, necessitating activation measures to enhance their mineralization potential. Furthermore, in carbon mix, carbon dioxide has slow solubility and diffusion rates within concrete, and the presence of CaCO₃ particles fills the pores, obstructing further carbon dioxide diffusion. Consequently, the carbon mix also shows an inactive carbon dioxide storage capacity. To improve the carbon dioxide storage capacity of the carbon mix, it is essential to develop suitable additives or enhance carbonation devices and processes. However, these improvements will likely lead to increased costs and energy consumption.

CHALLENGES ENCOUNTERED ND FUTURE ASPECTS

Bibliometric analysis has various limitations even though it might reveal research trends and knowledge systems. The thoroughness and timeliness of the results can be affected by variables like database coverage, language preferences, and delays in citation accumulation. Furthermore, quantitative indicators might not adequately convey the research's deeper significance. To enable more thorough and impartial information integration and theoretical growth, future review studies should incorporate a greater variety of literature sources and qualitative analytic techniques.

The bibliometric analysis presented in this article indicates that mineralization objects,

methods, material properties post-carbonation, and environmental assessments are of significant interest in MC-SM research. Various evaluation methods exist in this field, including carbon dioxide sequestration rate, mineralization degree, and mineralization efficiency, which can lead to misunderstandings and confusion. To mitigate this problem, it is essential to carefully evaluate the strengths and weaknesses of different assessment methods and to establish a more universally accepted and applicable evaluation approach. Furthermore, advancements in Life Cycle Assessment are needed to extend its scope beyond the “cradle” to the “gate” and to address the impacts of allocation methods and uncertainty analysis on the Life Cycle Assessment results for carbonated products [28].

Scalability, economic viability, and environmental benefits present hurdles for the technical implementation of MC-SM technology. For example, steel corrosion may result from the lower pH of carbonated ordinary Portland cement-based concrete compared to uncarbonated ordinary Portland cement-based concrete. The magnesium-based cement system is more expensive in terms of raw materials and has a lower pH than the ordinary Portland cement system, which makes it unsuitable for passivating steel bars. Furthermore, the establishment of standardized carbonation protocols is complicated because the efficiency of carbonation is impacted by the variety in characteristics of solid waste sources (such as steel slag, red mud, fly ash, and recycled concrete).

Moreover, significant costs and energy consumption are associated with carbon dioxide capture, purification, transportation of carbonized materials, and energy-intensive curing processes. For instance, producing a carbonated concrete block is approximately 35% more expensive than

producing one with ordinary Portland cement. Many technologies remain in prototype or pilot stages, hindered by high costs and immature process chains. The large-scale commercial application of mineralized cement is further impeded by the absence of relevant standards.

CONCLUSION

The research should focus on improving carbon dioxide sequestration capacity by exploring carbonation mechanisms and optimizing process parameters. Additionally, a comprehensive life-cycle assessment of MC-SM is essential. This work lays the groundwork for advancing large-scale mineral carbonation using solid materials.

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