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Critical Review

A Comprehensive Review of Mineral Carbonation of Civil Engineering Materials: A Bibliometric Analysis

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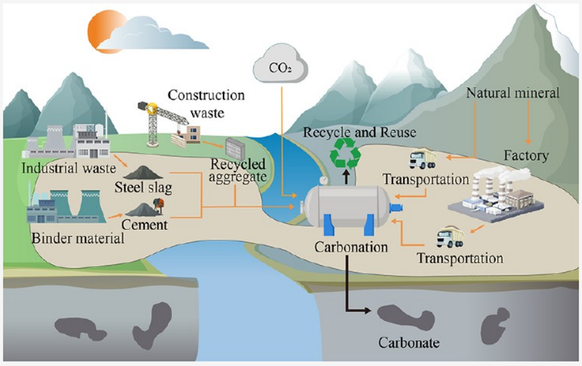
ABSTRACT: Mineral carbonation of civil engineering materials (MC-CEM) has received increased attention in reducing CO_2 emissions. This work comprehensively describes the research trends and hotspots in MC-CEM based on bibliometric analysis. Articles were collected in the Web of Science core database from 2001 to 2024 and analyzed in detail. The results show that MC-CEM is a hot research topic, with hot-topic words changing quickly. China and Switzerland reveal the most publications and the highest average citations, respectively. Under carbonation curing, the strength and durability are usually improved due to carbonate formation. Concerning CO_2 storage capacity, carbide slag, reactive magnesium oxide cement, and β-C_2S are active, while fly ash, mine tailings, and carbon mix are inactive. Slow kinetics and low carbonation influence the large-scale industrial application of MC-CEM. Finally, challenges and prospects in MC-CEM are illustrated. In future studies, it is necessary to enhance the CO_2 sequestration capacity by investigating the carbonation mechanisms and optimizing the process parameters. A comprehensive life-cycle assessment of MC-CEM is also needed. This work provides the basis for the development of large-scale mineral carbonation by using civil engineering materials.



KEYWORDS: *mineral carbonation, civil engineering materials, bibliometrics, CO_2 storage capacity, solid wastes*

1. INTRODUCTION

The concentration of CO_2 in the atmosphere has been increasing since the industrial revolution, with global annual emissions exceeding 36 billion tons and continuing to rise. The urgency of reducing greenhouse gas emissions has reignited people's interest in cement-based materials for carbon sequestration.[1] The carbon capture, utilization, and storage (CCUS) technology[2] is a promising strategy for storing CO_2, including ocean storage, geological storage, and mineral storage.[3] According to the International Energy Agency, CCUS needs to account for 20% of emissions reduction to meet the goal of halving greenhouse gas emissions by 2050.

In ocean storage, captured CO_2 is transported by ships or pipelines to the deep-sea floor (1000 or 3000 m) for storage. Yet, it presents the advantage of high cost and ocean acidification.[3,4] Meanwhile, high concentrations of CO_2 in seawater can kill sea beings, causing dissolved CO_2 to return to the environment eventually.[5] Besides, ocean storage is still in the experimental stage. In conventional geological storage, large-scale supercritical CO_2 is injected into porous and permeable geological formations.[5] Yet, the geological formations are dynamic, and risks (CO_2 leakage and stratigraphic deformation) may occur. Moreover, the reaction time takes thousands of years due to low reactivity between the geological rock layer and CO_2. Compared to the first two, mineral storage of CO_2 is permanent, environmentally safe, and almost does not require monitoring. Therefore, mineral carbonation has received widespread attention in recent years,[6] with its schematic diagram shown in Figure 1a.

Mineral carbonation was proposed by Seifritz in 1990,[7] defined as the reaction of raw materials containing metals such as Ca and Mg with CO_2 to form insoluble carbonates. According to the literature survey of our research group, raw materials of mineral carbonation can be divided into natural ores and civil engineering materials containing Ca and Mg.[8] The latter includes Ordinary Portland cement system,[9] magnesia cement system,[10,11] industrial solid waste,[4] construction solid waste,[12,13] mining tailings,[14,15] etc. However, existing literatures[4,9−13,15,16] on mineral carbonation typically focuses on one of the topics above, and a comprehensive review of carbon sequestration of civil engineering materials by bibliometric analysis is lacking. Table S1 demonstrates this work's differences and innovations relative to these studies.

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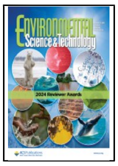

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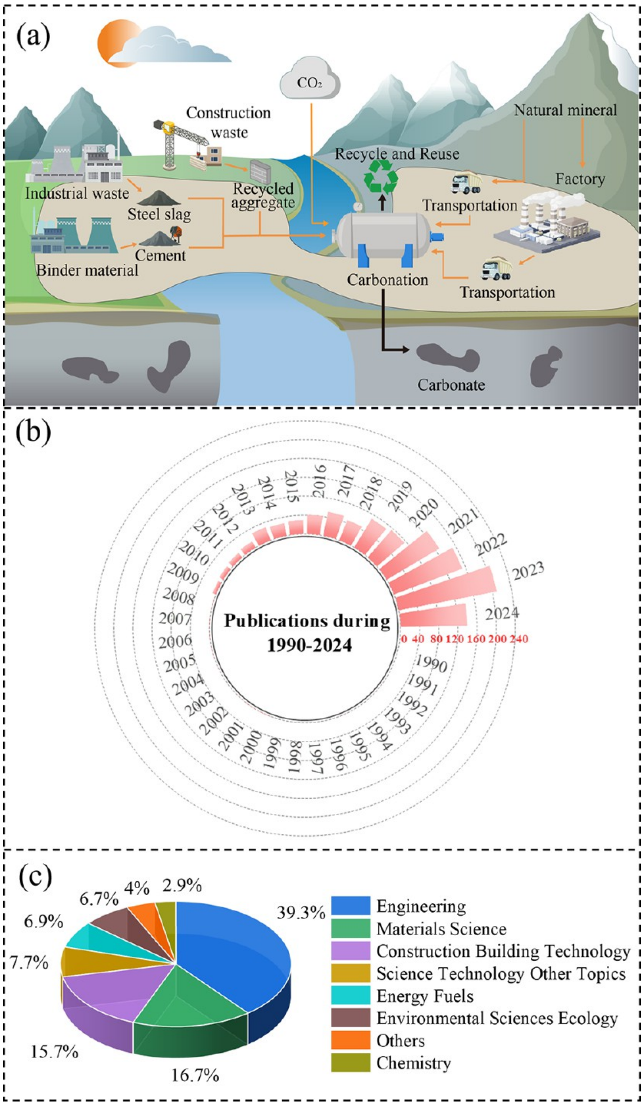


Figure 1. (a) Schematic diagram of mineral carbonation, (b) yearly publications, and (c) discipline classification in MC-CEM research from January 1990 to May 2024.

Although Lin et al.[3] conducted scientific quantitative analysis on the mineral carbonation of natural minerals, steel slag, blast furnace slag and cement-based materials. Yet, the carbonation of solid wastes (e.g., mine tailings, red mud, carbide slag and recycled concrete powder) was not considered in the report.[3] Moreover, carbon mix (transporting CO_2 during concrete mixing) is usually not illustrated in review of mineral carbonation.

Meanwhile, reactive magnesium oxide cement (RMC)[17] has received increasing research interest due to its advantages over OPC (lower calcination temperature and higher theoretical CO_2 sequestration rate[18]). Compared to reactive MgO cement, magnesium oxychloride cement (MOC), magnesium sulfate cement (MOS) and magnesium phosphate cement (MPC) may demonstrate different performance development after carbonation.[19,20] Some scholars have conducted reviews on carbonation of RMC,[10,11] but reviews on carbonation of MOC, MOS, and MPC are lacking. Unlike previous reviews,[4,9−14,16] this study proposes the following novel contributions: (1) To our knowledge, this study represents the first comprehensive investigation to systematically expand the research scope across the full spectrum of civil engineering disciplines, including not only cement (OPC and RMC) and solid waste but also MOC, MPC, MOS, and carbon mix. (2) A comprehensive bibliometric analysis was conducted on mineral carbonation of civil engineering materials (MC-CEM, with all abbreviations in this paper listed in Table S2). Suitable journals for obtaining important information in MC-CEM research were provided by the fuzzy comprehensive evaluation method[21] (Section 2.3). (3) The CO_2 storage capacity of 17 types of civil engineering materials was collected from 62 articles, which were normalized and compared (Section 3.7), facilitating the selection of civil engineering materials with high carbon sequestration potential.

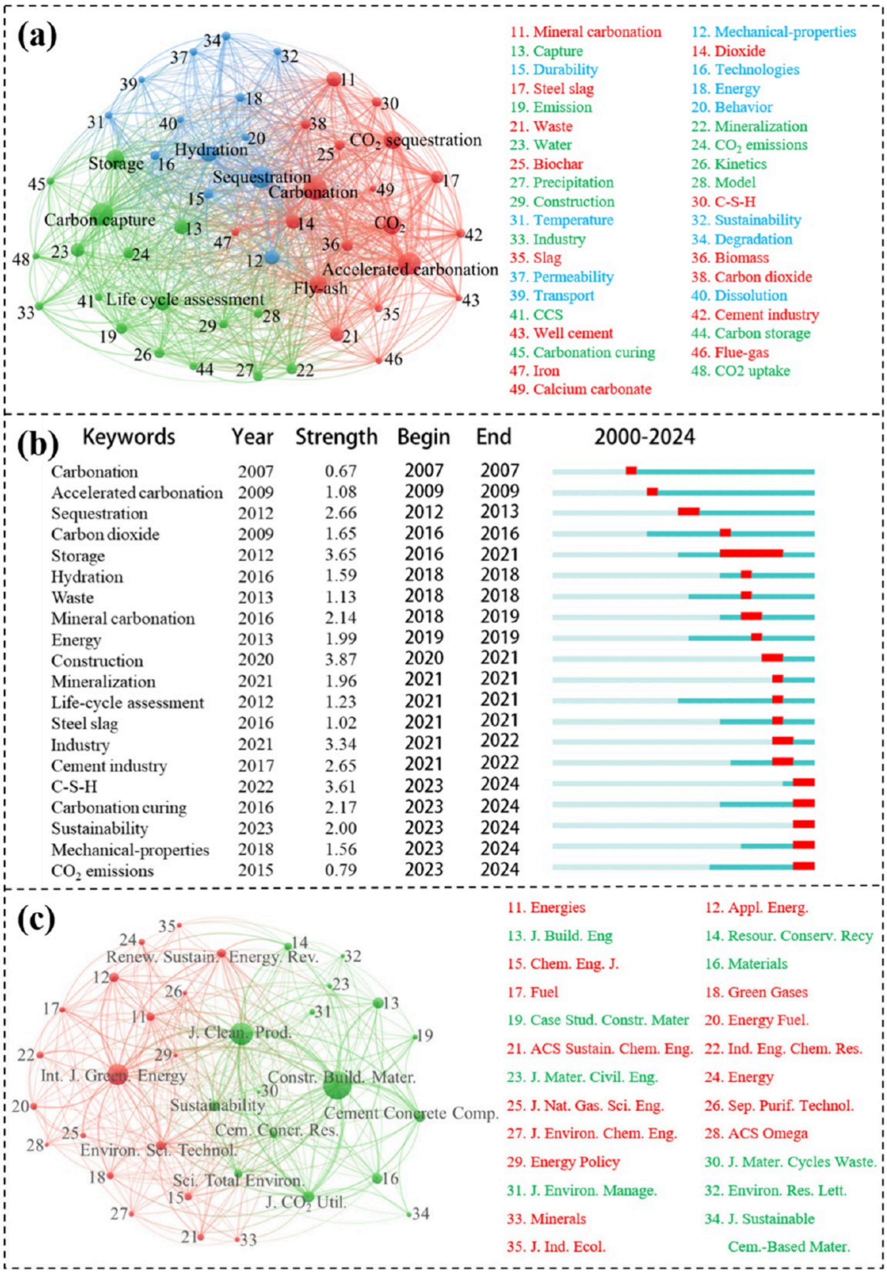
To provide a comprehensive understanding of the progress in MC-CEM, the related articles from 2000 to 2024 were collected and a bibliometric analysis was conducted. The latest research progress and the challenges encountered were illustrated. The results show that most materials' early strength, durability, and other mechanical properties have improved after carbonation.[22−27] Some constructive progress in carbon sequestration technology has occurred. However, a series of new problems have arisen, e.g., low carbonation efficiency,[28] immature craftsmanship[29] and excessive energy consumption.[30] To attract more researchers' attention and address these issues, this review aims to help researchers understand the development trends, advantages and disadvantages of mineral carbonation, and provide a scientific basis for scientific research decision-making and evaluation.

2. BIBLIOMETRIC ANALYSIS

2.1. Sample Overview. The method of bibliometric analysis is illustrated in Section S1. Figure 1b illustrates yearly publications in MC-CEM research from January 1990 to May 2024. Before 2008, the initial phase was characterized by a limited yearly publication number. From 2008 onward, the yearly publication numbers gradually and consistently increased. Meanwhile, from 2019 to 2023, the academic community initiated an extensive investigation into mineral carbonation, resulting in a notable surge in published works. This growth trend is consistent with the increasing attention paid to environmental issues and climate change. This indicates that mineral carbonation is emerging as a prominent and promising research area. Moreover, according to the search results, Berger et al.[31] were the earliest to use CO_2 to accelerate the hydration of calcium silicate in 1972. The earliest proposal on the CO_2 fixation potential of calcium silicate concrete was made in 2006 by Shao et al.[32]

As shown in Figure 1c, MC-CEM research is multidisciplinary, with the predominant domains of Engineering, Materials Science and Construction Building Technology. These three domains belong to "Technology" (one of the five primary research areas in Web of Science). It suggests that the MC-CEM research focuses on the practice, which aligns directly with requirements to alleviate the world's greenhouse effect.

2.2. Analysis of Keywords. The visual analysis of keywords is employed to identify research trends and emerging topics within specific fields.[33,34] The minimum number of occurrences was set to 25 to create a co-occurrence network map. Common keywords such as "cement" and "concrete" were removed, and some keywords with similar meanings were merged. Subsequently, a co-occurrence network analysis was



Keywords	Year	Strength	Begin	End
Carbonation	2007	0.67	2007	2007
Accelerated carbonation	2009	1.08	2009	2009
Sequestration	2012	2.66	2012	2013
Carbon dioxide	2009	1.65	2016	2016
Storage	2012	3.65	2016	2021
Hydration	2016	1.59	2018	2018
Waste	2013	1.13	2018	2018
Mineral carbonation	2016	2.14	2018	2019
Energy	2013	1.99	2019	2019
Construction	2020	3.87	2020	2021
Mineralization	2021	1.96	2021	2021
Life-cycle assessment	2012	1.23	2021	2021
Steel slag	2016	1.02	2021	2021
Industry	2021	3.34	2021	2022
Cement industry	2017	2.65	2021	2022
C-S-H	2022	3.61	2023	2024
Carbonation curing	2016	2.17	2023	2024
Sustainability	2023	2.00	2023	2024
Mechanical-properties	2018	1.56	2023	2024
CO_2 emissions	2015	0.79	2023	2024

Figure 2. (a) Visual analysis of keywords co-occurrence, (b) top 20 keywords, and (c) mainstream journals that published MC-CEM.

conducted on the remaining 49 keywords, as illustrated in Figure 2a.

The red, green and blue clusters represent the "materials", "methods", and "performance" in MC-CEM research. The red cluster reveals the most keywords and is distinctly related to other keywords, indicating an extensive research scope of carbonizable materials. In addition to ordinary Portland cement (OPC), it also includes magnesium-based cement,[18,35] steel slag,[36,37] red mud,[38,39] fly ash,[40,41] recycled aggregates,[42,43] etc.

The green cluster focuses on the commonly used methods in MC-CEM research, such as carbonation models and kinetics analysis. The hydration/carbonation mechanism in MC-CEM research was explored, and influencing factors (e.g., temperature, liquid−solid ratio, time, etc.) were summarized.[44] Meanwhile, life-cycle assessment (LCA) was also used to evaluate the environmental impact of the products in MC-CEM research.[45,46] The blue clustering focuses on studying the properties of materials after mineralization, such as mechanical and durability properties. Although it has the fewest keywords, it is an extension of red and green and has significant importance.[43,47]

Figure 2b shows the top 20 keywords. "Year" represents the year in which a keyword first appeared. "Begin" and "End" reflect the starting and ending years when a keyword exists as a hot-topic keyword. "Storage" lasts the longest (from 2016 to 2021), indicating it is a key topic in MC-CEM research. Moreover, the iteration speed of hot-topic keywords is very fast. Specifically, except for "Storage", the highlighting time of all keywords is within two years, indicating that MC-CEM is a hot research topic and has made significant progress in recent years.

In addition, "Carbonation" became a hot-topic keyword since 2007, which is close to the time (2006) when the concept of CO_2 fixation potential of concrete was proposed[32] (illustrated in Section 2.1). The above-highlighted words also reflect the development trend of MC-CEM. Among them, the current hotspots in MC-CEM research are mineralization methods (carbonation curing,[48] accelerated carbonation,[49] etc.) and mechanism study (chemical changes and microstructural evolution of materials during carbonation process[50]). Among them, "carbonation curing" primarily focuses on enhancing performance of civil engineering materials by the curing means of carbonation.[51,52] In contrast, "accelerated carbonation" aims for CO_2 sequestration through optimized carbonation conditions.[53] In some cases, it serves to evaluate durability (i.e., carbonation resistance) of civil engineering materials.[54] Nevertheless, these two definitions share many similarities, and both fall within the scope of carbonation mineralization in this review.

The quantitative analysis of the top 10 keywords sorted by average normalized citations is listed in Table 1. The top 3 keywords are "cement industry", "waste", and "life-cycle assessment". It suggests that the carbonation of cement-based materials, waste, and environmental impact assessment in MC-CEM have received considerable attention from researchers.

Table 1. Quantitative Analysis of the Top 10 Keywords Sorted by Average Normalized Citations in MC-CEM Research

keywords	occurrences	avg. citations	avg. normalized citations	avg. pub. year
cement industry	29	51	3.36	2019
waste	48	71	2.45	2020
life-cycle assessment	76	55	1.73	2020
CO_2 emissions	43	61	1.61	2020
sequestration	131	36	1.55	2020
carbon dioxide	30	57	1.47	2018
sustainability	36	25	1.44	2022
accelerated carbonation	134	32	1.41	2021
steel slag	59	39	1.38	2020
carbonation curing	26	21	1.30	2021

2.3. Analysis of Journal Sources.

Journal source visualization, as a fundamental bibliometric method,[55] provides the academic community with a systematic knowledge landscape and research orientation, enabling intuitive identification of core journals, their knowledge dissemination pathways and academic influence centers.[56] Analyzing journal sources also enables readers to obtain the optimal information more efficiently and ascertain which journal their work may be best suited for publication. Figure 2c illustrates the visualization of mainstream journals that published MC-CEM. The node size reflects the number of published articles, and the connecting lines represent mutual citations. A minimum threshold of 5 journal publications and a minimum citation count of 20 was set, resulting in a total of 35 journals that met the specified criteria. The top 3 journals in MC-CEM research are *Constr. Build. Mater.* (119 articles), *J. Cleaner Prod.* (81 articles), and *Cem. Concr. Res.* (74 articles). Moreover, these three journals belong to the same green cluster (engineering materials), meaning they have a higher degree of interdependence and a higher frequency of citation among their publications.[57]

Table 2 presents a quantitative analysis of the top 15 journals based on average normalized citations. To identify the most suitable journals in MC-CEM, the top 15 journals were analyzed using the fuzzy comprehensive evaluation method.[21] The factor set U concludes that the influencing factors are U1, normalized average citation U2, publication number of mineralization topics U3, and other factors U4: publication quality, journal reputation, open access, etc. The weight vectors are 0.3, 0.3, 0.2, and 0.2 for U1, U2, U3, and U4, respectively. A fuzzy evaluation matrix was established and the final results (appraisal value) are shown in Table 2. *Cem. Concr. Res.* has the highest average normalized citation (3.28), far exceeding other journals. *Cem. Concr. Res.*, *J. Cleaner Prod.*, and *Renewable Sustainable Energy Rev.* reveal the top 3 appraisal values, indicating they are very suitable for readers to obtain important information in MC-CEM research.[21]

2.4. Analysis of Article Regions.

Geographic analysis facilitates understanding the spatial distribution and dissemination of scientific knowledge.[58] By examining research output, collaboration networks, and knowledge flows from a geographic perspective, it is possible to uncover the contributions and influence of different regions in scientific

Table 2. Quantitative Analysis of Mainstream Journals in MC-CEM Research

journal	number of articles	IF	avg. citations	avg. normalized citations	avg. pub. year	appraisal value
Cem. Concr. Res.	13	10.9	126	3.28	2020	0.15
Renewable Sustainable Energy Rev.	18	16.3	55	2.55	2020	0.10
Sci. Total Environ.	16	8.2	43	2.13	2022	0.08
Appl. Energy	20	10.1	78	1.86	2018	0.06
Resour. Conserv. Recycl.	14	11.2	37	1.65	2021	0.06
Cem. Concr. Compos.	24	10.8	34	1.63	2020	0.08
Fuel	9	6.7	34	1.55	2019	0.04
J. Cleaner Prod.	81	9.7	59	1.50	2019	0.13
J. CO_2 Util.	28	7.2	33	1.43	2021	0.05
Chem. Eng. J.	14	13.3	14	1.31	2021	0.08
Environ. Sci. Technol.	20	10.8	73	1.20	2017	0.07
J. Sustainable Cem.-Based Mater.	7	4.7	5	1.07	2023	0.01
Energy Policy	5	9.3	96	0.93	2009	0.04
Sep. Purif. Technol.	6	8.1	9	0.91	2022	0.03
Constr. Build. Mater.	119	7.4	23	0.90	2021	0.03

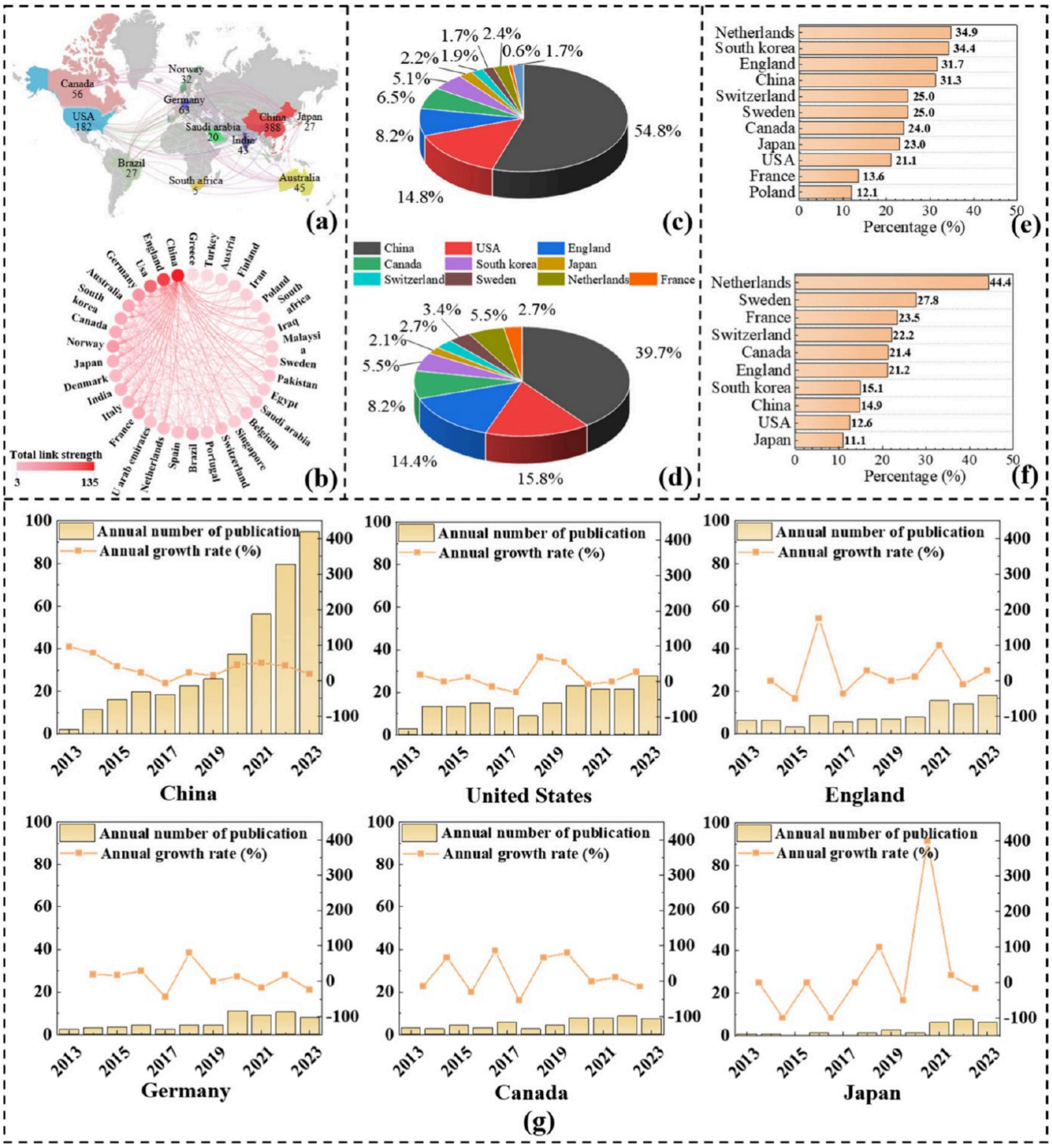


Figure 3. (a and b) International collaboration and cooperation intensity among the mainstream countries in MC-CEM research. Proportion of publications of each country to the total number of publications in the journal: (c) top 15 journals; (d) top 5 journals. Proportion of top 15 or top 5 journals publications to the total number of publications in countries: (e) top 15 journals; (f) top 5 journals. (g) Number of annual publications in various countries.

Table 3. Quantitative Analysis of Article Regions in MC-CEM Research

country	number of articles	total citations	normalized citations	avg. citations	avg. normalized citations	avg. pub. year
Switzerland	18	1744	35	97	1.92	2020
England	99	5287	162	53	1.63	2019
Japan	27	829	42	31	1.57	2021
Sweden	18	885	28	49	1.55	2016
France	17	1167	26	69	1.55	2019
Canada	56	2859	84	51	1.49	2019
Portugal	22	1064	28	48	1.28	2021
South Korea	53	2102	68	40	1.28	2019
Malaysia	16	1223	20	76	1.25	2019
USA	182	8040	227	44	1.25	2018

endeavors, providing a basis for formulating strategies for international cooperation. Parts a and b of Figure 3 show the international collaboration and cooperation intensity among the mainstream countries in MC-CEM research. China leads in publications with 388 papers and maintains the closest collaborative ties with other countries, underscoring its significant role in MC-CEM research. However, the average normalized citations of China (1.05) are not ranked in the top 10 (Table 3).

Besides, the United States and the United Kingdom ranked second (182 papers) and third (99 papers) in article number, respectively, with average citations also reaching the top 10 (10th and 2nd in Table 3, respectively). Moreover, the close ties among countries demonstrate that mineral carbonation has become a global effort to mitigate greenhouse gas effects.

Parts c and d of Figure 3 show the contributions of different countries to the top 15 and top 5 journals in MC-CEM research. Overall, China, the United States, the United Kingdom, and Canada contribute the most, accounting for over 70% of the total. Meanwhile, when the range changes from the top 15 journals (Figure 3c) to the top 5 journals (Figure 3d), China shows the most significant decrease, while the United States, the United Kingdom, and Canada demonstrate varying degrees of improvement.

Parts e and f of Figure 3 show the proportion of publications in the top 15 (Figure 3e) and top 5 journals (Figure 3f) to the total number of publications in each country. The Netherlands reveals the highest proportion of publications in the top 15 and 5 journals. Moreover, when the range changes from the top 15 journals to the top 5 journals, the proportion of publications in The Netherlands increases, indicating that The Netherlands contributes high-quality and widely recognized papers. Similar situations are seen in Sweden and France.

Figure 3g shows the yearly publication number of six typical countries in MC-CEM research. China has the highest number of publications, which has steadily increased in recent years, due to China's dual carbon strategy proposed in 2020 and considerable investment in scientific research funds. The European Union, the United States, and Japan have already achieved carbon peaks in 1990, 2007, and 2013, respectively, and have successfully proposed achieving carbon neutrality by 2050. Therefore, these countries' average yearly publication growth rate is lower.[59,60]

2.5. Analysis of Article Citations. Co-citation analysis measures the similarity between the literature by analyzing the number of times two or more articles are cocited, which reflects the knowledge connections between publications.[61,62] Identifying these classic works through citation analysis is essential, as they reflect key developments in the field.[63] Citation studies help identify major scientific breakthroughs, track emerging or trending topics, and pinpoint core knowledge markers.

To study the most influential publications in MC-CEM research, the citation analysis on 38 highly cited articles (>150 times citation) was conducted.[64] Figure 4 shows the citation co-occurrence network, and the top 10 articles are summarized in Table 4. Figure 4 and Table 4 show that Scrivener et al.'s article[65] has the highest number of citations and the most vital connections. This article summarizes potential economically feasible solutions for reducing CO_2 in the cement-based materials industry. Among them, the scheme of carbonation-cured concrete was mentioned, and the special clinker used for carbonation has a lower cost and smaller CO_2 emissions than ordinary cement clinker. Besides, Pan et al.[2,66] studied the mineralization carbonation of waste, which was consistent with the visual keywords. Other highly cited research focuses on ecological cement and CO_2-cured cement-based composite materials.

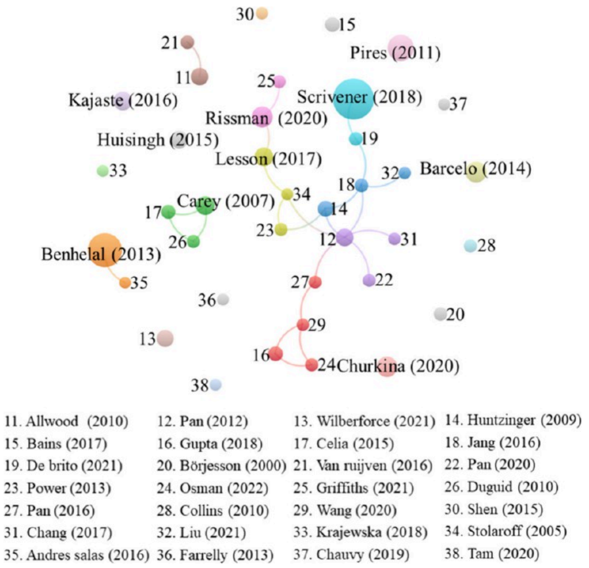


Figure 4. Analysis of coauthorship in MC-CEM research.

3. RESEARCH PROGRESS ON MINERAL CARBONATION OF CIVIL ENGINEERING MATERIALS

MC-CEM is a promising method to alleviate the greenhouse effect, including the carbonation of cementitious material (Section 3.1), industrial solid waste (Section 3.2), construction solid waste (Section 3.3), mine tailings (Section 3.4), carbon mix (Section 3.5), and other materials (Section 3.6). Moreover, it should be noted that carbon sequestration of biochar, asphalt, plastic, and wood is not illustrated in this work due to its mechanistic divergence from the defined mineral carbonation.

3.1. Cementitious Material. *3.1.1. Portland Cement System.* In the CO_2-cured Portland cement-based system, CO_2 reactions with calcium silicate (eqs 1 and 2)[16] and CSH (eq 3).[74] The carbonation of adequately moistened C_2S and C_3S occurs within a few minutes to hours, and the rapid generation of C−S−H accounts for the early strength gain. Meanwhile, the paste matrix comprises C_2S, C_3S, C−S−H, $CaCO_3$, and $Ca(OH)_2$. The carbonation products in different systems are summarized in Table S3. Except for calcite, a gel-like amorphous phase CSH with a lower CaO/SiO_2 ratio than ordinary C−S−H was reported in the CO_2-cured calcium silicates system. Several studies refer to this gel phase as decalcified C−S−H or polymerized silica gel,[75] exhibiting morphological similarities to C−S−H obtained from C_3S hydration.[76]

$$3CaO\cdot SiO_2 + (3-x)CO_2 + yH_2O \rightarrow xCaO\cdot SiO_2\cdot yH_2O + (3-x)CaCO_3 \quad (1)$$

$$2CaO\cdot SiO_2 + (2-x)CO_2 + yH_2O \rightarrow xCaO\cdot SiO_2\cdot yH_2O + (2-x)CaCO_3 \quad (2)$$

$$C{-}S{-}H + CO_2 \rightarrow CaCO_3 + SiO_2 + H_2O \quad (3)$$

The accelerated development of the strength of CO_2-cured OPC-based materials has been extensively reported.[25,77−80] As shown in Figure 5a, due to microstructure densification, the early strength of carbonated cement paste/mortar/concrete is

Table 4. Top 10 Articles Sorted by Normalized Citations

article	title	total citations	normalized citations
Scrivener et al. (2018)[65]	ecoefficient cements: potential economically viable solutions for a low-CO_2 cement-based materials industry	1164	16.7
Rissman et al. (2020)[67]	technologies and policies to decarbonize global industry: review and assessment of mitigation drivers through 2070	394	9.9
Churkina et al. (2020)[68]	buildings as a global carbon sink	379	9.5
Benhelal et al. (2013)[69]	global strategies and potentials to curb CO_2 emissions in the cement industry	872	9.1
de Brito et al. (2021)[70]	past and future of sustainable concrete: a critical review and new strategies on cement-based materials	194	7.3
Liu et al. (2021)[71]	fundamental understanding of carbonation curing and durability of carbonation-cured cement-based composites: a review	160	6.0
Kajaste et al. (2016)[72]	cement industry greenhouse gas emissions—management options and abatement cost	317	5.2
Huisingh et al. (2015)[73]	recent advances in carbon emissions reduction: policies, technologies, monitoring, assessment, and modeling	290	4.9
Pan et al. (2020)[2]	mineralization and utilization by alkaline solid wastes for potential carbon reduction	182	4.6
Pan et al. (2012)[66]	CO_2 capture by accelerated carbonation of alkaline wastes: a review on its principles and applications	288	4.2

significantly higher, with 1-d strength under CO_2 curing ~55% of 28-d strength under water curing. However, as further hydration is hindered by the high carbonation degree, the strength increase rate in the later stage decreases under CO_2 curing. The late strength of specimens with/without CO_2 curing is close to each other. Specifically, the difference in 28-d strength with/without CO_2 curing is <10%.[22−24] Moreover, CO_2-cured mortar exhibited a 180-d strength slightly higher (~5%) than wet-cured mortar.[79] Current research on CO_2-cured concrete faces persistent challenges in scalability and cost-effectiveness.[80] High expenses arise from CO_2 capture, purification, and energy-intensive curing processes, while industrial adoption requires specialized pressurized reactors for flue gas adaptation. Additionally, carbonated OPC-based concrete exhibits lower pH levels compared to its uncarbonated counterpart, potentially increasing the risk of steel corrosion.[81]

3.1.2. Magnesium-Based Cement System. Magnesium-based cement has been attracting increasing attention in the last couple of years.[82,83] Common magnesium-based cement includes reactive magnesium oxide cement (RMC), magnesium oxychloride cement (MOC), magnesium phosphate cement (MPC), and magnesium sulfate cement (MOS).[10,84,85]

3.1.2.1. Reactive Magnesium Oxide Cements (RMC). RMC was first proposed in 2001,[17] with MgO typically >90%. It exhibits some advantages over OPC. (1) Lower calcination temperature:[82,83,86] 700−1000 °C vs 1450−1550 °C of OPC. (2) Significant strength improvement after carbonation,[87] so it is promising for applications such as porous masonry units and precast concrete products. (3) Higher theoretical CO_2 sequestration rate.[18,88] Besides, magnesium-based cement is also favored for applications more compatible with lower pH environments (nuclear waste isolation, concrete containing bacteria or pH-sensitive fibers).[10] The carbon products in the CO_2-cured RMC system are hydrated magnesium carbonates (HMCs), including nesquehonite ($MgCO_3 \cdot 3H_2O$), hydromagnesite ($4MgCO_3 \cdot Mg(OH)_2 \cdot 4H_2O$), dypingite ($4MgCO_3 \cdot Mg(OH)_2 \cdot 5H_2O$), and artinite ($MgCO_3 \cdot Mg(OH)_2 \cdot 3H_2O$).[10,89−91] As shown in Figure 5b, the mortar with 100% RMC demonstrates a much higher CO_2 sequestration rate than OPC-based mortar (46%[89] vs 21%[90]).

Furthermore, the factors that affect the strength and carbon sequestration rates of CO_2-cured RMC-based materials include CO_2 concentration and pressure, relative humidity, curing temperature and time, water-cement ratio, and additives (hydration agents, carbonation agents, and nucleating agents).[10,88,92,93] A higher carbonation degree means denser carbon products (HMCs), leading to decreased porosity and increased strength of CO_2-cured RMC-based materials. Generally speaking, a higher CO_2 concentration/pressure and water cement ratio, as well as appropriate relative humidity (e.g., ~78% RH for w/c of 0.5−0.7[94]), are beneficial for improving the carbonation degree within a specific time (e.g., within 1−3 d). Specifically, within an appropriate range, higher RH promotes the carbonation reaction and microstructure densification.[94] Yet, excessive humidity is not conducive to enhancing the carbonation degree and strength because CO_2 transport is slower in water than in air.[10] Additionally, a long carbonation time also leads to degraded strength due to excessive HMCs formation.[95] For instance, the compressive strength of RMC-based material decreased by ~11.1% at 28 d than at 14 d under 20% CO_2 curing.[95]

Nonetheless, RMC illustrates some disadvantages than OPC system. (1) Higher cost in preparation process,[96,97] derived from lower-temperature magnesite calcination but requiring high purity. (2) Lower pH (10.3−11.2[98] vs OPC: 12.4−13.5[99,100]) causes the steel bars to easily depassivated. (3) Higher carbon emissions during production than limestone calcination (1.1 vs 0.78−0.83 t/t[101]). Nevertheless, considering the higher carbon storage capacity of RMC, its net carbon emissions are much lower than OPC[101] (see Section 3.7 for details). Except for RMC, MOC,[10] MOS,[102] and MPC[103] also exhibit higher CO_2 emissions than OPC cement, due to the production of MgO and other necessary materials. However, some approaches can help to reduce the CO_2 emissions of these cements, for instance, using MgO derived via wet-process routes in MOC production[10] or incorporating tundish deskulling waste in MPC formulations.[103] Furthermore, when carbon storage capacity is taken into account, RMC probably demonstrates the lowest net carbon emissions among all magnesium-based cement systems, due to its high carbon sequestration efficiency.[102]

3.1.2.2. Magnesium Oxychloride Cement (MOC). MOC is formed through the reaction of MgO with $MgCl_2$ solution,[104] and its hydration phase development at room temperature is illustrated by eqs 4 and 5.[105] Figure 5c illustrates the

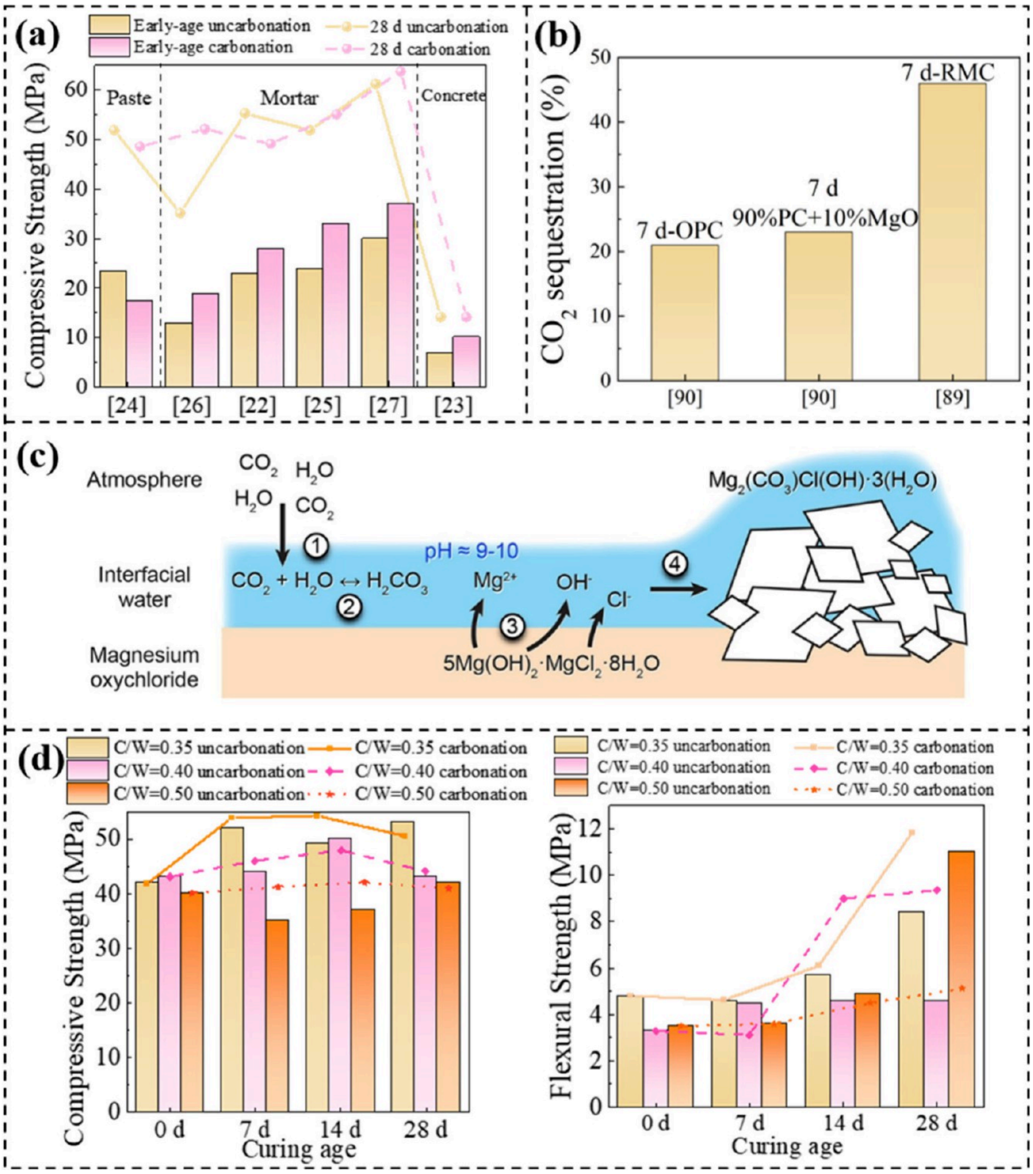


Figure 5. (a) Compressive strength of concrete with/without carbonation (data summarized from refs 22–27). (b) Carbon sequestration rate of the OPC system,[90] 10% RMC system,[90] and 100% RMC system.[89] (c) Schematic diagram of the MOC carbonation mechanism. Reprinted with permission from ref 104. Copyright 2017 Elsevier. (d) Influence of carbonation on the mechanical properties of MOS-based paste. Adapted with permission from ref 20. Copyright 2019 Elsevier.

carbonation mechanism of MOC.[104] Due to the formation of a dense microstructure and low degradation degree of carbonation products,[19] the strength, toughness and water resistance of CO_2-cured MOC-based materials are improved. It was reported that ∼20−40% of the emissions during the MOC board manufacturing process will be reabsorbed and stored within 15 years of service, which is consistent with the potential carbon offset achieved through carbonation of OPC.[106]

$$5MgO + MgCl_2 + 13H_2O = 5Mg(OH)_2\cdot MgCl_2\cdot 8H_2O \tag{4}$$

$$3MgO + MgCl_2 + 11H_2O = 3Mg(OH)_2\cdot MgCl_2.8H_2O \tag{5}$$

3.1.2.3. Magnesium Sulfate Cement (MOS). Magnesium sulfate cement (MOS) is an air-dried magnesium cement prepared by mixing lightly burned MgO and magnesium sulfate aqueous solution.[107,108] It has the advantages of being lightweight and having low energy consumption, good adhesion, and excellent insulation performance. Compared with MOC, MOS does not absorb moisture and has no Cl^-, so steel bars do not easily depassivate.[109]

Figure 5d shows the MOS-based paste's compressive and flexural strength after carbonation. Overall, CO_2 curing improves the strength of MOS-based paste by 11% at 28 d, and a small w/c (0.35) is beneficial for strength regardless of the curing regime and curing time. However, the compressive strength of MOS-based materials first increases and then decreases with increased CO_2 curing time.[20] The specimens exhibit the largest strength at 7 or 14 d rather than 28 d. This is consistent with the phenomenon in CO_2-cured RMC systems that a suitable CO_2 curing time is vital.[95]

3.1.2.4. Magnesium Phosphate Cement (MPC). MPC is based on the acid−base reaction between dead-burnt MgO

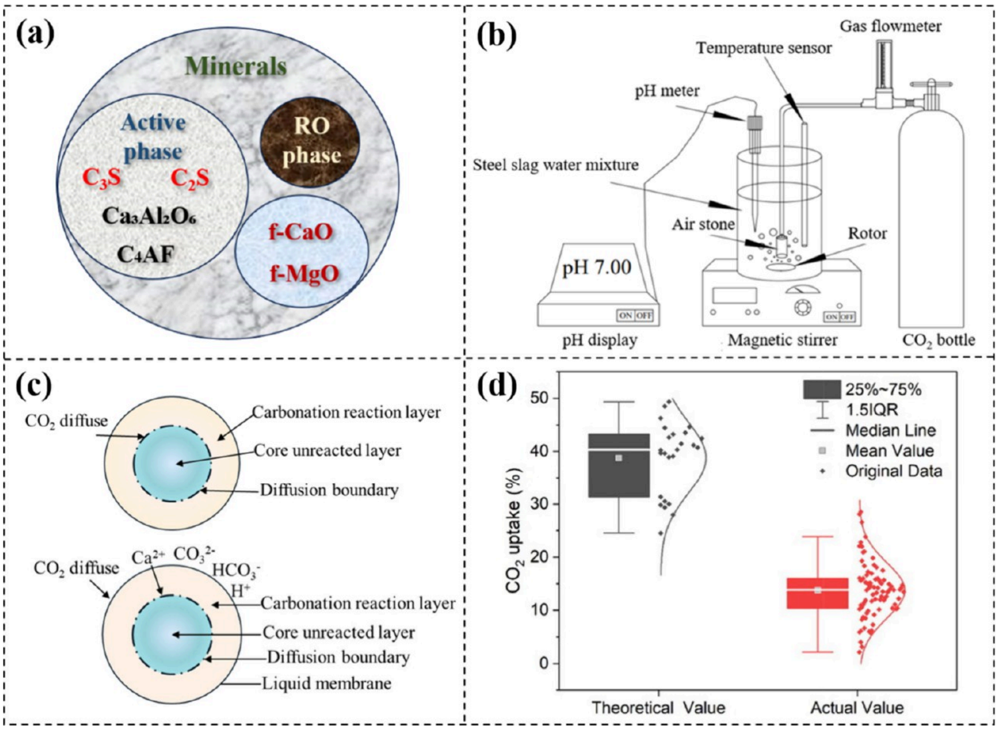


Figure 6. (a) Mineral composition of SS, (b) device used in wet carbonation of SS, (c) SS carbonation reaction model, and (d) summary of CO_2 uptake of SS. Adapted with permission from ref 124. Copyright 2022 Taylor & Francis.

and a soluble acid phosphate (e.g., ammonium and potassium phosphate) to form magnesium phosphate salts ($NH_4MgPO_4\cdot 6H_2O$ or $MgKPO_4\cdot 6H_2O$).[10] It is widely known as fast-hardening cement and is mainly used to repair roads and concrete structures rapidly.[110,111] Its hydration process is similar to MOC, and many dead burned MgO particles may be unreacted. The carbonation products of CO_2-cured MPC systems are the same as those of CO_2-cured RMC systems, e.g., nesquehonite, hydromagnesite and dypingite.[112] As carbonation products fill pores, the porosity decreases, forming a denser cement matrix. Consequently, the CO_2-cured MPC-based concrete exhibits a 3 d/28 d compressive strength of 113%/33% higher than air-cured MPC-based concrete, respectively.[112] However, the wide application of MPC is limited due to several reasons.[10] (1) MPC containing potassium phosphate demonstrates strength loss when exposed to water. (2) MPC incorporated with ammonium phosphate has the issue of releasing ammonia gas during the exothermic hydration process. (3) Phosphate resources are scarce and are in high demand as agricultural fertilizers.

As indicated above, MOC,[106] MOS,[20] and MPC[112] demonstrate lower CO_2 storage capacity than RMC[89] due to their smaller content of carbonizable $Mg(OH)_2$ and denser microstructure at early age, with details illustrated in Section 3.7. Regarding the challenges in engineering applications of magnesium-based cement systems, the uncarbonated/carbonated magnesium-based cement systems have a low pH environment,[98] affecting their use in steel-reinforced structural applications.[10] Their durability, corrosion resistance and volume stability need to be thoroughly investigated to facilitate wider applications. Meanwhile, accelerated carbonation demands concentrated CO_2, incurring expenses for gas sourcing, storage, and energy-intensive systems.[113]

3.2. Industrial Solid Waste.

Industrial production brings a large amount of industrial solid waste containing Ca and Mg, which could be used for mineral carbonation. Mineral carbonation of industrial solid waste includes steel slag, red mud, calcium carbide slag, coal gangue, fly ash, gypsum, and ground granulated blast furnace slag.[12] The composition of different solid wastes varies, resulting in their different carbonation mechanisms.[114−116]

3.2.1. Steel Slag.

As shown in Figure 6a, steel slag was composed of f-CaO, f-MgO, RO phase, and active phase (C_2S, C_3S, etc.).[114,115,117] According to different sources, steel slag (SS) can be subdivided into basic oxygen furnace slag (BOFS), electric arc furnace slag (EAFS), ladle furnace slag (LFS), argon oxygen decarburization slag (AODS), and induction furnace slag (IFS).[118] The carbonation method of SS includes direct carbonation and indirect carbonation. The former can be further divided into dry carbonation and wet carbonation[119] (Figure 6b) according to the operating humidity.[4] Under CO_2 curing, the expansion effect of SS can be eliminated by reducing the content of f-CaO and f-MgO (Figure 6c), and the performance of SS can be improved.[120−123] Scholars have conducted much research on the carbonation of SS. The theoretical maximum CO_2 sequestration rate of SS can be calculated according to the chemical composition of SS:

$$\text{max } CO_2 \text{ uptake} = 0.785(CaO - 0.7SO_3) + 1.09Na_2O + 0.93K_2O \quad (6)$$

Regarding the actual CO_2 sequestration rate of SS, there are many methods for evaluating CO_2 sequestration rate of SS, including the weighing[124] and TG analysis methods.[125−127] The weighing method is fast and simple, but its accuracy is insufficient. In comparison, the TG analysis method was widely utilized due to its higher accuracy. Equations 7−10 were used by Moon et al.,[125] Liu et al.,[126] and Li et al.,[127] respectively.

There are slight differences in the numerator and denominator, resulting in limited differences between the results.

$$\mathrm{CO_2\ uptake} = \frac{\Delta m_{\mathrm{CO_2}}}{m_{105^\circ\mathrm{C}}} \times 100 \tag{7}$$

$$\mathrm{CO_2\ uptake} = \frac{\Delta m_{\mathrm{CO_2}}}{m_{900\ ^\circ\mathrm{C}}} \times 100 \tag{8}$$

$$\mathrm{CO_2\ (\%)} = \frac{\Delta m_{\mathrm{CO_2}}}{m_{105\ ^\circ\mathrm{C}}} \times 100 \tag{9}$$

$$\mathrm{CO_2\ uptake\ (\%)} = \frac{\mathrm{CO_2\ carbonated\ (\%)} - \mathrm{CO_2\ initial\ (\%)}}{100 - \mathrm{CO_2\ carbonated\ (\%)}} \times 100 \tag{10}$$

where Δm_{CO_2} is the mass difference caused by CO_2 curing and $m_{105\ °C}$ and $m_{900\ °C}$ is the mass of SS powder at 105 and 900 °C, respectively.

During carbonation of SS, free calcium oxide (f-CaO) and calcium silicate (C_3S and C_2S) in SS react with CO_2.[128−130] Researches show that temperature, carbonation time, CO_2 pressure, particle size, gradation, and water content influence carbonation of SS,[36,131−134] with details illustrated in Section S2. As shown in Figure 6d, SS's actual CO_2 sequestration rate is far lower than the theoretical value.[124] Therefore, scholars attempt to improve the CO_2 sequestration rate of SS by optimizing carbonation parameters,[44] increasing the specific surface area of SS,[135] using additives[136] or developing an improved wet carbonation process.[44]

Regarding engineering applications, Li et al.[137] proposed that carbonation curing of SS is a potential replacement for cement for construction material production, especially suitable in Shandong Province, China. Meanwhile, cross-departmental collaboration and policy economic incentives are key to technology implementation. However, carbonation of SS faces some challenges. The inherent chemical heterogeneity of SS complicates accurate modeling of dissolution kinetics and carbonation efficiency.[138] While accelerated carbonation can achieve a CO_2 storage capacity of more than 150 g of CO_2 per 1 kg of SS under specific conditions, the operational costs and energy consumption during the process negatively impact economic viability and CO_2 captured carbon of SS.[44,139,140]

3.2.2. Red Mud. Red mud is a highly alkaline industrial byproduct from alumina production.[141] Although efforts have been made to use red mud in construction, environmental management, mining, agriculture, and other fields, only 2−3% of red mud has been effectively utilized or processed.[29] Because the red mud contains mineral components with Ca and Mg, the red mud can be carbonated. During this process, the red mud's alkalinity is reduced, offering a pretreatment method for red mud utilization. As shown in Figure 7a, Si et al.[142] established a model to link the parameters with the carbonation of red mud, in which C_{max} is maximum carbon sequestering capacity; C_{act} is actual carbon storage, and C_{pot} is potential additional carbon sequestration capacity. Meanwhile, Zhang et al.[143] found that the essence of red mud carbonation reaction is a liquid−liquid reaction (Figure 7b).

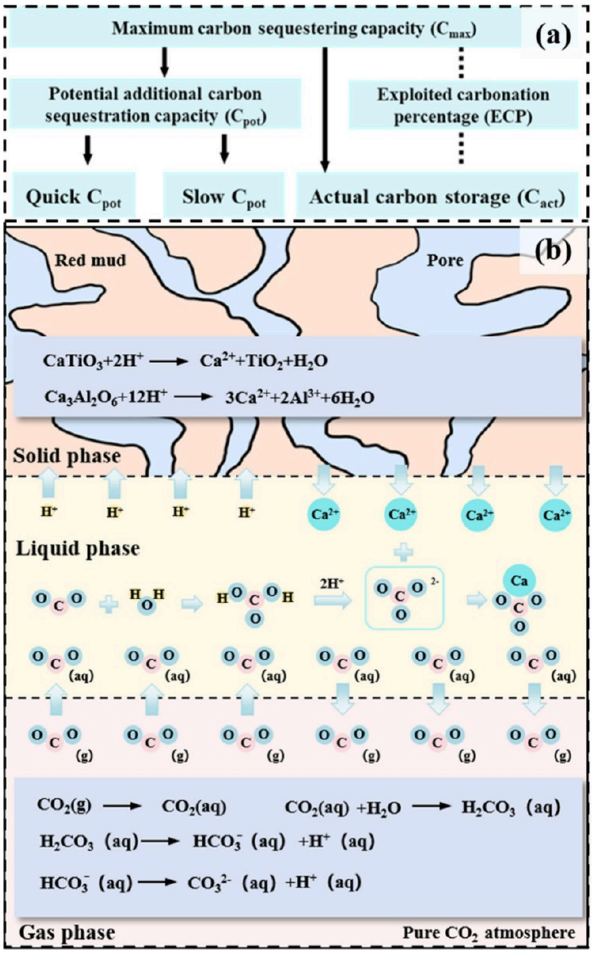


Figure 7. (a) Parameters for evaluating carbon storage of red mud. Adapted with permission from ref 142. Copyright 2013 Elsevier. (b) Carbonation mechanism of red mud. Adapted with permission from ref 143. Copyright 2024 Elsevier.

Table 5. Study on Direct Aqueous Carbonation of Red Mud

parameters	carbonation effect	ref
liquid-to-solid ratio (L/S) = 10 mL/g; reaction time = 5 h	CO_2 uptake = 70.2 g/kg; pH down to 8.45; alkalinity down to 178 mg/L	38
pressure = 3.5 bar; reaction time = 3.5 h; L/S = 0.3 kg/kg	CO_2 uptake = 53.0 g/kg	39
L/S = 10 kg/kg; temperature = 20 °C; CO_2 concentration = 15.00%	CO_2 uptake = 41.5 g/kg	146
L/S = 10 kg/kg; temperature = 25 °C; reaction time = 3 h	CO_2 absorption = 15.3%	147
L/S = 1.5 mL/g; pressure = 16 bar, reaction time = 2.5 h; temperature = 30 °C	pH down to 6.4; CO_2 saturation weight gain rate = 3.07%	148
L/S = 10:1 kg/kg; temperature = 30 °C; reaction time = 1 h; CO_2 concentration = 99.9%	number of CO_2 molecules, H_2CO_3 molecules, HCO_3, CO_3^{2-}, and H^+ in the solution increases	143

As shown in Table 5, scholars have carried out some research on the carbonation of red mud. Overall, the effect of direct carbonation of red mud is not ideal, so scholars proposed a joint carbonation method (calcium substances are introduced) to improve the carbonation degree of red mud. Li et al.[144] proposed the calcification carbonation method, in which lime and heated red mud are used to obtain a new red mud system with the main components of calcium silicate and $CaCO_3$. Zhang et al.[145] used CaO to prepare red mud-based cementitious materials and found that CaO activated gibbsite, diaspore, and silica components in red mud. In addition, the

Table 6. Study on Carbonation of Coal Gangue

modification conditions	carbonation conditions	carbonation effect	ref
calcined at 600−1100 °C for 2 h, added to a 15−35% NaOH solution, and then reacted with $Ca(OH)_2$ to obtain the porous adsorbent material	20 °C; atmospheric pressure; CO_2 flow rate = 20 mL/min	36.69 mg/g	157
Mg_x-Cu_{1-x}-SiO_3-type adsorbent prepared by the hydrothermal method	25 °C; 10^2 kPa	16.73 cm^3/g	158
mesoporous silica adsorbent material (M-SiO_2) was prepared	atmospheric pressure; 25 °C; CO_2 flow rate = 30 mL/min	83.5 mg/g	159
porous adsorbent materials were prepared using gangue ash, NaOH, cetyltrimethylammonium bromide and polyethyleneimine	atmospheric pressure; 30 °C; carbonation for 30 min	0.125−1.742 mmol/g	160
four new gangue silicate nanosorbent materials—$MgSiO_3$, $MnSiO_3$, $CuSiO_3$, and $ZrSiO_4$—were prepared using the hydrothermal method	140 °C; pH = 10; carbonation for 10 h	7.82−17.93 cm^3/g	161

carbonation of CaO in red mud made red mud particles bond, improving the compressive strength.

It should be noted that the age of RM and sampling details (ambient weather conditions and depth of sampling) also influence mineralization of RM, but these parameters are usually ignored and unreported.[149] Moreover, carbonation of red mud faces scientific and technoeconomic barriers. The reported CO_2 storage capacity remains low (<50 g/kg[146−148]), hampered by complex composition and poor reactivity of RM. Additionally, to achieve economic viability, assessing smelter emission cost-effectiveness by location and maximizing refinery flue gas use is necessary. The initial investment costs, operational expenses, revenue from byproducts, potential carbon credits, and scalability benefits need to be evaluated to assess engineering feasibility.[149]

3.2.3. Carbide Slag. Carbide slag (CS) is a byproduct of the hydrolysis of calcium carbide during acetylene production, with the primary chemical component of $Ca(OH)_2$. The smelting and volatilization processes involved in calcium carbide production result in a lower content of other components (e.g., CaO, SiO_2, $A1_2O_3$, Fe_2O_3, etc.).[150,151] As an alkaline solid waste, the mechanism of accelerating carbonation of carbide slag is shown in Figure S2. The following mechanisms control the overall rate and degree of carbonation reaction: (1) the rate at which Ca^{2+} is transported to the reaction site; (2) CO_2 diffusion rate; (3) dissolution rate of $Ca(OH)_2$; (4) pore clogging; (5) the product precipitates and covers the surface.[152]

Table S4 lists the study of carbide slag carbonation under different conditions. Yang et al.[153] used carbide slag for CO_2 capture by high-temperature calcium ring technology and found that the CO_2 conversion rate of carbide slag reached 18.13% at ambient temperatures. In the work of Liu et al.,[154] fly ash was added to carbide slag and carbonated in a fixed-bed reactor. The carbonation efficiency reaches 55% under 15% flue gas, 550 °C temperature, and 6 kg/h water injection. Altiner[155] raised the reaction temperature in the stirred tank and added sodium oleate, and the carbonation efficiency of carbide slag reached 94.65% under optimal conditions. Nonetheless, there are few studies on CO_2 capture by carbide slag in high-pressure reactors nowadays, and the effects of pressure and liquid−solid ratio on the reaction rate in the carbonation reaction process are unclear.

3.2.4. Coal Gangue. Coal gangue is a coassociated mineral of coal, produced in coal mining and washing. 0.15−0.2 t of coal gangue is produced per 1 t raw coal.[28] The chemical composition of coal gangue is diverse, including Al, Si, C, etc. The main chemical components of coal gangue include SiO_2 (40−70%), Al_2O_3 (13−40%), Fe_2O_3, CaO, MgO, TiO_2, K_2O, etc., with the total amount of SiO_2 and Al_2O_3 reaching 60−90%.[156] Kaolinite and quartz are commonly found in coal gangue. Other possible crystalline minerals include illite, chlorite, muscovite, feldspar, pyrite, siderite, hematite, calcite, etc. Coal gangue also contains a certain amount of amorphous substances, mainly moisture, carbonaceous, and weathered substances, etc.

The study on carbonation of coal gangue[157−161] is summarized in Table 6. Although progress has been made in carbonation of coal gangue, several issues remain to be solved: (1) Coal gangue has low coal quality and a large content of ash and impurities, resulting in low carbonation degree and high energy consumption and treatment costs. (2) Due to the coal gangue's varied composition and properties, the carbonation products' quality and performance are unstable, limiting their industrial applications. (3) Large amounts of water/gas/solid waste are generated during carbonation.

3.2.5. Fly Ash. Fly ash is widely used in producing civil engineering materials, mine landfills, and agricultural activities, with a utilization rate of <60%.[162] Nowadays, fly ash has attracted much attention as a mineral carbonation material with huge output.[163,164] After carbonation, the stability of fly ash is increased, and its application in civil engineering can be expanded.[165] The carbonation of fly ash is categorized into direct and indirect methods. Indirect carbonation is costly, and the presence of carbonate in fly ash reduces the carbonation effectiveness. So, most research focuses on direct carbonation, which can be further divided into gas−solid carbonation and wet carbonation methods, with reaction equations shown in Table S5. Generally, the wet carbonation method is more efficient than the gas−solid method.[166,167]

The carbonation degree of fly ash could be enhanced by optimizing various parameters.[164,166,168] Specifically, Dananjayan et al.[166] found that the best carbonation condition of fly ash is carbonation for 10 min at 30 °C, 3 bar pressure, the liquid−solid ratio of 0.2, and 100% CO_2 concentration, with the carbonation efficiency reaching 67.87%. Ji et al.[168] proposed that the influence of temperature, gas flow rate and solid−liquid ratio on the carbonation efficiency of fly ash is limited. Liu et al.[164] put forward the opposite viewpoint that increasing the temperature, CO_2 content and $H_2O(g)$ content improves the carbonation efficiency of fly ash. However, the effect of CO_2 concentration is less significant than that of temperature and $H_2O(g)$.

However, the optimal parameters for carbonation efficiency vary significantly among different types of fly ash and its CO_2 storage capacity is low (<50 g/kg[164,166,168]). Many mineral phases in fly ash (magnesium, calcium, silicon, iron, etc.) lead to a complex carbonation process, hindering stable efficiency improvements. Furthermore, inadequate characterization of fly ash's overall chemical and mineralogical composition con-

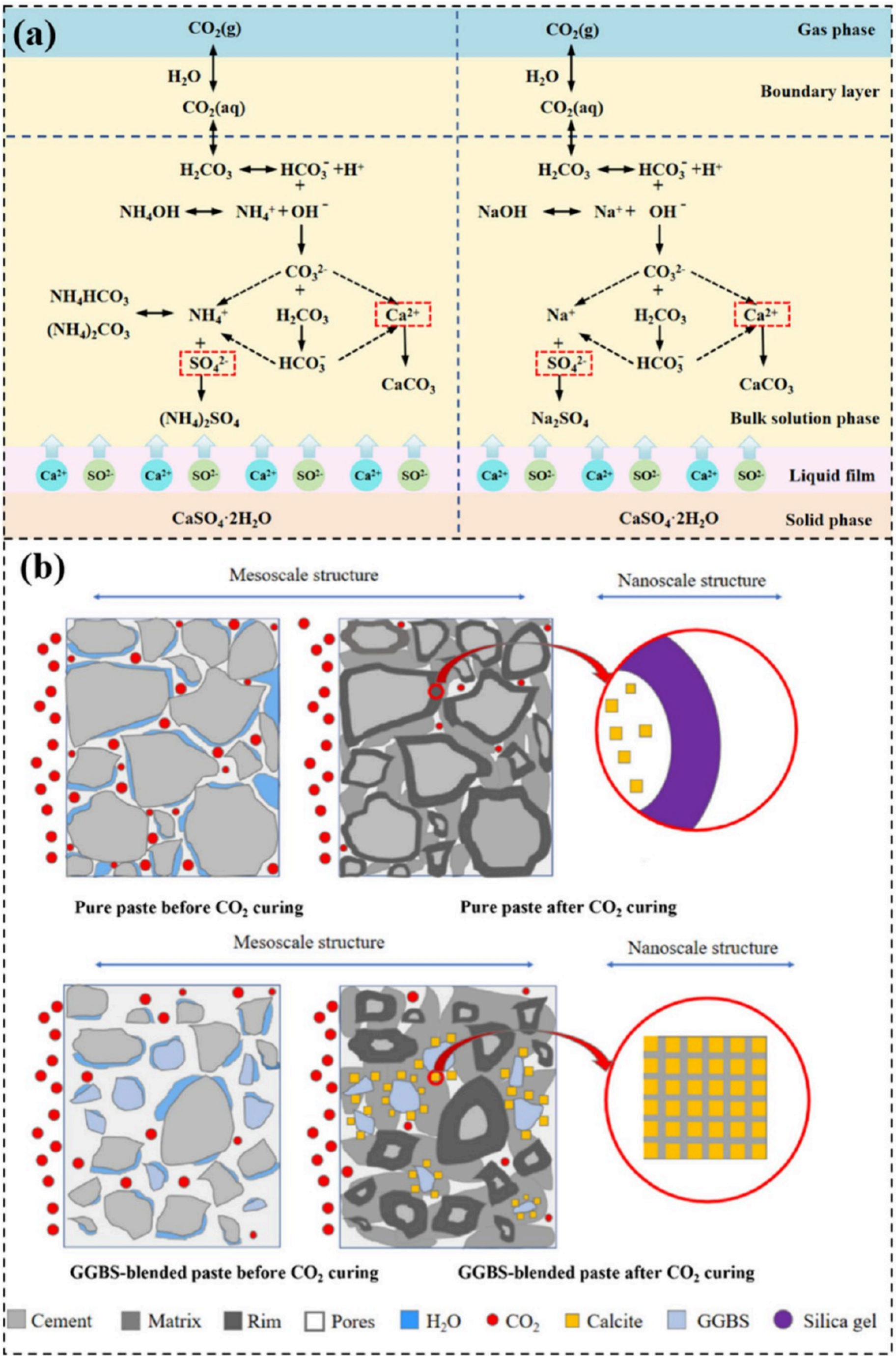


Figure 8. (a) Schematic diagram of the carbonation process of FGDG using NH_4OH and NaOH as basic media. Adapted with permission from ref 175. Copyright 2018 Elsevier. (b) Schematic diagram of the effect of GGBS on the CO_2 curing process. Adapted with permission from ref 176. Copyright 2023 Elsevier.

strains a deeper understanding of the carbonation of effective carbonation strategies.[169]

3.2.6. Gypsum. Byproduct gypsum (BG) includes flue gas desulfurization gypsum (FGDG), phosphogypsum (PG), titanium gypsum (TG), and red gypsum (RG) according to different industrial processes. The main component of BG is $CaSO_4{\cdot}2H_2O$, and its purity depends on the production process. Specifically, the purity of FGDG, PG, and RG is ~95%,[170] 90%,[171] and 75%,[172] respectively. Gypsum exhibits a rapid carbonation rate and high conversion efficiency under moderate conditions during the direct hydration mineral carbonation process.[170,173] Meanwhile, NH_4OH and NaOH are usually used as basic media for mineral carbonation of FGDG. However, the carbonation mechanism varies in different basic media.[170,174]

As shown in Figure 8a, the reactions in FGDG-NH_4OH-CO_2 and FGDG-NaOH-CO_2 systems are

$$CaSO_4{\cdot}2H_2O(s) + CO_2(g) + 2NH_4OH(aq) \rightarrow (NH_4)_2SO_4(aq) + CaCO_3(s) + 3H_2O \quad (11)$$

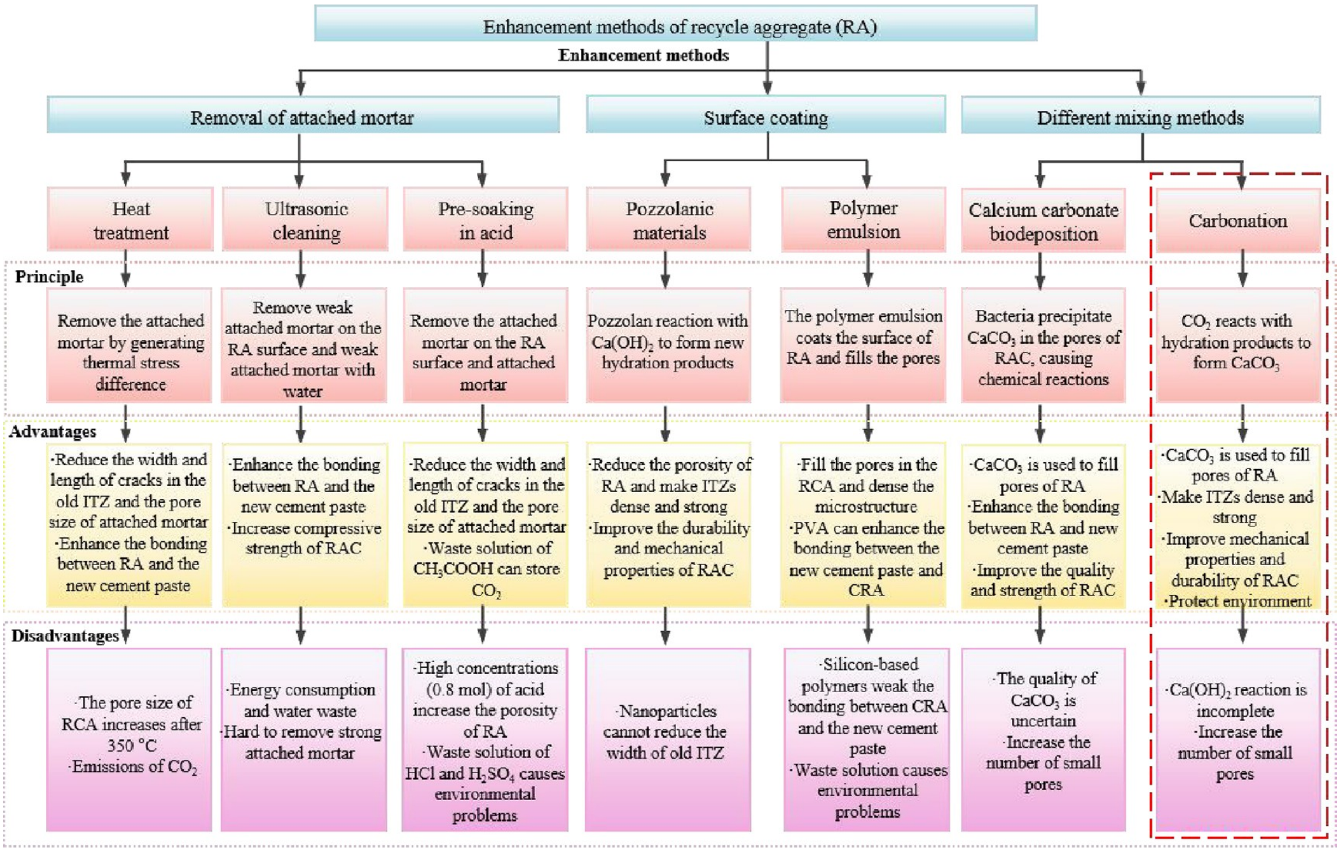


Figure 9. Enhancement methods of RA. Adapted with permission from ref 205. Copyright 2020 Elsevier.

$$CaSO_4 \cdot 2H_2O(s) + CO_2(g) + 2NaOH(aq) \rightarrow Na_2SO_4(aq) + CaCO_3(s) + 3H_2O \quad (12)$$

In the direct carbonation of BG, the product $CaCO_3$ obtained contains few impurities.[170,171] $CaCO_3$ prepared from BG not only achieves the resource utilization of BG but also saves the natural resources of $CaCO_3$. Meanwhile, the crystallization kinetics of $CaCO_3$ and the direct carbonation method for synthesizing adjustable-purity $CaCO_3$ are also hot research topics. Mineral carbonation of FGDG is considered feasible. However, metal impurities may significantly affect the effectiveness of CO_2 conversion, causing potential environmental leaching and influencing the reuse prospects of the carbonate product.[177] Moreover, the reason for the low carbonation degree of RG at room temperature and atmospheric pressure is still unclear. The carbonation process of PG and RG in the presence of other bases (e.g., NaOH) still needs to be studied. Moreover, BG direct aqueous solution carbonation method is the most promising CO_2 storage technology due to its simple process and fast carbonation speed, but the related research is not yet comprehensive.

3.2.7. Ground Granulated Blast Furnace Slag. Ground granulated blast furnace slag (GGBS) could replace OPC in concrete mixtures.[178−180] Adding GGBS influences the further hydration and microstructure of concrete under CO_2 curing, affecting its performance. In the study by Zhang et al.,[181] 25% GGBS was used to replace cement for preparing paste, which was cured under 3% CO_2 concentration, 50% relative humidity and 23 °C temperature for up to 12 weeks. Compared to control group, GGBS increased the carbonation area of the paste, enhancing the carbonation degree from 60.7% to 78.8% in the carbonation zone.

As shown in Figure 8b, in CO_2-cured cement paste without GGBS, the matrix and edges surrounding the cement particles contain $CaCO_3$ and silica-rich gel, mainly distributed in the matrix and edges, respectively. In addition, the $CaCO_3$ crystals are surrounded by an amorphous layer. In contrast, GGBS remains unreacted in CO_2-cured cement paste with GGBS, and small diamond-shaped $CaCO_3$ precipitates on GGBS. Due to the filler effect, adding GGBS increases the carbonation degree and calcite content. However, the loose microstructure of carbonation products and nanopores causes decreased compressive strength of CO_2-cured mortar.[176,182,183]

The carbonation of GGBS faces some challenges. For example, the carbonation efficiency of GGBS is limited by material composition and process conditions.[184] Cost and deployment barriers arise from alkaline activators, customized carbonation equipment, and industrial waste transportation expenses. Moreover, lab-scale accelerated carbonation dominates current studies, while practical issues like CO_2 transport efficiency and long-term durability data gaps impede engineering applications.

3.3. Construction Solid Waste. The recycled material from crushed construction and demolition waste (CDW) can be classified into three forms based on the particle sizes: recycled coarse aggregate (RCA, 5.00−31.5 mm), recycled fine aggregate (RFA, 0.15 mm−5.00 mm) and recycled powder (RP, <0.15 mm).[185,186] RCA and RFA both belong to aggregate, which are illustrated in Section 3.3.1. For RP, it includes recycled concrete powder[187−189] (also termed "recycled concrete fines"[190,191] by some scholars; unified as

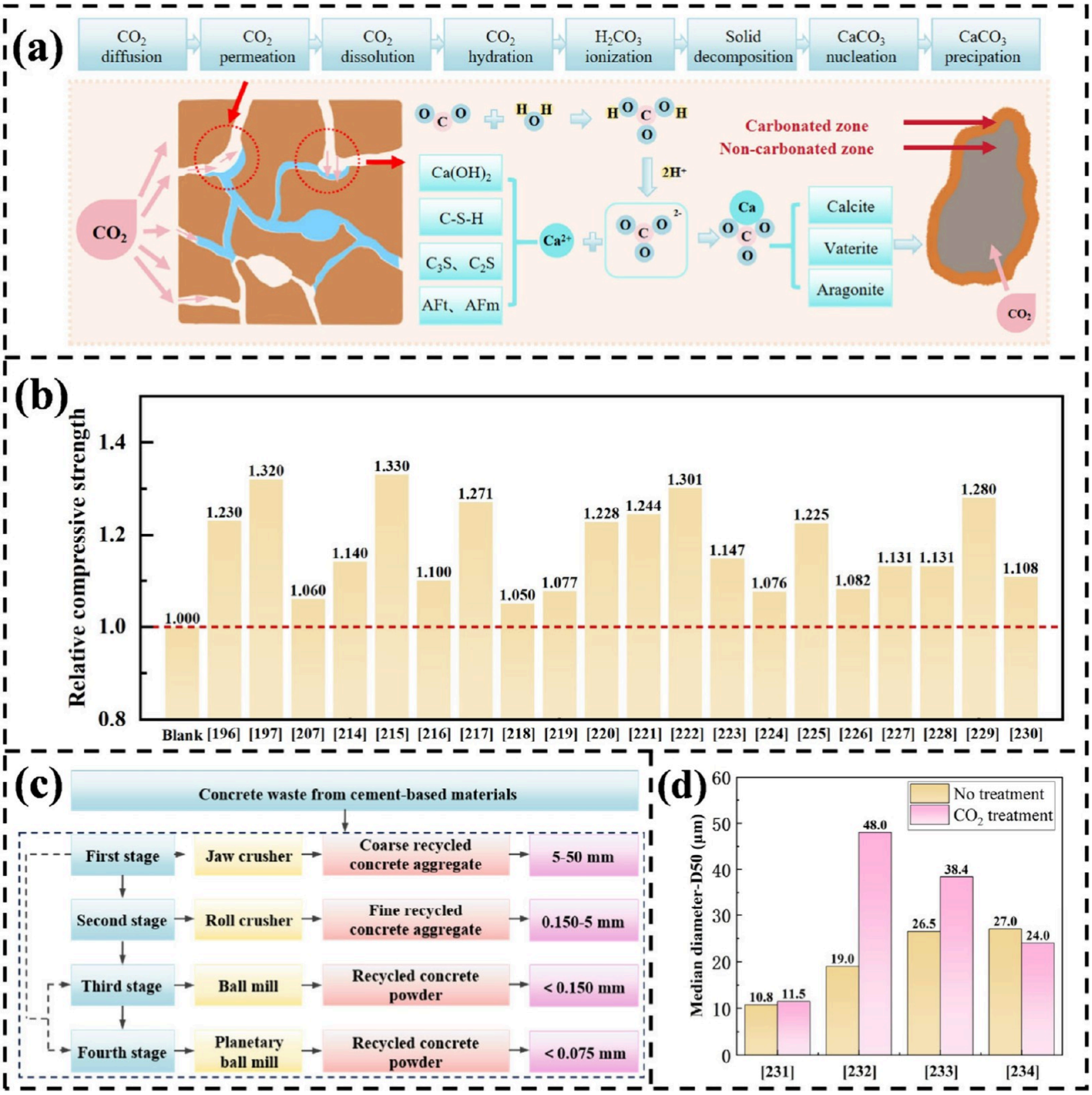


Figure 10. (a) Reaction processes of carbonation-treated RA. Adapted with permission from ref 207. Copyright 2021 Elsevier. (b) Relative compressive strength of the CRAC (data summarized from refs 196, 197, 208, and 214−230). (c) Production process of recycled concrete powder (RCP). (d) D50 of RCP before/after carbonation (data summarized from refs 231−234).

recycled concrete powder in this review), recycled brick powder and recycled glass powder. The widespread use of RA and RP reduces the demand for natural materials, making them more sustainable.[192−195] Meanwhile, they presented the potential for mineral carbonation.[196−199] Moreover, compared to small-sized recycled construction solid materials (RA and RP), large original concrete waste before aggregate recycling process has the disadvantages of inconvenient carbonation testing and difficulty in evaluating effects of carbonation. Hence, relevant literature is scarce, and it is not illustrated in this review.

3.3.1. Recycled Aggregate. In the past two decades, extensive research has been conducted on the performance of RA (including RCA and RFA) and recycled aggregate concrete (RAC). RA has adhesive old mortar and multiple interfacial transition zones (ITZ), so its performance is inferior to natural aggregates.[200,201] Additionally, the workability, mechanical properties, and durability of RAC are worse than those of natural aggregate concrete with the same water−cement ratio.[202,203] Therefore, modification techniques for RA have been proposed to improve its performance in recent years (Figure 9), and the carbonation method is more green and sustainable than other modification technologies.[204]

Compared to RCA, RFA has a smaller particle size and more higher amount of residual cement paste, so it reveals a higher carbonation degree and better modification effect.[185,206] Except for aggregate parameters (particle size and amount of residual cement paste), the curing parameters, including relative humidity, gas pressure, CO_2 concentration, temperature and CO_2 flow rate, are also key factors influencing the carbonation efficiency of RA.[206] Besides, the mechanical properties, elastic modulus, resistance to chloride ion

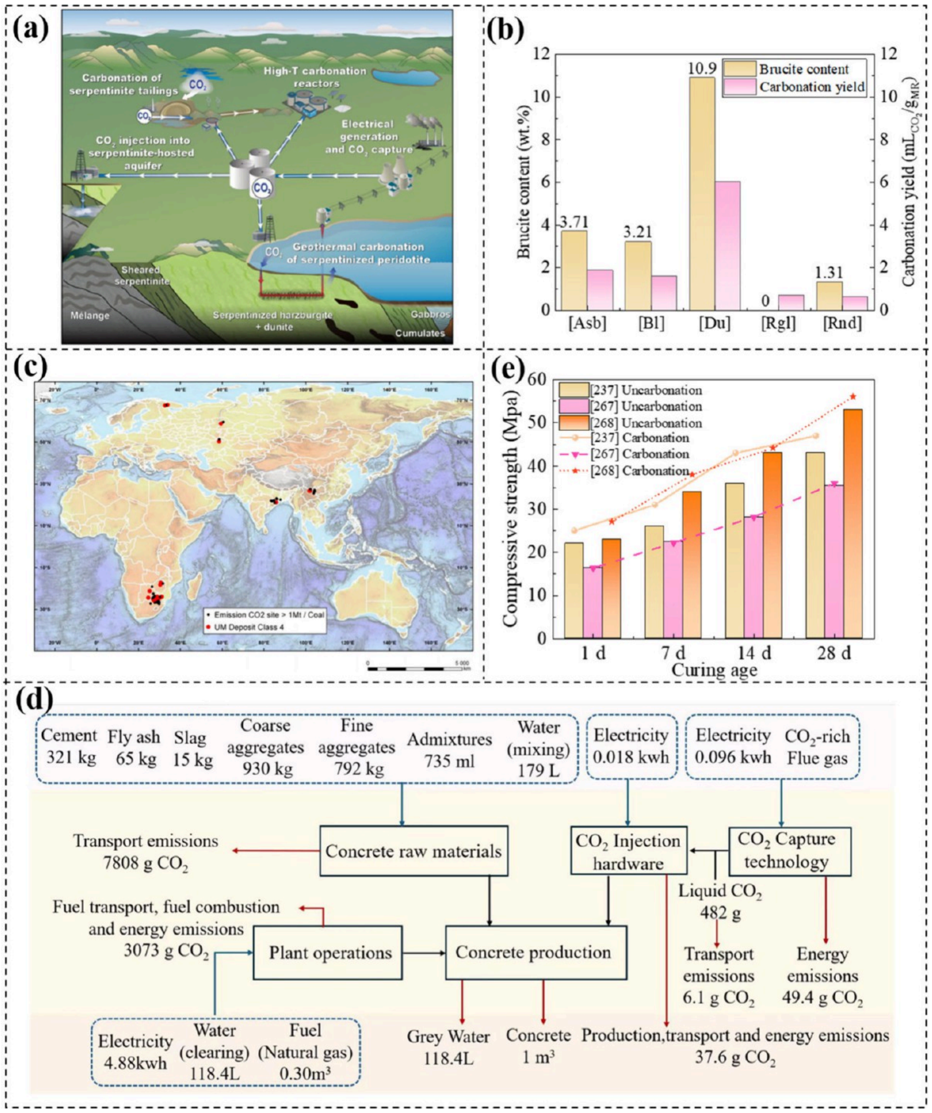


Figure 11. (a) Geoengineering system designed for the carbon capture and storage process through mineral carbonation. Adapted with permission from ref 264. Copyright 2013 mineralogical Society of America. (b) Carbonation yield and brucite content of ultramafic tailings. Adapted with permission from ref 265. Copyright 2014 Elsevier. (c) Map of the location distribution for coal-fired power plants and ultramafic rock deposits. Adapted with permission from ref 266. Copyright 2011 Elsevier. (d) Overall input and output in the process of carbon mix. Adapted with permission from ref 237. Copyright 2016 Elsevier. (e) Influence of carbon mix treatment on the compressive strength of concrete (data summarized from refs 237, 267, and 268).

penetration and frost resistance of CRAC are superior to those of RAC.[207] Specifically, compared to RAC, CRAC has a gas permeability 43.5% smaller[208] and a chloride ion diffusion coefficient 41−46% smaller.[196]

As shown in Figure 10a, when RA is exposed to a CO_2 environment, CO_2 permeates through pores and cracks and then reacts with CSH, C_3S, and C_2S,[209−211] resulting in the formation of $CaCO_3$ and silica gel.[212] Carbonation reactions occur rapidly in the initial stage and then slow down. As $CaCO_3$ indicates higher hardness and density than C−S−H and $Ca(OH)_2$, RA exhibits decreased crushing index, water absorption rate and increased density.[207,213]

Figure 10b shows the compressive strength of recycled aggregate concrete (RAC) and carbonated recycled aggregate concrete (CRAC). The compressive strength of CRAC is 5.0−33.0% higher than RAC. Additionally, the compressive strength of CRAC increases with the increase of CRA content,[196,197] and the 28 d compressive strength of CRAC is close to that of NAC,[196] indicating that CO_2 curing of RA overcomes the negative impact brought by RA at a high substitution degree.

3.3.2. *Recycled Powder.*

As shown in Figure 10c, when producing RA, particles smaller than 150 μm are also obtained, known as recycled concrete powder (RCP).[187] RCP contains $CaCO_3$, SiO_2, AFt, AFm, CH, C−S−H, and unhydrated cement particles.[188,189] However, RCP obtained from RA production has a low reactivity, which limits its use as an inert filler under high content.[235]

Carbonation is a promising treatment for enhancing the performance of RCP.[236,237] With the formation of $CaCO_3$, RCP becomes less porous, and the formation of stable calcium aluminum hemicarbon (CAHC), calcium aluminum monocarbon (CAMC), and AFT is also promoted, thereby improving the microstructure of the cement matrix. As shown in Figure 10d, Mehdizadeh et al.[198] found that the D50 of untreated RCP and CO_2-treated RCP were similar, but their particle size range was different. Some scholars[190,191,238] aimed to improve the carbonation efficiency of RCP by adding additives or optimizing carbonation method or parameters. Teune et al.[190] used 0.2−1.0 wt % of triethanolamine (TEA) to enhance calcium separation from RCP as $CaCO_3$, resulting in an approximately 2-fold increase in calcium precipitation below 63 μm and increased sequestrated CO_2 amount by 16−21 wt %. Jiang et al.[191] reported that by elevating carbonate concentrations and temperatures during RCP carbonation, more $CaCO_3$ was obtained (9.4−10.4 vs 4.5 wt % in the reference group).

Recycled brick powder (RBP) could be modified through CO_2 treatment to enhance the performance of RBP-based mortar.[199] Sahoo et al.[199] found that carbonated RBP revealed increased specific surface area and reaction activity, reduced mesoporous volume and average pore size. With 25% and 75% of manufactured sand replaced by RBP, mortar with carbonated RBM has 28 d compressive strength 19−21% higher than uncarbonated RBP. Furthermore, RBP demonstrates high porosity, facilitating CO_2 diffusion and precipitation of carbonation products. Consequently, adding RBP greatly enhances the carbon sequestration potential of cement-based materials.[199]

There is little research on the direct carbonation of glass powder (GP). Some scholars have investigated the synergistic effect of GP addition and carbonation curing on cement paste and found that adding GP benefits the carbonation reaction. Specifically, in the cement system with a GP/OPC ratio of 25%, the CO_2 absorption rate increased by 28.6% after 1 d of carbonation curing. Adding GP reduced the size of calcite crystals induced by carbonation, possibly due to the inhibition of crystal growth by local SiO_2 precipitation. Meanwhile, the carbonation curing of cement slurry mixed with GP helps to compensate for strength loss.[239]

Poon et al.[185] proposed that economic and environmental benefits are crucial for scaling carbonation of RA and RP. Financial viability depends on the balance between income from carbonated products and carbon markets, and costs including transport, processing, and carbonation operations. While some regions may achieve net profits, others show negative CO_2 balances due to policy and market disparities. Contradictory conclusions often arise from transportation distance,[185] which significantly affects both cost and CO_2 emissions. Thus, optimizing the locations of demolition, recycling, CO_2 sources, and construction sites is essential. Additionally, finer materials like recycled concrete powder exhibit higher CO_2 uptake and profitability than RA.[191,238] Pilot-scale studies tailored to local conditions are needed to validate feasibility and establish integrated recycling chains.

3.4. Mine Tailings.

Mine tailings (MT) are the leading waste of the mining industry, generating about 10−15 billion tons annually.[240] In natural minerals, the small-particle MT reduces the cost of mineral crushing and exhibits carbonation potential[3] (Figure 11a). However, the carbonation degree of MT is low. Its carbonizable substances are magnesium/calcium oxide and magnesium/calcium silicate:[15,241] $(Mg, Fe)_2[SiO_4]$, $3MgO\cdot 2SiO_2\cdot 2H_2O$, $Mg_3Si_2O_5(OH)_4$, and $CaSiO_3$, which requires a deposit of mafic/ultramafic (M/UM) rocks. The advantages of using MT for mineral carbonation are as follows: (1) The MT is of large reserves and it is widely distributed and readily available raw materials;[242] (2) Its particle sizes are fine, reducing the cost of crushing processing;[243] (3) Mineral carbonation of MT is permanent and almost does not require monitoring.[8]

Carbonation of MT can be divided into in situ carbonation (IMC) and ex situ carbonation (EMC). IMC involves injecting concentrated CO_2 into underground reservoirs and/or ore deposits.[244,245] While in areas lacking geological storage sites, EMC is more applicable, which can be carried out in independent factories or realized by modifying existing industrial processes.[246,247] The method of carbonation of MT can be divided into dry carbonation and wet carbonation. The carbonation kinetics of the dry carbonation method are slow and the conversion efficiency is low, which limits its large-scale application. However, this method is feasible for developing potential CO_2-absorbing solid adsorbents (e.g., calcined limestone).[248−250] In contrast, the wet carbonation method is more effective and one of the earliest mineral carbonation methods studied at the pilot scale.[243] It can be carried out in contact with either pure CO_2 or CO_3^{2-}-containing flue gas, where the latter reduces the capture and purification cost.[251−256]

However, due to significant differences in rocks' chemical and physical characteristics, there are distinct differences in the mineralization potential of different tailings. Li et al.[15] found that magnesium iron/ultramafic tailings with Mg/Si and Mg/Fe ratios >1.5 are easily carbonated. Chrysotile,[257] nickel,[258,259] chromium,[260,261] diamond,[262,263] and platinum group element (PGE)[262,263] containing carbonizable magnesium components, including brucite $[Mg(OH)_2]$, olivine $[(Mg, Fe)_2(SiO_4)]$, and serpentine $[(Mg, Fe, Ni)_3Si_2O_5(OH)_4]$, so they are suitable for mineral carbonation technology in ultramafic rock deposits.

The theoretical possible reaction equation during the carbonation of mine tailings is

$$CaSiO_3 + CO_2 \leftrightarrow CaCO_3 \tag{13}$$

$$Mg_2SiO_4 + 2CO_2 \leftrightarrow 2MgCO_3 + SiO_2 \tag{14}$$

$$CaMgSi_2O_6 + 2CO_2 \leftrightarrow CaMg(CO_3)_2 + 2SiO_2 \tag{15}$$

$$Mg_3Si_2O_5(OH)_4 + 3CO_2 \leftrightarrow 3MgCO_3 + 2SiO_2 + 2H_2O \tag{16}$$

$$Mg_3Si_2O_5(OH)_4 + 3CO_2 \leftrightarrow 3MgCO_3 + 2SiO_2 + 2H_2O \tag{17}$$

$$Mg(OH)_2 + CO_2 \leftrightarrow MgCO_3 + H_2O \tag{18}$$

The carbonation reaction of MT is related to mineral solubility. The order of dissolution rates of different minerals in water at room temperature/pressure is $Ca(OH)_2$ (portlandite) > $Mg(OH)_2$ (brucite) > $CaSiO_3$ (wollastonite) > Mg_2SiO_4 (forsterite) > $Mg_3Si_2O_5(OH)_4$ (serpentine) > $MgSiO_3$ (enstatite). Meanwhile, as shown in Figure 11b, the carbonation rate of mine tailings is also positively correlated with brucite contents, in which [Bl] is Black Lake mine, [Asb] is Asbestos mine, [Du] is Ni−Cu Dumont mine project, [Rgl] is Raglan Ni−Cu−EGP mine, and [Rnd] is Renard mine project. Jacobs et al.[269] invented the mineral carbonation potential calculator by estimating the modal mineral abundance of ultramafic rocks. Furthermore, as shown in Figure 11c, Picotet al.[266] evaluated factors including the characteristics and quantities of mining waste, CO_2 emissions, and the distance between the mines and emission sites to

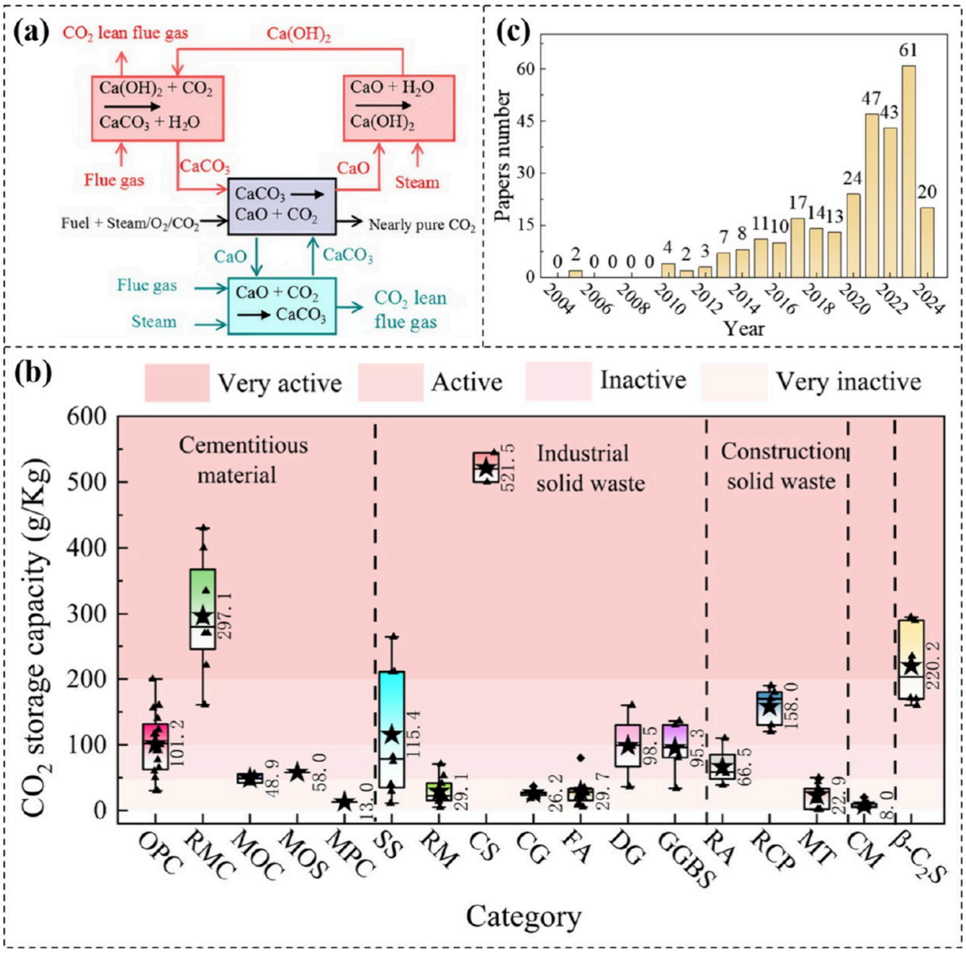


Figure 12. (a) Reaction equation in the production and carbonation of CaO. Adapted with permission from ref 291. Copyright 2016 Elsevier. (b) CO_2 storage capacity of different civil engineering materials (literature sources illustrated in Section S3). (c) Papers number related to LCA of MC-CEM from January 2004 to May 2024.

identify eight mineral deposits with significant carbonation potential in an ultramafic rock environment (where, in this assessment, rcoal-fired power plants have CO_2 emissions exceeding 1 million tons annually and are within 300 kilometers of "super-large" super-magnesium rock deposits).

As the carbonation degree of untreated tailings is generally lower, the mechanical activation, thermal activation, and chemical activation methods are adopted to accelerate it.[270] The thermal activation dehydrates the hydrosilicates (e.g., serpentine), but it does not affect igneous minerals (e.g., olivine). Meanwhile, the process must be precisely controlled to prevent mineral recrystallization due to overheating.[271] Chemical activation dissolves divalent cations from silicates, but it may pollute the environment.[272] Mechanical activation reduces the particle size of MT and destroys the crystal structure, but it consumes a high level of operating energy.[273] Nevertheless, mechanical activation could be a practical option when mechanical activation is part of the complete grinding process of MT, as the tailings already have a fine particle size at the initial stage.[274] At present, the most effective pretreatment method for olivine is mechanical activation in a high-speed stirred mill, while for serpentine, the best method is thermal activation at ~650 °C and chemical activation using $(NH_4)_2SO_4$.[275]

Currently, researchers have proposed many laboratory-scale strategies to overcome the main challenges of MC of mine tailings.[269,272,276−279] Nevertheless, little effort has been devoted to the pilot-scale and large-scale application of these strategies.[256] Li et al.[280] reported that the cost of processing and CO_2 sequestration takes up 83.4% of the total cost. In the pilot project of Mineral Carbonation International in Australia,[281−283] serpentine was used as the feedstock to capture the CO_2 emitted from the Kooragang Island power plant.[281] However, their current carbonation yield (35%) is significantly below the level required to upscale the process, even when considering the possible sale of the MC products. Li et al.[273] reported that a hypothetical nickel mining operation in British Columbia, Canada, had the potential to sequester 23.8 million tons of CO_2 per year over its 28-year mining lifespan. Under optimized conditions with a 60% CO_2 sequestration efficiency, the operating cost was ~$107 per ton of CO_2 avoided, too high for large-scale application. Existing pilot projects have low carbonation rates (e.g., in Australia[281−283] and Japan[284,285]) and high costs,[273] making large-scale application difficult.

3.5. Carbon Mix.

Carbon mix is the technology of transporting CO_2 into premixed concrete during transportation by a mixer truck, and its reaction is similar to that of OPC system (Section 3.1). Figure 11d shows the overall input and output in carbon mix, during which 963 g of CO_2 has been captured, cement usage is reduced by 17 kg, sand increases by

14 kg, and overall material transportation emissions are reduced by 124 g of CO_2.

Canadian company CarbonCure proposed a complete route of carbon mix:[286] A tank of liquid CO_2 was connected to a gas control system and injector. The liquid CO_2 was metered for injection into the truck, which converted into a mixture of CO_2 gas and solid CO_2 "snow". The CO_2 was delivered into the fresh concrete at a specified flow rate over a fixed injection interval, whereupon it reacted with the hydrating cement during initial mixing. The concrete was then subjected to assessment and testing.[237]

As shown in Figure 11e, the concrete treated with carbon mix reveals an improved early strength (3−17%). The concrete treated by carbon mix illustrates reduced initial and final setting time, accelerated hydration rate and strength development.[237] The process of carbon mix can reduce the amount of cement used. Although reducing the amount of cement may lead to a decrease in strength, it is possible to restore the compressive strength performance after carbonation.[268]

However, other aspects of consumption, such as the energy and materials consumed in implementing (construction equipment, CO_2 capture, transportation equipment), and the relatively small amount of CO_2 consumed in this way, the feasibility of this method remains to be carefully evaluated. Further, the biggest problem in carbon mix is the slow solubility and diffusion speed of CO_2 in concrete. In addition, the carbonation product $CaCO_3$ particles fill the pores of the concrete matrix, making CO_2 diffusion more difficult.[210] Besides, the workability of concrete under carbon mix may deteriorate. This degradation effect is insignificant/significant at low CO_2 concentrations (<0.3 wt % of cement[237])/high CO_2 concentrations (>0.5 wt % of cement[287]), respectively. Therefore, suitable additives are needed to overcome this drawback.

3.6. Other Materials. *3.6.1. Calcium Oxide.* The production of lime (mainly composed of CaO) is related to calcining limestone, which is one of the sources of anthropogenic CO_2 emissions. Lime could absorb CO_2, allowing 22% of the CO_2 emitted during lime production to be absorbed within 5 years.[288] At present, the carbonation of lime in civil engineering includes carbonation of sand lime bricks (SLB), lightweight lime concrete, autoclaved aerated concrete (AAC), mortar and water column building materials.[288−290]

Figure 12a illustrates the development of an effective method for capturing CO_2. During the CaO regeneration step, CaO is generated through the decomposition of $CaCO_3$. In a steam-rich environment, the heat from fuel combustion facilitates the decomposition of $CaCO_3$. Subsequently, the highly reactive CaO sorbent and nearly pure CO_2 exhaust are produced. The highly reactive CaO could be used to absorb CO_2 by direct carbonation (CaO carbonation) and indirect carbonation ($Ca(OH)_2$ carbonation).[291] Moreover, the Calera company[292] used CaO to prepare highly active $CaCO_3$ by CO_2 curing. Different crystal forms of $CaCO_3$ can be used as mineral admixtures. For example, active spherical $CaCO_3$ could react with water to transform into rod-shaped structural networks with bonding ability.

3.6.2. Calcium Silicate. Wollastonite ($CaSiO_3$) has great potential to form stable carbonates and is considered one of the most suitable natural minerals for sequestering CO_2.[7,293] $CaSiO_3$ demonstrates a high CaO content of 48.3%, making it an appropriate source of calcium. Besides, $CaSiO_3$ illustrates a faster reaction rate than $MgSiO_3$.[294] Wollastonite carbonation is divided into three steps.[30] (1) $CaSiO_3$ and gaseous CO_2 dissolve in the solution and release ions. $CaSiO_3$ particles react with H^+ and dissociate into Ca^{2+} and $SiO_2(aq)$. The dissociating rate of $CaSiO_3$ depends on the H^+ concentration and the maximum solubility of Ca^{2+}.[295−297] (2) Ca^{2+} reacts with dissolved CO_2 to form low-solubility $CaCO_3$. (3) The reaction products ($CaCO_3$ and SiO_2) precipitate once the solution is saturated.

The dissolution rate of wollastonite has a significant impact on the carbonation reaction. The dissolution of wollastonite is in turn influenced by the formation of SiO_2 and carbonate layers, which create resistance to ion diffusion as these layers hinder the ions' migration through them.[14] The diffusion coefficient of H^+ in each layer is the key to calculating the overall dissolution rate of wollastonite. The formation of SiO_2 and carbonate layers, as well as their resistance effects, occurs simultaneously.

3.7. CO_2 Storage Capacity of Civil Engineering Materials. Normalizing the CO_2 storage capacity of different civil engineering materials facilitates a deeper understanding of their mineralization potential,[298] with literature sources illustrated in Section S3. The CO_2 storage capacity of different civil engineering materials is related to their CaO/MgO content and form of existence. As shown in Figure 12b, according to the difference in CO_2 storage capacity, civil engineering materials can be classified into very active (>200 g/kg), active (100−200 g/kg), inactive (50−100 g/kg), and very inactive materials (<50 g/kg). This classification method follows the approach established by Zhang et al.[299]

Carbide slag (CS), reactive magnesium oxide cement (RMC), and β-C_2S belong to very active materials, due to their high content of carbonizable components. Specifically, carbide slag has a high content (85−95%) of $Ca(OH)_2$,[150−152] so it demonstrates the highest CO_2 storage capacity (average value = 517.8 g/kg[14]). RMC and β-C_2S also have high content of carbonizable MgO and C_2S, respectively.

Concerning cementitious material, RMC illustrates a much higher CO_2 storage capacity than OPC (average value: 297.1 vs 101.2 g/kg), consistent with research.[10,18] It should also be noted that the CO_2 emissions generated during the production of both OPC and RMC are non-negligible.[300] Although RMC production accounts for higher carbon emissions than OPC, its superior carbonation capacity results in a 73% reduction in net carbon emissions compared to OPC.[101] However, the CO_2 storage capacity of MOC, MOS, and MPC is distinctly lower than RMC. This is because RMC system reveals a higher content of carbonizable $Mg(OH)_2$ (e.g., ∼24% in RMC system[301] vs 12% in MPC system[302] after a period of CO_2 curing). Besides, compared to RMC, the early strength of MOC, MOS, and MPC systems is higher and the microstructure is denser. Therefore, CO_2 diffusion in MOC, MOS, and MPC systems is hindered and CO_2 storage capacity is lower.

Regarding solid waste, fly ash (FA) and mine tailings (MT) demonstrate a very inactive CO_2 storage capacity, so necessary activation measures are needed to stimulate their mineralization potential.[303] Additionally, in carbon mix (CM), CO_2 demonstrates slow solubility and diffusion speed in concrete, and $CaCO_3$ particles fill the pores, hindering subsequent CO_2 diffusion. Thus, carbon mix also shows inactive CO_2 storage capacity. It is necessary to develop suitable additives or improve carbonation devices and processes to enhance CO_2

storage capacity of carbon mix. However, all these steps above will lead to an increase in costs and energy consumption.

3.8. LCA in Mineral Carbonation of Civil Engineering Materials. LCA is a powerful tool for environmental management and analysis, quantifying resource consumption and environmental emissions across the entire product lifecycle.[304,305] LCA has emerged as a focal point in current research. As shown in Figure 12c, a marked increase in the number of studies on LCA of MC-CEM over the past decade could be observed.

Concerning cementitious materials, Xiao et al.[306] proposed that CO_2 mineralization in concrete materials could mitigate up to 15% of CO_2 emissions from cement clinker production, and theoretically almost 60% is possible. Van Roijen et al.[307] hold that the benefits of hydrated cement carbonation in direct air capture were oversimplified. They argue that approximately 13.8 billion metric tons of CO_2 were reabsorbed by cement globally between 1930 and 2015. Ruan et al.[101] proposed that the net CO_2 emissions of RMC were ∼73% lower than those of OPC when considering the carbonation capacity of cement. Jungclaus et al.[308] proposed that concrete mixtures incorporating high SCM substitutes should be prioritized in CCUS.

In terms of industrial solid waste, it was reported that direct and indirect carbonation sequestration potential of alkaline industrial waste is 4.02 Gt CO_2 per year globally.[2,309] Feng et al.[310] proposed that over 4 billion tons of industrial solid waste are generated annually in China alone and if fully carbonated, these solid wastes could capture over 200 million tons of CO_2 as carbonates. Pan et al.[311] found that the steel industry can simultaneously solve waste management and CO_2 emissions problems by using the high-gravity carbonation process, achieving a circular economy. Di Maria et al.[312] prepared carbonated SS-pressed blocks and found that their environmental impact was reduced by 70% to OPC-based blocks with the same compressive strength. Regarding construction solid waste, Li et al.[313] compared the global warming potential of carbonated recycled cementitious materials-based mortar and OPC-based mortar, and found the former represents a 75.2% reduction smaller. Meanwhile, Lei et al.[314] proposed that using CRA to replace RA partly saves environmental costs and reduces CO_2 emissions, offering significant environmental and economic benefits.

Concerning mine tailings, Gras et al.[315] argue that nickel tailings in Canada can capture ∼16% of the CO_2 annually emitted by its planned mining operation. In that case, 15 Mt of tailings produced each year produced 127.7 kt CO_2, and 21 kt CO_2 can be potentially sequestered via passive mineral carbonation. Vogeli et al.[263] found that mineral carbonation of platinum group element tailings can theoretically account for 43.6% of the 95% pure CO_2 produced by the synthetic fuels industry annually in South Africa. Khoo et al.[316] found that mineral carbonation of serpentine minerals by flue gas reduces 215 kg of CO_2 emissions for every 1 MWh of electricity produced by the power plant.[317]

4. CHALLENGES AND FUTURE PERSPECTIVES

Although bibliometric analysis can help reveal research trends and knowledge structures, its results still have certain limitations. For instance, the coverage of databases, language preferences, and the lag in citation accumulation may affect the comprehensiveness and timeliness of the analysis.[61] Moreover, quantitative indicators cannot profoundly reflect the profound connotations of the research.[55] Future review studies can combine broader literature sources and qualitative analysis methods to achieve more in-depth and impartial knowledge integration and theoretical construction.

Based on the results of the bibliometric analysis in this article, mineralization objects, mineralization methods, material properties after carbonation and environmental assessment have attracted widespread attention in MC-CEM research. There are many evaluation methods in MC-CEM research, e.g., CO_2 sequestration rate,[318] mineralization degree,[319] and mineralization efficiency,[15,320] which may cause misunderstandings and confusion during evaluation. To address this issue, it is necessary to comprehensively consider the advantages and disadvantages of various evaluation methods and develop a more widely accepted and applicable evaluation method. Meanwhile, technical improvements of LCA need to be made so that it is not limited to the system boundary from the "cradle" to the "gate" but also focuses on the impact of allocation methods and uncertainty analysis on the LCA results of carbonated products.

The engineering application of MC-CEM technology faces challenges in scalability, economic and environmental benefits.[80] For example, the pH value of carbonated OPC-based concrete is smaller than that of uncarbonated OPC-based concrete, which may lead to steel corrosion.[81] Compared to OPC system, magnesium-based cement system has a lower pH, not conducive to passivation of steel bars; its raw material costs are also higher. Additionally, characteristics instability in solid waste sources (e.g., steel slag,[138] red mud,[321] fly ash,[169] and recycled concrete[185]) affects the carbonization effect and hinders developing of standardized carbonation protocols. Moreover, Nonnegligible expenses and energy consumption arise from CO_2 capture, purification, transportation of carbonized materials, and energy-intensive curing processes. For example, producing a carbonated concrete block is ∼35% more expensive than that with OPC.[306] Many technologies remain in prototype/pilot stages, hindered by high costs and immature process chains. Large-scale commercial application of mineralized CEM is hindered as relevant standards are lacking.

Thus, performance-based metrics for product standards and comparisons need to be developed to support the inclusion of carbon-storing civil engineering materials. Nevertheless, before confidence in using these materials, the application may be initially more suitable for non- or low-load-bearing applications.[322] Optimize the locations of demolition, recycling, CO_2 sources, and construction sites for economic and environmental benefits.[185] The engineering application of MC-CEM can be advanced by uniting all its stakeholders (governmental supervisors, plants/companies, and research institutes) through the establishment of information platforms,[137] strengthened collaboration through forums[306] and effective economic incentives. Cross-departmental collaboration and policy economic incentives are necessary for implementation of MC-CEM technology.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.5c03561.

Method of bibliometric analysis, factors affecting the carbonation of steel slag, and literature sources of Figure 12b and CO_2 storage capacity of different civil engineering materials (Sections S1−S3), this work's differences and innovations relative to these studies, nomenclature, carbonation products in different systems, study of carbonated carbide slag, and reaction equation of carbonation of fly ash (Tables S1−S5), and research framework of bibliometric analysis and carbonation mechanism of CS (Figures S1 and S2) (PDF)

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Notes

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