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Review

Cold-bonded artificial aggregate: Processing, performance and perspectives

Jehangeer Raza [a], Vilma Ducman [b,*], Narinder Singh [a], Francesco Colangelo [a], Priyadharshini Perumal [c], Ilenia Farina [a]

[a] *Department of Engineering, University of Naples 'Parthenope', 80143 Naples, Italy*
[b] *ZAG Ljubljana, Dimičeva 12, 1000 Ljubljana, Slovenia*
[c] *Fiber and Particle Engineering Research Unit, University of Oulu, Finland*

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ABSTRACT

This article reviews cold-bonded artificial aggregates (CBAAs) produced from industrial wastes such as bottom ash, fly ash, slags, sludges, and other waste-derived precursors. It provides a detailed examination of the factors influencing their performance and the treatments used to enhance their properties. CBAAs are broadly classified into two groups based on binder type: cement-based aggregates and alkali-activated aggregates (AAAs). Cement-based CBAAs develop their required properties through hydration and pozzolanic reactions. Reported compressive strengths typically range from 1 to 12 MPa, while water absorption ranges from 10 % to 25 %. These properties vary significantly with precursor type, mix proportions, curing conditions, and surface treatments. In AAAs, cement is replaced by an alkaline binder to provide a more environmentally sustainable alternative. The properties of AAAs result from the formation of N-A-S-H or C-A-S-H gels, with commonly reported compressive strengths between 2 and 8 MPa. Performance can be improved by selecting the appropriate precursor type and optimizing the ratio of precursor to alkaline activator. Carbonation further enhances the properties of CBAAs by reducing porosity by up to 30 %, increasing compressive strength by a factor of two to three, and capturing CO_2 through the formation of $CaCO_3$. Environmental assessments indicate that converting industrial waste into aggregates can lower greenhouse gas emissions and keep leaching parameters within regulatory limits. This review also identifies ongoing challenges, such as precursor variability, energy demand, and scaling production, and outlines future directions to support the commercial deployment of CBAAs.

1. Introduction

Industrialization, urbanization and growing populations put substantial pressure on natural resources and cause climate change through the emission of greenhouse gases (GHGs) as well as the destruction of natural habitats. The population growth increases the construction activities to fulfill the demand for housing and infrastructure. Concrete is an extensively utilized construction material globally for many applications in transportation infrastructure as well as in commercial and residential buildings [1]. Concrete is composed of about 75–85 % by mass of differently sized aggregates [2] . The ever-growing construction industry uses nearly 50 billion tonnes of aggregates annually [3]. *The Global Material Resources Outlook 2060* projected that materials use will be more than double, increasing from 79 Gt in 2011 to 167 Gt in 2060, with non-metallic minerals such as sand, gravel, and limestone accounting for more than half of the total materials [4]. This situation forces the scientific community around the world to conserve natural resources and make palpable efforts towards a sustainable construction industry. At the same time, industries generate a huge number of wastes in the form of ashes, slags, sludges etc. which pollute the environment and disturb the ecological system [5,6]. Some of the industrial wastes can be used directly in construction while others need to be treated to either enhance their performance or reduce their negative environmental effects [7].

A sustainable construction industry requires alternative aggregates to produce concrete because the extraction of natural resources is already restricted to protect nature in some countries or regions [8,9]. The direct use of solid waste as an aggregate substitute in concrete is the most desirable option and materials like recycled aggregates fulfill this condition [10]. Recycled aggregates are waste from the construction industry (CDW – construction and demolition waste is defined as waste material by United States Environmental Protection Agency 2019 [11]) that is reused in the same industry. Using waste as artificial aggregates is one way of reducing their negative impact on the environment and provides suitable substitute materials for construction. The aggregates

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* Corresponding author.
E-mail address: vilma.ducman@zag.si (V. Ducman).

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Acronyms

AAs	Artificial aggregates
CBAAs	Cold-bonded artificial aggregates
CB-CBAAs	Cement-based cold-bonded artificial aggregates
AA-CBAAs	Alkali-activated cold-bonded artificial aggregates
LWC	Lightweight concrete
PEC	Porous eco-concrete
CDW	Construction and demolition waste
SCMs	Supplementary cementitious materials
LCA	Life cycle Assessment
GHGs	Greenhouse gases
GWP	Global warming potential
ASTM	American society for testing and materials
SEM	Scanning electron microscopy
EDXS	Energy-dispersive X-ray spectroscopy
BSE	Backscattered electrons
XRD	X-ray Diffraction
XCT	X-ray Computed Tomography
C-S-H	Calcium silicate hydrate
C-A-S-H	Calcium aluminosilicate hydrate
N-A-S-H	Sodium aluminosilicate hydrate
ITZ	Interface transition zone

OPC	Ordinary Portland cement
MSW	Municipal solid waste
MSWI	Municipal solid waste incineration
MSWI-FA	Municipal solid waste incineration fly ash
MSWI-BA	Municipal solid waste incineration bottom ash
MWBA	Municipal woody biomass ash
WPFA	Wastepaper fly ash
GGBFS	Ground granulated blast furnace slag
BFS	Blast furnace slag
BOFS	Basic oxygen furnace slag
SS	Steel slag
SF	Silica fume
RM	Red mud
FGD	Flue gas desulfurization
MP	Miscanthus powder
CSW	Concrete slurry waste
RCP	Recycled concrete powder
EU	European union
LEED	Leadership in energy and environmental design
BREEAM	Building Research Establishment Environmental Assessment Method
OSHA	Occupational safety and health administration

for concrete manufacturing are categorized into three types, as seen in Fig. 1.

Artificial aggregates (AAs) are usually produced by the process of granulation or pelletization. Other methods like pressure compaction or crushing are also used to prepare artificial aggregates. In the process of pelletization the fine solid waste is placed in a granulator together with a suitable binder and provided with a sufficient amount of liquid, and due to the cohesive and tumbling force, the particles bind together and form somewhat rounded pellets/granules [6]. Initially, the strength of artificial aggregates is very low, but curing them in controlled environments makes them stronger where the curing environment parameters have a significant impact on their final performance. Based on the curing temperature, artificial aggregates are divided into two groups; high temperature artificial aggregates cured at around 1000 °C and cold bonded artificial aggregates cured at ambient temperature [12]. High temperature treatment is the most common method used for the preparation of artificial aggregates and was introduced on a larger scale in the 1950s in the United Kingdom (UK) [8]. At the higher temperature, the particles of the granulate fuse together at their points of contact [13] and complex physical and chemical reactions occur [8], where gases are released, some of which are trapped in the structure, reducing the density and providing better sound and thermal insulation characteristics [14,15]. The final strength of high temperature artificial aggregates depends on the characteristics of the precursor materials and the firing temperature [8], as these conditions define aggregate's performance [16]. The promising properties, like strength, porosity and air permeability make the AAs a suitable choice for concrete production [17]. The eventual advantage of high temperature artificial aggregates is also a higher production rate because the aggregates are treated at high temperature for shorter duration time. However, the disadvantages of these aggregates are their high energy consumption and the eventual release of hazardous gases during curing.

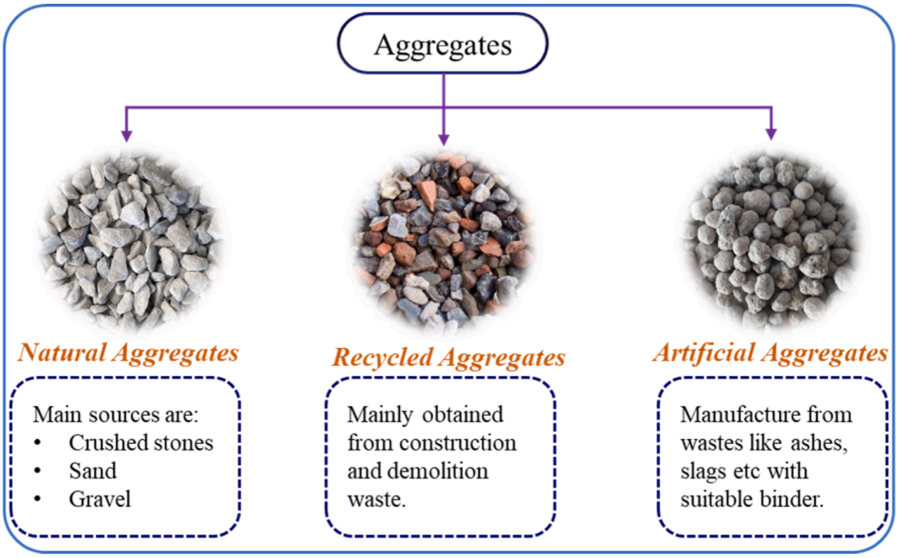


Fig. 1. Aggregate types used for concrete production.

CBAAs are considered effective and sustainable as they are cured at room temperature, making them more environmentally friendly in comparison to high temperature processes. Additionally, CBAAs which contain Ca-based carbonate-able compounds react with CO_2, which is advantageous because it not only sequesters CO_2 but also improves the aggregate's performance. Using alternative binders such as alkali activators can further improve environmental performance [18]. However, there are some economic implications, as the production of artificial aggregates is more expensive than that of natural aggregates, due to the need for machinery and skilled labour [19]. However, incentives from governments and supportive polices could help to stimulate the market and encourage investors and consumers towards its use.

In this review article, cold bonded artificial aggregates (CBAAs) are divided into three groups based on their binder: cement-based, alkali-activated and other binder-based artificial aggregates and are discussed in detail. The factors affecting the microstructure of cold bonded artificial aggregates and their relationship with physio-mechanical and environmental properties are analyzed. The performance enhancement and environmentally friendly approaches like carbonation are also discussed in detail and its effect on the overall performance of artificial aggregates is elaborated. Finally, arguments are developed for the critical evaluation of future challenges, barriers and perspectives that artificial aggregates will face while upscaling to a commercial scale. Recommendations for future research and the upscaling of production to a commercial scale are given.

2. Methodology

This review focuses on research published over the past two decades on cold-bonded artificial aggregates. Articles were identified through Web of Science, ScienceDirect, and Google Scholar using the keyword "lightweight artificial aggregates." An initial dataset of approximately 350 articles was collected using the publication filter 2006–2025. Only peer-reviewed journal articles written in English were considered. Titles and abstracts were screened, and studies on high-temperature or sintered aggregates were excluded. Given the breadth of the field and the increasing volume of published research, the selection aimed to be as comprehensive as possible, ensuring that all major types of cold-bonded aggregates and the full diversity of materials, processes, and performance assessments were represented. A final set of approximately 180 articles specifically addressing cold-bonded aggregates was examined in this study. Most of the included studies were published in the last six years (2019–2025), highlighting the rapid recent advancement of the field. Geographically, China is the leading contributor, followed by South Korea and India, as shown in Fig. 2, which presents countries with 20 or more publications in the selected period.

The review is structured as: Cement-based aggregates are discussed first, with emphasis on the influence of precursors on chemical and physical properties, their proportion in the aggregate matrix, and the effects of binder type and curing conditions. Surface treatments and their impact on aggregate performance are evaluated. Alkali-activated aggregates are then examined in detail, focusing on precursor type, alkaline solution concentration, and curing regimes. Environmental aspects of cold-bonded aggregates are reviewed through two main lenses: leaching behavior and life cycle assessment. Finally environmental techniques such as carbonation are discussed in terms of both aggregate performance and CO_2 sequestration potential.

3. Artificial aggregates

Aggregates prepared from wastes such as ashes, slags, and sludges, using a suitable binder and fluid, are known as artificial aggregates. Artificial aggregates are broadly classified into two types based on curing temperature, as depicted in Fig. 3.

Synthesized fresh granules or pellets (artificial aggregates) fired at temperatures above 800 °C are called high temperature artificial aggregates [20]. The properties of high temperature aggregates are primarily influenced by the precursor's characteristics, curing temperature and duration, and heating and cooling rate [21,22].

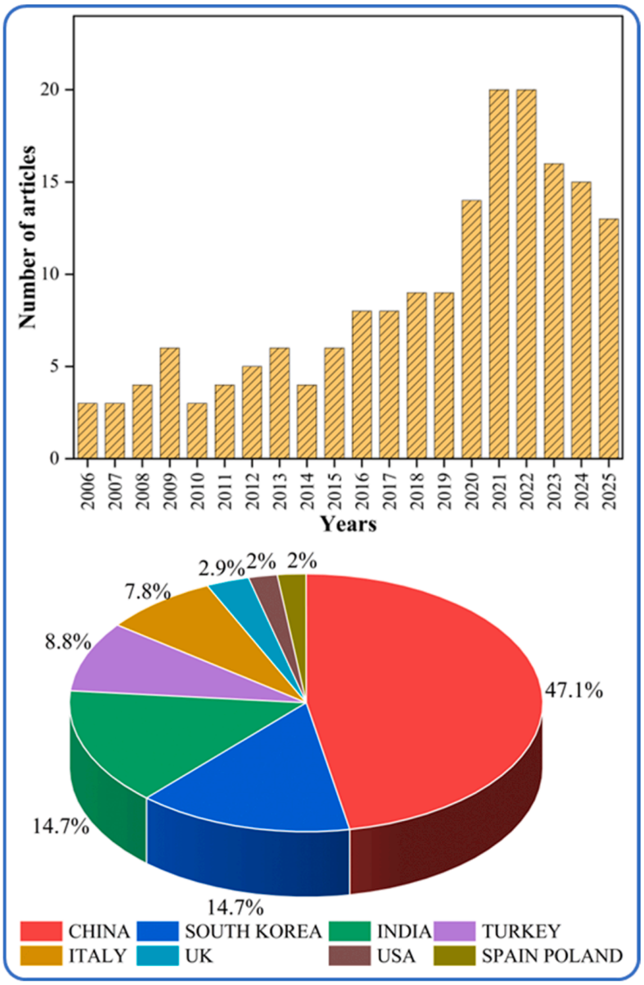


Fig. 2. Distribution of paper over the years and location.

3.1. Cold bonded aggregates

3.1.1. Cement-based aggregates

Cement-based aggregates are those in which cement is used as the binder. The aggregates gain strength from cement hydration products and the pozzolanic activity of precursor materials such as fly ash and slags. Both reactions enhance strength, durability, and resistance to chemical attack due to the formation of C-S-H gel [23,24], followed by pozzolanic activity which is slower than the hydration reaction [25].

3.1.1.1. Physio-mechanical performance. The important properties to consider when selecting aggregates for construction applications are mechanical strength, water absorption, and density. The properties of cement-based cold-bonded artificial aggregates (CB-CBAAs) are related to their porosity and microstructure, which depend on the physical properties and chemical composition of the precursor materials [26], binder (type and content), curing conditions [27] and surface treatment [28]. The curing conditions significantly affect aggregate performance; high temperature and humidity curing favour the development of hydration products and enhance the physical performance of aggregates [27,29,30]. In addition to the curing conditions, surface treatment of cement-based aggregates significantly enhances their physio-mechanical properties [26,28,31].

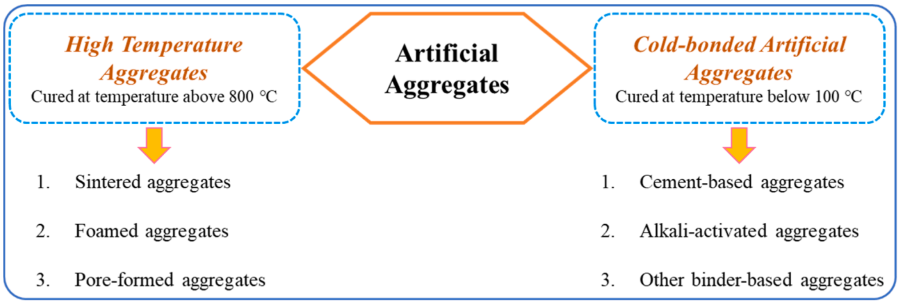


Fig. 3. Categories and sub-categories of artificial aggregates.

The following are some of the main factors which affect the physio-mechanical performance of cement-based artificial aggregates:

3.1.1.2. Precursor materials. The properties of cement-based artificial aggregates depend on the bonding forces developed through cement hydration and pozzolanic reactions. Therefore, selecting suitable raw materials and their appropriate proportions for aggregate preparation is a crucial step. The chemical composition influences the formation of hydrated products and subsequently affects the properties of CB-CBAAs. Various industrial waste materials, such as ashes (fly and bottom), GGBFS, and silica fume, are commonly used as precursor materials for the preparation of CB-CBAAs. Among these, fly ash is the most commonly used raw material for the preparation of cement-based aggregates due to its physical and mechanical properties [32]. According to ASTM C 618, fly ashes are divided into two classes: Class F fly ash, which contains less than 10 % CaO, and Class C fly ash, which contains more than 10 % CaO. The CaO content affects the formation of hydration products [33] which in turn affects the performance of the aggregates. Fig. 4 shows that aggregate made from fly ash with lower CaO content has a denser microstructure and improved physical and mechanical performance compared to that with high CaO content. Higher CaO content imparts some cementitious properties of the precursor; in such cases, the addition of activators such as gypsum, NaOH, Na_2SO_4, or water glass is required to enhance binding [26]. SiO_2 is also important because it plays a critical role in the formation of C-S-H, and precursors with a high percentage of SiO_2 have shown better performance. The CaO

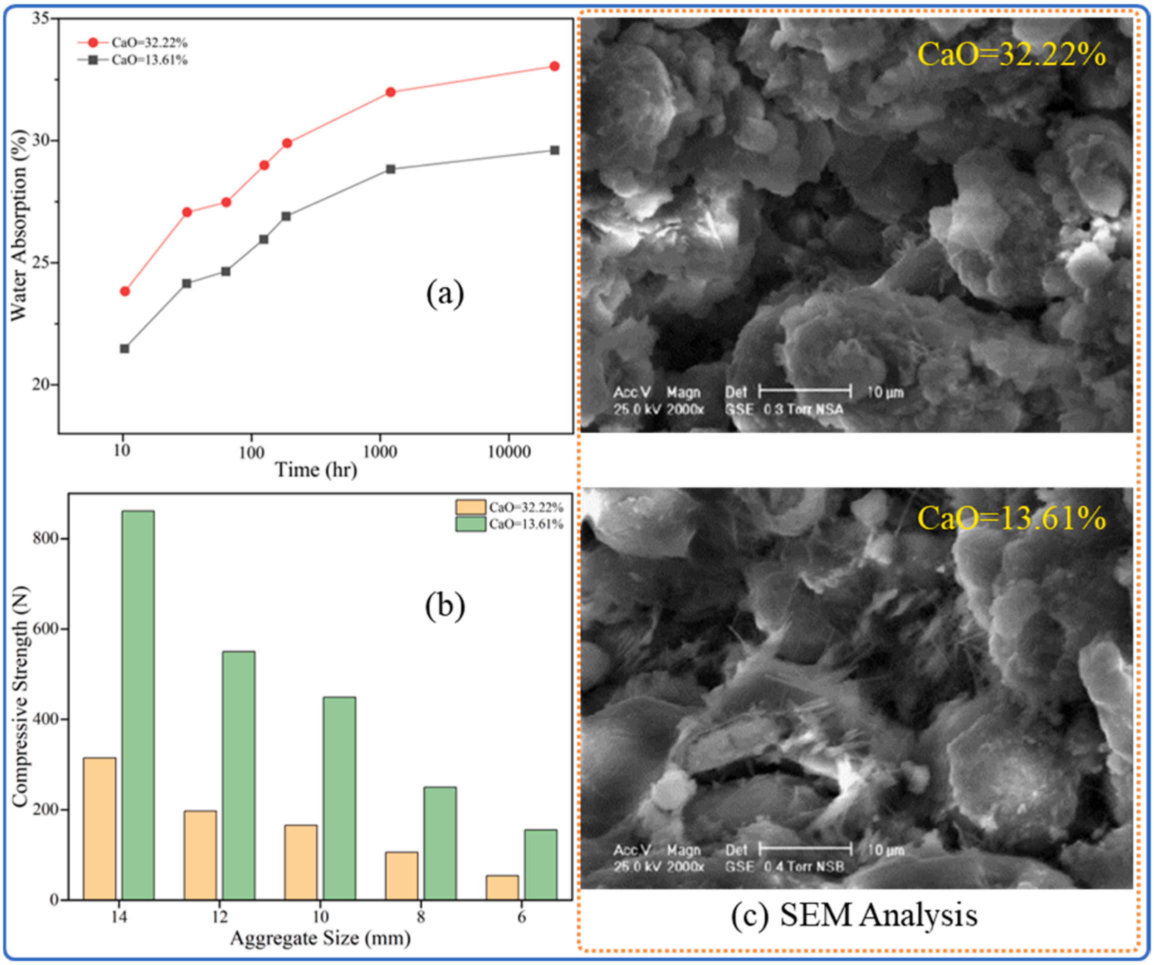


Fig. 4. Effect of CaO content on (a) water absorption, (b) Compressive strength and (c) morphology of aggregate, reproduce. Adapted with permission from Ref. [26] Copyright (2007) Elsevier B·V.

to SiO_2 ratio is crucial for the formation of the hydration product (C-S-H); if this ratio is unbalanced, it negatively affects the formation of the hydration product, which in turn impacts the microstructure and ultimately the strength of the aggregates. Excess CaO leads to the formation of weak $Ca(OH)_2$ phases, resulting in a more porous structure and aggregates with inferior performance. Higher SiO_2 content favors later-stage pozzolanic activity and develops a more refined pore structure, but due to lower CaO, it limits the formation of C-S-H and disrupts the reaction kinetics, ultimately affecting early-stage strength. Therefore, it is important to select the binder and precursor with a balanced chemical composition, rather than choosing a precursor with either higher CaO or SiO_2 content [26,34].

The physical characteristics of precursor materials also influence the properties of aggregates. Aggregates produced from fly ash with a lower specific weight (2.4) and higher specific surface area (3928 cm^2/g) have approximately 14 % lower water absorption, 3.3 % lower specific gravity, and 30 % higher mechanical strength compared to those made from fly ash with a specific weight of 2.56 and a specific surface area of 3206 cm^2/g [26,34]. Due to the lower specific surface area, some ash particles remain unreacted and do not participate in the hydration reaction, resulting in a porous structure and poorer aggregate performance. Generally, it was observed that mechanical strength improves by increasing the amount of cement in aggregate matrix [35].

Mechanical activation is a pre-treatment technique used to increase the fineness of precursors to achieve higher pelletization efficiency and improved properties [36] as it increases ash reactivity to form more reaction products [37]. Impact and shearing effects were used for size reduction in a centrifugal mill. Aggregates produced with activated fly ash have 22.73 % lower water absorption and 33.61 % higher mechanical strength compared to those made with non-activated fly ash [37]. In another ash pre-treatment, the municipal solid waste incineration fly ash was washed twice as a pre-treatment step, and through this process, some of the chlorides and sulfides leached out of the incineration fly ash [38,39]. The washed fly ash aggregates have a smooth surface, while others have cracks due to the presence of chlorides and sulphates in the fly ash, as shown in Fig. 5.

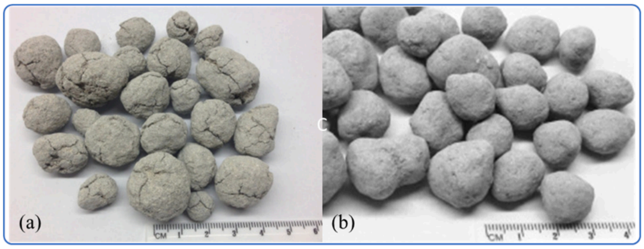


Fig. 5. (a) Unwashed fly ash and (b) washed fly ash-based artificial aggregates. Adapted with permission from Ref. [38] Copyright (2015) Elsevier B·V.

3.1.1.3. Aggregates matrix's. The composition of the precursors and binder impacts the physio-mechanical properties of aggregates. For example, in the literature, engineering muck-based aggregates were prepared by adding fly ash and slag as supplementary materials. The results, presented in Table No. 1 clearly demonstrate that physio-mechanical performance improves with an increase in cement content [40]. The aggregate's performance was found to be closely related to hydration activity. Replacing a portion of cement with fly ash reduces aggregate performance due to a decrease in hydration products, as fly ash has lower hydration activity compared to cement [41]. When cement is replaced with slag, the physio-mechanical performance also declines; however, it remains superior to that of aggregates containing fly ash, due to the high CaO content of slag [40]. Slag hydration is generally enhanced in the presence of calcium hydroxide (alkaline environment), but Ca and Mg in the pore solutions of the pastes may inhibit the consumption of Al, while the high Al content in turn inhibits the consumption of Ca, resulting in the slow formation of hydrates [42].

Generally, binder content (percentage) plays a decisive role in the microstructure and performance of aggregates. However, higher binder content increases density, cost, and environmental impacts without providing proportional improvements in aggregate strength. Therefore, optimal binder content should be used to balance the reactivity of precursors, microstructure, sustainability, and technological performance of aggregates. Table 1

In the case of cement-based aggregates, it was observed that the size of the aggregates also affects the physio-mechanical performance, as larger aggregates are more likely to have a porous structure, which is considered responsible for poor aggregate performance, as shown in Fig. 6 [31,43]. The better performance of the small-sized aggregates is due to the release of water during the final compaction phase of granulation, while, due to the high porosity of larger aggregates, water becomes trapped in these pores [38]. The relationship between aggregate size is not necessarily proportional, as another study found that the strength of aggregates increases with size. This is because the aggregate matrix contains more material and more hydration products, which develop a better microstructure (not compacted) with more closed pores that resist the applied mechanical load [26]. An example of the performance of such aggregate, as discussed above, is shown in Fig. 4, where it is clearly observed that the mechanical strength increases with the size of the aggregate. Another study observed mixed trends regarding the size, porosity, and performance of aggregates [31], while yet another study found that size has no significant influence on the strength of aggregates [12].

From the above discussion, it can be inferred that the hydration products followed by pozzolanic activity determines the mechanical and physical properties of the CB-CBAAs. It is critical to choose the appropriate material and percentage because an excess of one material can result in unreacted materials and impair the performance of the aggregate. There are three important parameters to consider for LWAs: strength, reduced density, and microstructure. The microstructure determines the type of porosity – either closed (structural) porosity or open (interconnected) porosity. In both cases, the density of the aggregates decreases. However, closed porosity enhances aggregate performance, while open porosity negatively affects performance by causing high water absorption, lower strength, and poor solidification or stabilization of hazardous content. Generally, as the size of the LWAs increases, their open porosity also increases, which decreases aggregate performance. However, if a high binder content and reactive precursors are used in a controlled pelletization process, the size will not affect aggregate performance. Conversely, with lower binder content, poorly reactive precursors, and uncontrolled pelletization, an increase in LWA size leads to increased open porosity, resulting in poor aggregate performance.

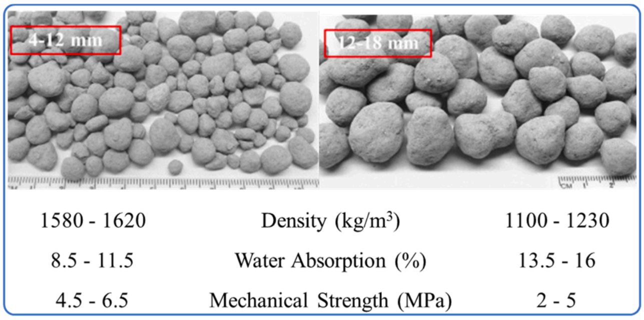
3.1.1.4. Curing condition. The usual curing conditions in the cold bonding method are ambient conditions, which require a longer time for the aggregates to gain sufficient strength but reduce production rates. To increase production capacity and improve the properties of the aggregates, various accelerated curing conditions have been applied. The curing condition has three important parameters that affect the performance of the aggregates: (1) temperature, (2) humidity, and (3) time. Curing methods such as ambient conditions, immersion in water at ambient or specific temperatures (below 100 °C), steam curing, autoclave curing, or sealing the aggregate in a plastic bag are used in the literature for aggregate curing [30]. The high temperatures accelerate the formation of hydration products, which embed the particles in the aggregate matrix. Fig. 7 shows the cross-sectional image displaying the various contents in the aggregate prepared from MSWI-BA, GGBFS, and cement, and cured under different conditions, along with physio-mechanical properties.

The morphology of the hot water-cured aggregate confirms the presence of various hydration products, such as binding gels and

Table 1
Composition and physio-mechanical performance of aggregates [40].

Samples No.	Aggregates composition by wt.%				Mechanical Strength (MPa)	Loose Bulk density (gm/cm^3)	Water Absorption (%)
	Cement	Fly ash	Slag	Engineering muck (Heavy silty clay)			
C8	8	/	/	92	6.06	1.16	13.66
C16	16	/	/	84	8.76	1.13	12.98
C24	24	/	/	76	11.77	1.12	12.57
F8C16	16	8	/	76	10.41	1.09	16.67
F16C8	8	16	/	76	6.52	1.05	20
F24C0	0	24	/	76	1.22	0.91	23.31
S8C16	16	/	8	76	10.61	1.13	14.17
S16C8	8	/	16	76	7.86	1.125	15.15
S24C0	0	/	24	76	1.71	1.12	17.6

Note: “C” denotes cement, “F” represent fly ash and “S” stands for slag.



1580 - 1620	Density (kg/m^3)	1100 - 1230
8.5 - 11.5	Water Absorption (%)	13.5 - 16
4.5 - 6.5	Mechanical Strength (MPa)	2 - 5

Fig. 6. Effect of aggregates size on its performance. Adapted with permission from Ref. [38] Copyright (2015) Elsevier B·V.

ettringite (needle-shaped). These products are responsible for a denser structure and improved performance [44] as shown in Fig. 8.

At ambient conditions, more time is required to form sufficient hydration products [45]. As the curing period increases, more hydration products are formed, developing a dense microstructure that provides strength to the aggregates [35].

3.1.1.5. Surface treatment. Surface treatment of cold-bonded artificial aggregates can be performed to enhance their physio-mechanical and leaching characteristics [38], and to increase the utilization of the waste materials [28]. Various surface treatment techniques, such as immersing the aggregates in slurry for a specific period [26], spraying particular materials [29], and double step cold bonding [28] have been used to improve aggregate performance. For example, fly ash-based artificial aggregates with OPC as the binder were surface treated by placing them for 30 min in water glass (Na_2SiO_3) which significantly enhanced the aggregates' physio-mechanical performance, as shown in Fig. 9 along with SEM micrographs [26]. From the mechanical strength results, it can

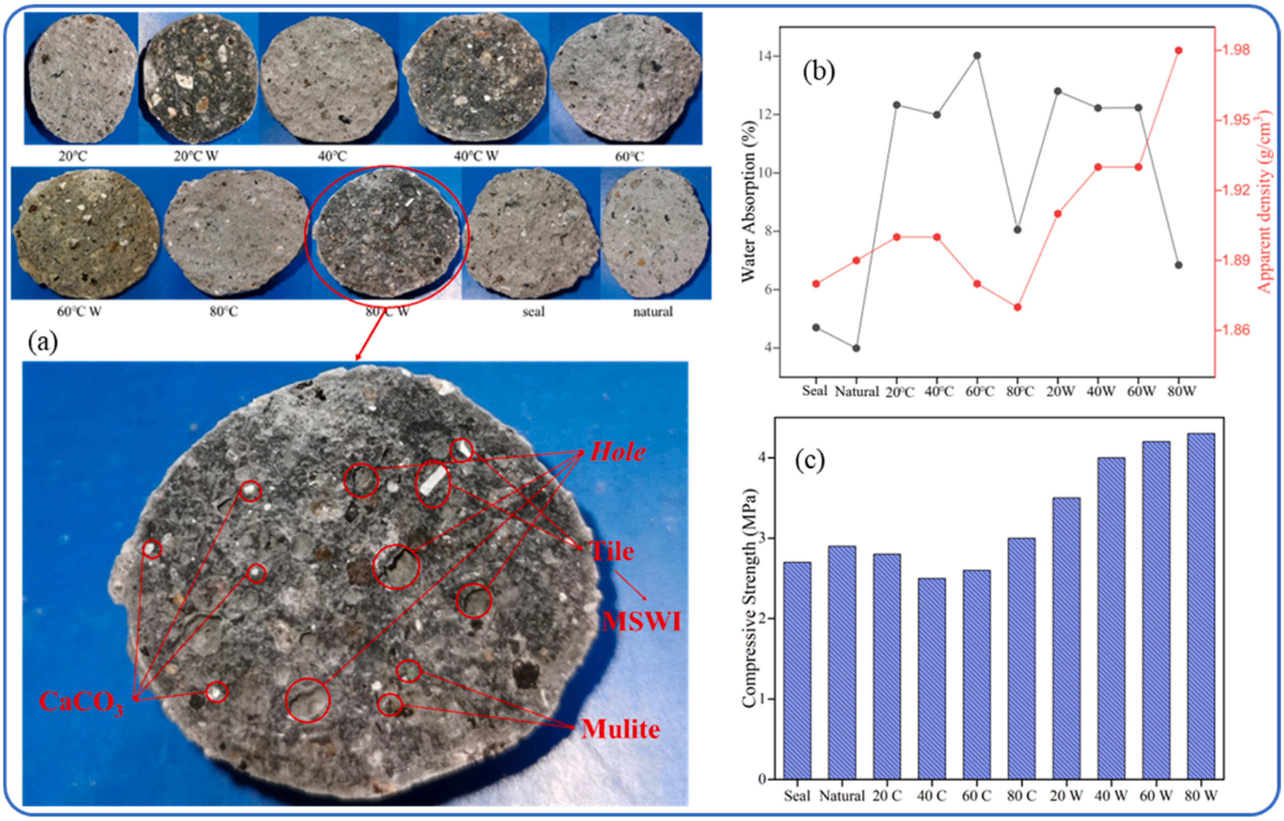


Fig. 7. (a) Cross sectional image, (b) water absorption and density and (c) mechanical strength of aggregates cured at various temperatures. Adapted with permission from Ref. [27] Copyright (2022) Elsevier B·V.

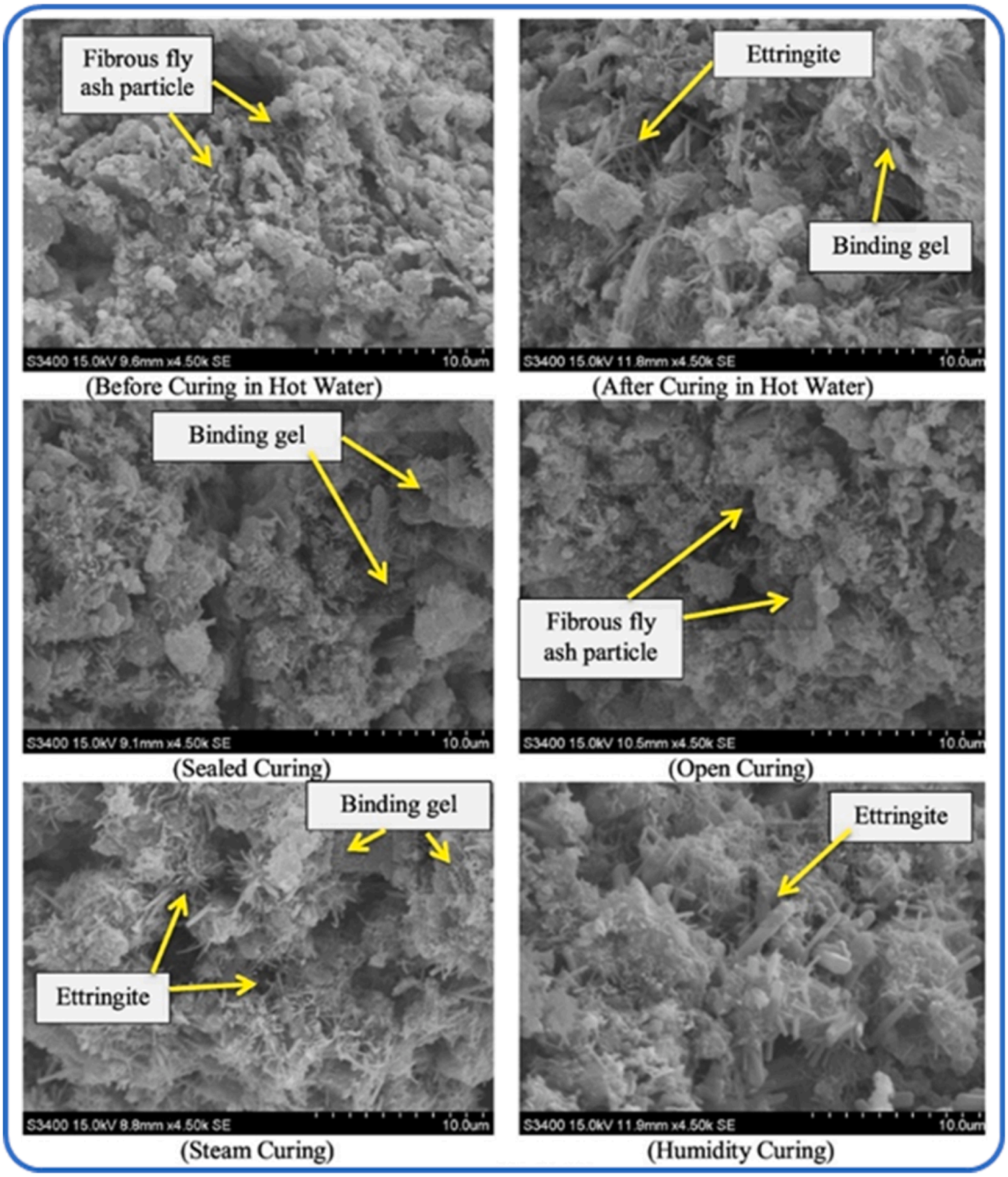


Fig. 8. SEM analysis of aggregates cured at various regimes. Adapted with permission from Ref. [30] Copyright (2021) Elsevier B·V.

be observed that increasing the size of the aggregate improves the mechanical strength and enhances the effect of the surface treatment. This is because larger aggregates have more pores, allowing more alkaline solutions to penetrate the aggregate matrix and form geopolymer gel, which accumulates in these pores. SEM analysis confirms that the surface of the treated aggregate is smoother compared to the untreated aggregate, as the water glass fills the open porosity and results in the formation of a glassy surface with higher Si content [26].

If an extra material is used for surface treatment, an additional outer layer is developed over the surface of the aggregate through various techniques, such as soaking in slurry [46], spraying slurry on aggregates [29] and multiple pelletization [28]. An illustrative and optical image is shown in Fig. 10 for such treated aggregates.

In the case of multiple pelletization, the aggregates were first cured for a certain period to gain strength, as the fresh pellets do not have sufficient strength. In the literature, two types of multiple pelletization were performed on aggregates in two different studies. In the first method, the aggregates were cured for 28 days. After curing, the aggregates were placed again in the granulator operating at 45 rpm and set at 45°, sprayed with water to moisten them, and then the mixture prepared for the outer layer was slowly added along with continued water spraying [31]. In another study, aggregates were cured in sealed plastic bags for 24 h and then surface treated using a multiple pelletization method. In this method, aggregates were soaked in water for 5 min, then placed in a pelletizer rotating at 15 rpm, and a slurry mixture was slowly sprayed onto the aggregates [29].

In the literature, these two surface treatments were compared, and it was observed that immersion in slurry is an ineffective surface treatment, as it sometimes negatively affects the physio-mechanical properties of the aggregates by forming protrusions on the surface, resulting in lower filling capacity or having very little effect [29]. The slurry immersion method is low cost and requires less time, but it does not cover the entire surface of the aggregate and produces an irregular shape [46]. The effects of these surface treatments on physical and mechanical properties are listed in Table 2, along with a comparison of these methods. The thickness of the outer layer and the size of aggregates selected for treatment also affect performance. For example, in literature, CBAAs were produced with 80 % phosphogypsum, 15 % GGBFS, and 5 % OPC (aggregate matrix) and surface-treated with 45 % phosphogypsum, 50 % GGBFS, and 5 % OPC (outer layer) by multiple

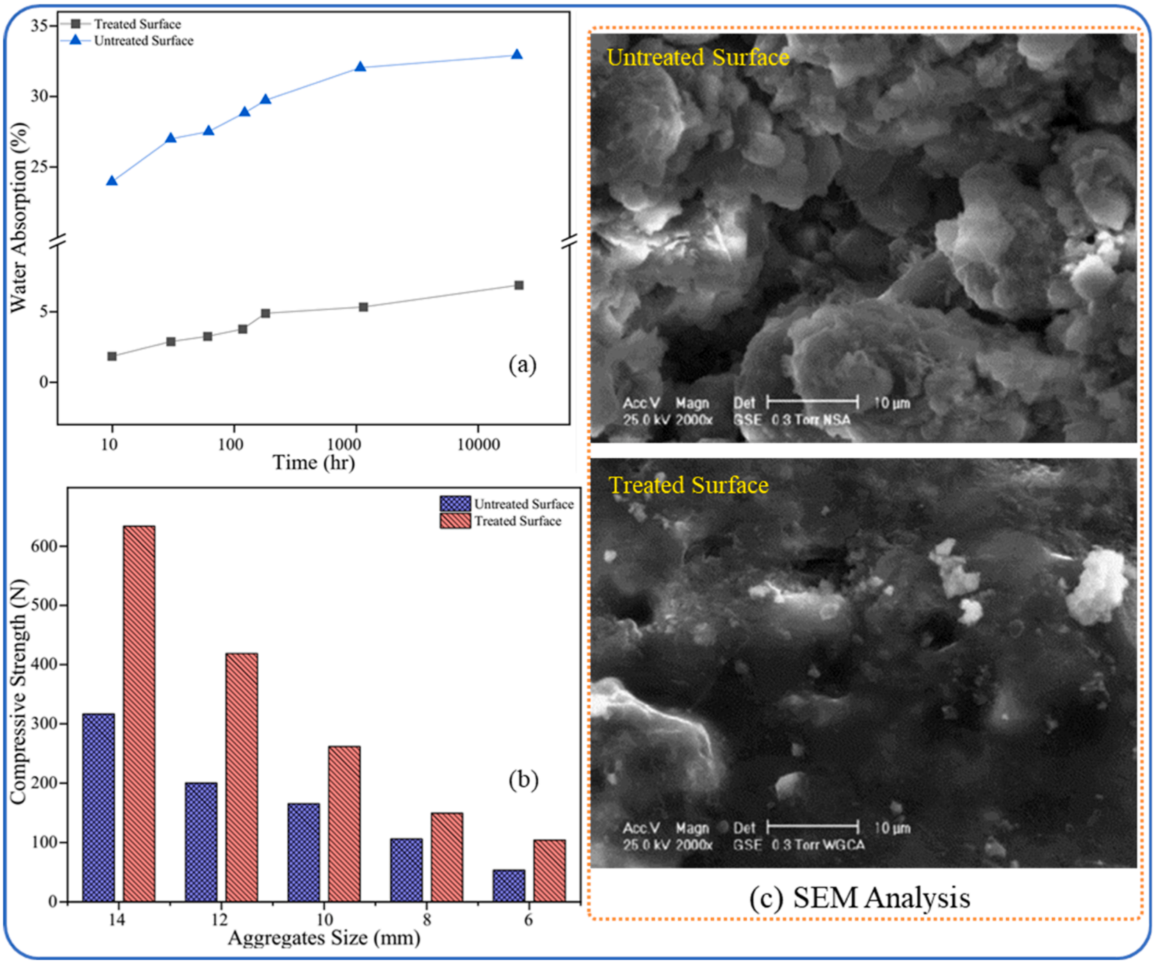


Fig. 9. SEM analysis of (a) untreated and (b) treated aggregate surface, EDS study of (c) treated and (d) untreated aggregate surface. Adapted with permission from Ref. [26] Copyright (2007) Elsevier B·V.

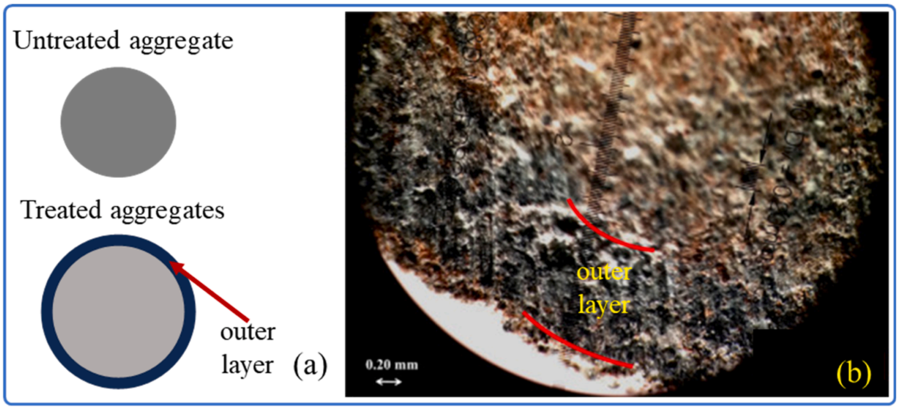


Fig. 10. (a) Illustrative diagram of single and double step cold bonded granules and (b) optical image of double layered aggregate. Adapted with permission from Ref. [38] Copyright (2015) Elsevier B·V.

pelletization. The 6 mm aggregates were coated with a 1 mm outer layer (P80–6); 5 mm aggregates had a 1.5 mm outer layer (P80–5), while a 2 mm outer layer was developed on 4 mm aggregates (P80–4). The author observed that P80–5 had a mechanical strength of 6.20 MPa, followed by P80–4 with 6.02 MPa, and P80–6 with 5.06 MPa, which was the lowest among the surface-treated aggregates. It was observed that inconsistent shrinkage between the aggregate and the outer layer affects the performance of the aggregates [46]. Summarizing Table 2 it seems that among the surface treatment methods, multiple pelletization was found to be the most effective, as it produces a uniform outer layer and a consistently densified aggregate surface. Slurry immersion is economical, simple, and timesaving, but it does not provide complete surface coverage, resulting in irregularly shaped aggregates and surface protrusions that negatively affect aggregate performance. However, it was observed that the thickness of the outer layer, aggregate size, material selection (shrinkage compatibility), and pelletization parameters

Table 2
Physio-mechanical properties of untreated and treated aggregates.

Aggregates composition	Outer layer composition	Surface treatment	UMS (MPa)	TMS (MPa)	UWA (%)	TWA (%)	Ref
5 %OPC + 15 %GGBFS + 80 %MSWI-FA	70 %Marble sludge + 30 %OPC	Multiple pelletization	1.33	1.95	16.81	11.50	[28]
10 %OPC + 10 %GGBFS + 80 %MSWI-FA			1.45	5.36	15.48	6.96	
15 %OPC + 5 %GGBFS + 80 %MSWI-FA			1.86	10.92	12.18	15.51	
18 %OPC + 49 %FA + 33 % expanded perlite particles	10 %Slica fume + 90OPC	Multiple pelletization	3.11	3.55	44	32	[29]
		Slurry immersion	3.11	2.77	44	42	
80 %Phosphogypsum + 15 %GGBFS + 5 % OPC	GGBFS + FA (1:1) with water glass solution at liquid-solid ratio of 0.7	Multiple pelletization	8.3	9.4	7.5	5.8	[46]
		Slurry immersion	8.3	8.6	7.5	6.15	

Note: UMS, TMS, UWA and TWA stand for untreated aggregates mechanical strength, treated aggregates mechanical strength, untreated aggregates water absorption and treated aggregates water absorption respectively.

influence the performance of surface-treated aggregates.

3.1.2. *Alkali activated cold bonded artificial aggregates (AA-CBAAs)*

The alkali-activated aggregates gain strength through the dissolution of reactive aluminum and silicate oxides in the presence of the alkaline solution and the formation of silicate-aluminate gel [16,47,48]. The gel hardens and binds the unreacted particles of the precursor materials to enhance the strength of aggregates [16]. Precursors (waste) containing aluminosilicate minerals are considered suitable for the preparation of alkali-activated aggregates [49]. Such AA are successfully prepared with alkali solutions derived from waste, which brings sustainability and economic benefits [50].

3.1.2.1. Physio-mechanical performance. The physio-mechanical properties of alkali-activated materials largely depend on the chemical composition of the precursor material, curing conditions, and the concentration of the activator solution. The SiO_2/Al_2O_3 ratio in the precursor material [51,52] and alkalis concentration contributes to the development of the polymerization gel, which affects the structure of the aggregate and determines its physio-mechanical performance [53]. High temperature ($\geq$ 80 °C) and 100 % humidity curing are favorable conditions for achieving greater and earlier strength, along with improved physical characteristics [50,54]. It should be noted that (prolonged) curing at high temperatures may contribute to the contraction in polymerization gel, as cracks appear in the aggregate matrix which affect its performance [55].

The factors which affect the physical and mechanical performance of alkali-activated aggregates are presented below.

3.1.2.2. Precursor materials. Waste materials containing chemically active aluminosilicate minerals are considered suitable precursors for the preparation of AA-CBAAs [49]. The most common raw materials are various industrial wastes (including ash of brickyard), agricultural wastes, municipal wastes [56]. The reactivity of the raw materials in the alkali medium is crucial and depends on the mineralogical and morphological properties, particle size distribution, and elemental composition. The mechanical performance of the AA-CBAAs was found to depend on the solubility of SiO_2 and Al_2O_3 [57,58], and their relationship is shown in Fig. 11.

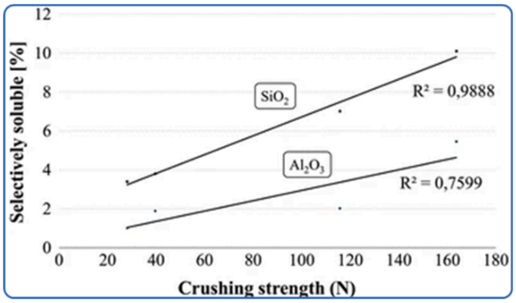


Fig. 11. Correlation between the selectively soluble SiO_2 and Al_2O_3 and crushing strength of aggregates. Reprinted with permission from Ref. [58] Copyright (2016) Elsevier B.V.

The amorphous phase of the reactive aluminosilicate dissolves in the presence of a high concentration of OH− ions (basic pH) and releasing Si^{4+} and Al^{3+} ions in the form of monomeric groups in the solution [59]. The chemical composition of the precursor material determines the rate of aluminosilicate dissolution [60]. Water is necessary for gel formation, as it facilitates the dissolution of solid aluminosilicates via alkaline hydrolysis, and the resulting aluminate and silicate species in the aqueous phase lead to the precipitation of N-A-S-H gel [61]. The water consumed in the dissolution process is released during condensation, and some time is required for the supersaturated aluminosilicate solution to form a continuous gel [61]. The addition of GGBFS enhances mechanical strength [51,62], as most studies observed that adding a certain amount of GGBFS (main components are CaO and SiO_2) to fly ash-based AA-CBAAs significantly improves physio-mechanical performance [16,51,62–64]. The high CaO content in the GGBFS enhances the binding ability of the precursor materials by contributing to the formation of C-S-H gel [62]. This leads towards more dense and compact structure with refined pore size of aggregate [65,66].

The effect of GGBFS on MWBA (municipal woody biomass ash)-based AA-CBAAs is shown in Fig. 12 which demonstrates that mechanical strength increases with higher GGBFS content due to reduced porosity and fewer interconnected pores (as seen in cross-sectional and SEM images) [51]. Recently, lithium slag-based AA-CBAAs were prepared, and it was again observed that increasing the percentage of GGBFS enhanced their performance [67]. Similarly, in another study, the characterization of mine tailing-based AA-CBAAs showed that increasing the slag content from 10 % to 30 % had a pronounced impact on their physical (23 % reduction in water absorption) and mechanical (enhanced by 22.38 %) properties [68]. At lower slag content ($\leq$ 15 %), the geopolymerization process did not progress optimally, as unreacted mica sheets remained in the geopolymer structure, while a higher percentage of slag resulted in a well-geopolymerized gel [68,69]. The same research group in another study analyzed the effect of different percentages of FA on mine tailing-based AAAs. It was found that increasing FA from 10 % to 30 % reduced water absorption by 20 % while increasing mechanical strength by 111 % [69]. Comparing both studies, it was concluded that the Si/Al ratios play a significant role in determining the performance of AAAs, which is closely related to the chemical composition of the precursor. For instance, in the same study [68], it was observed that increasing slag content up to 20 % significantly increased the Si/Al ratio, but further increases in slag content decreased the ratio, which negatively impacted aggregate performance. However, increasing FA content beyond 20 % improved the Si/Al ratio, thereby enhancing performance. In AAAs, various compositions of phosphorus tailings (PT), fly ash (FA), soda residue (SR), and granulated blast

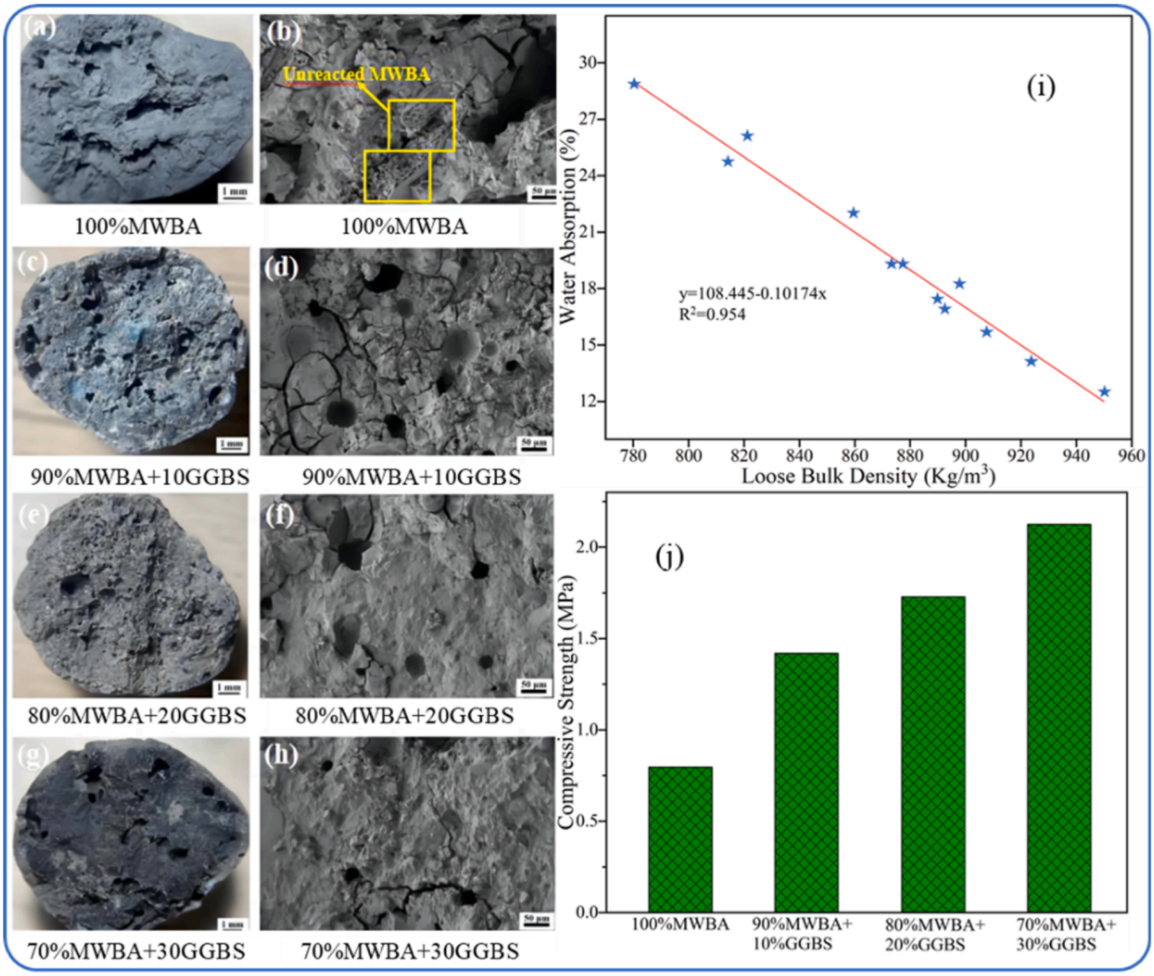


Fig. 12. (a, c, e, g) Cross Sectional, (b, d, f, h) SEM images, (i) loose bulk density relation with water absorption and (j) crushing strength of MWBA-based aggregates with different GGBS content. Adapted with permission from Ref. [51] Copyright (2023) Elsevier B·V .

furnace slag (GGBS) were studied [70]. It was observed that a mixture of 40 % PT, 20 % FA, 30 % SR, and 10 % GGBS exhibited superior performance compared to other compositions, as this combination provides the most suitable aluminosilicate content. Alkaline compounds play a dual role by breaking down the active Si-Al networks (silicon oxides and silicates) and enhancing the dissolution of ions [70]. This process supports the hydration reaction and promotes the development of gel-like structures, such as C-S-H, which in turn improves the stability, strength, and durability of the material [71].

Similarly, the addition of cement to AAAs enhances the performance of the aggregates because cement develops a C-S-H phase alongside the gel phase formed by alkali activation [64]. The presence of this additional phase results in a dense microstructure and enhances the bonding between particles in the aggregate matrix, leading to aggregates with improved performance. For example, the addition of cement increases the density by 15.24 %, while mechanical strength almost triples and water absorption is reduced by 35.06 % in sewage sludge ash and GGBFS-based aggregates [64]. Fig. 13 shows SEM micrographs with and

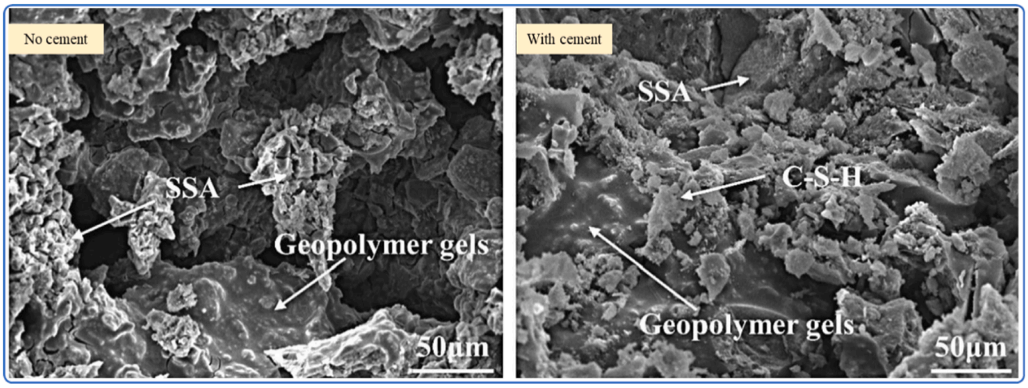


Fig. 13. SEM analysis of sewage sludge ash and GGBFS-based aggregates with and without cement and their respective physical and mechanical properties Adapted with permission from Ref. [64]. Copyright (2022) Elsevier B·V.

without the addition of 15 % cement. However, this contradicts the main aim of using alkali-activated aggregates, which is to reduce cement usage and protect the environment.

3.1.2.3. Activator solution. The alkaline solution is used as a binder and provides the moisture content required for granule formation. The most used alkaline solution contains sodium hydroxide (NaOH) and sodium silicate (Na_2SiO_3). The molarity of NaOH significantly affects the properties of the aggregates [72–74]. The activator solution in the aggregate matrix physically binds and encapsulates unreacted precursor particles in an equilibrium combination of C-(A)-S-H and N-A-S-H gels [75] as well as a spongy microstructure, which are aluminosilicate and alkali-activated slag – the primary stabilizing reaction products [76]. In the literature, the effect of different concentrations (0, 4, 6, 8, 10, 12) of NaOH was analyzed [74] and it was found that as the concentration increased from 4 to 8, the performance of the aggregates improved. The SEM micrographs, along with the apparent density, water absorption, and mechanical strength, are shown in Fig. 14. In the absence of NaOH in the aggregate matrix, geopolymer gel is absent and some unreacted precursor particles are present, which affect aggregate performance. Such reactions increase porosity and cause cracks to develop in the aggregates, which negatively affect aggregate performance [74]. The high content of NaOH delays the development of polymerization strength due to the hydrolysis and precipitation of Al^{3+} and Si^{4+} in solid aluminosilicate [77]. Rapid dissolution and re-precipitation rates are observed at higher NaOH concentrations, restricting the development of the polymerization gel and disrupting the suitable microstructure required for improved aggregate performance [53]. Such reactions cause increased porosity and even cracks in the aggregates [74]. Higher NaOH content favors the formation of N-A-S-H gel and magadiite [78] and retards the formation of high-strength gels such as C-S-H and C-A-S-H [79,80]. It should be noted that, irrespective of the raw material content, the alkaline solution affects the properties of the aggregates, and it was observed that these properties improved with increasing alkaline solution content within limits [51]. Recently, the effect of NaOH molarity (6 M –14 M) on lithium-slag-based AAAs was analyzed [67]. Water absorption decreased from 15.87 % to 5.88 %, while mechanical strength increased from 2.33 MPa to 10.87 MPa as the molarity increased from 6 M to 12 M. Further increases in molarity had only a slight effect on performance.

3.1.2.4. Curing conditions. The reaction mechanism under various

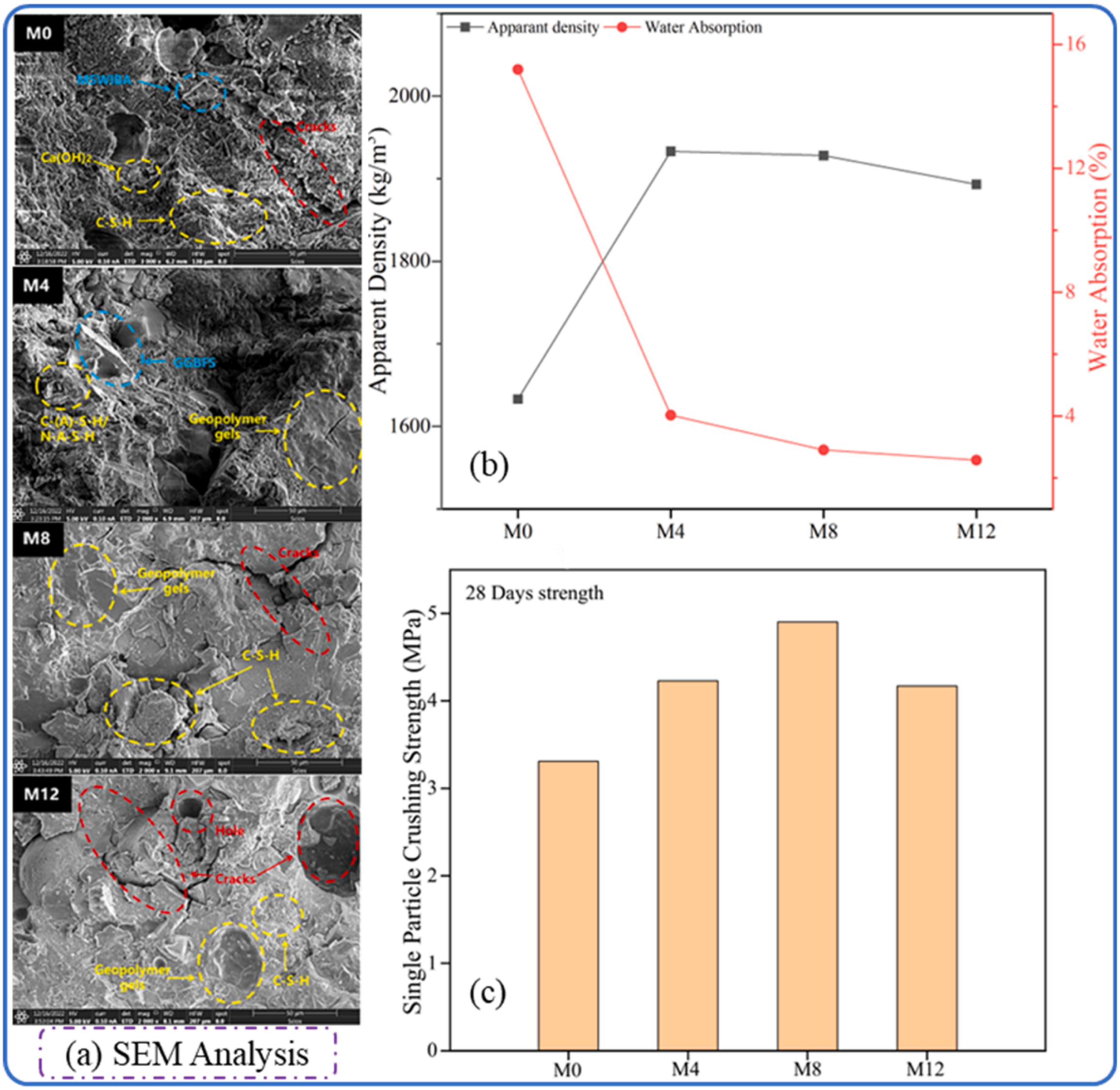


Fig. 14. (a) SEM micrographs, (b) apparent density, water absorption and (c) crushing strength of aggregates with various concentration and without NaOH Adapted with permission from Ref. [74]. Copyright (2023) Elsevier B·V.

curing conditions for alkali activated aggregates [54] is shown in Fig. 15. It was observed that as temperature and humidity increase, the formation of large polymerization gels improves compared to dry and room temperature curing [54]. The polymerized gel provides a dense and compact microstructure to the aggregate, resulting in better performance. However, it should be noted that prolonged curing at higher temperatures can cause defects or shrinkage in the polymer gel and cracks in the aggregate matrix, which adversely affect performance [55,59].

The SEM micrograph, bulk density, water absorption and cylinder compressive strength of the aggregates cured under various curing conditions [50] are presented in Fig. 16. The steam-cured aggregate, treated at 60 °C for 12 h, enhances condensation and depolymerization of precursor materials, which improves the crystallinity of hydration products and produces dense, compact structures [50].

Under ambient conditions, curing duration will be prolonged [81], while at higher temperatures it may be shorter. In addition to curing time, temperature, and humidity, pressure also affects the performance of alkali-activated aggregate [57]. High curing temperature ($\geq$ 80 °C) and humidity (100 %) are favorable conditions for achieving greater and earlier strength, along with improved physical characteristics [50,54]. It should be noted that prolonged curing at high temperatures disturbs the polymerization gel, as cracks appear in the aggregate matrix and affect its performance [55].

In summary, for alkali activated aggregates the SiO_2/Al_2O_3 ratio and activator concentration are crucial factors, as these define the development of the microstructure and polymerization gel, which determine the physio-mechanical characteristics. For accelerated curing, high temperature and humidity are recommended in the literature, but prolonged exposure impairs aggregate performance.

3.1.3. Other binder-based aggregates

For cold bonded aggregates the most commonly used binders are cement and lime [8]. Cement-based aggregates and alkali-activated aggregates were discussed in detail in the previous sections. This section will summarize other binder-based CBAs. The commonly used alternative binders, such as lime and bentonite, have shown poorer performance compared to cement or alkali activated cold bonded aggregates [38,82–85]. In one of the study, lime- and cement-based aggregates were compared, and it was observed that lime-based aggregates exhibited poor physio-mechanical performance and leaching behavior [38]. However, in the same aggregates, when 25 % coal fly ash was added and lime was reduced to 15 % with 60 % MSWI-FA, the performance of the aggregates improved as shown in Fig. 17.

However, sometimes higher “Ca” content hinders the formation of hydration products (i.e. C-S-H, C-A-S-H) or a lack of other suitable components such as Si and Al, which are required for the hydration reaction to react with “Ca” to form hydration products [26]. In the case of FA the crushing strength of aggregates increased, and the water absorption decreased with increasing amounts of either PC or $Ca(OH)_2$ up to 10 % and 5 %, respectively [86]. The hydration products are primarily responsible for the strength of cementitious materials while other Ca-based compounds assist in improving their characteristics. Fig. 18 presents the physio-mechanical performance and the SEM micrographs, from which it can be observed that cement-based aggregates have a dense structure with C-S-H phases that contribute to better aggregate performance, while calcium hydroxide-based aggregates exhibit porosity and some unreacted binder, which is responsible for poor physio-mechanical characteristics.

Lime has been shown to be a poor binder compared to cement when used alone as a binder in municipal solid waste incinerator (MWSI) applications, as only cement and coal fly ash/lime systems provided sufficiently high mechanical strength [38]. In another study, physio-mechanical properties showed the binder effect as follows: cement > cement + lime > lime [87]. However, it has been observed in the literature that adding GGBFS to a matrix of ash-based aggregates with lime as a binder result in better properties compared to cement, because lime activates and reacts with the higher CaO content in GGBFS. GGBFS is hydraulic in nature, hardens in the presence of water, and contributes to the overall strength of the aggregate [83].

Apart from the known binders, industrial waste was effectively utilized in the preparation of aggregates [88–90]. All these investigated materials belong to SCMs, meaning they contain all the essential chemical compounds required to carry out the pozzolanic reaction and form hydration products. The red mud (RM)-based aggregates were prepared using SCMs quarry dust I (limestone), quarry dust II (diabase), aluminum ash, FGD gypsum) as binders and successfully recycled 80 % of RM [91] . It was observed that increasing the content of SCMs from 15

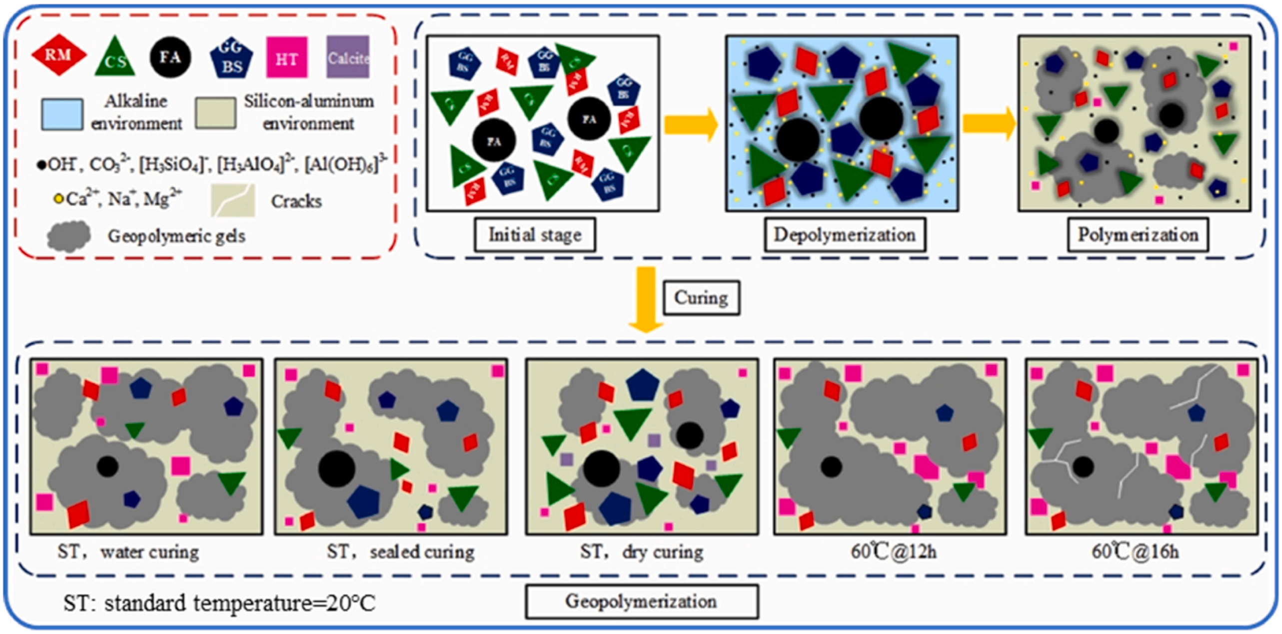


Fig. 15. Schematic diagram of reaction mechanism of alkali activated aggregates cured at various temperatures. Reprinted with permission from Ref. [54]. Copyright (2023) Elsevier B·V.

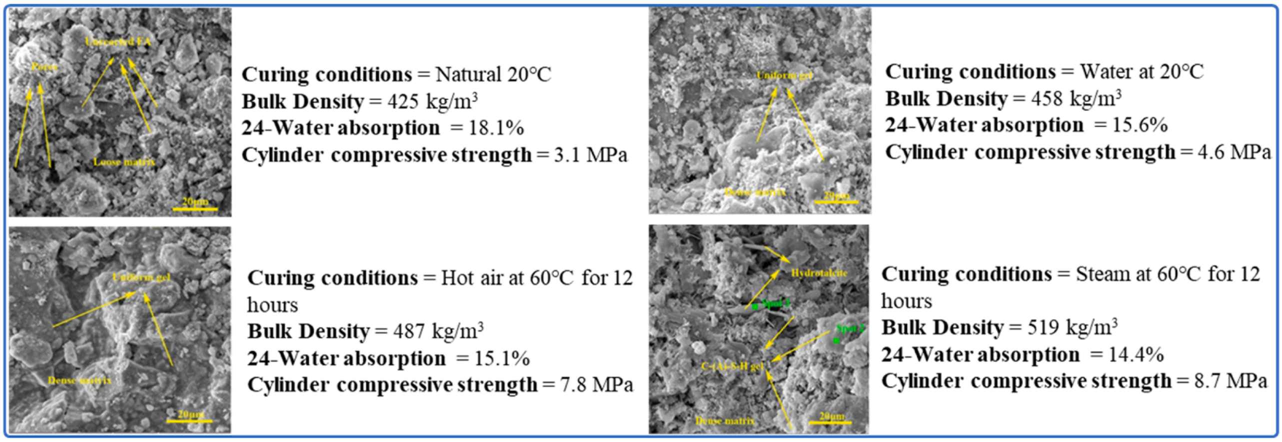


Fig. 16. Alkali-activated aggregates cured at different conditions with their respective SEM analysis, water absorption, bulk density and cylinder crush strength Adapted with permission from Ref. [50]. Copyright (2023) Elsevier B·V.

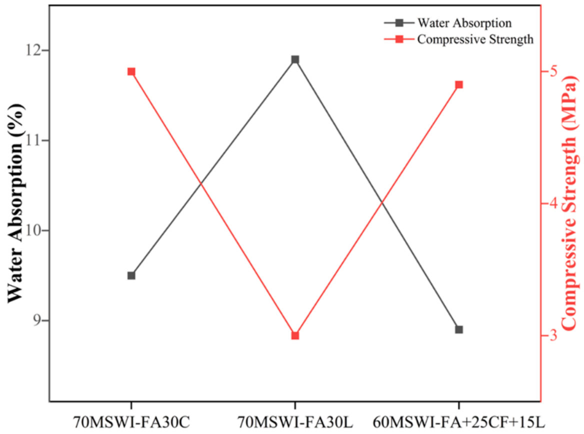
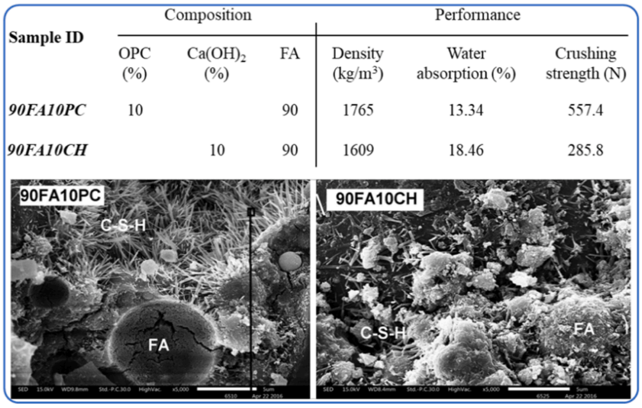


Fig. 17. Water absorption and mechanical strength of cement and lime-based aggregates Adapted with permission from Ref. [38]. Copyright (2015) Elsevier B·V.



Sample ID	Composition			Performance		
	OPC (%)	$Ca(OH)_2$ (%)	FA	Density (kg/m³)	Water absorption (%)	Crushing strength (N)
90FA10PC	10		90	1765	13.34	557.4
90FA10CH		10	90	1609	18.46	285.8

Fig. 18. (a) water absorption, density and (b) crushing strength along with SEM analysis of aggregates with OPC and CH. Adapted with permission from Ref. [86]. Copyright (2018) Elsevier B·V.

% to 30 % enhances the physio-mechanical performance of aggregates, with similar trends found for pelletization efficiency. Similarly, CBAs were successfully prepared from steel slag (SS) and miscanthus powder (MP). Aggregates prepared with 90 % SS and 10 % MP showed better water absorption and mechanical performance than those prepared with 100 % SS; however, according to the EN 13055 standard, these aggregates are not classified as lightweight [90].

4. Environmental performance

Environmental assessment of artificial aggregates is as important as evaluating their mechanical and physical properties. To examine the environmental impacts of artificial aggregates, life cycle assessment (LCA) and analysis of heavy metal leaching from the aggregate matrix are two key parameters to be considered.

4.1. Life cycle assessment

Life cycle assessment (LCA) is a tool used to analyze the environmental impacts and resources consumed over the entire life cycle of a product or service. LCA comprehensively considers and assesses all aspects of human health, the environment, and resources [92]. LCA has four major phases which are illustrated in Fig. 19.

The sustainability of construction materials, including aggregates, has become a major concern in recent years due to the depletion of natural resources and high demand [9,94] . For a sustainable approach, it is crucial to consider technical, environmental, and economic aspects. Traditionally, research has concentrated on the development of novel construction materials with a focus on technical properties [95]. It is now recognized that environmental and economic aspects are also important factors to consider when introducing and industrializing new construction materials [96], as well as traditional materials such as concrete, brick, pavement, and aggregates, in order to quantify, evaluate, compare, and improve their environmental performance over their life cycle [97]. To select sustainable materials and effective waste management solutions, recent studies have focused on comparative life cycle assessment analyses of natural and artificial aggregates [94,98]. To determine an effective waste management solution, three disposal scenarios for fly ash were evaluated: landfill disposal, recycling as a raw material for cement, and use as a precursor for artificial aggregate (AAs) production [99]. The results presented in Table 3 indicates that using fly ash for AAs is the most effective option, as it results in lower environmental impacts, whereas landfill disposal poses the highest environmental threat, followed by cement manufacturing. The same group of researchers conducted another comparative study on sewage sludge management solutions (incineration, landfill, AAs), where results (Table 3) showed that its use for AAs is the most effective solution [100]. In the same study, the author compares AAs with natural aggregates (sea sand) and finds that AAs have a 27 % lower GWP compared to natural aggregates [100]. The high environmental impact of natural aggregates is due to transportation (48.6 %) and processing (47.2 %). A detailed

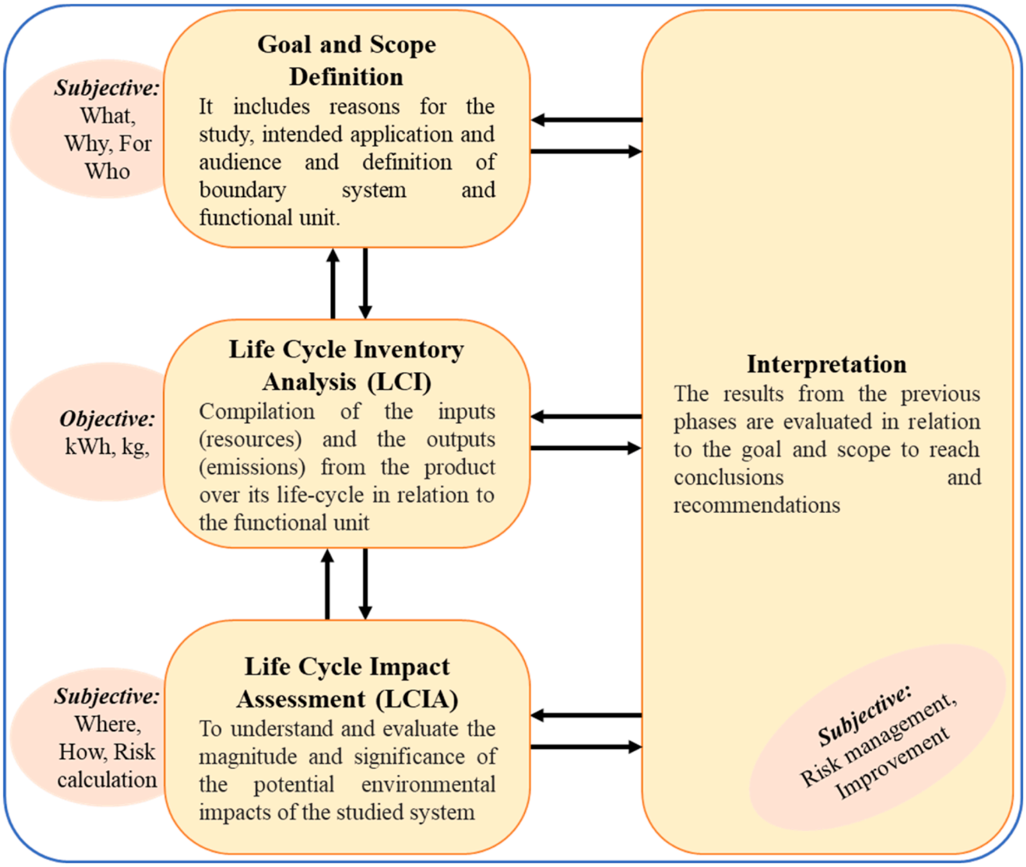


Fig. 19. Four phases of life cycle assessment (LCA). Adapted with permission from Ref. [92,93]. Copyright (2009) Elsevier B·V.

Table 3
Indicator of environmental consequences for each fly ash management scenario [99].

	AP (kg SO_2_Eq)	GWP (kg CO_2_Eq)	EP (kg PO_4^-_Eq)	HTP (kg 1,4-DCB_Eq)	POCP (kg CH_4_Eq)	ODP (kg CFC-11_Eq)	Ref
Landfill	2.94×10^{-2}	420	8.09×10^{-2}	0.6	6.07×10^{-4}	2.10×10^{-7}	[99]
Cement	3.38×10^{-1}	284	3.54×10^{-2}	5.99	7.20×10^{-3}	4.47×10^{-6}	
LWA	3.58×10^{-1}	275	1.75×10^{-2}	0.248	1.30×10^{-2}	1.35×10^{-9}	
Incineration	9.68×10^{-2}	445	1.19×10^{-1}	1.33×10^{-1}	5.30×10^{-3}	1.97×10^{-6}	[100]
Landfill	4.65×10^{-2}	831	7.59×10^{-3}	7.06×10^{-2}	1.59×10^{-1}	1.17×10^{-6}	
LWA	2.73×10^{-2}	441	4.46×10^{-3}	4.15×10^{-2}	1.64×10^{-3}	3.41×10^{-7}	
NA	8.40	604	1.56	14.4	1.64×10^{-2}	1.57×10^{-6}	

comparison in the study validated the environmental benefits of the AAs [100].

AP: Acidification potential, GWP: Global Warming Potential, EP: Eutrophication potential, HTP: Human toxicity, POCP: Photochemical oxidation (summer smog), ODP: Stratospheric ozone depletion

As artificial aggregates are synthetic materials, their production requires various raw materials and processes, which directly or indirectly consume energy and natural resources, impacting human health and the ecological system [101], Life cycle assessment (LCA) can effectively evaluate the extent and significance of potential environmental impacts of artificial aggregates and identify hotspots where improvements and management are needed. Comparative studies in the literature include comparisons among artificial aggregates to select the most suitable option, as well as comparisons with natural and recycled aggregates, where the most appropriate solution is chosen based on both technical and environmental properties [28,94]. For example, artificial aggregates made from 98 % fly ash and 2 % hydrated lime were compared with natural aggregates (basalt) to assess their environmental impact using LCA [102]. Fig. 20 presents the normalized findings of environmental impacts for both types of aggregates, showing that fly ash-based aggregates outperform natural aggregates in all evaluated impact categories. It should be noted that the author studied only the processes involved in the manufacture of artificial aggregates and excluded transportation, whereas natural aggregates were considered from extraction to final size, including transportation. Additionally, the mechanical performance of aggregates was not included in this comparative study.

In summary, the most important factors to consider when evaluating the LCA of artificial lightweight aggregates are: (1) precursor material, (2) content and type of binder, (3) water consumption, (4) electricity consumption, and (5) transportation [94,101]. The use of cement as a binder in artificial aggregates plays a detrimental role in the environmental impacts of CBAAs [94]. For example, an LCA study of CB-CBAAs prepared from engineering muck (heavy silt clay) with fly ash revealed that the environmental impact (GWP) increased by 65 % when the cement content rose from 8 % to 24 % [40]. Altering the composition of the aggregate matrix also changes the requirements for water and electricity consumption, which ultimately influences the environmental impacts of aggregate [101]. For the preparation of artificial aggregates, precursors derived from waste materials such as fly ash sometimes

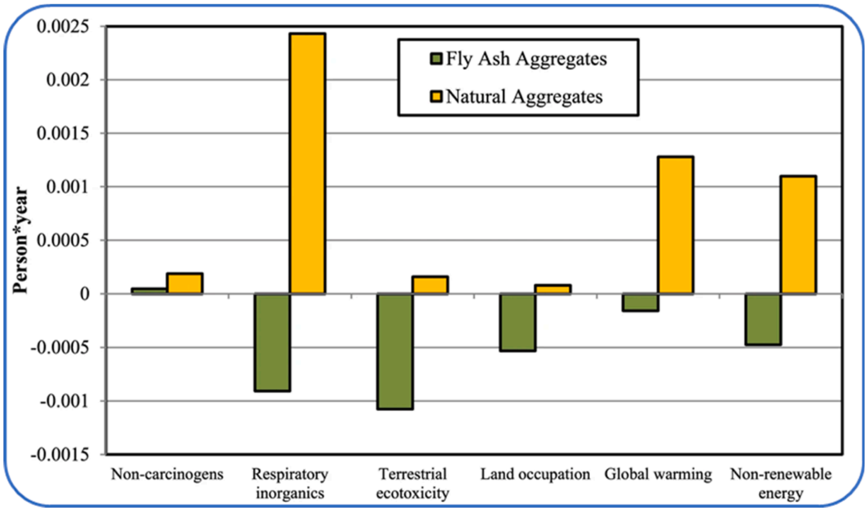


Fig. 20. Comparison of normalized results for FA-based and natural aggregates in terms of environmental impacts. Reprinted with permission from Ref. [102]. Copyright (2024) Springer.

require pre-treatment before use, either to reduce their particle size to a fine powder (mechanical activation) [36] or to wash them with water to remove certain soluble salts, chlorides, sulphates, and some heavy metals [38,101]. In these cases, the LCA analysis of CBAAs indicates that significant environmental impact is associated with pre-treatment, such as the washing of precursors like MSWI-FA [28,101]. The washing of MSWI-FA has a 76 kg CO_2 eq. impact, and 0.13 kg CO_2 eq. for its drying, while the granulation of aggregates has a 9.2 ± 0.2 kg CO_2 eq. environmental impact [101]. These pre-treatment processes involve the consumption of water, energy, and time, as well as additional infrastructure, and the resulting wastewater also requires treatment; if discharged directly into the environment, it may have detrimental effects. In a follow-up study to [101], LCA was performed on surface-treated (double-bonded) aggregates, and the results showed that the aggregates' environmental impact increased by 3–4 % in terms of CO_2 emissions and freshwater consumption [28]. Only a few studies exclusively focus on the LCA of CBAAs; therefore, more attention should be given to the LCA of various types of CBAAs to evaluate their environmental impacts.

4.2. Leaching analysis

Leaching is a process in which a liquid (water or another chemical solution) passes through the solid matrix, dissolving and carrying away hazardous heavy metal ions into the surrounding environment. Fig. 21 schematically shows how the leaching analysis of AAs is performed in the laboratory. Leaching analysis is one of the tools used to determine the environmental impact of aggregates by analyzing the release of hazardous contents (heavy metals and salts) into the environment.

Many waste materials are used in the preparation of artificial aggregates, and these materials contain various hazardous heavy metals. The formation of the C-S-H phase in cement-based artificial aggregates may encapsulate these heavy metals and prevent their escape [104], as C-S-H has lower water permeability [105]. The highly alkaline environment in cement-based aggregates favors the formation of less soluble heavy metal compounds, thereby decreasing leaching [106]. Additionally, heavy metals may also be captured in the hydrate lattice and are strongly bound in the cement matrix [104]. For example, the leaching results for powdered MSWI-BA and MSWI-BA-based aggregates, shown in Fig. 22 indicate that converting waste materials into aggregates significantly reduces leaching behavior [107]. The aggregates solidify and stabilize the heavy metal content within the aggregate matrix and decrease its leaching into the environment [91]. The physicochemical encapsulation by hydration products (Aft, C(N)-A-S-H gel, and U-phase) is the primary reason for heavy metal solidification [108,109]. Phosphogypsum, a hazardous waste material, releases 128.99 mg/L in its raw form, but this amount is reduced to 0.35 mg/L when converted into cement-based aggregates [110]. The formation of ettringite and C-S-H gel in cementitious materials effectively adsorbs and solidifies phosphorus impurities [111].

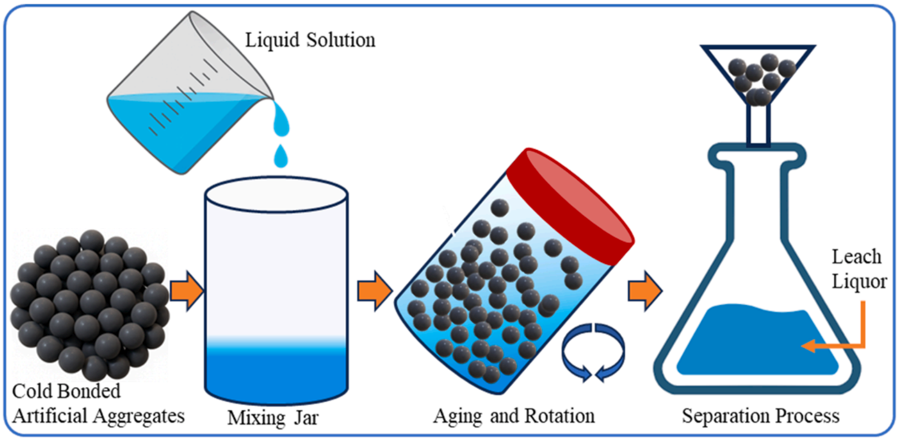


Fig. 21. Schematic diagram for leaching process of artificial aggregates. Adapted with permission from Ref. [103] Copyright (2020) Scientific Reports.

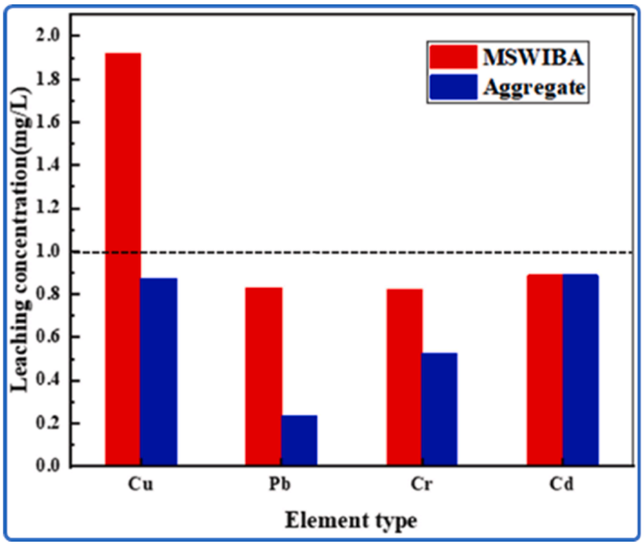
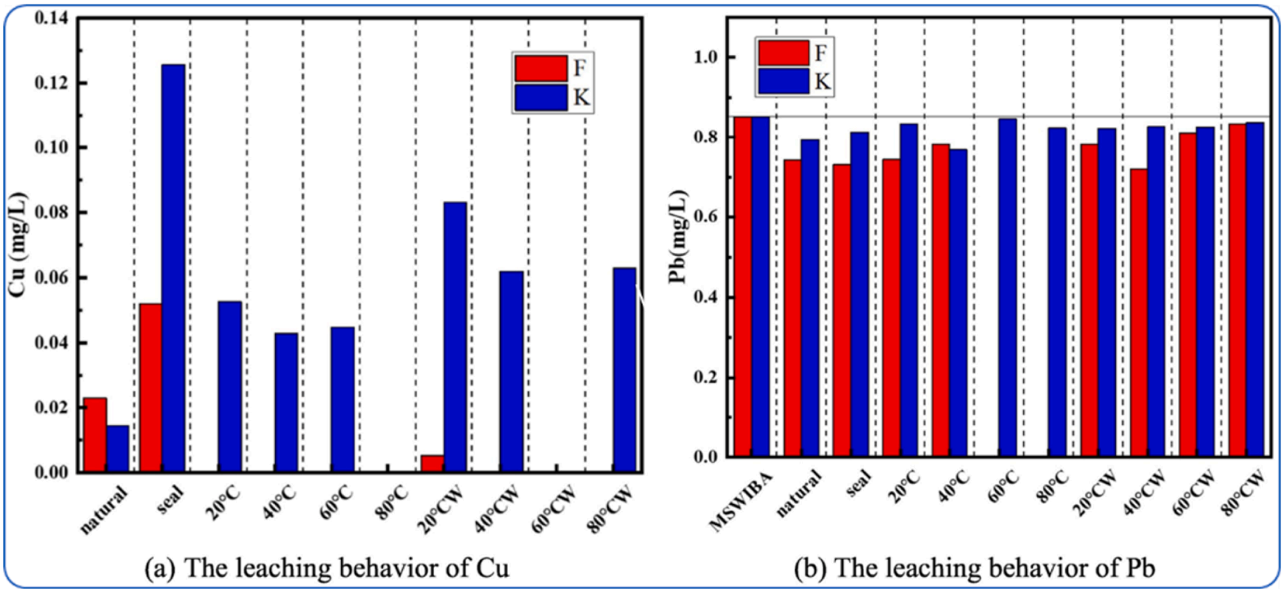


Fig. 22. Leaching behavior of (a) powder MSWIBA and MSWIBA-based aggregate. Reprinted with permission from Ref. [107]. Copyright (2022) Elsevier B·V.

The leaching performance of two cement types, CEM III/B and CEM I/B, was compared, revealing that CEM III/B demonstrates superior performance due to its chemical composition, which includes a higher percentage of CaO and SiO_2. This facilitates the development of increased hydration products (C-S-H) [112], as also supported by [113]. Fig. 23 summarizes the effect of the type of precursor used for aggregate preparation – fly ash-based aggregates (F group) compared with GGBFS-based aggregates (K group)—and various curing methods on the leaching behavior of the aggregates [27]. The formation of secondary hydration products in fly ash-based aggregates contributes to improved leaching behavior compared to GGBFS-based aggregates. The ettringite (AFm), which are double-layer hydroxides of calcium aluminum/iron hydrate, react with hazardous heavy metal ions and form interlayer hydroxyl groups [114]. This is also confirmed by XRD results, where dioptase ($CuSiO_3H_2O$) and dolerophanite (Cu_2O (SO_4)) were observed [27]. An increase in curing temperature favors the formation of hydration products and other compounds with heavy metals [27] which leads to decrease in Cu leaching. Greater formation of hydration products (Aft and Al gels) and enhanced pozzolanic activity result in a denser microstructure, which effectively reduces heavy metal migration [88].

Usually, as the age of cement-based artificial aggregate increases, the leaching of heavy metal ions decreases. However, for some heavy metals such as Cr, Ni, Zn, As, and P, leaching first decreases during the initial 14 days and then increases [115]. It has been suggested that with longer curing times, the water content in the aggregate matrix is either consumed in further reactions (hydration or pozzolanic) or evaporates, leading to the development of more interconnected pores [115]. This situation allows the aggregate matrix to absorb CO_2 from the surroundings, which reacts with C-S-H, breaking the gel structure and releasing previously stabilized heavy metal ions from the aggregate matrix [116].

Besides the curing conditions and chemical composition of the precursor materials, pH also has a significant effect on the solidification and stabilization of heavy metals. A lower pH provides a favorable environment to reduce leaching, and vice versa. The literature suggests that high $CaCO_3$ and lower $Ca(OH)_2$ content in the aggregate matrix contribute to lowering the pH, which facilitates the solidification of heavy metal ions such as copper [27,117]. Additives such as nano-silica also lower the pH of the CB-CBAAs matrix, which improves leaching, particularly of sulphates and molybdenum [118]. The origin (location, source, process, etc.) of the precursor materials also affects the leaching behavior of artificial aggregates [38,119]. Physical properties such as the fineness of the precursor materials also influence the formation of hydration products; a larger surface area results in more hydration, leading to higher initial strength and improved stabilization of hazardous contents [104]. Aggregate physio-mechanical performance is closely related to leaching behavior, as both depend on the porosity and microstructure of aggregates [38]. Surface treatment, such as double-step pelletization, reduces the leaching of heavy metals compared to untreated aggregates; the treatment material fills the pores and develops a dense structure, which reduces leaching [28,31,38,120]. Surface treatments like double bonding via multiple pelletization provide physical barriers around the aggregate surface to prevent the leaching of heavy metal ions [20]. For example, the leaching behavior of CB-CBAAs prepared from phosphogypsum (80 %) and slag (15 %) improved by 43 % when surface treated with an outer layer (50 % slag, 45 % phosphogypsum, 5 % OPC) developed by dual-step granulation [121]. In another study by the same research group, double-step



(a) The leaching behavior of Cu

(b) The leaching behavior of Pb

Fig. 23. Leaching behavior of (a) Cu and (b) Pb elements from F and K group aggregates. Reprinted with permission from Ref. [27]. Copyright (2022) Elsevier B·V.

pelletization surface treatment was compared with slurry soaking for the same aggregates, but in this case, the material used for surface treatment was alkali-activated fly ash-slag. It was observed that slurry soaking reduced sulphate leaching by 86 %, while multiple pelletization reduced it by 90 % [46].

Alkali-activated aggregates more effectively solidify and stabilize contaminants in waste materials used as precursors, compared to the same waste in powder form [58,74]. Alkali-activated cement effectively solidify/stabilize hazardous content in its matrix as compared to the Portland cement [122]. The type of precursor material (chemical composition) affects the leaching behavior of alkali-activated materials; for example, leaching of oxyanionic species (As, Cr, Mo, and V) in geopolymer with higher sodium content as compared to calcium because such composition favors oxyanions dissolution [123]. Also, the high pH of alkali-activation increases leaching of oxyanionic species which however, contradict the statement that some precursor like fly ashes have high pH prior to alkali activation [58]. But on the other side the leaching of elements like Pb, Cd and Ni decrease with increasing the pH and also the BFS presence showed better solidification/stabilization as compared to the fly ash [124]. The NaOH concentration affects the leaching behaviors of alkali-activated aggregates, as shown in Fig. 24 [74]. Aggregates with higher NaOH concentration demonstrate better environmental stability. The higher alkaline environment help in AA reaction and promotes formation of various hydroxides, carbonates, silicates, and aluminates [125]. In alkali-activated aggregates, the presence of the alkali activator results in the formation of polymer gels that encapsulate heavy metals and create a robust polymer molecular chain backbone [126].

The type of precursor affects leaching behavior; for example, blast furnace slag (BFS)-based geopolymers have been found to be better than fly ash-based geopolymers [124]. The main reason is the superior buffering capacity of BFS-geopolymers, due to the formation of C-A-S-H gel instead of N-A-S-H gel in FA-based geopolymers [124]. The type of heavy metal (i.e. Cr, Pb, Ni) and ion charge (anion or cation) affect the leaching process and its interaction with the precursor materials in the aggregate matrix [127]. Curing conditions influences the leaching mechanism of the heavy metal ion, as they affect the rate of chemical reactions and the formation of crystal growth [128]. The type of heavy metal in the precursor materials also shows dependence on the curing conditions [129,130]; for example, in steam curing at 80 °C, copper forms a stable compound [129] but arsenic leaching increases because high temperature and humidity increase its diffusion and migration to the surface [131]. However, curing conditions that encourage the formation of a dense structure affect the physical retention of the heavy metal in the aggregate matrix [132].

The inclusion of cement in alkali-activated aggregate enhances leaching performance by forming additional hydration products alongside geopolymer gels [133]. In literature, studies have been conducted on the leaching analysis of various alkali-activated waste materials, such as ashes and slags, for civil applications [134,135] but studies on alkali-activated aggregates are very limited. Further research should be conducted on the leaching behavior of alkali-activated aggregates to identify specific influencing factors. In the field of alkali-activated materials, most existing research has focused on bulk materials, while relatively few studies have systematically addressed aggregate behavior. Although insights from the broader field of alkali-activated materials can be partially applied to aggregates, such knowledge transfer is not always directly appropriate. Aggregates represent a specific branch of alkali-activated materials, characterized by properties such as porosity and high specific surface area, which may significantly influence leaching behavior.

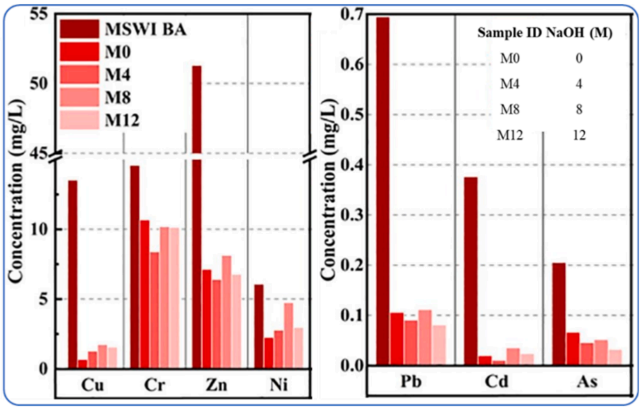


Fig. 24. Effect of NaOH concentrations on MSWIBA+GBBFS-based alkali activated aggregates leaching behavior along with raw MSWIBA. Adapted with permission from Ref. [74]. Copyright (2023) Elsevier B·V.

4.3. *Environmentally friendly and performance-boosting approach*

An environmentally friendly and performance-enhancing approach, known as carbonation or accelerated carbonation, is applied to cold-bonded artificial aggregates. In this process, CO_2 reacts with the aggregate materials, where it is sequestered and forms compounds that improve the performance of the aggregates. The carbonation treatment of artificial aggregates is valuable as it reduces the curing period, increases strength, and stores carbon dioxide [136]. This approach contributes positively to addressing climate change [90,137], as global warming remains an ever-growing concern. Wastes such as steel slag, incinerated ashes, and paper ashes are suitable materials for the preparation of carbonated aggregates [138,139]. The carbonation methods applied to artificial aggregates are broadly divided into two types: the first is “post-granulation carbonation”, where fresh aggregates are treated in a CO_2 environment; the second is the “during and post-granulation carbonation” approach, in which CO_2 is supplied both during the granulation process and after granulation to the fresh granules. Both methods are explained in the following sections.

4.3.1. *Post granulation-carbonation*

The post-granulation carbonation approach, also known as “accelerated carbonation”, has been found useful in the literature and applied to aggregates to increase strength and absorb CO_2. The main purposes are to enhance the properties of the aggregates and to sequester CO_2. The carbonate-able phase in the artificial aggregates absorbs CO_2 to form stable compounds that improve performance [140]. The oxides present in the waste-based aggregates act as a medium to capture CO_2 [141,142]. The calcium silicate phase and CH react with CO_2. An illustrative diagram of post carbonation is presented in Fig. 25.

The post-carbonation process occurs in four steps [50]:

Step 1: The gaseous CO_2 reacts with H_2O and forms H_2CO_3 solution, a process facilitated by the pores in the artificial aggregates.
Step 2: H_2CO_3 immediately decomposes into H^+, HCO^{3-}, and CO^{3-} ions, which lowers the pH value.
Step 3: The CH phase releases Ca^{2+} and SiO^{4-} as calcium silicate dissolves.
Step 4: The dissolution of the CH phase and carbonation of calcium silicate results in the formation of $CaCO_3$, which accumulates on the surface of the aggregates.

The following are the possible reactions that occur when aggregates are placed in a CO_2 environment, depending on the precursor materials [50,143].

$$Ca(OH)_2 + CO_2 \leftrightarrow CaCO_3 + H_2O \tag{1}$$

$$CO_2 + H_2O \leftrightarrow H_2CO_3 \tag{2}$$

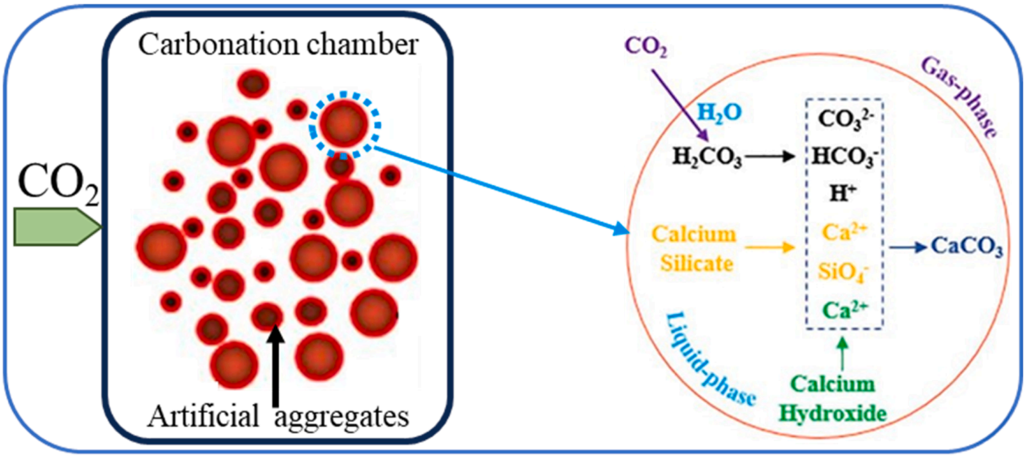


Fig. 25. Carbonation process of artificial aggregates. Adapted with permission from Ref. [50]. Copyright (2023) Elsevier B·V.

$$Ca(OH)_{2H_2CO_3} \leftrightarrow CaCO_3 + 2H_2O \tag{3}$$

$$3CaO{\cdot}2SiO_2{\cdot}3H_2O + H_2CO_3 \leftrightarrow CaCO_3 + 2SiO_2 + 6H_2O \tag{4}$$

CO_2 sequestration reduces the environmental impact of CBAAs and improves the performance of aggregates [50]. For example, Table 4 lists the properties of carbonated and uncarbonated aggregates prepared from wastepaper fly ash (WPFA), clearly indicating that their physical and mechanical properties are enhanced [144].

Fig. 26(a) shows the optical images of the aggregates, clearly indicating that the aggregates have numerous pores (black spots) and a less densified surface before carbonation, while after carbonation, the aggregates exhibit a denser surface [144]. This densification occurs due to the formation of $CaCO_3$, which blocks these pores and forms a layer on the outer surface of the aggregate [115], as illustrated in the schematic diagram of pre- and post-carbonation aggregates in Fig. 26 (c). The internal structure of artificial aggregates is porous due to the entrapment of air bubbles during granulation [144] or due to physical and chemical characteristics of the precursor. The microstructure, with interconnected pores, has detrimental effects on performance; therefore, several treatment techniques are applied to aggregates to mitigate the effects of porosity and improve aggregate performance, such as treatment with CO_2, which reacts with calcium to form $CaCO_3$ and fills these pores to develop a dense microstructure [115]. Exposure of cementitious materials to a CO_2 environment enhance the performance [143], but prolonged carbonation has adverse effects [145]. The SEM micrographs of internal structures of aggregates, along with illustrations, are shown in Fig. 26 (b,d), which had pores and cracks before carbonation, but $CaCO_3$ formation fills these pores and cracks after carbonation.

Precursor materials with a higher content of calcium-based compounds were found to be more effective for carbonation because they react with CO_2 to form $CaCO_3$ [146]. Along with the hydration products in aggregate matrix, calcite precipitation [144,147] acts as binding medium. In general, for CB-CBAAs, as the cement content increases, the strength of the aggregates increases. However, when subjected to carbonation, the strength decreases because higher cement content leads to greater CO_2 absorption, forming a strong and dense layer on the surface of the aggregates which hinders further CO_2 absorption and impairs additional carbonation [148]. In addition, the carbonation reaction is initially very intense due to the higher cement content. It is exothermic in nature increases the temperature of the curing chamber, creating thermal stresses in the aggregate matrix that lead to expansion, affecting the internal structure and causing cracks on the surface of the aggregates, thereby reducing mechanical strength [148]. For example, the mechanical strength of CBAAs prepared from MSWI-FA with 10 % cement was 2.24 MPa, increasing to 3.14 MPa after carbonation. However, when the cement percentage increased to 50 %, the strength dropped from 5.25 MPa to 4.09 MPa after carbonation [148]. Precursors with a higher Ca/Si ratio will form more C-(A)-S-H gel in the aggregate matrix, making it more vulnerable to decalcification during carbonation and resulting in more carbonaceous holes and cracks, which affect the original aggregate structure and negatively impact its strength [74]. Hence, for carbonation, precursors with higher calcium-based compounds are desirable but should be added in an optimum amount to the aggregate matrix to maximize the carbonation effect.

Table 4
Physical and mechanical properties of artificial aggregates before and after carbonation [144].

Specifications	Units	Non-carbonated LWA	Carbonated LWA
Aggregate form	–	Round	Round
Granular class	mm	2–16	2–16
Absolute density	(kg/m^3) (±15 %)	2105	2370
Bulk density	(kg/m^3) (±15 %)	897	902
Compressive strength	MPa (±10 %)	3.15	7.6
Porosity	%	29	21
Water absorption (24 h)	% (±10 %)	27.9	19.8

The addition of organic precursors such as miscanthus powder increases CO2 uptake by the aggregate matrix but decreases its strength if added above the optimum amount [90]. These materials have high intra particle porosity which allows to absorb more water and act as intermedia for CO_2 activation which accelerate carbonation and lead higher CO_2 sequestration [90,149]. However, the amount should be optimised because higher levels of miscanthus powder-like materials themselves have poor mechanical characteristics. The SEM micrographs, together with the schematic microstructure and physio-mechanical characteristics of steel slag (SS) and miscanthus powder (MP)-based aggregate, are presented in Fig. 27. The microstructure shows that the addition of MP increases porosity and allows more CO_2, but also decreases the bonding between slag particles, which reduces aggregate performance [90].

Precursors with a porous morphology generally enhance CO_2 sequestration but do not necessarily improve aggregate performance. When porous material such as biochar (at 5 %, 10 %, 15 %, and 20 %) was added to CB-CBAAs prepared from red mud, the carbonation effectively decrease water absorption from the range of 17.96 %–20.46 % to 16.71 %–17.04 %, and increase bulk density from the range of 1.85 g/cm^3–1.98 g/cm^3 to the range of 2.02 g/cm^3–1.83 g/cm^3. However, mechanical strength decreased from 2 to 1.3 MPa to 1.6–1.3 MPa [150]. The mechanical strength decreases with the addition of biochar because the surface of the aggregates becomes denser after carbonation due to

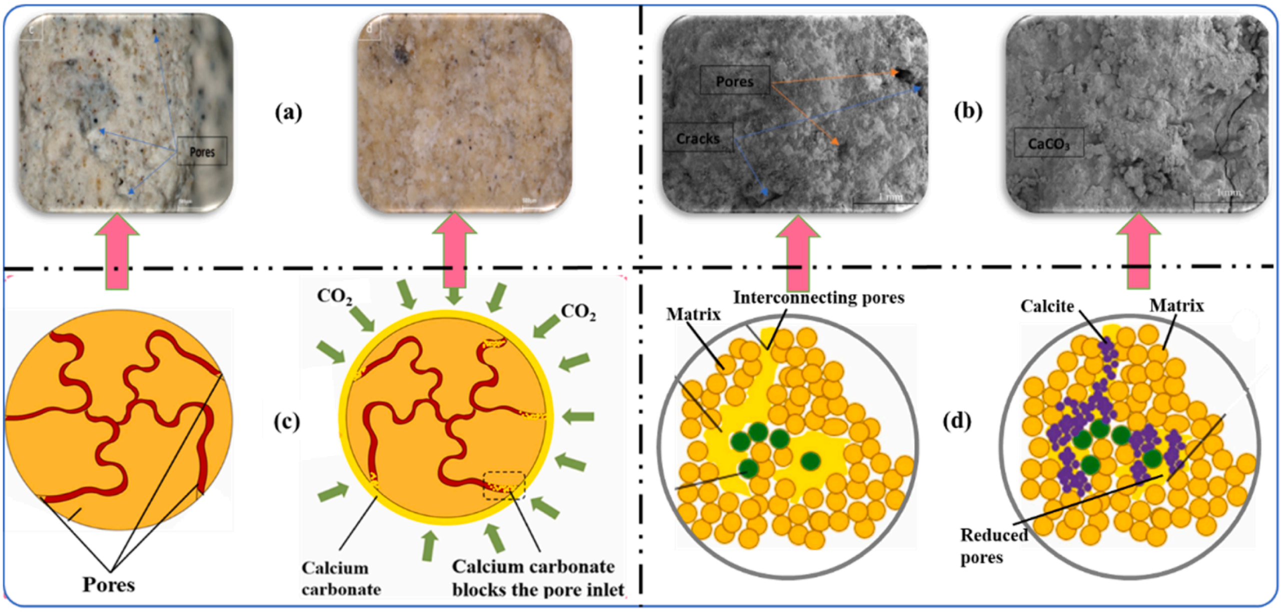
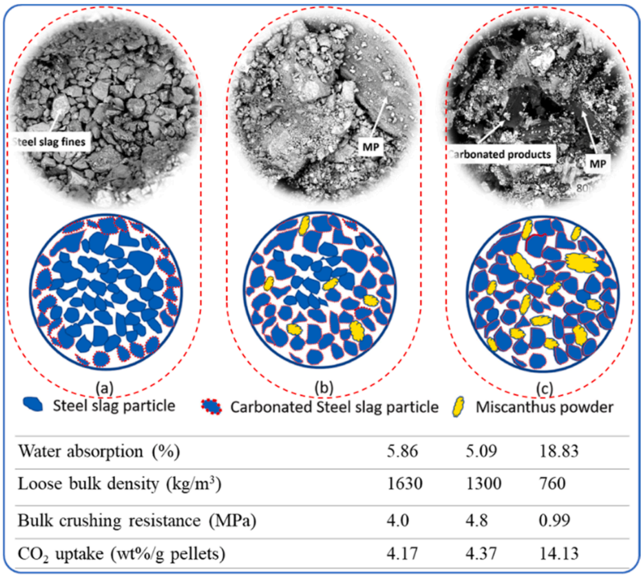


Fig. 26. (a) Optical images, (b) SEM micrographs. Adapted with permission from Ref. [144] Copyright (2022) Elsevier B·V, and (c,d) illustrative diagram of outer surface and internal structure of aggregates pre- and post-carbonation. Adapted with permission from Ref. [115]. Copyright (2024) Elsevier B.·V.



Water absorption (%)	5.86	5.09	18.83
Loose bulk density (kg/m^3)	1630	1300	760
Bulk crushing resistance (MPa)	4.0	4.8	0.99
CO_2 uptake (wt%/g pellets)	4.17	4.37	14.13

Fig. 27. SEM morphology with schematic inner structure of (a) 100 %SS, (b) 90 %SS10 %MP and (c) 65 %SS35 %MP aggregates along with physical mechanical and CO_2 uptake characteristic. Adapted with permission from Ref. [90]. Copyright (2021) Elsevier B·V.

the formation of carbonation products such as calcite. This gives the aggregates a core-shell-like structure, as the calcite forms a dense layer on the surface of the aggregates, causing the densities inside and outside the aggregates to vary and resulting in non-uniform behavior under compressive load [150].

Carbonation was found to be most favorable for enhancing the performance of cement-based aggregates because of the presence of calcium-based components in the aggregate matrix, which react with CO_2 to form $CaCO_3$, while in alkali-activated aggregates, the polymerization gel is responsible for their performance, and the accumulation of carbonation products causes it to shrink and become more vulnerable to defects [74]. Before and after carbonation, SEM and EDS analysis of alkali-activated aggregates is shown in Fig. 28 which clearly indicates that with the formation of carbonation products, cracks appear in the gel [74]. Secondly, the change in the polymer structure occurs due to the formation of sodium carbonate by extracting Na+ from the alkaline aluminosilicate geopolymer matrix, which disturbs the original structure of the aggregate and adversely affects its performance [151].

Papers on the effect of carbonation on leaching behavior are scarce; however, it is argued that the accumulation of carbonation products physically retards the migration of heavy metal ions [136]. This is the area that should be further explored.

4.3.2. *Factors affecting carbonation*

The CO_2 concentration, temperature, humidity, time, and pressure within the carbonation chamber are considered carbonation parameters. These parameters influence the carbonation process and ultimately affect the aggregate's performance. In literature it was observed that faster carbon absorption were observed initially but as the time increase the rate of carbon absorption drops [115]. The slow carbonation in the later stage is due to the slower diffusion of CO_2 into the internal parts of the aggregate matrix [81]. In accelerated carbonation, CO_2 first encounters the aggregate's outer surface and then diffuse slowly along the internal pore network [152]. The development of reaction products densifies the structure, forming more tortuous pore network and slows down the rate at which CO_2 diffuses through the aggregate matrix [81, 153]. Increasing carbonation time will increase carbonation depth, as shown in the phenolphthalein images in Fig. 29 (a) for concrete slurry waste(CSW)-based aggregates [115]. However, the carbonation efficiency (rate) drops significantly. It is evident from Fig. 29 (b) that a high amount of CO_2 is absorbed in the first 7 days, but this decreases to 12–37 % after 56 days for various CSW-based aggregates [115].

Lengthy carbonation is ideal for sequestering CO_2, but when assessing the performance of aggregates, it can occasionally have a detrimental impact on the aggregate's properties. In the literature, it was observed for red mud-based aggregates that as the carbonation duration increases, the density increases and water absorption decreases, but mechanical performance declines after a certain carbonation period [50].The increase in density and drop in water absorption are due to formation of $CaCO_3$ [154], which accumulates in the pores present in the aggregate matrix. For example, for red mud and fly ash-based aggregates, after 6 h of carbonation, a bulk density of 975 kg/m^3 was recorded, which increased to 1019 kg/m^3 after 24 h of carbonation,

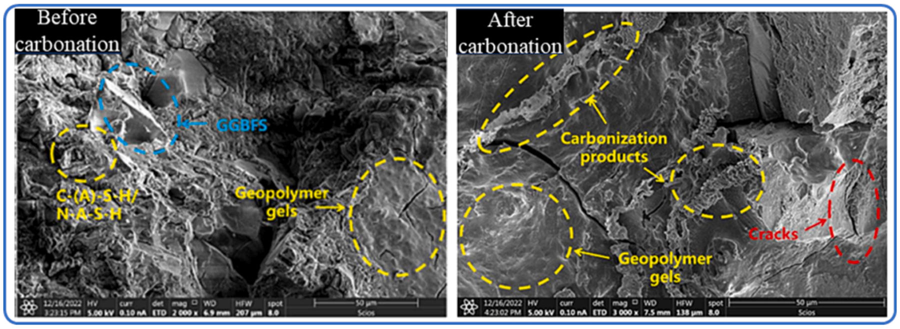


Fig. 28. SEM/EDS analysis (a) before and (b) after carbonation. Adapted with permission from Ref. [74]. Copyright (2023) Elsevier B·V.

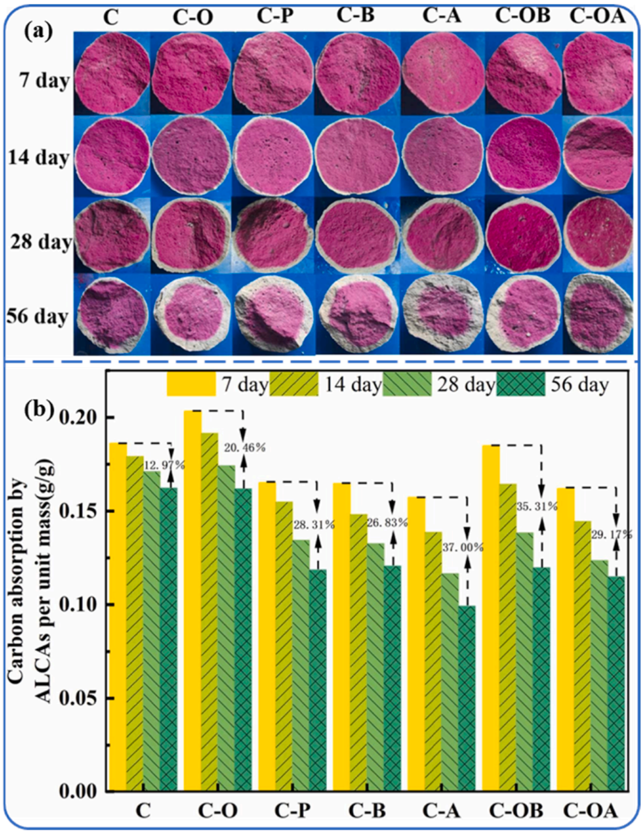


Fig. 29. Phenolphthalein pictures and absorbing content of CO_2 for CSW-based aggregates cured for 7, 14, 28 and 56 days. Adapted with permission from Ref. [115]. Copyright (2024) Elsevier B·V.

while 24-hour water absorption decreased from 10.9 % to 7.8 %. Mechanical performance improves as carbonation time increases from 6 h (7.6 MPa) to 12 h (8.1 MPa) but then decreases to 7.8 MPa as carbonation time increases to 24 h. The decrease in strength with increased carbonation time was attributed to the shrinkage of gel-like products, which causes micro-cracks and reduces strength [50].

The curing temperature in the carbonation chamber also affects the formation of carbon-based products. The temperature during carbonation influences the dissolution rate of precursor constituents, CO_2 solubility, nucleation, growth, morphology, and mineralogy of the carbon-based products [155]. At higher temperatures (but still $\leq$ 100 °C), the dissolution rate of Ca-based compounds from precursor materials such as slag increases [156,157]. It should be noted that the solubility of CO_2 in water decreases at higher temperatures, which might negatively affect carbonation [158]. An increase in temperature therefore leads to opposing effects on two main steps of the carbonation reaction: (1) the leaching of Ca^{2+} and Mg^{2+} from the precursor, and (2) CO_2 dissolution in water. For stainless steel slag-based aggregates, carbonation at high temperatures (60 °C) showed that initially, calcium dissolution from the slag was higher compared to CO_2 dissolution, resulting in the dissolved CO_2 precipitating as $CaCO_3$ due to the higher concentration of Ca^{2+} ions [155]. Such a situation in the aggregate matrix disrupts $CaCO_3$ formation and results in a less dense microstructure with many small pores, which negatively impacts the physio-mechanical performance of the aggregates. At lower temperatures, CO_2 dissolution is higher compared to Ca-silicate, resulting in a higher CO_2 concentration in the aggregate matrix than Ca^{2+} ions. In this scenario, $CaCO_3$ precipitation occurs on slag Ca-rich particles. Such slow and controlled diffusion results in a dense microstructure with a small number of large pores, which imparts better physio-mechanical properties to the aggregates [155]. Fig. 30 schematically details the leaching of Ca^{2+} and CO_2 dissolution, alongside the development of microstructure, with BSE-SEM images of aggregates cured in CO_2 at two different temperatures.

Similarly, CO_2 concentration affects carbon uptake, the formation of carbonation products, and the development of the microstructure, but it has been argued that its effect is not as significant as that of temperature. A high rate of carbonation reaction has been observed at higher CO_2 concentrations, and When CO_2 concentration is lower, the rate of carbonation is not as high, but over time, a high conversion rate is achieved, producing more carbonation products [155].

The humidity during post-carbonation curing affects the solubility of CO_2 and thus determines the formation of $CaCO_3$. Carbonation is an exothermic process which rises temperature and cause to evaporation of water from aggregates and also the some water content used during hydration reaction or absorbed by amorphous silica which cause to reduces the intensity of carbonation [155]. In one study, the wet carbonation method involved placing aggregates in a porous basket and immersing them in water at a 10:1 liquid-to-solid ratio. CO_2 was injected into the water at a flow rate of 0.2 L/min. The aggregates were then placed in a drying chamber for 24 h at 50 °C and 50 % relative humidity [147]. The author observed that bulk density increased by 4 %, water absorption decreased by 14 %, and mechanical performance improved by 8 % in biomass ash and slurry waste-based aggregates. The wet carbonation process was found to be efficient, as water-dissolved CO_2 penetrated the porous aggregate structure, where small $CaCO_3$ particles formed, filling the pores and creating a denser structure [147]. However, the duration of such carbonation is short, so some pores remain partially unfilled, which may slightly affect the aggregate's performance [147]. By comparing dry carbonation with wet carbonation, the latter was found to be more effective. For example, natural curing, dry carbonation, and wet carbonation were compared for CBAAs prepared from recycled concrete powder (RCP) and GGBFS. It was observed that mechanical strength increased by 25–40 % with dry carbonation and by 70–90 % with wet carbonation, compared to natural curing, while the effect on physical properties was not as significant as on mechanical properties. The authors observed that wet carbonation produced dense microstructures because water helps CO_2 penetrate the internal pores of

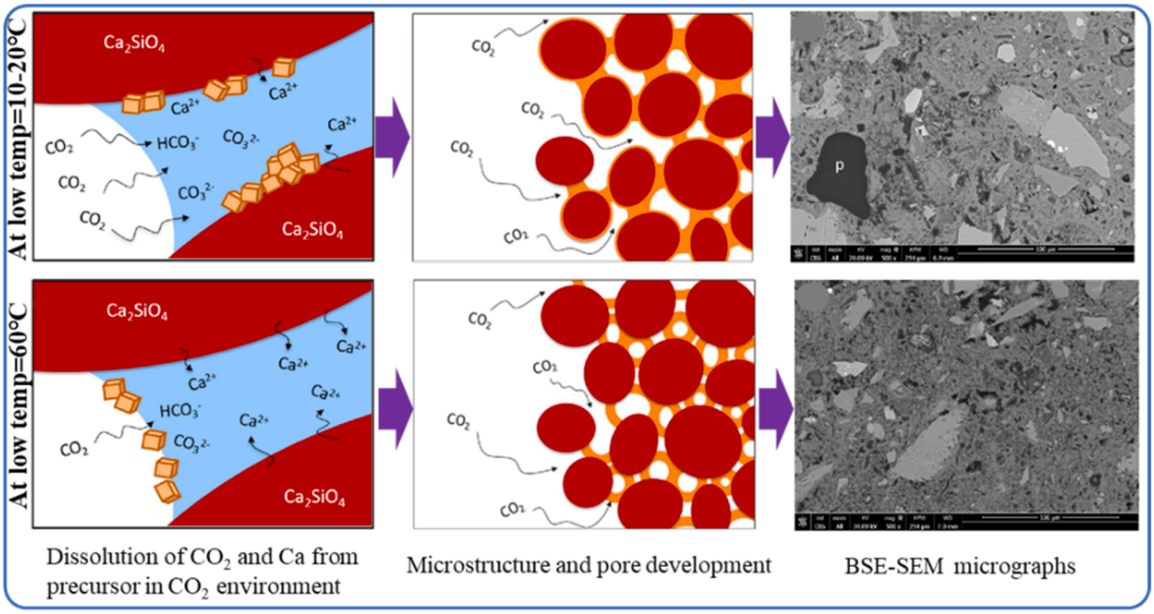


Fig. 30. Schematic diagram of Ca^{2+} leaching and CO_2 dissolution to develop microstructure along with BSE-SEM images. Adapted with permission from Ref. [155]. Copyright (2020) Elsevier B·V.

the aggregates and react with Ca to form $CaCO_3$ [146].

The application of pressure for a short duration during CO_2 curing has minimal effect on the physio-mechanical properties of aggregates, but this effect is more pronounced compared to wet carbonation. Biomass ash and slurry waste-based aggregates are placed in a sealed container, vacuumed to 0.5 bar pressure, and then CO_2 gas is injected at 0.1 bar while maintaining 50 % relative humidity in the chamber [154]. The author observed that bulk density and mechanical strength increased by 8 % and 9 % respectively, while water absorption decreased by 23 %. However, if high pressure is maintained for a longer period, it forces CO_2 to penetrate the innermost parts of the aggregates, ensuring the formation of more calcite to fill the pores and develop a denser microstructure with improved physio-mechanical properties [147].

4.4. *During and post granulation-carbonation*

As the name suggests, carbonation occurs during the granulation process, and after granulation, the aggregates are treated in a carbon environment. An illustrative diagram is presented in Fig. 31. The primary function of carbonation during granulation is to increase the reactivity of precursor materials and binders, decrease pores, and promote the growth of connective pores, thereby enhancing post-granulation carbonation and ultimately improving aggregate performance [159]. However, carbonation is an exothermic reaction that releases heat and requires additional water for granulation. The CO_2 concentration should be optimal to prevent the formation of aggregates with defective microstructure, which would affect the aggregate's performance [139,159]. Very few studies were found on such carbonation treatment; therefore, this article presents two such studies.

Case Study 1 [159]: Different concentrations of CO_2 ($x = 0$, 10, 20, 34, 60, 80, 100 %) were applied during granulation of BOFS-based aggregates and are divided into two groups, CxC and CxA, where "x" represents the CO_2 concentration during granulation. The CxC group aggregates are cured in a carbonation chamber for 4 days at 20 °C, 65 % relative humidity, and 20 % CO2 concentration. After 4 days, the aggregates are further cured in air under ambient conditions. The second group, CxA aggregates, are cured under ambient conditions. The influence of granulation carbonation on the mechanical performance (at 4, 14, and 28 days) of aggregates cured in a carbon environment and under ambient conditions is depicted in Fig. 32 (a) and water absorption after 4 days is shown in Fig. 32 (b). Considering the effect of CO_2 concentration during granulation, the results show that aggregates prepared with 40 % CO_2 concentration during granulation perform better due to the early development of $CaCO_3$ [160,161]. The higher concentration of CO_2 during granulation disrupts the microstructural development of

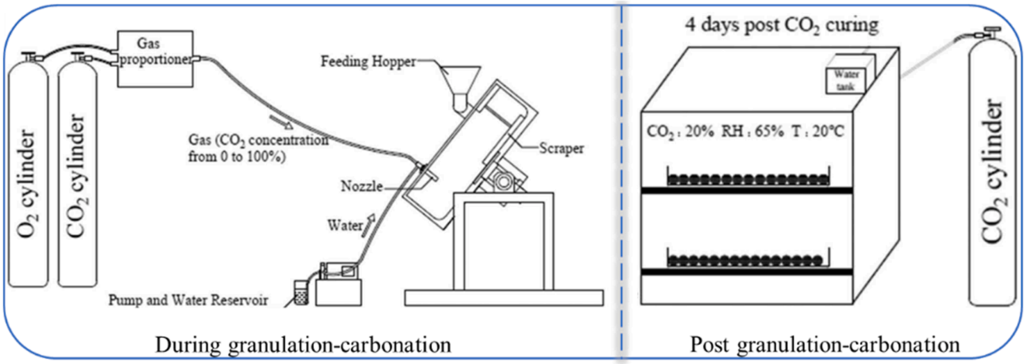
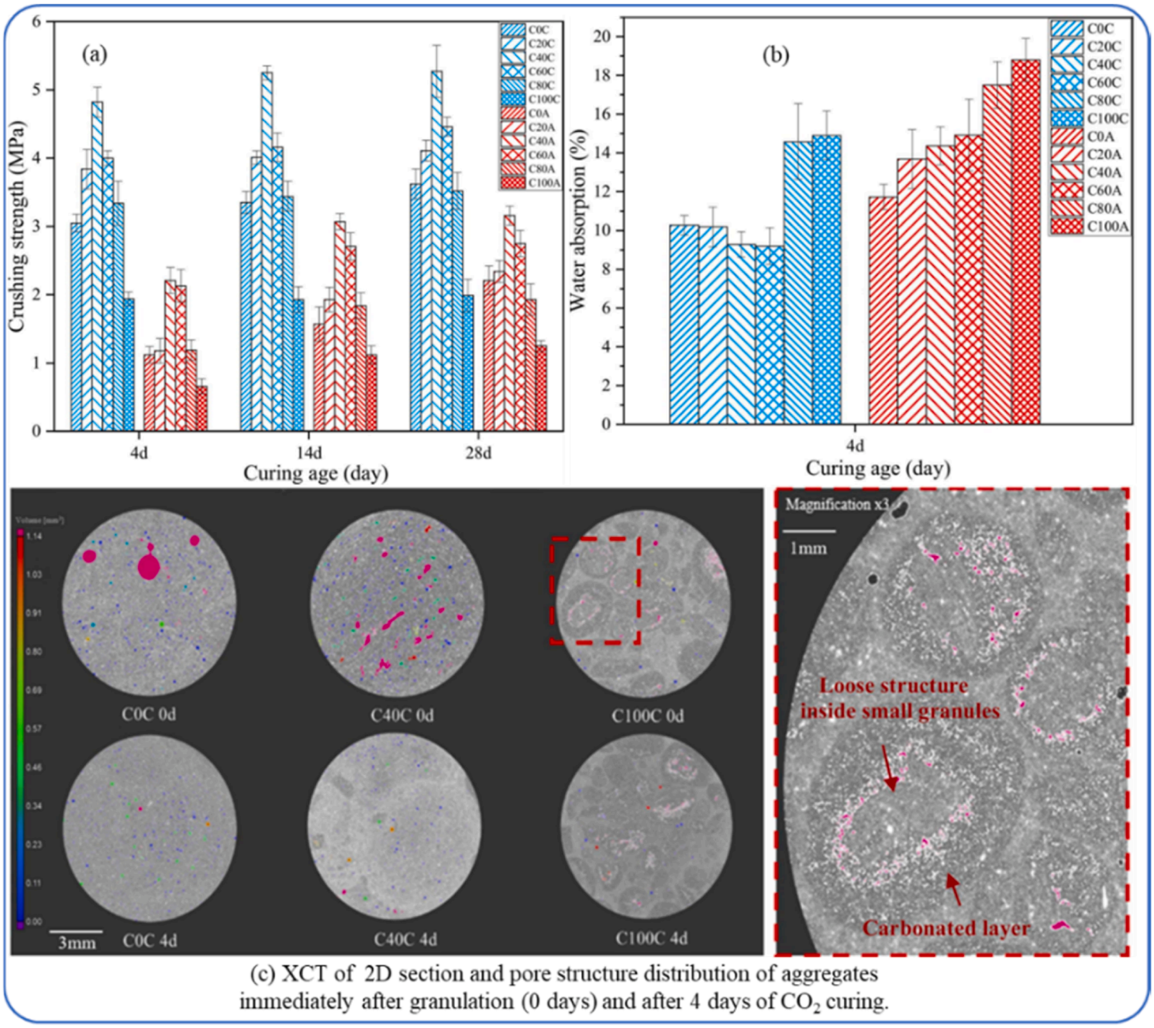


Fig. 31. Illustrative diagram of during and post granulation-carbonation. Adapted with permission from Ref. [159]. Copyright (2022) Elsevier B·V.



(c) XCT of 2D section and pore structure distribution of aggregates immediately after granulation (0 days) and after 4 days of CO_2 curing.

Fig. 32. (a) Crushing strength at 4, 14 and 28 days, (b) water absorption after 4 days and XCT-images of aggregates. Adapted with permission from Ref. [159]. Copyright (2022) Elsevier B·V.

aggregates, as the exothermic nature of the carbonation reaction leads to water evaporation, accelerating the drying of the aggregates and hindering the formation of hydration products, ultimately resulting in inferior aggregate performance [139]. Also, higher CO_2 concentration during granulation causes rapid precipitation of $CaCO_3$, forming a passive layer that prevents further carbonation [155,162].

If the effect of the post-granulation curing regime is considered, aggregates cured in a carbonated environment have shown better performance compared to those cured under ambient conditions. In post-granulation carbonation, the continuous availability of CO_2 allows complete reaction of CO_2 with $Ca(OH)_2$, C_3S, and C_2S present in BOFS, forming $CaCO_3$ and C-S-H. This results in a denser structure, enhancing mechanical strength and reducing water absorption [163,164]. Compared to room temperature-cured aggregates, post-granulation carbonated aggregates contain a higher proportion of these products, resulting in a denser microstructure. The X-ray computed tomography images shown in Fig. 32 (c) (0 day and 4 days CO_2 curing) clearly indicate that aggregates prepared with 40 % CO_2 concentration (C40C) during granulation have more pores (1.15 % connected and 3.03 % total porosity) when measured directly after granulation. The interconnected pores facilitate the diffusion of CO_2 into the aggregate matrix during post-granulation carbonation and ensure its reaction with the precursor materials, which reduces the pores and increases the carbon sequestration capacity. In contrast, for aggregates prepared with 100 % CO_2 concentration (C100C) during granulation, the XCT image shows that their structure consists of granules of various sizes with a carbonated layer inside the aggregate matrix. At higher CO_2 concentrations, CO_2 reacts with BOFS particles and hinders their reaction with the binder and the formation of hydration products. Aggregates with such a structure, regardless of the curing regime post-granulation, negatively affect the physico-mechanical performance of the aggregate.

Case Study 2 [139]: Another study prepared basic oxygen furnace slag (BOFS)-based aggregates and treated them under three different curing regimes; the performances are presented in Fig. 33. The author compared the effects of the three curing regimes on the aggregates:

1) During and post granulation-carbonation (CC): 99.9 % CO_2 concentration during granulation, followed by curing for 4 days in a controlled environment with 20 % CO_2 concentration, 65 % relative humidity, and a temperature of 20 °C.
2) Post carbonation (AC): aggregates prepared in an ambient environment, then cured for 4 days in a controlled environment with 20 % CO_2 concentration, 65 % relative humidity, and a temperature of 20 °C.
3) Ambient conditions (AA).

After curing for 4 days in the CO_2 environment, the aggregates were cured at 50 % relative humidity and 20 °C in air. Their physio-mechanical performance was studied after 7, 14, and 28 days, as shown in Fig. 33. It was observed that no significant improvement

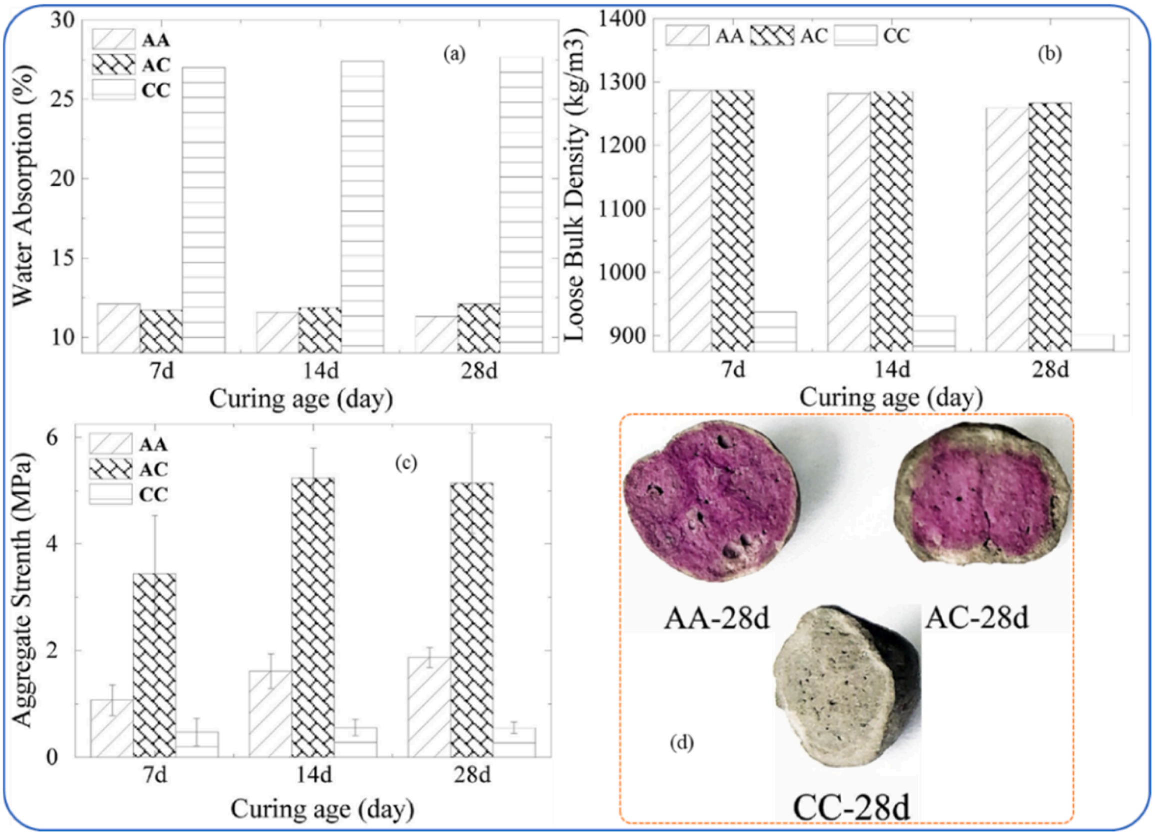


Fig. 33. (a) water absorption, (b) loose bulk density, (c) Mechanical strength and (d) Cross sectional image of artificial aggregates cured at various conditions. Adapted with permission from Ref. [139]. Copyright (2020) Elsevier B·V.

occurred after 14 days.

Among the aggregates, post-granulation carbonated (AC) aggregates have shown better overall performance compared to the other two types. This performance difference is attributed to the microstructure, as AC-treated aggregates have a denser and more compact structure than AA- and CC-treated aggregates, resulting in lower porosity. Additional carbon-based compounds, along with hydration products, were found in the AC aggregates, contributing to their dense structure. Synchronous carbonation during and after granulation results in aggregates with poorer performance because carbonation is an exothermic process that causes water to evaporate during granulation [139]. Lower water content causes some particles to remain unreacted, voids form in the aggregate matrix, and an increase in pores in the structure, all of which contribute to poor performance. The addition of CO_2 during granulation was found to enhance CO_2 sequestration and rapidly initiate the formation of $CaCO_3$ and C-S-H products, which produce a dense microstructure and improved mechanical properties. However, a high CO_2 concentration is not favorable because it causes water evaporation and a rapid reaction with precursor materials, leading to the development of small granules with a carbonated layer within the aggregate matrix and reduced aggregate performance. Researchers need to further explore synchronized post- and during-granulation carbonation in terms of the optimum ratio between water addition and CO_2 concentration to improve aggregate properties and optimize CO_2 sequestration. Application of this procedure to different precursor- and binder-based aggregates is recommended, with subsequent performance and microstructure analysis.

5. Future perspective-critical evaluation

The topic of artificial aggregates is currently receiving significant attention worldwide, with a focus on environmental management, resource conservation, and sustainable construction practices. In the future, several important issues remain regarding the enhancement of their performance and the transition from laboratory settings to commercialization, which will be discussed in detail in the following sections.

5.1. Applications

CBAAs are produced from agricultural and industrial waste, promoting a circular economy and providing alternative materials for the construction sector to support sustainability. The controllable porosity of CBAAs affects their water absorption capacity, density, mechanical properties, and their ability to chemically and physically immobilize hazardous content, making them suitable for various applications. These include lightweight concrete (LWC), porous eco-concrete, CO_2 sequestration, energy storage, and more.

The CBAAs have higher porosity and lower particle density, making them suitable for preparing light-weight concrete (LWC) for use in structural and non-structural applications. LWC, due to its light weight are considered most suitable for high-rise buildings to reduce the structural dead load [165] and improve thermal and noise insulation [166]. The AAs reduce the autogenous shrinkage of LWC due to their higher water absorption and stable desorption ability which help in effective internal concrete curing [167]. However, concerns remain regarding the durability and strength of LWC, which depend on the mechanical performance of AAs, usually observed on lower side [32]. Literature reports that novel techniques such as compression casting significantly improve the performance of LWC [168]. The AAs show strong potential for CO_2 sequestration, which helps to improve their physical and mechanical characteristics, while also contributing to the reduction of GHG emissions [50,136,169,170]. However, it should be noted that the carbonation parameters, such as CO_2 concentration, temperature, and pressure, affects the performance of the AAs [170], which requires further exploration and study.

The incorporation of phase change materials (PCMs) into construction materials enhances thermal energy storage capacity and improves the thermal comfort of the building envelope, helping to reduce the energy consumption of electrical appliances, particularly heating, ventilation, and air conditioning (HVAC) systems [171]. The microencapsulation of PCMs in the AAs offers significant advantages. The pelletization process used to produce AAs allows precise control of their size and ensures uniform dispersion of the PCMs throughout the aggregate matrix [172]. Furthermore, the surface treatment of such aggregates provides an additional outer layer, which prevents leakage of the PCM [172]. However, it is important to note that incorporating PCMs in AAs compromises their strength, as they do not react with the binder or other precursors, leading to increased porosity of the aggregate [172]. This is a critical aspect and future perspective, as it simultaneously addresses the recycling of waste materials and the use of PCMs to reduce the energy consumption of buildings and maintain thermal comfort.

The emerging and innovative application of AAs is porous eco-concrete (PEC). The porous nature of this concrete offers a sustainable method to prevent slope runoff provides, vegetated slope protection [173], reduces dust, noise and water seepage, and can serve as a water purification filter [174]. Recently, the use of PEC for vegetation concrete has increased significantly for planting on slopes along roadsides, dams, and for the restoration of landfill sites to conserve soil, protect water resources, and enhance environmental aesthetics [175,176]. The porous nature of AAs aids in retaining moisture and regulating temperature, thereby increasing plant compatibility and highlighting their potential use in PEC [176]. Additionally, as AAs are synthesized aggregates, their preparation allows the incorporation of fertilizers into their rounded pellets, enabling them to act as nutrient carriers for plants and to release these nutrients slowly and in a controlled manner [177]. However, such attractive and innovative applications significantly impact the mechanical performance of concrete, which is the primary requirement.

AAs are beneficial in many respects due to their engineered synthesis and properties; some of these were highlighted in the above paragraphs. However, it should be noted that while engineering the properties of AAs, the basic mechanical strength is compromised. Therefore, the field of AAs is of great importance, as it helps the ecological system in various ways, from the utilization of hazardous waste to plantation and CO_2 sequestration.

5.2. Barries

For a sustainable future and to combat climate change, it is essential to reduce the over-exploitation of natural resources and pollution. To achieve these goals, materials should be recycled (such as recycled aggregate) and waste materials should be transformed into value-added products (such as lightweight artificial aggregate), supporting the implementation of a circular economy. However, cultural, economic, technological, and regulatory barriers hinder the large-scale use of materials like artificial aggregates [178]. As artificial aggregates are newly developed materials, further research is needed because, from a technological perspective, their performance is unpredictable, while the construction industry requires high-quality materials [179,180]. To enhance their properties, additional processes are required to prepare them, which may burden the economy [178]. Due to technological and economic concerns, the market is also cautious about these materials regarding their durability and effectiveness compared to traditional aggregates [178]. The legal and regulatory framework is also a major concern, as there are either no regulations or insufficient regulations for commercializing and using these materials. Developing a regulatory framework is not straightforward, and regulations vary between countries, adding complexity to the supply chain and making it difficult for the industry to scale up [178,180]. It is important to create an environment that encourages the use of these materials in the construction industry, which requires coordinated efforts from policymakers, researchers, and industry participants.

5.3. Precursors

The most commonly used precursor materials for producing artificial aggregates are industrial and agricultural wastes such as ashes, sludges, slags, sediments, red mud, and cement kiln dust [6]. Among these wastes, fly ash is frequently used to produce artificial aggregates due to its properties, availability, and cost. However, it should be noted that fly ash is primarily used in the production of cement and concrete, followed by bricks and blocks [181]. Additionally, fly ash has many other applications in various fields, including environmental remediation, water treatment, pollutant gas absorption, zeolite synthesis, superhydrophobic powder, and others [182,183]. These divers increase fly ash market demand, which challenges its availability for producing artificial aggregates.

To reduce CO_2 emissions, thermal power plants will gradually be replaced by renewable energy sources in the future, which will also affect the availability of ash. The phased decommissioning of coal-fired power plants is one measure that will reduce ash availability. Geographical location also affects the availability of precursor materials.

5.4. Behavior in cement-matrix

Concrete is the primary matrix in which artificial aggregates are used and affect its fresh properties, mechanical performance, durability, and microstructure. The physio-mechanical performance of artificial aggregates depends on their microstructure, which ultimately affects the mechanical strength and durability of the cement matrix. The use of artificial aggregates in concrete might enhance its workability, thermal insulation behavior, and interface transition zone (ITZ) due to their shape, surface texture, and internal porosity [8]. Artificial aggregates form a continuous and tighter bond with cement, developing an indistinguishable ITZ compared to other aggregates. This may be due to four reasons [8]: (1) the aggregate and cement matrix have similar compositions (both are cementitious materials); (2) water stored in the aggregate provides the necessary internal curing during subsequent hydration of the ITZ; (3) mechanical interlocking forms between the aggregate and matrix due to the penetration of cement paste into the aggregate, resulting in a close bond; (4) a pozzolanic reaction occurs between the fly ash particles (the outer layer of the AA) and calcium hydroxide within the ITZ region. However, despite the development of a strong ITZ, there are additional problems associated with the use of artificial aggregates in the cement matrix. Variability in the characteristics of precursors and the processes used for the preparation of artificial aggregates may result in inconsistent properties, making their use in the cement matrix difficult [8]. Chloride penetration is also a major concern associated with the utilization of artificial aggregates, which limits their application, where sintered aggregates have shown better resistance to chlorination [184]. The durability of concrete with artificial aggregates remains a major concern. Therefore, further research is required to prepare sustainable, strong, and durable aggregates and to reduce the use of excessive energy (as in sintered aggregates) and cement. The performance of artificial aggregates in concrete mixtures requires further investigation, with particular emphasis on identifying the factors that influence their compatibility and long-term durability.

5.5. Economics

Likewise, in addition to the physio-mechanical and durability performance of artificial aggregates, cost is also an important factor. Cost and properties determine the market and attract consumers to the product. The economics of the product involve several costs, including initial investment capital, production or operational costs, and regulatory costs. Establishing a production facility for artificial aggregates requires high initial investment capital to acquire land, construct facilities, and purchase machinery and technology. Operational costs include raw materials, energy consumption, and skilled labor [28,96]. Licenses and permits include technology for controlling and monitoring pollution in accordance with government policies and regulations [28, 96]. Environmental regulation for artificial aggregates is necessary because they are produced from potentially hazardous materials. Compared to natural aggregates, the initial cost of artificial aggregates is higher due to energy consumption, technology requirements, and regulatory requirements. However, on the sustainability index, artificial aggregates perform better than natural aggregates, although factors such as binder use, energy consumption, and distance from raw material sources can reduce these benefits [94]. Market or social acceptance of artificial aggregates depends on the type of aggregate, production process, physio-mechanical performance and most importantly market conditions and behaviour [94].

Research and development are important areas that require substantial investment to further explore the potential of artificial aggregates and to establish a pathway for their commercialization from laboratory scale. In the literature, laboratory-scale cost analysis has been examined, but publicly available cost analysis at the commercial scale – from initial investment in facility development to delivery to the end-user – is lacking for artificial aggregates. The initial investment required to establish an artificial aggregates facility is a major concern, as market and social acceptability are currently insufficient to attract investors. Therefore, the government should develop policies to promote and ensure the use of artificial aggregates in the construction industry, such as through green procurement. Social awareness is also as important as other factors; people should be educated about sustainable approaches. These measures will increase investor interest in establishing larger production facilities for artificial aggregates.

5.6. Legislation

Legislation plays a crucial role in the commercialization of artificial aggregate production from laboratory to industrial scale. Various international, national, and regional guidelines ensure quality, environmental compatibility, and safety in the production of building materials. Waste materials are used to produce artificial aggregates, and the European Union encourages these practices. Green building certification schemes such as LEED (Leadership in Energy and Environmental Design) and BREEAM (Building Research Establishment Environmental Assessment Method) promote the use of sustainable materials, including artificial aggregates. Artificial aggregates must meet several criteria from production to final use. Occupational Safety and Health Administration (OSHA) standards, Construction Products Regulation (EU) No. 305/2011, and the updated Construction Products Regulation (EU) No. 2024/3110 adopted in 2025 must be followed to legally place products on the market. Legislation is essential to ensure that man-made aggregates produced from (hazardous) waste comply with environmental, technical, sustainability, and safety guidelines. It also encourages manufacturers to innovate and improve artificial aggregate production to align with sustainability goals. For innovative building materials, compliance with these legal measures and regulations can be challenging. Therefore, governments and international bodies should provide incentives to manufacturers to attract greater investment.

6. Future recommendation

Artificial aggregates require ongoing investment in research and development to further enhance their performance. To ensure sustainability, low-carbon binders should be identified, and further research should focus on precursor materials to improve both technological and environmental performance. Energy-efficient and renewable energy-integrated plants should be used in aggregate production to reduce indirect greenhouse gas emissions. In addition to technological characteristics, environmental performance should be thoroughly assessed to determine the environmental impact of artificial aggregates and identify areas for improvement. For carbon sequestration, artificial aggregates should be developed to directly absorb greenhouse gases when placed in chimneys or pollution pathways, thereby capturing pollutants and preventing emissions into the environment. Regulatory authorities should establish additional protocols for artificial aggregates to ensure their quality and performance and to promote their use. These protocols should be incorporated into national and international building codes, and governments should provide incentives such as tax relief or subsidies to encourage investment and adoption.

At a commercial scale, integrating artificial aggregate production plants with thermal power plants will reduce transportation needs, and hot flue gases should be used for accelerated curing of cold-bonded aggregates. As flue gases contain CO2, certain types of aggregates will uptake CO_2, reducing the environmental impact of thermal power plants. This technology should be seriously considered, as it offers a sustainable approach in which all waste from the power plant is recycled.

Alternatively, a renewable energy-based production plant for artificial aggregates could be established, using solar panels to generate electricity for plant operations and concentrated solar systems to provide the high temperatures required for aggregate sintering. Both approaches will significantly address the energy and indirect environmental challenges associated with artificial aggregate production, but some concerns remain. For example, the renewable energy approach requires more land and may necessitate building the plant in a different location, increasing transportation needs, while integrating a thermal power plant with the production facility requires advanced technology.

7. Conclusions

In this review article, various types of cold-bonded aggregates are discussed, and the main findings are as follows:

1. Cold-bonded aggregates are suitable as they are cured at ambient conditions or at slightly elevated temperatures ($\leq 80\ ^{\circ}C$). Based on the type of binder used, they are categorized into two main groups: cement-based and alkali-activated aggregates. In cement-based aggregates, hydration products define aggregate performance. The microstructure of cement-based aggregates is highly porous, but increasing the cement content can reduce this porosity, although this has negative cost and environmental impacts. Surface treatment is an effective method that significantly reduces porosity and improves physio-mechanical properties. Alkali-activated aggregates are shown as promise alternative, but the optimal combination of precursors and alkali solution concentration must be determined for each case.
2. These aggregates are made from industrial waste which might contain significant impurities in the form of heavy metals. The vitrified outer layer of high-temperature aggregates locks impurities within the aggregate matrix, while hydration products may encapsulate them in cement-based aggregates. In alkali-activated aggregates, the polymerization gel solidifies and stabilizes these impurities, but this area requires further detailed investigation to make them suitable for construction applications.
3. Carbonation is an effective method to improve the performance of cold-bonded aggregates and to sequester CO_2. In alkali-activated aggregates, carbonation products can shrink the polymerization gel, which is responsible for aggregate strength. However, further research is needed to determine effective carbonation parameters such as temperature, moisture, and CO_2 concentration, and to provide economically viable solutions.
4. Artificial aggregates have significant potential to reduce pollution, reduce economic burdens and promote sustainability in the

construction sector. However, their future still faces considerable challenges and obstacles in social, economic, environmental, technological, and regulatory aspects.

Despite many challenges, artificial aggregates have strong potential to move from laboratory to commercial scale, as they successfully convert waste into value-added materials. The growing demand for sustainable building materials, green building certification, and carbon footprint credits are driving artificial aggregate technology forward. With advances in technology and integration with renewable resources, more environmentally friendly aggregates can be produced. Ongoing research in the field of artificial aggregates is helping to develop and produce aggregates with improved technological properties.

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Informed consent statement

Not applicable.

Data availability statement

Data is contained within the article.

CRediT authorship contribution statement

Jehangeer Raza: Writing – original draft, Visualization, Methodology, Data curation, Conceptualization. **Vilma Ducman:** Supervision, Formal analysis, Conceptualization. **Narinder Singh:** Investigation, Data curation. **Francesco Colangelo:** Validation, Data curation. **Priyadharshini Perumal:** Supervision, Investigation. **Ilenia Farina:** Writing – review & editing, Supervision.

Declaration of interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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