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Review

An Overview of Methods to Enhance the Environmental Performance of Cement-Based Materials

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Abstract: Urbanization and demographic growth have led to increased global energy consumption in recent years. Furthermore, construction products and materials industries have contributed significantly to this increase in fossil fuel use, due to their significant energy requirements, and consequent environmental impact, during the extraction and processing of raw materials. To address this environmental problem, architectural design and civil engineering are trying to implement strategies that enable the use of high-performance materials while minimizing the usage of energy-intensive or toxic and dangerous building materials. These efforts also aim to make buildings less energy-consuming during their useful life. Using waste materials, such as Construction and Demolition Waste (CdW), is one of the most promising approaches to address this issue. In recent years, the European Union (EU) has supported recovery strategies focused on using CdW, as they account for more than 30% of the total waste production in the EU. In this regard, reuse techniques—such as incorporating concrete fragments and bricks as road floor fillers—have been the subject of targeted scientific research. This review will outline various strategies for producing green cement and concrete, particularly emphasizing the reuse of Construction and Demolition Waste (CdW).

Keywords: construction and demolition waste; sustainability; recycling strategies; waste management; cement

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1. Introduction

Cement is a fundamental component of construction material used globally; specifically, Ordinary Portland Cement (OPC) is the primary building material for worldwide housing and infrastructure, making it incredibly significant. It is a binding agent that holds together the aggregates, such as sand and gravel, to form mortar or concrete, the world's most widely used building material [1]. According to the International Energy Agency (Figure 1), the global demand for cement was estimated to be 4.3 Gt in 2021, and is expected to continuously increase by 2030 (30% more respect 2020), driven primarily by the construction of infrastructure and housing in developing countries [2]. Concrete is a ubiquitous material that has played a crucial role in modern construction and infrastructure development. Its versatility, durability, and low cost have made it a go-to building material for many applications, from high-rise buildings and bridges to sidewalks and retaining walls. However, its impact on the built environment and the environment is multifaceted, with both positive and negative consequences.

On the one hand, concrete structures are designed to withstand the forces of nature, such as storms and floods, which makes them safer and more reliable than other materials. This means that people who live and work in buildings made from concrete are better protected from the effects of natural disasters [3,4]. In addition, its use has significantly impacted urbanization, since more extensive and complex structures have created larger cities and urban areas. Conversely, the growth of urban areas has also had negative consequences, such as increased air pollution, the destruction of wetlands, traffic congestion, and the loss of green spaces and other habitats. Furthermore, cement production is one of the leading sources of greenhouse gas (GHG) emissions, contributing about 8% of global carbon dioxide (CO_2) emissions [5,6]. Its production requires substantial heat, using up to 4 GJ of energy per ton of clinker. To produce 1000 kg of cement, roughly 120 kg of coal, with an energy content of 27.5 MJ/kg [7], is utilized, while electricity consumption ranges between 90 and 120 kWh/ton [8]. Therefore, there is a need to develop more sustainable and eco-friendly alternatives to traditional cement and cement-based materials.

Various measures are being taken to reduce the energy and carbon footprint and enhance sustainability to mitigate the adverse impact of cement-based materials on the environment. An encouraging advancement is the employment of carbon capture technology in cement production, which can substantially decrease the emission of carbon dioxide into the atmosphere [9]. Another option is the replacement of significant amounts of Portland cement with less impacting materials, reducing the effects of mining and protecting biodiversity while using natural resources efficiently. It is also crucial to replace raw materials such as limestone and clay with waste from other industrial activities that can be used in cement-making, and would otherwise be sent to landfills. This would result in "green cement" with a lower amount of clinker, an innovative and sustainable solution offering a more environmentally friendly alternative to OPC [10]. It is produced using innovative manufacturing processes and raw materials that significantly reduce energy consumption and GHG emissions compared to conventional OPC. Alternative raw materials can be used instead of traditional ones, including contaminated soil, waste from road cleaning, and other materials containing iron, aluminum, and silica. Some specific examples of such waste materials are coal fly ash and blast furnace slag [11,12]. The demand for green cement is rapidly increasing due to the urgent need to reduce the construction industry's energy and carbon footprint. This industry is one of the most energy-demanding, and thus contributes heavily to global GHG emissions, accounting for approximately 40% of total energy-related CO_2 emissions [13]. Finally, the environmental impact of cement-based materials can be mitigated through sustainable production practices and the use of alternative materials. Recycling concrete waste, bricks [14] and clay into fine and coarse aggregates reduces landfill waste and the extraction of natural raw materials, promoting eco-friendly development. High-quality recycled coarse aggregates can replace up to 100% of natural aggregates in concrete, improving environmental performance. Methods like acid treatment and CO_2 curing enhance the quality of recycled aggregates. Recycled sand from coarse aggregates offers a cheaper and sustainable alternative to natural sand, supporting high-value recycling. This innovation improves the mechanical strength and durability of recycled mortar, boosting sustainable construction practices [15].

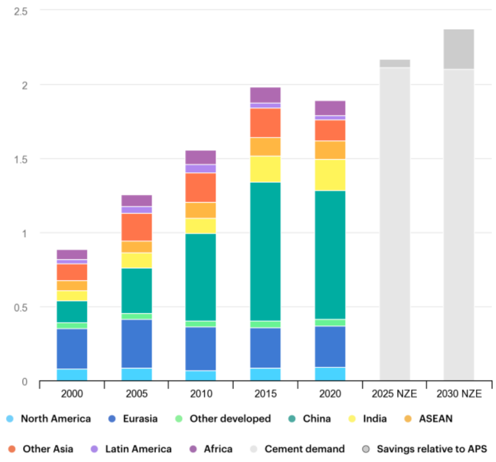

Figure 1. The global demand for cement [16].

2. Carbon Sequestration in Cement-Based Materials

Carbon sequestration in the cement industry is a complex and perplexing topic, with many factors at play, since cement production is one of the largest contributors to carbon dioxide emissions [17]. This makes it a prime target for carbon sequestration efforts, which aim to capture and store carbon dioxide emissions safely and permanently.

At its core, carbon sequestration involves capturing carbon dioxide emissions before they are released into the atmosphere and storing them to prevent their release. This can take many forms in the cement industry, including carbon capture and storage (CCS) technology. It involves capturing carbon dioxide emissions from cement production and storing them underground or elsewhere [18,19]. However, while carbon sequestration may seem like a straightforward solution to the carbon emissions problem in the cement industry, many challenges and uncertainties make it a highly complex and challenging endeavor. For example, the effectiveness of CCS technology in the cement industry has yet to be thoroughly tested or proven, and there are concerns about the safety and sustainability of storing carbon dioxide underground [20]. Moreover, the costs associated with carbon sequestration in the cement industry are significant, and it is unclear who will bear these costs. CCS encounters various technical and financial impediments that must be surmounted to enable its widespread implementation. A crucial financial challenge is that CCS is not profitable, necessitating substantial capital investment. In addition to financial barriers, there are also significant technical challenges associated with CCS, such as the uncertainty surrounding long-term CO_2 leakage rates.

Furthermore, some countries may not have an adequate geological storage capacity for CCS, which may increase transportation and injection costs, particularly for offshore storage. This limitation applies to several countries, including the UK, Norway, Singapore, Brazil, and India [21,22]. Cement producers may be reluctant to invest in CCS technology due to the high costs and uncertainties. At the same time, governments and other stakeholders may be hesitant to provide the necessary funding and support [22]. Furthermore, various technical and logistical challenges are associated with carbon sequestration in the cement industry. For example, capturing and storing carbon dioxide requires significant energy and resources, which can offset the emissions reductions achieved through sequestration. In addition, the process of storing carbon dioxide underground can be highly complex and requires careful monitoring and management to prevent leaks or other environmental risks. Despite these challenges, many experts believe that carbon sequestration is essential for reducing carbon emissions in cement and other carbon-intensive sectors. With suitable investments and policies in place, it may be possible to overcome the technical, financial, and logistical hurdles associated with carbon sequestration and achieve meaningful emissions reductions. However, it is crucial to recognize that the cement industry has no one-size-fits-all solution to carbon sequestration. Depending on the specific circumstances and challenges individual cement producers and regions face, different approaches may be required. For example, some producers may find switching to alternative cement production methods that produce fewer emissions more cost-effective. In contrast, others may need to rely on CCS technology to achieve emissions reductions. Ultimately, the success of carbon sequestration in the cement industry will depend on various factors, including technological advancements, policy support, and public awareness and engagement. While there are no easy answers or quick fixes, the proper calculation of economic and environmental costs for carbon sequestration will be crucial in the choice of transitioning to a low-carbon economy [23].

3. Green Cement

Green cement is a term referring to cement obtained through innovative manufacturing processes and/or using raw materials that significantly reduce energy consumption and GHG emissions compared to traditional Portland cement. There are currently several methodologies for making “green cement”, such as the use of pozzolanic materials (fly ash, slag, etc.) as a replacement for Portland cement [24], and in this work we focus our attention on: geopolymer binders and calcium sulfoaluminate (CSA) cement and cementitious materials with improved self-healing ability.

3.1. Geopolymer Binders

Geopolymer binders are a wide class of binders, generally referred to as alkali-activated materials. They are characterized by the presence of a highly alkaline solution, typically sodium hydroxide or sodium silicate, where aluminosilicates dissolve. These materials have been gaining popularity in recent years since they need less energy than Portland cement for their production. In fact, while Portland cement production requires around 1450 °C, alkali-activated materials require lower temperatures for clay calcination, and commonly use wastes from other industrial process as raw material; for instance, ground granulated blast furnace slag (GGBFS), fly ashes [25], stone muds [26] and even municipal solid waste incineration residues [27]. This can reduce the amount of waste going to landfills and reduces the need for new materials to be mined or extracted. The production of GGBFS is estimated at around 300 million tons per year [28], which is very large even if still much lower than what would be needed to completely substitute Portland cement. Thus, a widespread substitution of OPC with geopolymer-based binders would require using very large amounts of extracted raw materials.

Nevertheless, geopolymer binders seem very interesting due to their interesting set of properties (Figure 2). First, they present good durability, avoiding some of the issues of OPC, for instance alkali-aggregate reaction, which can cause it to break down over time. They also have a higher resistance to chemicals and can be used in harsh environments, such as in the construction of chemical plants or wastewater treatment facilities. Geopolymers also have the potential to reduce economic and environmental costs, since their production is less energy-intensive than traditional cements and requires a lower amount of fossil fuels. The possibility of using waste materials that would otherwise be disposed of further reduces the need for new raw materials and leads to cost savings for the construction industry, and potentially even for consumers [29]. However, some studies [30] suggest that a standard concrete is less expensive than one based on geopolymers, even if this last presents a lower environmental impact.

One of the challenges facing the widespread adoption of geopolymer binders is the lack of standardization and regulation. Traditional Portland cement is regulated and standardized by organizations such as ASTM International and the European Committee for Standardization, ensuring consistency and quality. On the other hand, geopolymer binders do not have the same standardization level, making it difficult for contractors and engineers to use them in construction projects. However, efforts are underway to standardize and regulate them. In 2019, the International Union of Laboratories and Experts in Construction Materials, Systems, and Structures (RILEM) published guidelines for testing and characterizing geopolymer binders. These guidelines aim to provide a standardized approach to the testing and characterization of these materials, which can help to ensure consistency and quality in their use [31].

At the beginning of the development of geopolymers, another important issue was the high temperature needed for the curing. This issue was partially solved using calcium-containing waste materials, which can react at low temperature thanks to the presence of alkali as an activator. This is similar to what happens in Type III cements, where slag is used as a supplementary cement material, and may require activation by portlandite formed by the hydration of Portland cement. Anyway, some issues related to workability and shrinkage are still yet to be solved in order to allow a significant commercial penetration of geopolymer binders.

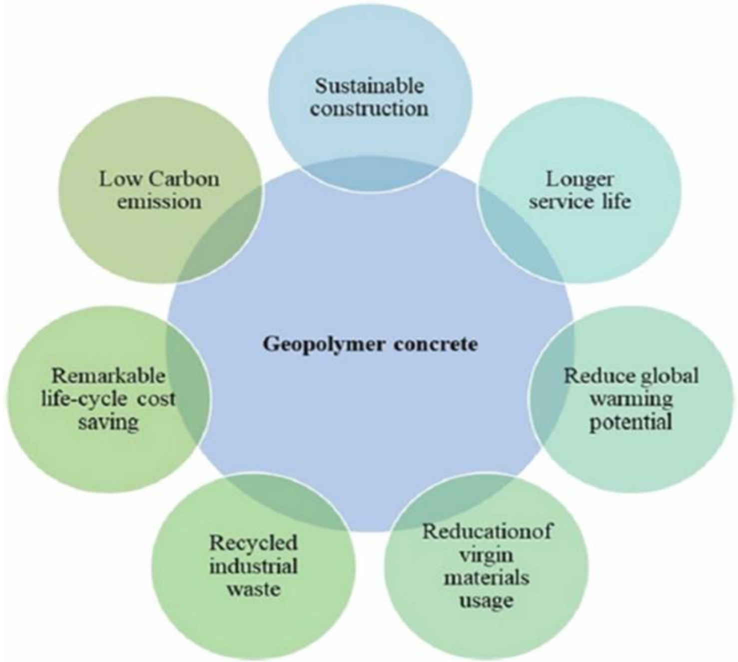

Figure 2. Usefulness of geopolymer concrete in construction [32].

3.2. Calcium Sulfoaluminate Cement

Another approach that was explored in recent years to reduce the energy footprint of concrete regards the use of alternative cements, based on less energy-intensive raw materials. One possibility is to use Calcium Sulfoaluminate Cement (CSA cement), a class of specialty cements that are composed of calcium sulfoaluminate ($4CaO{\cdot}3Al_2O_3{\cdot}CaSO_4$), dicalcium silicate ($2CaO{\cdot}SiO_2$) and gypsum ($CaSO_4{\cdot}2H_2O$) [33,34]. CSA cement is a type of hydraulic cement first developed in the late 1950s [35]. It is made starting from a mixture of calcium sulfate, alumina, and limestone. Unlike OPC, which requires long curing times to achieve its strength, CSA cement can set and harden rapidly, often within hours. This property makes it an attractive option for construction projects that require quick turnaround times. In addition to its rapid setting time, CSA cement offers several other benefits over OPC.

For one, its production seems to emit less CO_2 than Portland cement [36,37]. This is due to the specific composition of CSA cement, which requires less limestone as a raw material, and to a lower clinkering temperature, at 1250–1350 °C instead of 1450 °C for Portland cement. Ren and coworkers [38] suggest that the higher costs of CSA cement with respect to Portland are related to the higher costs of alumina-containing raw materials, and that the use of waste material can lower both the cost and the environmental impact of the CSA cement. It must be considered that Chinese LCA normalization was used in these papers, and that scale factors also contribute to its higher cost with respect to Portland cement. In any case, CSA cement seems to be able to significantly reduce the environmental impact of concrete [39].

Another advantage of CSA cement is its high early strength. It can achieve up to 50 MPa strength within 24 h of casting. This property makes it ideal for projects requiring rapid construction or bearing heavy loads soon after casting. For instance, CSA cement has been used in constructing runways and other infrastructure projects where quick turnaround times are essential; additionally, it has good resistance to chemical attacks, making it suitable for projects in harsh environments [40]. Despite these advantages, CSA cement is not yet widely used in construction. One reason is that it is still more expensive than Portland cement, as discussed above. Moreover, it is not yet as well-understood as OPC, and its properties can vary depending on the specific mixtures used. This variability makes it difficult to predict its behavior in different applications. Another big challenge facing its extensive adoption is the disputable availability of raw materials. In contrast to OPC, which uses readily available materials such as limestone, clay, and gypsum, CSA cement requires bauxite or another alumina-bearing mineral, or waste materials of suitable composition, which may not be widely available in every part of the world. It must be remembered that the produced amount of Portland cement is enormous and that every alternative must be considered in relation to the availability of the resources used for its production.

3.3. Cementitious Materials with Improved Self-Healing Ability

The high annual cost of maintenance and the growing concern about the safety and sustainability of infrastructure in Europe have increased interest in the development of self-healing cementitious materials and preventative repair methods. The appearance of small cracks (less than 300 microns in size) in concrete is inevitable, but does not necessarily cause the collapse of structures. However, small cracks weaken the functionality, accelerate degradation, and reduce the service life and sustainability of such structures. The enormous development that concrete technology has undergone in recent decades has led to the development of materials with extremely low porosity, but has not prevented the intrinsic risk of cracking. On the contrary, high-performance concretes are even more fragile and sensitive to cracking in a short time, compared to concretes that have a lower compressive strength. This behavior has given rise to the study of methodologies that can heal these fissures, which can be divided into passive methods, applied manually after an inspection and allowing only superficial cracks to be sealed, and active methods, incorporated into fresh concrete and therefore allowing both internal cracks and surface cracks to be healed. The latter techniques are also called self-healing techniques (Figure 3).

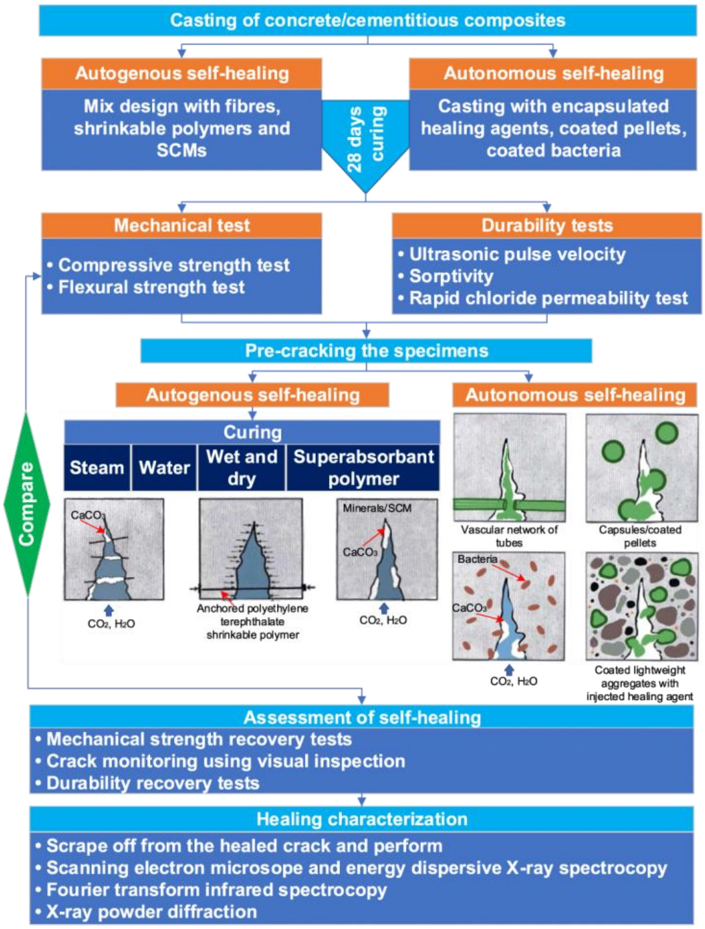

Figure 3. Self-healing framework [41].

The autogenous ability of concrete to repair itself is based on traditional constituents of the cement matrix, but can also be induced by targeted additions to the mix. This phenomenon was observed for the first time in 1836 by the French Academy of Sciences when the autogenous repair of concrete was highlighted in pipes, water retention structures, etc. [42]. The mechanisms that contribute to the autogenous repair of a crack, when it has formed and is exposed to water, are mainly twofold: the continuous hydration of anhydrous cement grains and the precipitation of calcium carbonate crystals ($CaCO_3$) on the edges of the crack, following the chemical reaction between the calcium ions (Ca^{2+}) of the cement matrix and the carbonate ions (${CO_3}^{2-}$) available in water, or carbon dioxide from air in contact with the damaged area. The autogenous repair induced by the continuous hydration of anhydrous cement grains is very useful, as the new hydration products have similar mechanical properties as the primary C-S-H gel that are in any case higher than those of calcium carbonate precipitates. However, the conditions for the formation of secondary C-S-H are different because the nucleation and growth take place on the edges of the cracks, and not in the bulk of the cement paste, as well as the fact that the water/cement (w/c) ratio could be much higher in the case of water arriving from the external environment. Autogenous repair is effective for small cracks between 10 and 100 microns, sometimes up to 200 microns, but only in the presence of water. The type of cement seems not to be important, but the clinker content determines the release of Ca^{2+} ions and the subsequent ability of the matrix to form calcium carbonate-based precipitates. On the contrary, the silicate additions have an effect depending on their nature and quantity in the mixture, related to the characteristic pozzolanic reactions with the Portlandite, which affects the duration of the self-repair mechanisms [42–44]. Furthermore, concretes with high mechanical resistance, prepared with a low w/c and a high binder content, contain many anhydrous cement grains that can potentially produce significant quantities of C-S-H. Finally, the age of the concrete also has an influence, as for short times, more anhydrous grains are available, while for longer times the formation of $CaCO_3$ as a self-healing agent will prevail. To conclude, autogenous repair can be favored by specific additions, such as the addition of blast furnace slag and fly ash, or porosity-reducing additives acting through crystallization (the so-called crystalline admixtures), which react with water to form insoluble precipitates (based on modified C-S-H and a calcium-based hydrated compound) in the pores and superabsorbent polymers with the ability to swell in the presence of liquids (swelling up to 1000 g/g).

The autonomous repair of concrete is based on the incorporation of various microcapsules (<1 mm), whose rupture releases a repairing agent contained in it so as to seal the crack. Various polymeric capsules have been studied (based on: urea, melamine or phenol and formaldehyde [45–47], polyurethane [48] resin, polystyrene [49], polyvinyl alcohol [50], acrylates [51], or even pig gelatine/gum acacia [52] or silica [51]) containing various repairing agents (epoxy, polyurethane or acrylic resin, calcium silicate, colloidal silica or calcium sulpho-aluminate). However, polymeric shells and epoxy-based cargos are the most widely investigated systems. Microcapsules significantly affect the viscosity of the fresh mix, while after 28 d of curing, the compressive strength and the elastic modulus show a consistent decrease with the increasing concentration of microcapsules [52]. The literature survey shows that the mechanical recovery rate of cracked samples is roughly proportional to the content of the microcapsules [43].

Macrocapsules based on fibers (with external diameter 1 mm and length 100 mm) or glass tubes (with external diameter 3 mm and length 100 mm) filled with cyanoacrylate resin or sodium silicate have been also studied [53,54]. However, glass tubes may be subjected to the silica alkali reaction. To avoid this inconvenience, ceramic capsules [55], extruded EVA [56], PLA, PMMA, PEG [57], or cement tubes [58] with external diameters up to 8.4 mm and lengths up to 5 cm, have also been tested. When incorporated in a self-compacting concrete, cementitious macrocapsules (with a of 1.6 vol%) showed a compressive strength that was not significantly influenced by the presence of the tubes, whatever their orientation with respect to the load direction [59]. The average sealing efficiency ranged from 54 to

74% for the samples containing cementitious capsules. However, although promising, these technologies based on the micro- and macroencapsulation of repairing agents present the limit of the actual durability of the encapsulated repairing agents over time. Finally, microcapsules are also limited to one-time use, contrarily to macrocapsules.

Thus, thirty years ago [60], vascular networks (VN) based on hollow fibers were proposed: they operate according to the same healing mechanism as capsule-based systems, but with the advantage of a continuous external supply of a healing agent to the damaged zones within concrete. Moreover, the delivery of a healing agent through a vascular network can be done under pressure, further increasing the efficiency of repair, and with the potential to allow multiple healing cycles. Recently [61], thanks to the ability of additive manufacturing to produce complex geometries, ductile-porous 3D-printed VN were investigated. Load regains up to 56% and stiffness recovery up to 91% were achieved with a polyurethane resin as the healing agent. Combining traditional fabrication techniques like extrusion or injection molding for manufacturing linear pipes and additive manufacturing to produce branched parts seems a good compromise to save time and money in view of the possible large-scale diffusion of VN in the future.

A last very interesting solution for the autonomous repair of concrete involves the use of ureolytic bacteria capable of decomposing urea into ammonium/ammonia (Equation (1)) and carbonate ions. If calcium ions are available in sufficient quantities, then calcium carbonate precipitates (Equation (2)) [62]:

$$CO(NH_2)_2 + 2H_2O \xrightarrow{\text{ureolytic bacteria}} 2NH_4^+ + CO_3^{2-} \tag{1}$$

$$Ca^{2+} + CO_3^{2-} \rightarrow CaCO_3 \tag{2}$$

Theoretically, one mole of calcium carbonate can be formed if one mole of urea is present. However, the process is strongly controlled by the enzyme that produces urease (the kinetics of the reaction are 10^{14} times faster when it is present).

The precipitation of calcium carbonate is also induced by the reduction in nitrates in oxygen-poor environments via denitrifying microorganisms (Equation (3)):

$$5HCOO^- + 2NO_3^- \rightarrow N_2 + 3HCO_3^- + H_2O \tag{3}$$

The self-healing of concrete due to the precipitation of $CaCO_3$ by bacteria is based on the following parameters: adequate pH value and specific nutrients for bacterial cells. For nitrate-reducing bacteria, calcium formate and calcium nitrate are used as nutrients. After 56 days of immersion in water, the sealing of cracks with a width of up to 480 ± 16 µm could be observed [63]. Nitrate reduction can also lead to the production of nitrite ions, known corrosion inhibitors of steel reinforcement (Equation (4)):

$$2HCOO^- + 2NO_3^- + 2H^+ \rightarrow 2CO_2 + H_2O + 2NO_2^- \tag{4}$$

The bacteria *Pseudomonas aeruginosa* and *Diaphorobacter nitroreducens* can survive in mortars if inserted into expanded clay granules, diatomaceous earth or activated carbon, for example. The company Basilisk, a spin-off of Delft University in the Netherlands, markets some bacteria of the *Bacilli* genus capable of producing spores that can remain dormant for up to 200 years [64]. When fed with calcium lactate, they precipitate calcium carbonate (Equation (5)):

$$CaC_6H_{10}O_6 + 6O_2 \rightarrow CaCO_3 + 5CO_2 + 5H_2O \tag{5}$$

The carbon dioxide from the reaction can in turn react with portlandite to produce additional calcium carbonate. Aerobic metabolic degradation requires the presence of oxygen; therefore, a limited availability of this gas decreases the quantity of precipitated calcium carbonate. However, the absence of oxygen slows down the rate and risks of corrosion of steel reinforcement, and the presence of aerobic bacteria will still prolong the service life of reinforced concrete structures in environments favorable to corrosion.

3.4. Potential Benefits, Applications and Challenges

As discussed in the previous paragraphs, two possible “green” cements are being envisaged in order to reduce the environmental impact related to Portland cement production. They could be used in a wide range of applications, including concrete, mortar, and grout, making them a versatile alternative to traditional cement. It is important to consider the specific characteristics of these cements in order to understand their potential advantages and drawbacks in construction and building applications.

Durability and chemical resistance seem to be improved with respect to OPC, allowing their use also in harsh environments, while the high strength of CSA and high-calcium geopolymers makes them interesting for structural applications. This reinforces the interest in these materials, which was initially based mostly on their lower embodied energy and carbon footprint, and the possibility of using waste materials, either for the cement paste preparation (in the case of geopolymers) or as raw materials during the clinkering process (in the case of CSA). Workability issues are to be considered, but the two cements cover both the short and long setting ranges, such that for fast curing applications, CSA would be preferred, while geopolymers are preferred in slow curing.

On the whole, the use of green cement could simultaneously benefit society, the environment and the construction industry, thanks to reduced energy consumption and carbon footprint, improved waste materials use and an improvement in circularity. Another significant issue raised in particular for geopolymers is the health and safety of cement industry workers, but high-calcium geopolymers allow one to avoid the use of dangerous highly alkaline solutions, obtaining acceptable materials from the safety point of view.

The use of these green cements in reinforced concrete is still hampered by the scarce knowledge about the corrosion behavior of steel inside both CSA and geopolymer-based concrete [65,66]. New types of fibers, either natural or artificial, are being proposed, but the possibility of substituting steel into reinforced concrete is not yet scientifically substantiated, in particular due to the difficulty of preparing reinforcements that are continuous, strong, tough, and durable [67,68].

Another important issue related to green cement is that, currently, its production is typically more expensive than traditional cement due to the scale factor, and the requirement of alternative raw materials and innovative manufacturing processes. Specific resources may not always be available, secondary raw materials from waste have often varying and non-homogeneous compositions, and legislation must be amended to allow for the recycling of waste in construction materials. Processing plants must be built and upscaled, which is difficult due to the competitiveness of traditional cement, which is a cheap and well-known material with a very standardized production process. All these issues make green cement less attractive to cost-conscious builders and developers who prioritize cost over sustainability. The request to reduce the construction industry’s energy consumption and carbon footprint has increased the demand for green cement, but currently, it remains a niche product, even if the global market for environmentally sustainable cement is expected to expand significantly in the coming years, driven by rising interest in sustainable and environmental friendly materials. In fact, a significant push toward the use of this cement comes from governments and regulatory bodies worldwide, which are promoting its use through policies and incentives. For example, the European Union aims to reduce GHG emissions from the construction sector by 60% by 2050, creating more demand for green cement [69].

4. Construction and Demolition Waste—CDW

Sustainable construction practices have emerged as a response to the significant natural resource consumption associated with traditional building and construction technologies. These practices aim to repurpose industrial waste and by-products to minimize construction’s environmental impact and protect valuable resources. The waste materials resulting from construction and demolition operations are collectively referred to as CDW. Due to their enormous volume, generating adverse environmental and economic effects, they are regarded as one of the more significant challenges in the construction industry. Indeed, the escalating amount of waste generated and its disposal process negatively impacts the environment and society; this category of waste represents a significant portion of global waste, accounting for between 30% and 40% of the total solid waste, with a global net use rate between 20% and 30% [70]. In the world context, the United States' recovery rate is approximately 70%, while in China, it remains low, at less than 5% [71]. In the United States, total CDW was estimated to be 600 million tons in 2018 [72], while China generates approximately 2.4 billion tons of CDW every year, which accounts for roughly 40% of the total urban waste produced in the country [73]. The rapid urbanization in China has led to increased CDW generation, resulting in significant pressure placed on waste management systems and a severe "garbage siege" phenomenon prevalent in many urban areas [74]. The low utilization of CDW can be attributed to several factors, including the lack of reuse and recycling design for buildings, insufficient recovery facilities in some areas, and low demand for some materials due to regulatory restrictions. The competitiveness of CDW recycling can be improved through intrusive measures, such as increasing raw material prices or imposing taxes, but also by simply establishing end-of-waste criteria for specific CDW fractions. In the European Union, for example, approximately 3 billion tons of waste are generated each year, with one-third of this amount originating from construction and demolition activities [75,76], with an average recovery rate of almost 50%. Nevertheless, it varies significantly among member states, ranging from 10% to 80%. For instance, Italy has a recovery rate of almost 80%, France at 48%, Spain at around 40%, and Germany at 34% [77].

4.1. CDW Composition

Depending on the source and separation methods, CDW primarily comprises inert mineral materials with varying amounts of other components. However, definitions and compositions of CDW can vary from state to state. It can be broadly classified into five categories: metal, concrete and mineral, wood, miscellaneous, and unsorted mixed fractions [78]. More specifically, these waste materials may include concrete, bricks, tiles, ceramics, wood, glass, plastic, bituminous mixtures and tars, ferrous and non-ferrous metals, soils, stones, insulation materials, gypsum-based materials (such as plasterboards), chemicals, waste electronic and electrical equipment (WEEE), packaging materials, and hazardous substances. Hazardous substances commonly found in building materials include asbestos, lead-based paints, phenols, polychlorinated biphenyls (PCBs), and polycyclic aromatic hydrocarbons (PAHs). Many substances, such as insulation, roofs, tiles, and fire-resistant sealing, are often used in conjunction with concrete to complete the structure and finishes. The composition of CDW varies significantly depending on factors such as local typology, construction techniques, climate conditions, economic activities, and technological advancements in the area [79]. Additionally, the composition of CDW changes over time due to aging buildings and low-quality structures built between the 1960s and 70s, which are now reaching the end of their lifespan and require demolishing [80]. Therefore, defining a standard composition representative of a large region is one of the biggest challenges to face. Figure 4 depicts the average composition of CDW.

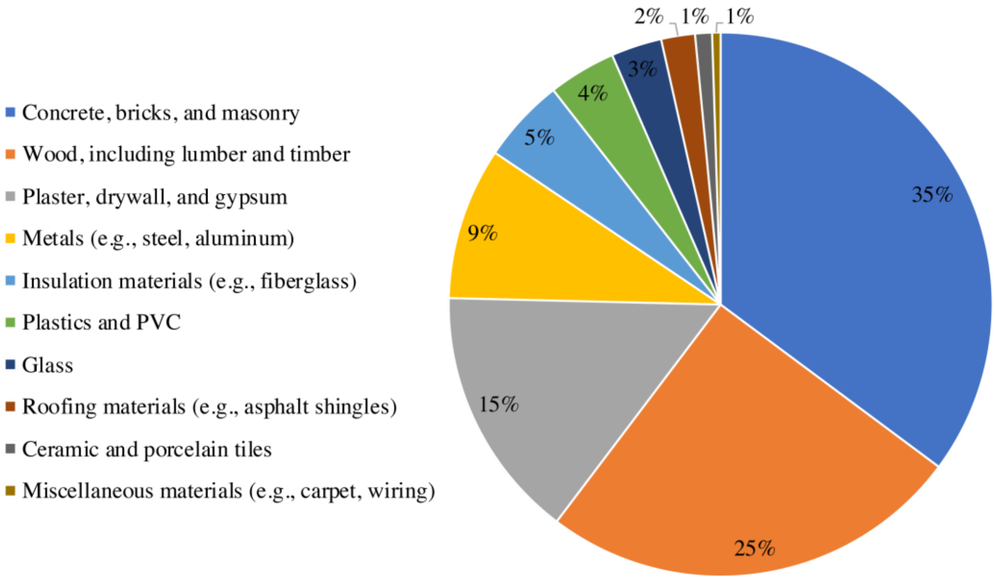

Figure 4. CDW average composition.

4.2. CDW Processing

Recycled aggregate (RA) production involves various processing procedures, including crushing, screening, sorting, and washing [81]. Each of these procedures is critical in ensuring that the resulting material is of high quality and suitable for construction projects. Generally, two types of plants are used to process CDW into recycled aggregates: stationary and mobile. The first one recycles CDW using fixed equipment, while mobile recycling machinery is transported to the worksite to recycle waste on-site [82]. Both plants use the same equipment, such as screens, crushers, and magnetic separators. Stationary plants have the disadvantage of being located far from the demolition site, but are generally more productive than mobile plants. The increased transportation burden is compensated by the product’s better quality and the plant’s higher capacity. Further, stationary plants can also process natural aggregates and have a higher capacity than mobile ones, leading to lower processing costs for recycled aggregates due to economies of scale.

The first step in processing recycled aggregates is crushing, which involves using a crusher to break down the waste material into smaller pieces. The crusher used in this process can be either primary or secondary. The primary crusher is used to break down larger pieces of waste material, while the secondary crusher is used to further reduce the crushed material size [83]. After the crushing process, the material is then screened to remove any contaminants that may be present. The screening process involves passing the crushed material through a series of screens with different mesh sizes. The screens are designed to separate the material into different sizes, with the larger pieces being returned to the crusher for further processing [84]. Once the material has been screened, it is sorted (manually or using automated equipment) to remove non-aggregate materials, such as plastics and wood. This process is critical to ensuring the resulting material is of high quality and suitable for construction projects. However, not all unwanted particles are expected to be removed.

The final step in the processing of recycled aggregates is washing. This process involves the use of water to remove any remaining contaminants that may be present in the material. The washed material is then dried to remove any excess moisture before being used in construction projects. Still, this step is not always carried out, because it also presents significant challenges when it comes to disposing of the resulting mud, including high costs and complex administrative procedures. For this reason, it is customary to find recycled aggregate shielded with fine dust generated during the grinding process [85].

Strict quality control procedures are required after the crushing process to ensure that the RA produced are high-quality and suitable for construction projects. These procedures involve monitoring the production process, testing the material, and inspecting the final product.

5. Recycled Sand from CDW in Mortar and Concrete

In the last few decades, there has been significant research on the properties of recycled sand (RS) obtained from construction and demolition waste (CDW) and its potential application in the production of concrete and mortar. Numerous studies have shown that RS can be used effectively as a substitute for normal aggregates in cement-based materials despite some implementation problems, mainly in mechanical strength and workability areas. Using fine crushed concrete as a substitute for traditional aggregates in concrete leads to a reduction in strength of 15–30%, depending on the replacement level (ranging from 25 to 100%) [86]. However, using a fixed rate of water-to-cement ratio leads to decreased compressive strength and increased drying shrinkage, but improves the resistance to chloride ion penetration compared to control concrete [87]. Other studies argue that using fine recycled concrete aggregates (FRCA) does not significantly impact the mechanical properties of concrete in replacement ratios up to 30% [88]. The effects of using recycled fine aggregates (RFA) on the properties of concrete containing either natural or recycled coarse aggregates [89] could reduce the compressive strength of the resulting concrete, regardless of whether the coarse aggregate is natural or recycled. Still, using RFA had a more significant impact on the properties of concrete containing recycled coarse aggregates, which exhibited lower compressive strength and higher water absorption than concrete made with natural coarse aggregates. The researchers also noted that using RFA led to a decrease in the density of the concrete. Overall, the study suggests that using RFA in concrete can be a viable option for reducing the environmental impact of concrete production. Nevertheless, careful consideration of the properties of the recycled aggregates and adjustments to the mix design are necessary to ensure that the resulting concrete meets the required performance standards.

Research was also conducted on the use of recycled sand (RS) in the manufacturing of mortars. The properties and amount of the fine aggregate used strongly influence mortar's rheological properties and workability [90]. The total quantity of material finer than 0.08 mm in the dry mix can be used as a control parameter for the workability of mortars, as it affects the water requirement and potential shrinkage of mortars with natural sand, recycled sand, or a mixture of them [91]. In this context, some authors [92] investigated the influence of washed recycled sand (to remove the fine fraction excess) when used as a partial replacement in mortars, concluding that washing the recycled aggregate could enhance its quality and enable one to obtain a better mechanical performance with respect to standard mortar. The binder type used in mortar mixtures also affects their mechanical properties. Hydrated lime, a combination of lime and natural pozzolan, or a mixture of lime, natural pozzolan, and cement made by adding RS to lime-based mortars, with the help of superplasticizer at 1% by weight of cement (bwoc), can improve compressive strength, especially at early ages. This improvement may be attributed to the reaction between the lime and the silica constituents of the raw materials in the sand [93].

On the other hand, some have observed [94] an increase in the mechanical properties of cement lime mortars by up to 60%, from 5 MPa to 8 MPa, by increasing the amount of recycled aggregates up to 100%. Nevertheless, a decrease in mechanical properties has been seen in pure cement mortars, from 25 to 15 MPa, for samples with substitutions up to 100%. Ledesma et al. [95] investigated the maximum feasible use of RS obtained from ceramic masonry waste in producing eco-mortars at 0%, 25%, 50%, 75% and 100%. The researchers found that using RS decreased the compressive strength of the eco-mortars by almost 12%. Still, this decrease was within acceptable limits for non-structural applications. The researchers also observed that incorporating recycled sand increased the eco-mortars' water demand and air content. Nevertheless, using a superplasticizer effectively improved the workability and strength of the mortars while minimizing the water demand. The study concluded that the maximum feasible use of recycled sand from ceramic masonry waste in eco-mortars is around 50%, beyond which the decrease in strength becomes significant. Finally, the influence of the saturation state and replacement percentage or fraction of natural sand with recycled sand on the properties of mortars led to greater water absorption and smaller slumps, but not better mechanical properties, which were superior when adding dried RS, as this absorbs water only during the preparation and curing stages [96]. Regarding the substitution fraction, it was observed that the compressive strength of mortars with RS decreased linearly as the replacement percentage of RS increased.

As a general remark on the substitution of recycled sand in mortar and concrete, it seems that the typical effect is a decrease in mechanical properties. This is reasonable, since RS is less pure and more porous than standard sand. However, the fact that the pores of the sand can absorb part of the water can alter the results, giving the impression that the mechanical properties increase due to the use of RS, when in fact the effect is a reduction in water available for cement hydration, i.e., the reduction in water-to-cement ratio.

6. Improving the Microstructural Properties of Recycled Aggregates

As discussed in the previous paragraph, mortars made with recycled aggregates have poor mechanical performance. In fact, mixed recycled aggregates from CDW contain various constituents, such as natural aggregates, cement, bricks, tiles, glass, small amounts of metal, and other minor organic and inorganic impurities. This mixed composition contributes to lowering the performance of cement-based materials containing recycled aggregates. Due to the presence of CDW, compressive strength, as well as tensile and shear strength, are reduced due to higher porosity, crushing index, micro-cracks in the interfacial transition zones, contamination, and variances in quality.

The presence of micro-cracks in the interfacial transition zones due to non-homogenous recycled aggregates can result in the penetration of harmful reactive substances such as sulfate ions, which can react with the hydration products of the cement [97,98]. This reaction produces gypsum and ettringite, further weakening the recycled concrete aggregate due to the higher volume of these reaction products applying internal stresses. Therefore, improving the microstructural and mechanical properties of recycled concrete aggregate has become crucial to enhancing its applicability and usefulness when producing recycled concrete [99–101].

The existing literature indicates that there are six major methods available to enhance the properties of recycled aggregates. These methods can be categorized into two groups: the "improve by removing" category, whereby weaker parts of the recycled aggregate, for instance cracked cement zones, are removed by chemical and thermal processes; and the "improve by adding" category, where the aggregate is reinforced by the addition of mineral admixtures, or by self-healing, carbonation, sequential mixing, or fortification by coating and infiltration processes [102–104].

The chemical approach involves the use of strong acids, such as hydrochloric (HCl) and sulfuric acid (H_2SO_4), to dissolve certain hydration products in the cement. This method effectively removes loose and cracked mortar from the recycled aggregates (RA), reducing water absorption and improving concrete performance [105,106]. However, Tam et al. [106] showed that using acid-treated recycled concrete aggregate (RCA) in concrete allows for a maximum replacement of only 30% of the natural coarse aggregate. Furthermore, strong acids pose safety risks and introduce harmful chemicals into the concrete. In the thermal approach, instead, the RA is heated to a high temperature of over 400 °C to remove the hydration products and weaken the residual mortar, which is then mechanically removed from the natural aggregate. Not surprisingly, this method requires a significant amount of thermal energy and may produce fine powders that attach to the surface of the RA, negatively impacting its quality. The mechanical rubbing process can also cause new micro-cracks to form, further weakening the RA [107].

In the "improve by adding" category, instead, one way to improve the RA quality is to strengthen the adhered mortar. Shi et al. [103] investigated using both pozzolan slurry (including silica fume, nano-SiO_2, and fly ash slurries) and CO_2 treatment as enhancement methods for RCA. Their findings show that concrete made with treated RCA had a compressive strength that was increased from 17% to 55%. Polymer emulsions also effectively reduced RCA water absorption by 5% to 30% [108]. Zhan et al. [109] used a carbonation process that enhanced the properties of the recycled aggregates. With this method, the water absorption of RCA was reduced by 20%, down to 24%. Furthermore, the durability of concrete made with treated RCA was significantly improved compared to untreated RCA. In addition, mineral precipitation, which leverages the activity of bacteria to precipitate calcium carbonate on the surface of the RCA [110], can also significantly reduce water absorption by 13%, down to 17%, and enhance the microstructure of the RCA. On the other hand, this method is costly and not practical for widespread use.

From the coating and infiltration point of view, as stated by Tam et al. [111], a limited number of research studies are available that focus on identifying methods to improve the microstructural properties of recycled aggregates. In their review, they explain that research studies in this area have been superficial and incomplete, leaving an extensive knowledge gap that requires further investigation and filling. Given the abundance of available chemical varieties, there is potential for developing numerous chemicals and solutions to treat and improve the microstructural properties of recycled concrete aggregate. Figure 5 provides an illustration summarizing the improvement methods.

An interesting approach to improving the RA involves crystallization technology. This method, known as crystalline waterproofing, is widely spreading in concrete applications, and involves active substances that react with the hydration products or dehydrated cement particles in the concrete to produce additional reactants in the form of crystals [112–115]. These crystals then effectively block off the pores in the concrete, decreasing its overall permeability [116,117]. Recent studies have shown that the use of this additive does not contribute to the improvement of concrete compression strength, but significantly increases durability [118]. The increase in durability also appears to be due to the closure of capillary pores resulting from the formation of ettringite on crack surfaces [119]. No effects on workability have been detected [120]. This innovative approach offers a promising alternative for reducing porosity and improving the waterproofing properties of concrete structures, and could be applied to RA to partially fill the present pores.

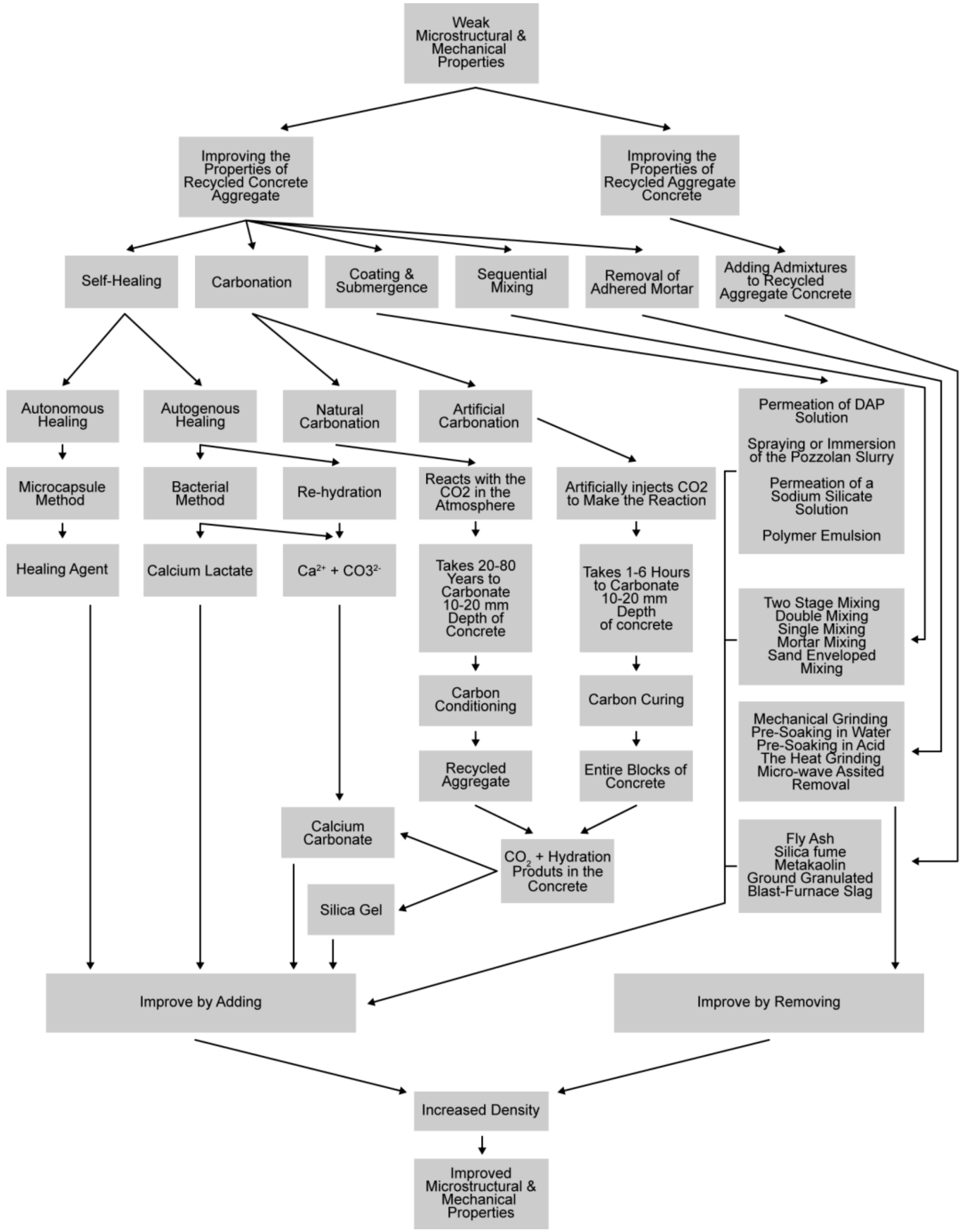

Figure 5. An overview of the enhancement techniques and their interconnections [111].

7. Conclusions

This overview emphasizes the need to address the environmental impacts of construction materials used in sustainable urban development, given the industry's high energy consumption and carbon emissions.

- Sustainable improvements in cement-based materials can be pursued via two strategies: using eco-friendly cements and fully exploiting Construction and Demolition Wastes (CDW) as aggregates, whilst considering techniques to improve recycled aggregates' properties.
- Architectural and civil engineering efforts to promote high-performance, eco-friendly materials are crucial. The EU's support for CDW recovery strategies highlights the importance of managing construction waste.

- The potential use of green cement and crystallizing agents to enhance cement sustainability. Green cement reduces energy use and carbon footprint, unlike traditional Portland cement. Crystallizing agents improve concrete durability and self-healing, reducing maintenance needs. Their use enhances both the environmental performance and longevity of cement-based materials.
- Using CDW, green cement, and crystallizing agents offers a path toward a greener construction industry. Continued research and collaboration are essential to expand these strategies and create a sustainable built environment for future generations.

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