The enduring legacy of limestone: from an ancestral pillar of construction to a modern precursor in synergy with recycled concrete.

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DOI: https://doi.org/10.21041/ra.v16i1.838

ABSTRACT

Facing the urgent need to reduce the environmental impact of Portland cement, this review explores the potential of limestone and pulverized recycled concrete (PRC) as pillars for sustainable binders. The historical and current role of limestone is analyzed, from its ancestral use to its application in modern cements, LC3, and, crucially, as a precursor in alkali-activated cements (AAC). PRC as a precursor in AACs is also examined, highlighting its contribution to the circular economy. The environmental and performance advantages of AACs based on these materials are discussed, as well as key challenges, including long-term durability, raw material variability, and the need for standardization. It is concluded that both resources are strategic, requiring focused research for their effective implementation.

Keywords: limestone; Portland cement; recycled concrete; alkali-activated cements; sustainable cements.

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Contribution of each author

In this work, José Iván Escalante García is the only author, therefore, he participated in all the activities to carry out its content.

Discussions and subsequent corrections to the publication

Any dispute, including the replies of the authors, will be published in the third issue of 2026 provided that the information is received before the closing of the second issue of 2026.

El legado continuo de la caliza: de pilar ancestral de la construcción a precursor moderno en sinergia con el concreto reciclado.

RESUMEN

Ante la urgencia de reducir el impacto ambiental del cemento Portland, esta revisión explora el potencial de la piedra caliza y el concreto reciclado pulverizado (CRP) como pilares para aglomerantes sostenibles. Se analiza el rol histórico y actual de la caliza, desde su uso ancestral hasta su aplicación en cementos modernos, LC3 y, crucialmente, como precursor en cementos activados alcalinamente (CAA). Se examina también el CRP como precursor en CAA, destacando su contribución a la economía circular. Se discuten las ventajas ambientales y de desempeño de los CAA basados en estos materiales, así como los desafíos clave, incluyendo la durabilidad a largo plazo, la variabilidad de las materias primas y la necesidad de estandarización. Se concluye que ambos recursos son estratégicos, requiriendo investigación focalizada para su implementación efectiva.

Palabras clave: caliza; cemento Portland; concreto reciclado; cementos alcalinos; cementos sustentables.

O legado contínuo do calcário: de pilar ancestral da construção a precursor moderno em sinergia com o concreto reciclado.

RESUMO

Diante da urgência de reduzir o impacto ambiental do cimento Portland, esta revisão explora o potencial do calcário e do concreto reciclado pulverizado (CRP) como pilares para aglomerantes sustentáveis. Analisa-se o papel histórico e atual do calcário, desde seu uso ancestral até sua aplicação em cimentos modernos, LC3 e, crucialmente, como precursor em cimentos álcali-ativados (CAA). Examina-se também o CRP como precursor em CAA, destacando sua contribuição para a economia circular. Discutem-se as vantagens ambientais e de desempenho dos CAA baseados nesses materiais, bem como os desafios-chave, incluindo a durabilidade a longo prazo, a variabilidade das matérias-primas e a necessidade de padronização. Conclui-se que ambos os recursos são estratégicos, requerendo pesquisa focada para sua implementação efetiva

Palavras-chave: calcário; cimento Portland; concreto reciclado; cimentos álcali-ativados; cimentos sustentáveis.

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1. INTRODUCTION

The 21st century poses a significant challenge for the construction industry: reconciling its development with environmental sustainability. Population growth and ongoing urbanization, particularly in less economically developed regions (United Nations, World Urbanization Prospects, 2018), drive a high demand for infrastructure, which consequently generates a considerable environmental impact. This impact is largely attributed to the production of essential materials such as Portland cement (PC), the primary binder in modern concrete and responsible for approximately 8% of global anthropogenic CO_2 emissions (Ortega-Zavala et al., 2019; Schneider, 2019). PC production is highly intensive in both energy and natural resource consumption

Given this scenario, adopting cementitious materials and construction practices with a lower ecological footprint is imperative. In this search, limestone, an abundant geological resource with a building legacy of over 10,000 years (Courland, 2011), emerges as a fundamental pillar. Its current role is multifaceted, ranging from its use as an aggregate and essential raw material for PC clinker production to its role as a versatile supplementary cementitious material (SCM) in composite cements and innovative Limestone Calcined Clay Cements (LC³) (Scrivener et al., 2018). Furthermore, it shows promising potential as a reactive precursor, either alone or in combination, in alkali-activated cement (AAC) technology (Ortega-Zavala et al., 2019)

Concurrently, the rising generation of construction and demolition waste (CDW), which reaches approximately 10,000 Mt/year globally (Chen et al., 2021), represents both a major environmental challenge and an opportunity for circular economy models. Within CDW, concrete from demolished structures—especially those originally constructed with limestone aggregates—is a valuable precursor for new binders. Transforming this pulverized recycled concrete (PRC) into an active component for alternative cements, such as AACs, addresses disposal problems and reduces the depletion of virgin resources (Rodriguez-Morales et al., 2024).

This perspective article analyzes the evolving and multifaceted role of limestone and the emerging contribution of limestone-based PRC in developing low-environmental impact cements. It explores the trajectory of limestone from historical uses and its current function in the PC industry to its incorporation into innovative sustainable systems. Additionally, the potential of PRC as a precursor is examined. Finally, the article discusses the inherent advantages of these materials, the technological and durability challenges yet to be addressed, and the research perspectives required for their effective integration into a truly sustainable construction industry

2. LIMESTONE: FROM AN ANCESTRAL RESOURCE TO A KEY COMPONENT IN MODERN CEMENTS

Limestone ($CaCO_3$) is a material that has been used in construction since ancestral civilizations. Archaeological evidence at Göbekli Tepe (present-day Turkey) suggests that limestone-derived products were already being utilized more than 11,000 years ago, demonstrating a fundamental relationship with human constructive development (Courland, 2011). The vast presence of these rocks is remarkable; it is estimated that globally, 15.2% of the ice-free land surface (equivalent to 20.3 million km²) corresponds to carbonate rocks, of which 9.4% are continuous and 5.8% are discontinuous or mixed with evaporites (Goldscheider et al., 2020), as shown in Figure 1. Given this widespread availability, limestone has consequently served as the basis for the production of binders and construction materials for millennia.

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2.1 Historical and traditional uses: the era of lime

The true technological leap in using limestone as a binder is attributed to the Roman civilization, which perfected the calcination process at 800–900°C to produce calcium oxide (CaO, quicklime); this compound, upon hydration, generates calcium hydroxide (Ca(OH)₂, slaked lime). This material hardens by reacting slowly with atmospheric CO_2, reverting to CaCO₃ and completing the "closed lime loop" (where the final product and the original rock are both CaCO₃), as illustrated in Figure 2. This cycle is partly responsible for the durability of many ancient structures.

Roman ingenuity, however, went a step further by mixing slaked lime with reactive pozzolanic materials, such as volcanic ash or even finely ground ceramic fragments10. These additions reacted chemically with the Ca(OH)₂ in the presence of moisture, forming hydraulic compounds — primarily hydrated calcium silicates and aluminates (see Table 1)— which provided Roman mortars and concretes with superior mechanical strength, underwater stability, and exceptional durability over centuries.

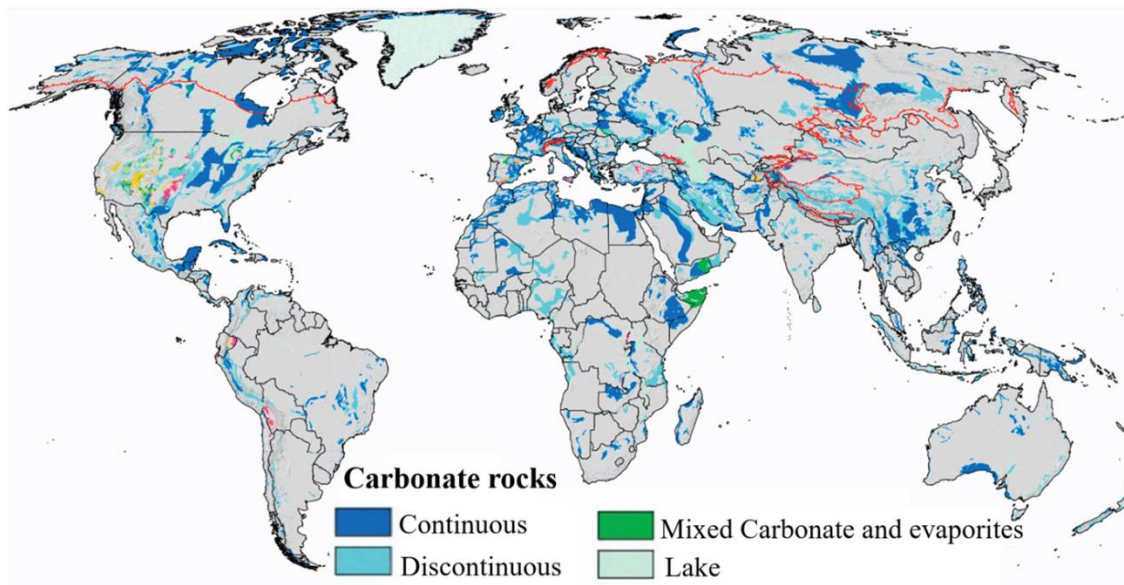Figure 1. Distribution of carbonaceous rocks. Modified from (Goldscheider, et. al., 2020).
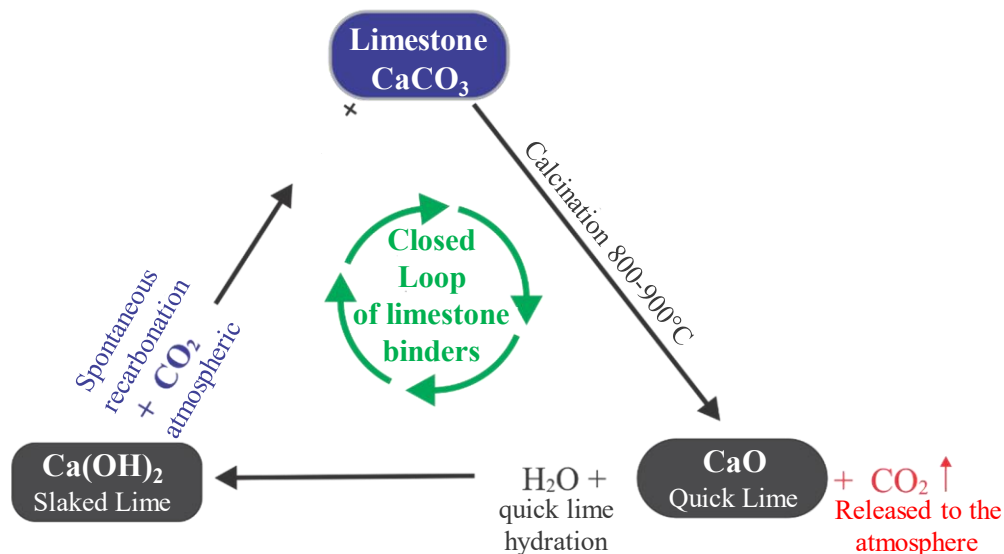

Figure 2. Closed loop of lime-based cements (limestone).

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Table 1. Pozzolanic activity reactions for the formation of cementitious products

Active ingredients in pozzolanas	Chemical activator	Cementitious reaction products	
		Formula	Common condensed formula *
SiO_2		$CaO \cdot SiO_2 \cdot H_2O$	C-S-H[+]
Al_2O_3	$+ \quad Ca(OH)_2 + H_2O \quad \rightarrow$	$CaO \cdot Al_2O_3 \cdot H_2O$	C-A-H
SiO_2, Al_2O_3		$CaO \cdot SiO_2 \cdot Al_2O_3 \cdot H_2O$	C-A-S-H

* without a specific stoichiometry.
[+] C-S-H gel is the main compound that contributes to mechanical strength and cohesion.

Despite the development of modern, fast-hardening hydraulic cements, lime-based binders remain in use. Their lower initial strength and slower strength development relative to Portland cement (PC) limit their massive structural use today. Nonetheless, their specific properties—high water vapor permeability, good workability, flexibility, and compatibility with ancient construction systems—make them valuable in specific niches. Most notably, they are indispensable in the restoration and conservation of architectural and cultural heritage, where material and aesthetic compatibility with the original substrates is paramount (Saba et al., 2019).

2.2 2.2 Limestone in the Portland cement era: A multifaceted protagonist
With the invention and mass production of Portland cement (PC) since the 19th century, limestone has established itself as an irreplaceable mineral, taking on diverse and fundamental roles in the production chain of modern concrete and construction materials. Its abundance and chemical composition turned it into the cornerstone of the global cement industry. Its main applications are:

- **Essential raw material for PC clinker:**
  o Limestone is the primary source of CaO, constituting ~80% of the raw mix for PC clinker manufacturing.
  o During the clinkerization process at ~1450°C, $CaCO_3$ s decarbonates, releasing CO_2, and the resulting CaO reacts with silico-aluminous components (from clays, marls, etc.) to form the hydraulic phases of clinker: alite (C_3S), belite (C_2S), tricalcium aluminate (C_3A), and tetracalcium ferroaluminate (C_4AF).
  o The process is highly energy-intensive, consuming ~3.5 GJ per ton of clinker.
  o CO_2 emissions (originating from both decarbonation and fossil fuel combustion) range from ~0.8–0.9 tons of CO_2 per ton of clinker.

- **Predominant aggregate in mortars and concretes:**
  o Crushed limestone is extensively used as an aggregate (gravel and sand) in mortar and concrete formulations.
  o Given the massive global production of concrete—approximately 30,000 million tons annually—and the fact that aggregates can represent up to 75% of concrete volume, the amount of limestone used for this purpose is enormous.
  o Its broad availability, relatively low cost, and suitable physical-mechanical properties position it as the primary option for aggregates in numerous regions.

- **Supplementary cementitious material (SCM) and active addition:**
  o In recent decades, to reduce the environmental footprint of cement, finely ground limestone has gained prominence as an SCM.

- o International standards, such as the European EN 197-1, allow for the incorporation of up to 35% in PC.
- o Its primary mechanism of action is physical (filler effect), optimizing particle size distribution, providing nucleation sites for clinker hydration, and improving workability.
- o Although its chemical reactivity is considered limited, it can participate in the formation of hydrated carboaluminate phases, contributing marginally to strength.
- o This practice reduces the "clinker factor" of cement, thereby decreasing CO_2 emissions and energy consumption per ton of final product.
- o However, its use in high percentages is a subject of scientific debate regarding the long-term durability of reinforced concrete, specifically due to the potential reduction in carbonation resistance and the consequent increased risk of steel reinforcement corrosion.

2.3 Limestone in recent cementitious innovations: towards sustainability.
The growing pressure to decarbonize the construction industry has catalyzed research into new binders with a lower dependence on PC clinker. In this context, limestone contributes to sustainability with roles that go beyond being a simple addition:

- **Limestone Calcined Clay Ternary Cements (LC³):** A prominent innovation is the development of Limestone Calcined Clay Cements (LC³)[3]. In these formulations, limestone, in proportions of ~15% by weight (for 50% clinker), plays a fundamental active chemical role, surpassing its traditional function as a mere filler. Limestone reacts synergistically with clinker hydration products and with the reactive alumina from calcined clay (metakaolin, MK), leading to the formation of hydrated calcium carboaluminate phases (such as hemicarbonate AFm-Hc and monocarbonate AFm-Mc). These products, added to conventional C-S-H, densify the microstructure, resulting in mechanical strengths comparable to or better than PC and improvements in various durability aspects. LC³ cements allow for clinker content reductions of up to 50% compared to traditional PC, mitigating CO_2 emissions by ~30% (Scrivener et al., 2018).

- **Advances in cements with high limestone substitution and other SCMs:** To maximize clinker substitution, other innovative technologies, some already commercialized, utilize up to 50% limestone. These strategies are based on particle engineering to optimize packing and particle size distribution, the use of state-of-the-art dispersants, and the combination of limestone with other low-carbon footprint SCMs that provide additional reactivity (ECOCEM Global, 2024). The objective is to minimize clinker content without compromising—and even improving—the engineering properties of the final material.

- **Limestone as a precursor in alkali-activated cements (AAC):** Finally, the application horizon for limestone in alternative binders expands even further when considering its potential in systems that radically depart from Portland cement chemistry. Recent research has begun to explore the use of limestone as a precursor in alkali-activated cements (AAC), demonstrating that under high alkalinity conditions, $CaCO_3$ can exhibit reactivity and contribute to the formation of cementing phases (Ortega-Zavala et al., 2019). This fascinating role of limestone in AACs, whether as a sole precursor or in combination with other mineral or waste materials, will be explored in greater detail in the next section of this work.

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3. LIMESTONE AND PULVERIZED RECYCLED CONCRETE AS CONCRETE PRECURSORS FOR AAC

Beyond the optimizations in systems based on PC, the search for binders with a radically lower environmental footprint has driven the development of alternative technologies. Among them, alkali-activated cements (AAC, or geopolymers in some sub-types) represent a family of materials with considerable potential. Their appeal lies in the ability to drastically reduce CO_2 emissions associated with clinker and to utilize a wide range of industrial by-products, low-purity minerals, and wastes as raw materials, including those from the construction sector. In this context, versatile limestone and abundant pulverized recycled concrete (PRC) are interesting candidates as cementitious precursors

3.1 Fundamentals of alkali-activated cements (AAC)

AACs are inorganic binders whose formation chemistry differs fundamentally from that of PC. They are produced through the chemical reaction between a solid precursor (generally rich in reactive, amorphous, or vitreous SiO_2 and Al_2O_3) and a highly alkaline activator, provided either in solution ("two-part") or as a solid ("one-part"). Although the first observations regarding the alkaline activation of slags date back to the mid-20th century (Purdon, 1940), the pioneering and systematic work of Glukhovsky and colleagues laid the scientific foundations for this technology (Krivenko, 2017).

.The key components of AACs are:

- **The Precursor:** It is the main source of SiO_2 and Al_2O_3) and CaO that will constitute the structure of the hardened binder (Figure 3). Traditionally, industrial by-products such as ground granulated blast furnace slag and low-CaO fly ash, or natural minerals such as calcined clays (MK), have been used. However, the spectrum of potential precursors is much broader and includes less conventional materials such as waste glass and those that are the subject of this article: limestone ($CaCO_3$) and pulverized recycled concrete with limestone aggregates ($CaCO_3$, SiO_2, Al_2O_3). The reactivity of the precursor—determined by factors such as its mineralogy, degree of amorphicity, and fineness—is crucial for the development of AAC properties.

- **The Activator:** Its function is two-fold: (a) the high pH it provides (generally > 12) dissolves the Si, Al, and Ca ionic species from the precursor; and (b) it catalyzes the polycondensation or precipitation reactions that form the three-dimensional binding network. The most common activators are aqueous solutions of sodium hydroxide, sodium silicate ("water glass"), sodium carbonate, or their mixtures (Figure 3). In one-part systems, the activator is incorporated as a powdery solid alongside the precursor, requiring only the addition of water to initiate the reaction, which simplifies on-site handling by making it similar to Portland cement

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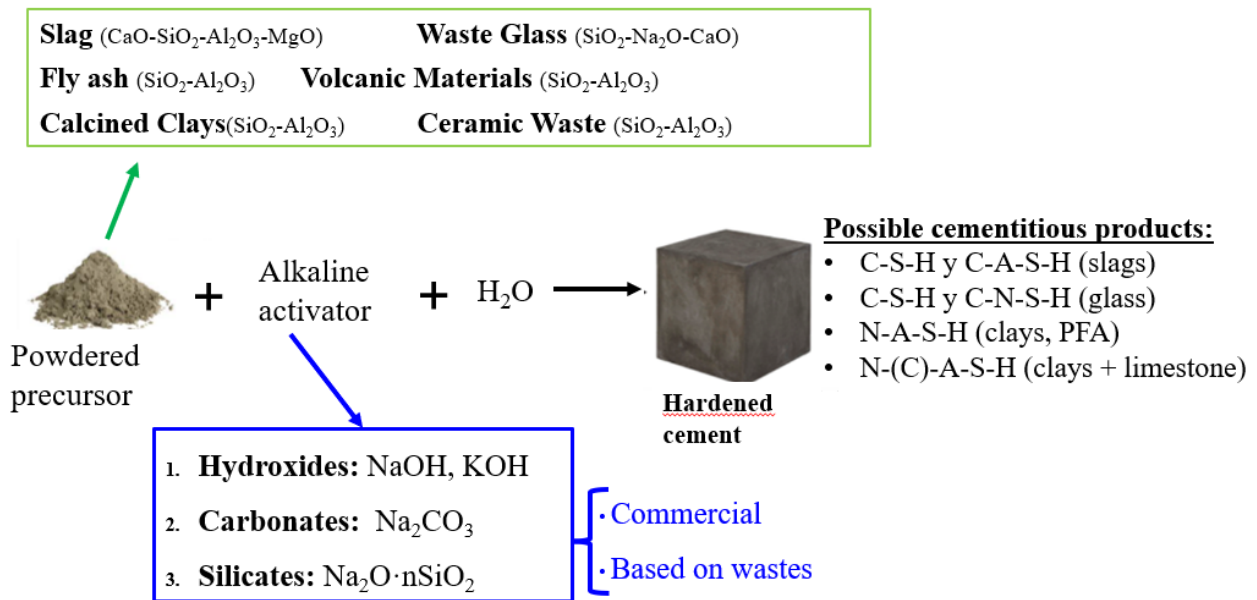
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Figure 3. Reaction scheme of "one-part" alkali-activated cements. In two-part cements, the alkaline activator is added pre-dissolved with the mixing water.

The chemical reactions involved are complex and depend on the precursor composition as well as the type and concentration of the activator, leading to the formation of amorphous or semi-crystalline inorganic gels that act as the binding matrix. In CaO-rich systems (based on slag or with the addition of limestone/PRC), the main product is a hydrated calcium (alumino) silicate gel referred to as C-(A)-S-H, which is somewhat similar to the C-S-H gel found in PC. In systems with low CaO (utilizing MK or fly ash), N-A-S-H type gels (geopolymers) are formed. The incorporation of calcium (e.g., through the addition of limestone) can modify the gel structure toward mixed N-(C)-A-S-H compositions, thereby affecting the final properties.

The primary motivation for studying and developing AACs lies in their potential advantages: a significant reduction in the carbon footprint by avoiding clinkerization, the ability to valorize large volumes of industrial waste and by-products, and the possible attainment of materials with superior durability in certain chemically aggressive environments.

3.2 Limestone as a precursor in AACCaliza como precursor en CAA

Although the use of limestone ($CaCO_3$) as a primary precursor in alkali-activated cements (AAC) is less explored due to its relatively low reactivity compared to amorphous aluminosilicates, its potential is considerable[6]. This interest is driven by its enormous global abundance, low cost, and the lack of a need for thermal treatment[7]. Initial studies on 100% limestone-based AAC have revealed that it can react in a strong alkaline environment, forming calcium silicate hydrate (C-S-H) and developing mechanical properties (see Figure 4), although its kinetics and degree of reaction require further investigation (Ortega-Zavala et al., 2019).

Ideal AACs depend on raw materials that are globally abundant, easily accessible, and low-cost for their precursors. Limestone meets these requirements as it is highly economical[10]. Metakaolin (MK), obtained from the calcination of kaolinitic clays, is also abundant and offers the advantage of a simple calcination process that is less energy-demanding (~ 0.35 GJ/t of clay) than the production of Portland cement (PC) clinker (Juenger et al., 2019)[11]. The confluence of these factors makes LS-MK blends feasible and highly advantageous precursors for AACs, a potential that has been extensively researched through statistical optimization to maximize both strength and sustainability indicators (Perez-Cortes & Escalante-Garcia, 2020a; Perez-Cortes & Escalante-

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Garcia, 2020b). The growing interest in this subject is reflected in several recent bibliographic reviews (Rakhimova, 2022; Ma et al., 2022; Rashad, 2022; Chan & Zhang, 2023)13.
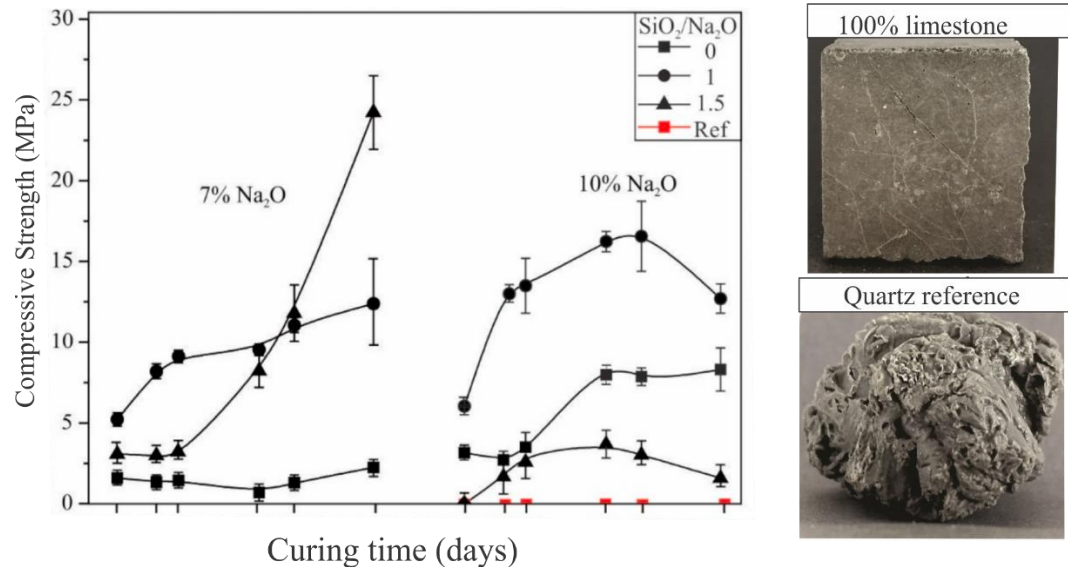

Figure 4. Compressive strength and photographs of alkali-activated limestone and quartz (red line) hardened cements. Adapted from (Ortega-Zavala et al., 2019).

Given the lower intrinsic reactivity of limestone, research has focused on its combination with more reactive precursors, primarily MK, seeking an optimal balance between performance, cost, and sustainability. Early works on MK-limestone systems reported variable results, reflecting the complexity of these interactions; for instance, while one study reported an increase in 28-day strength with 20% limestone (from 38.4 to 45.4 MPa, Yip et al., 2008), another showed only marginal increases with 6% and a reduction with 12% limestone (Aboulayt et al., 2017). However, a significant breakthrough came from statistical optimization. These more recent studies demonstrated that it is possible to systematically design mixtures with high limestone contents (up to 60–80%), which achieve notable compressive strengths (>50 MPa at 28 days, see Figure 5), comparable to systems based on 100% MK or 100% PC (Perez-Cortes & Escalante-Garcia, 2020a; Perez-Cortes & Escalante-Garcia, 2020b).

A key finding in these optimized systems—and one of their greatest advantages—is the significant reduction in the demand for alkaline activators[18]. For example, while a system composed of 100% MK can require up to 25% Na_2O (relative to the precursor mass), replacing 80% with limestone decreases this demand to just 4% while maintaining competitive mechanical strengths (see Figure 5). This principle of efficiency in low-alkalinity limestone systems extends to other binders; "one-part" cements that combine 49% limestone with PC and are activated with sodium silicate also reach excellent mechanical properties (Santana-Carrillo et al., 2022). This lower dependence on activators has positive economic and environmental implications by reducing the carbon footprint, energy consumption, and costs associated with both commercial activators and MK processing, as illustrated in the comparison in Figure 6.

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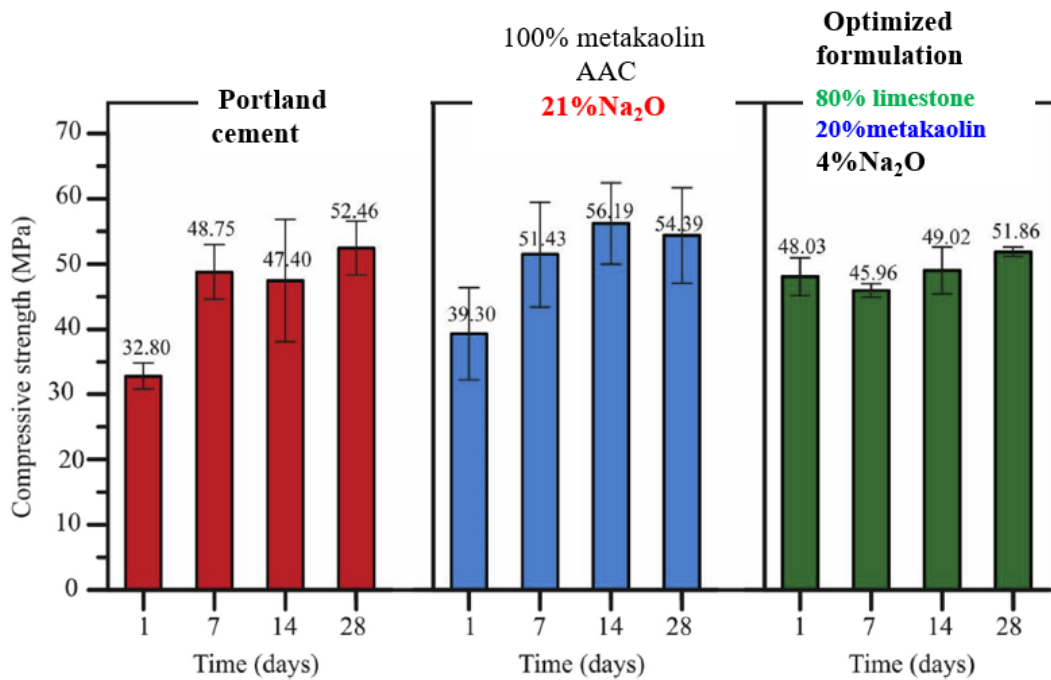
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Figure 5. Compressive strength comparison of 100% PC pastes, 100% metakaolin AAC, and optimized AAC formula with 80% limestone and 20% metakaolin. Adapted from (Perez-Cortes and Escalante-Garcia, 2020b).

Chemically, limestone surpasses its role as a mere diluent[22]. The Ca^{2+} ions released by its partial dissolution in the alkaline medium are incorporated into the binding gel. In MK-based systems, this modifies the composition of the N-A-S-H (geopolymeric) gel toward a mixed N-(C)-A-S-H gel (see Figure 7A), improving its three-dimensional structure, densifying the microstructure (see Figure 7B), and contributing to mechanical strength (see Figure 6). The presence of $CaCO_3$ can also lead to the formation of carboaluminates (Yip et al., 2008; Rakhimova et al., 2018). These optimized high-limestone systems also exhibit good durability against acids, sulfates, and moderate temperatures (300°C) (Perez-Cortes & Escalante-Garcia, 2023; Perez-Cortes et al., 2021). Furthermore, their application in concrete is promising, showing high strengths (up to 60 MPa at 7 days) with reasonable binder dosages (Escalante-Garcia & Perez-Cortes, 2018; see Table 2) and the ability to passivate steel reinforcement (Vázquez Leal et al., 2023).
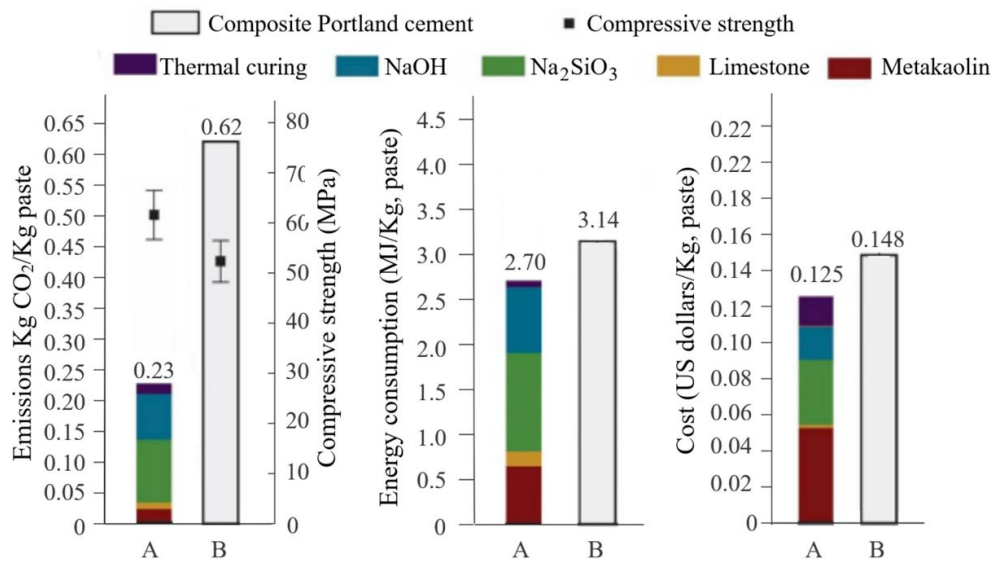
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Figure 6. Comparison of CO_2 emissions, energy consumption, and cost between: A - AAC (60% limestone, 40% metakaolin); and B - composite PC. Adapted from (Pérez-Cortés & Escalante-García, 2020a)

Limestone has also been combined, with interesting results, with other precursors such as blast furnace slag (Sakulich et al., 2009), fly ash (Gao et al., 2015), or recycled glass (Menchaca-Ballinas & Escalante-García, 2020). However, the limited availability of some of these precursors (such as slag) compared to limestone may restrict their large-scale applicability.

In summary, limestone is not an inert filler in AACs; it actively participates in reactions and modifies the microstructure. Crucially, it allows for a significant reduction in the amount of aluminosilicate precursor (such as MK, which requires calcination) and the demand for alkaline activators, making it a key component for formulating more sustainable and economical AACs.

Table 2. Mechanical properties of concretes prepared with 400 kg/m3 of binder and limestone aggregates (Escalante-Garcia and Perez-Cortes, 2018).

Formulation of the binder (wt. %)	Curing Temperature	Compressive Strength (MPa)	
		7 days	28 days
60% limestone, 40% metakaolin, 10.7% Na2O	20°C	47	57
60% limestone , 40% metakaolin, 8.5% Na2O	24h @70°C then at 20°C	33	40
30% limestone, 70% metakaolin , 16.9% Na2O	20°C	60	68

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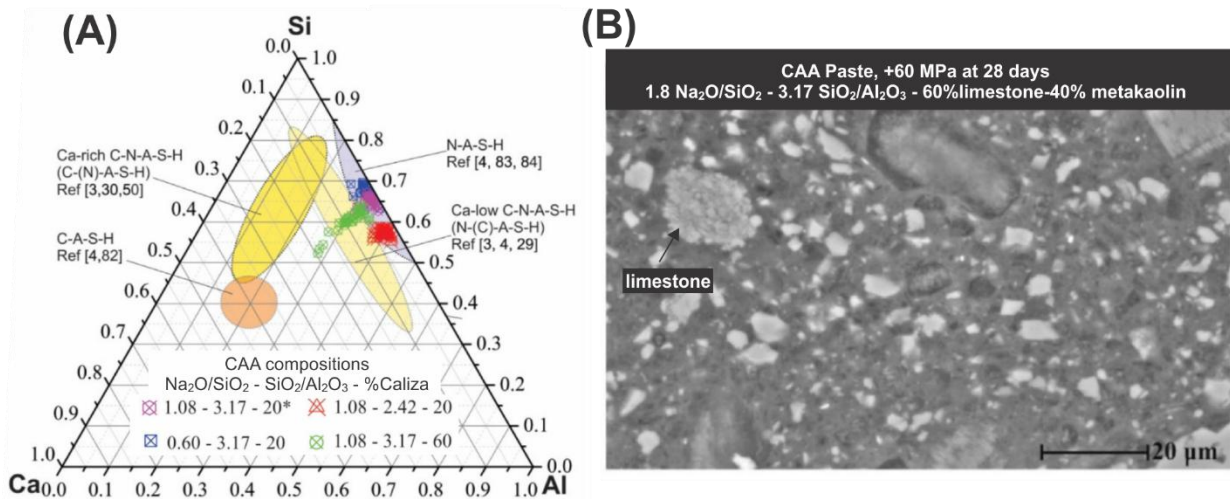
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Figure 7. (A) Al-Si-Ca ternary compositional diagram (normalized to 100%) of AAC (20 and 60% limestone, with the remainder being metakaolin). (B) Microstructure of AAC paste with 60% limestone and 40% metakaolin. Adapted from (Perez-Cortes and Escalante-Garcia, 2020c), which includes references for the chemical composition zones.

3.3 Concreto reciclado pulverizado como precursor en CAA

The management of construction and demolition waste (CDW) is a major global environmental challenge, with an annual generation of ~10,000 million tons, constituting ~30% of total worldwide solid waste, of which ~35% ends up in landfills (Chen et al., 2021). Recycling rates vary enormously, ranging from >80% in the Netherlands or Germany to <5% in many other regions, including densely populated countries (Özalp et al., 2016; Akhtar & Sarmah, 2018).

Currently, the most common way to recycle concrete from CDW is in crushed form as recycled aggregates to produce new concrete, helping to reduce the demand for natural aggregates. However, a higher-value valorization path aligned with circular economy trends is to use pulverized recycled concrete (PRC)—also referred to in the literature as PHC (pulverized hardened concrete)—not as a filler, but as a reactive precursor in alkali-activated cements (AAC). PRC is a complex material composed of hydrated PC paste (rich in C-S-H, portlandite, etc.), residual anhydrous PC, and fragments of the original aggregate (siliceous, silico-aluminous, or, as in many of the base studies for this work, calcareous). This composition provides an inherent potential for reacting in an alkaline medium.

Exploratory studies on the alkaline activation of PRC as a sole precursor (100% PRC) have demonstrated its feasibility, reaching compressive strengths of 12–20 MPa, especially with initial thermal curing (see Table 3, Rodriguez-Morales et al., 2024). Although these strengths may be sufficient for certain non-structural applications, they can be improved by combining PRC with other more reactive or complementary precursors. Promising results have been reported in binary or ternary mixtures:

- **With metakaolin (MK):** 55–75% PRC (with the remainder being metakaolin), reaching up to 30 MPa at 28 days (Table 3).
- **With waste glass:** Systems with 32–60% PRC (with the remainder being glass), reaching up to 46 MPa at 28 days (see Table 3).
- **With Portland cement (PC):** Systems with 45–65% PRC and 20–40% PC, achieving strengths of 30–55 MPa at 28 days (see Table 3). In these systems, PC can act not only as a precursor but also contribute to the initial alkalinity (see Table 2 and Figure 5, ibid.).

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Table 3. Compressive strength of AAC pastes formulated with PRC (pulverized recycled concrete) precursors and complementary precursors. The formulations incorporate different types of commercial and alternative activators.

	Pulverized recycled concrete (% peso)	Complementary precursor (% wt)			type of alkaline activator	%Na$_2$O relative to the precursor mass	Compressive strength 28-días, MPa	Curing regime [++]
		Metakaolin	PC	Waste glass				
1	55	45			Commercial Sodium metasilicate, Ms = 1	10	30	60-20°C
2	75	25			Commercial Sodium metasilicate, Ms = 1	12	28.5	60-20°C
3**	60			40	Commercial Sodium metasilicate, Ms = 1	7.5	32	60-20°C
4**	32			68	Commercial Sodium metasilicate, Ms = 1	7.5	46	60-20°C
5	45		40		15% commercial sodium silicate type G, Ms = 3.2	2.9	45.2	20°C
6	45		50		5% sodium silicate from waste glass, Ms = 3.5	1.2	55.8	20°C
7	85	-	-	-	15% commercial sodium silicate type G, Ms = 3.2	3.39	16	20
							12	60-20°C
8	80	-	-	-	20% sodium silicate from waste glass, Ms = 4	5%	12*	20°C
							19.8	80-20°C

Ms = modulus of the sodium silicate used as an activator.
* increased to 19 MPa after 90 days
** two-part cements (activator added in solution).
[++] 60-20°C or 80-20°C indicates the first 24 h of curing were at 60 or 80°C, then continued at 20°C.

Figure 8 compares CO$_2$ emissions, energy consumption, and cost between several PRC-based AACs compared to PC. The effectiveness of AACs in reducing emissions is evident. Additionally, promising research is being developed using combinations of PRC with other alternative precursors such as fly ash, waste glass, and ceramic industry waste.
A relevant aspect is the type of activator used. Various studies currently under development at the Cement and Environment Laboratory of Cinvestav Saltillo have improved the mechanical properties of AACs by optimizing the type of alkaline activator. In addition to commercial activators (hydroxides, sodium silicates), the use of alternative activators with a lower environmental impact, such as sodium silicates obtained from the thermochemical treatment of

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waste glass, has been investigated. These alternative activators can be as effective as commercial ones regarding strength development in PRC systems, but with a significantly lower carbon footprint and cost (see Table 3, items 7 and 8, and Figure 8), reinforcing the sustainability of this recycling route and promoting a circular economy.

Microstructurally, PRC-based AACs exhibit dense matrices with embedded and partially reacted PRC particles (see Figure 9), which is consistent with the measured mechanical performance.
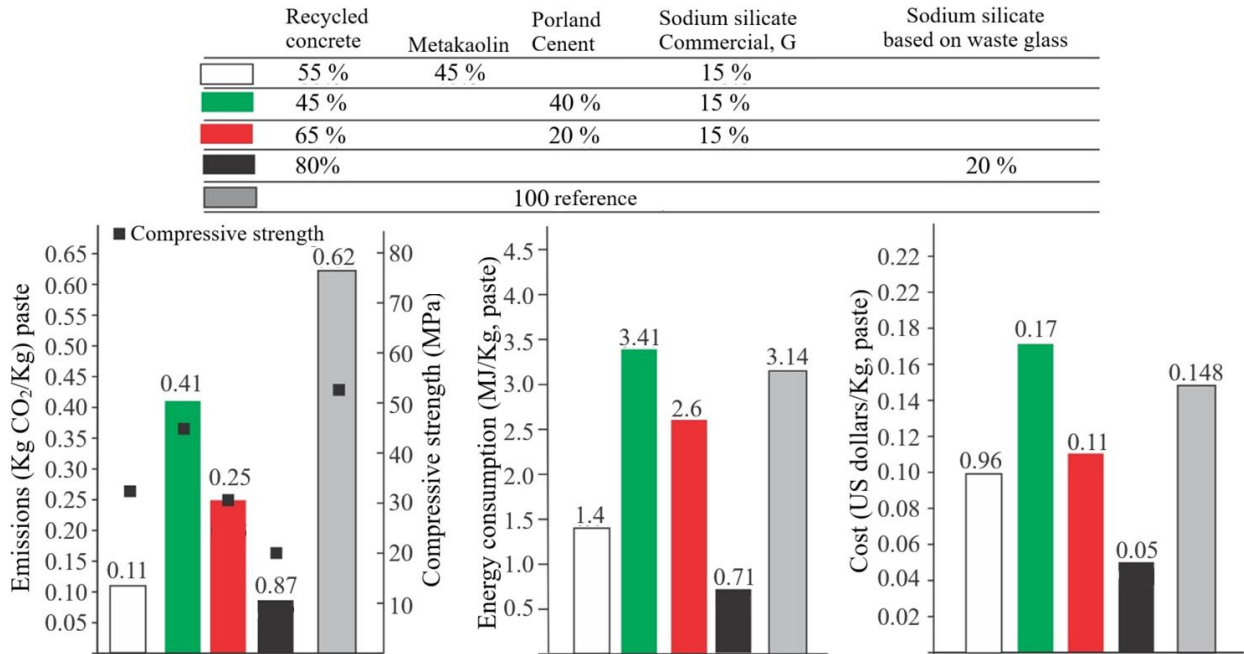
	Recycled concrete	Metakaolin	Porland Cenent	Sodium silicate Commercial, G	Sodium silicate based on waste glass
(white)	55 %	45 %		15 %	
(green)	45 %		40 %	15 %	
(red)	65 %		20 %	15 %	
(black)	80%				20 %
(gray)	100 reference				

Figure 8. Comparison of CO_2 emissions, energy demand, and cost of some PRC-based AACs and other supplementary precursors like Portland cement and metakaolin[38]. Includes data for 100% PRC with an alternative waste-glass-based alkaline activator.
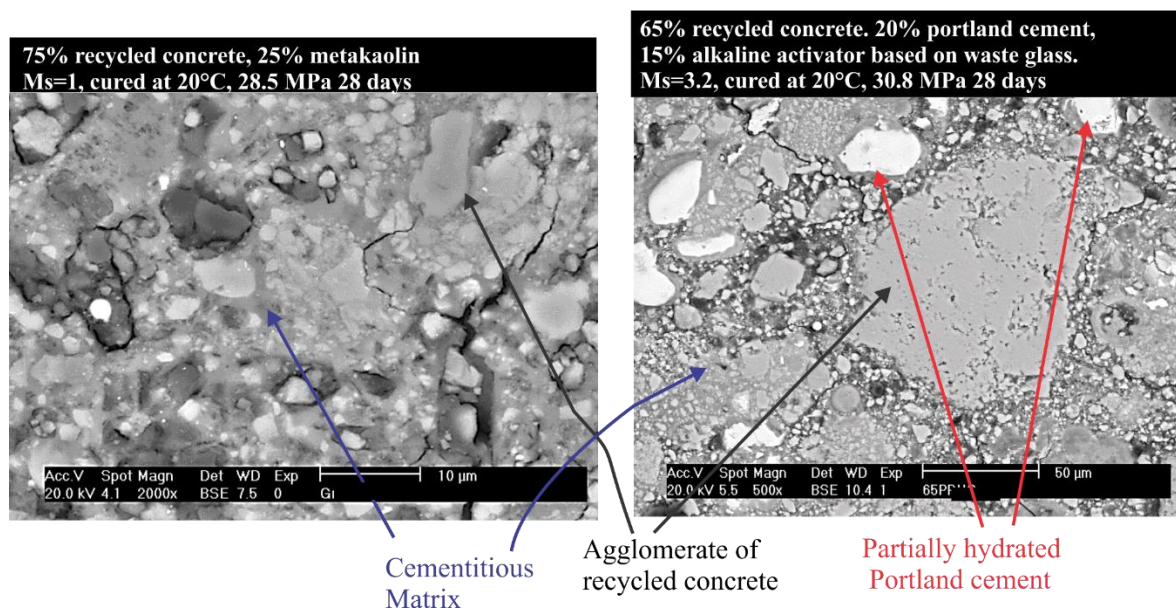

Figure 9. Microstructures of AAC using recycled concrete as a precursor. Scanning electron microscopy images, backscattered electrons (Escalante-Garcia J. I., 2024)..

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Advantages of PRC as a precursor. Using PRC as a precursor in AAC offers multiple environmental advantages:

- Transforms an abundant waste into a valuable resource.
- Reduces the demand for virgin raw materials (for both cement and aggregates).
- Decreases pressure on landfills.
- Contributes to closing the life cycle of construction materials.

Furthermore, the potential to use alternative alkaline activators derived from other waste streams (such as glass) opens paths toward an even more integrated circular economy. It is important to note that the valorization potential of CDW in AAC could extend beyond concrete, as other components such as ceramic bricks, tiles, or glass have also proven to be viable precursors in alkaline activation (Borrachero et al., 2022; Ahmari et al., 2012; Komnitsas et al., 2015), although more research is needed to understand the behaviour of more heterogeneous blends.

4. PERSPECTIVES, CHALLENGES AND OPPORTUNITIES

The previous sections have outlined the historical journey and the multifaceted current contribution of limestone in the world of cementitious materials, as well as the emerging potential of pulverized recycled concrete (PRC) as a valuable resource within the circular economy of construction, particularly in the context of alkali-activated cements (AAC). It has been demonstrated that both materials, individually or in combination, can be the basis for developing binders with adequate mechanical performance and, crucially, with a significantly reduced environmental footprint compared to traditional Portland cement (PC). However, the transition from promising research and initial applications toward broad and responsible adoption in the industry requires a critical evaluation of both the inherent opportunities and the substantial challenges that still persist. This final section focuses on this prospective vision, analyzing the potential derived from the availability of these precursors, the technical and implementation obstacles that must be overcome, and the future research lines that could pave the way toward their consolidation as pillars of truly sustainable construction

4.1 Abundance and potential of precursors: strategic resources for sustainability.
Limestone: a mineral resource of vast global availability
Limestone is one of the most abundant minerals, with vast deposits distributed globally (Goldscheider et al., 2020), which minimizes logistical and transport barriers compared to precursors with more localized availability, such as slag or certain fly ashes. Furthermore, its extraction and primary processing (crushing and grinding) are relatively simple processes with lower energy costs compared to the complex production of PC clinker.

Pulverized Recycled Concrete (PRC): An Expanding Anthropogenic Resource.
PRC constitutes an anthropogenic resource of colossal magnitude and constant growth, originating from the immense stock of existing concrete that, at the end of its useful life or through demolition, transforms into a massive waste stream. The annual generation of construction and demolition waste (CDW) exceeds 10,000 million tons, of which 30–40% is concrete (Chen, et al., 2021)9. Utilizing this waste stream as a precursor for new cements not only addresses the environmental problem of its final disposal but also provides a massive and distributed source of raw material, especially in urban environments with high CDW generation. Additionally, the use of PRC can offer economic advantages, reducing landfill costs and substituting virgin raw materials.

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Strategic environmental potential of limestone and PRC.
From an environmental perspective, the potential of these materials is equally significant. The utilization of limestone as an addition in PC or as a precursor in LC³ and AAC avoids CO_2 emissions by partially replacing clinker, including emissions from decarbonation and fuel consumption. For AACs with a high proportion of limestone, there is the added benefit of a lower demand for alkaline activators, reducing their environmental footprint. PRC, for its part, materializes circular economy principles: it transforms waste into a resource, conserves non-renewable raw materials, and avoids pollution from landfilling. Substituting conventional PC with these alternative materials can contribute significantly to global emission reduction targets (Shah et al., 2022). Studies on optimized limestone-metakaolin (MK) AACs have shown CO_2 reductions exceeding 60% and energy and cost savings on the order of 15% compared to PC (Perez-Cortes & Escalante-Garcia, 2020a).

In summary, the vast global availability, potential low cost, and inherent environmental benefits position limestone and pulverized recycled concrete as fundamental strategic resources. Their intelligent and efficient utilization is key to advancing toward the Sustainable Development Goals (SDG) and decoupling necessary infrastructure development from the environmental impact historically associated with cement production. Nonetheless, fully realizing this enormous potential requires addressing and resolving a series of technical and non-technical challenges, as discussed below.

4.2 Challenges in implementation and use.

Despite the undeniable potential of limestone and pulverized recycled concrete (PRC) as precursors for more sustainable cements, their transition from laboratory and pilot applications to widespread industrial adoption faces significant challenges that require a comprehensive approach.

Uncertainty regarding long-term durability.
One of the most critical obstacles, and a subject of intense debate, is the uncertainty concerning the long-term durability of cements with high contents of these materials, especially in alkali-activated cements (AAC). While there are documented concerns about the durability of Portland cement (PC) concretes with high limestone additions—particularly regarding lower carbonation resistance and the risk of steel corrosion (Panesar & Zhang, 2020)—the situation for AACs is more complex. Although specific laboratory studies on optimized limestone-metakaolin AACs show promising results against chemical attacks (acids and sulfates) and moderate temperatures (Perez-Cortes & Escalante-Garcia, 2023), and even a capacity for steel passivation (Vázquez Leal et al., 2023), robust evidence regarding their long-term behavior (decades) under real and diverse exposure conditions is still lacking. This absence of an extensive performance history, comparable to that of PC, generates hesitancy for their use in critical structural applications

Variability of precursor raw materials.
The inherent variability of raw materials is another major challenge. While limestone composition varies depending on the deposit, the heterogeneity of PRC is even more pronounced, as it originates from concretes of different ages, mix designs, aggregate types, and potential contamination from other demolition materials such as gypsum or plastics. This variability makes it difficult to guarantee consistency in precursor quality and, consequently, the predictability of AAC performance, demanding robust characterization methods and potential pre-treatment of the PRC.

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Technological challenges in alkali activation.
Alkali activation technology itself presents significant hurdles9. Conventional activators (NaOH and $Na_2O \cdot SiO_2$) involve considerable costs and environmental footprints, and their handling—especially in highly alkaline solutions used in two-part systems—poses occupational safety risks. Alternative activators, such as those derived from recycled glass, are promising, but their industrial-scale production and long-term validation are still under development. Furthermore, controlling the fresh-state properties of AACs (rheology, setting time, workability) can be more complex than with PC, requiring careful formulation adjustments to adapt to various construction techniques, including 3D printing (Perales-Santillán et al., 2024).

Obstacles to standardization, acceptance, and economic viability.
Finally, there are significant barriers regarding standardization and industry acceptance. The lack of technical standards, specifications, and widely recognized design codes for limestone and PRC-based AACs limits their use in civil engineering projects. Overcoming sector inertia and gaining the trust of designers, builders, and regulators requires solid technical data, the dissemination of success stories, and professional training. These factors are joined by economic and logistical considerations, such as the need for efficient supply chains for processed PRC and activators, and a comprehensive evaluation of life-cycle costs.

4.3 Future vision and research lines.
The challenges described, though significant, should not deter the exploration and development of these alternative materials; on the contrary, they set the roadmap for future research. The urgency of mitigating climate change and transitioning toward a circular economy in construction makes it imperative to continue moving in this direction. To achieve this, the following priority lines of research and development have been identified:

- **Understanding and ensuring long-term durability:**
  - Conduct extensive field studies, monitoring the behavior of real structures built with these cements under diverse and representative environmental conditions over prolonged periods (decades).
  - Deepen the fundamental understanding of the specific degradation mechanisms (carbonation, chloride ingress, sulfate attack, alkali-aggregate reaction, etc.) operating within these particular cementitious matrices.
  - Develop and validate reliable, reproducible accelerated testing methods that show a robust correlation with performance observed under real service conditions.
  - Detailed investigation of the interaction between these new binders and reinforcement steel, focusing on passivation mechanisms, critical contaminant thresholds, and corrosion rates in specific alkaline environments.

- **Advances in materials science and mix design:**
  - Precisely characterize the nature and evolution of the phases formed during the alkali-activation of limestone and PRC, and their impact on final properties.
  - Investigate how the inherent variability of precursors (especially PRC) affects reaction kinetics and microstructure, developing strategies to mitigate negative effects.
  - Design and optimize precursor blends (combining limestone, PRC, MK, and other SCMs) and activator systems (type, dosage, sustainable alternatives) to achieve specific engineering properties and improve material robustness.
  - Establish effective strategies for rapid characterization, pre-treatment, or beneficiation of PRC to ensure its quality as a precursor.

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- **Technological development and application optimization:**
  o Promote research, development, and industrial scaling of alkaline activators that are more economical, safer to handle, and have a lower environmental footprint.
  o Refine the control over rheological properties (workability, viscosity) and setting times to facilitate the use of these cements in a wide range of construction applications, including advanced methods such as additive manufacturing (3D printing).
  o Develop and adapt non-destructive testing (NDT) techniques for on-site quality control and health evaluation of structures made with these materials.

- **Standardization, implementation, and comprehensive evaluation:**
  o Foster a concerted effort at national and international levels for the creation of technical standards, product specifications, and design guides based on solid scientific evidence, facilitating the safe and reliable entry of these cements into the market.
  o Conduct complete, transparent, and comparative Life Cycle Assessments (LCA) and Life Cycle Cost Analysis (LCCA) to comprehensively document their environmental and economic benefits.
  o Drive the construction and monitoring of pilot projects and demonstration structures to validate performance in real conditions, build industry confidence, and promote widespread acceptance.

Addressing these research lines in a coordinated manner and with a future-oriented vision is essential. Progress in these areas will overcome current obstacles and unlock the considerable potential of limestone and recycled concrete-based cements, significantly contributing to a construction industry that is more sustainable, resource-efficient, and aligned with circular economy principles.

5. CONCLUSIONS: TOWARD A SUSTAINABLE FUTURE IN CONSTRUCTION WITH LIMESTONE AND RECYCLED CONCRETE.

The journey through the world of cementitious materials reaffirms a conviction: the construction industry is at a turning point, where innovation inspired by traditional resources and the intelligent valorization of waste are imperative for forging a more sustainable future. In this horizon, limestone and recycled concrete emerge not merely as alternatives, but as protagonists with transformative potential

- **Limestone: a millennial legacy with strategic relevance.**
  This work has highlighted the presence of limestone from its fundamental use at the dawn of civilization to its irreplaceable role in the Portland cement era, and more critically, its promising entry as a key component in cutting-edge, low-carbon cements such as LC³ and alkali-activated cements (AAC). Its extraordinary global abundance, low cost, and the valuable possibility of utilizing it without calcination in various applications establish it as an unavoidable strategic resource for sustainability.

- **Pulverized Recycled Concrete (PRC): The Rebirth of a Waste.**
  The enormous potential of PRC, obtained from the massive flow of construction and demolition waste, has been underscored to transcend its fate as mere waste. Its conversion into a viable precursor for AAC is a clear example of the circular economy in action, reinserting waste into the value chain and transforming it into raw material for new high-performance binders, thereby alleviating pressure on landfills and natural resources.

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- **Alkali-activation: a catalyst for sustainability.**
Alkali-activation technology is revealed as a particularly effective way to capitalize on the virtues of limestone and PRC. Systems based on a high proportion of limestone (in synergy with metakaolin) or PRC (alone or combined) are not only technically viable but have demonstrated encouraging mechanical and durability properties in laboratory settings. More importantly, they offer substantial environmental advantages: a drastic reduction in the clinker factor, lower demand for alkaline activators (especially with high limestone presence), and effective waste valorization. Innovation in activators derived from other waste streams, such as glass, promises to further enhance the sustainability of this technological route.

- **Challenges on the horizon: the pending task.**
Despite this optimistic outlook, the widespread adoption of these materials faces considerable challenges highlighted in this review. Consolidating confidence in their long-term durability under real service conditions is paramount. Likewise, managing the inherent variability of raw materials (particularly PRC), the development and scaling of activators that are simultaneously sustainable, safe, and economical, and the establishment of robust, harmonized technical standards and regulations constitute obstacles that the scientific and technical community must address with priority and rigor.

- **A call to joint action:**
In conclusion, limestone—as a natural resource or as the soul of recycled concrete—reaffirms itself as a key piece in the present and, above all, the future of sustainable cementitious materials. Its synergy with alkali-activation technology and other low-carbon footprint strategies traces a promising path toward a construction industry that is greener, more resource-efficient, and firmly anchored in the principles of the circular economy. To materialize the immense potential of these resources and overcome pending challenges, continuous research, innovative technological development, and close, decisive collaboration among all sector actors—from academia to industry and regulatory bodies—will be essential for the benefit of a more sustainable planet for future generations.

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