



CARBONATION OF CONCRETE: HISTORICAL DEVELOPMENT AND CHANGING INTERPRETATIONS

KARBONATYZACJA BETONU: ROZWÓJ HISTORYCZNY I ZMIENIAJĄCE SIĘ INTERPRETACJE

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Abstract

Carbonation of cement-based materials has been interpreted in different ways during the development of building materials and concrete technology. This paper presents a selective narrative review of the main milestones that changed the meaning of carbonation: from the useful hardening of lime binders, through the durability problem of reinforced concrete, to present discussions on blended and low-clinker binders, accelerated testing and CO₂ uptake. The review was based on historically important, methodologically relevant and currently influential literature. Particular attention was paid to the changing interpretation of carbonation in relation to binder composition, exposure conditions, reinforcement corrosion, testing methodology and the growing use of supplementary cementitious materials. The review shows that carbonation cannot be treated as a single, universal durability indicator. Its meaning depends on the type of binder, moisture conditions, pore structure, alkaline reserve, presence of reinforcement and the aim of assessment. The main conclusion is that concepts developed for ordinary Portland cement systems cannot be directly transferred to concretes rich in supplementary cementitious materials without additional interpretation and method-specific validation.

Keywords: carbonation, accelerated carbonation, concrete durability, reinforced concrete, supplementary cementitious materials, CO₂ uptake

Streszczenie

Karbonatyzacja materiałów na bazie cementu była różnie interpretowana na kolejnych etapach rozwoju materiałów budowlanych i technologii betonu. W artykule przedstawiono selektywny przegląd narracyjny najważniejszych kamieni milowych, które zmieniały znaczenie karbonatyzacji: od korzystnego twardnienia spoiw wapiennych, przez problem trwałości żelbetu, aż po współczesne dyskusje dotyczące spoiw mieszanych i niskoklinkierowych, badań przyspieszonych oraz pochłaniania CO₂. Przegląd oparto na literaturze o znaczeniu historycznym, metodologicznym i współczesnym. Szczególną uwagę zwrócono na zmieniającą się interpretację karbonatyzacji w odniesieniu do składu spoiwa, warunków ekspozycji, korozji zbrojenia, metod badawczych oraz rosnącego stosowania dodatkowych materiałów cementowych. Przegląd pokazuje, że karbonatyzacja nie może być traktowana jako jeden uniwersalny wskaźnik trwałości. Jej znaczenie zależy od rodzaju spoiwa, warunków wilgotnościowych, struktury porów, rezerwy alkalicznej, obecności zbrojenia oraz celu oceny. Główny wniosek jest taki, że pojęć wypracowanych dla układów opartych na cemencie portlandzkim nie można bezpośrednio przenosić na betony bogate w dodatkowe materiały cementowe bez dodatkowej interpretacji i walidacji właściwej dla danej metody badawczej.

Słowa kluczowe: karbonatyzacja; karbonatyzacja przyspieszona; trwałość betonu; żelbet; dodatkowe materiały cementowe; pochłanianie CO₂

1. INTRODUCTION

Carbonation has long been recognized as a key mechanism of hardening in lime binders exposed to atmospheric CO₂ [1]. In modern concrete technology, however, the same process is discussed mainly in the context of durability and reinforcement protection [1-9]. This change in meaning makes carbonation a useful subject for a review that connects the history of materials with current research on durability. The paper assumes that the interpretation of carbonation changed over time together with the development of binders, structural systems and durability criteria.

In lime-based systems, carbonation was a useful phenomenon. In Portland cement systems, it entered the field of cement chemistry. In reinforced concrete, it became linked with steel depassivation and the risk of corrosion. Modern low-clinker systems require a further reinterpretation, because carbonation resistance, corrosion behaviour and CO₂ uptake can no longer be discussed only on the basis of ordinary Portland cement (OPC) [2, 5-8].

Instead of covering all published studies on carbonation, this paper presents a selective historical synthesis focused on the main stages, key authors and changes in the basic research questions. This approach may be useful for readers interested not only in what is known about carbonation, but also in how and why its meaning changed with the development of concrete science, as summarized in Table 1.

2. REVIEW METHODOLOGY AND SCOPE OF THE PAPER

This paper was prepared as a selective narrative review. Its aim was to reconstruct the main stages in the development of knowledge on carbonation and to follow the changes in its scientific and engineering interpretation. The literature was collected by targeted searches in Scopus, Web of Science and Google Scholar from January 2024 to March 2026. The following search terms were used: carbonation, cementitious materials, durability, reinforcement corrosion, accelerated carbonation, natural carbonation, supplementary cementitious materials, blended cements and CO₂ uptake. To improve the completeness of the search, backward and forward citation searching, review of reference lists and consultation of standard historical and technical works often cited in this field were also used [1-8, 10, 11, 14, 16-22].

The collected material was assessed in relation to the aims of the article. The scope included the historical development of knowledge on carbonation from lime materials to reinforced concrete [1, 10, 11, 14-18], mechanistic explanations of the process

[2, 16, 21, 22], and its interpretation in relation to steel depassivation, corrosion risk and durability assessment [3, 4, 17-20]. The paper also includes current discussions on carbonation in blended and low-clinker systems, especially in relation to testing methodology and CO₂ uptake [2, 5-9, 12, 13].

The literature finally included in the paper was selected because of its relevance to the argument presented here and its ability to show wider changes in the understanding of carbonation. The aim was not to prepare a full list of all contributions to this field, but to indicate milestones showing important historical, methodological and interpretative changes [1, 2, 16-22]. Because this article is not a systematic review, no PRISMA diagram or bibliometric meta-analysis is presented. Nevertheless, the review was prepared with a clear scope, search logic, selection rules and limitations of the adopted approach.

3. BEFORE CARBONATION OF CONCRETE BECAME

A PROBLEM: LIME, MORTARS AND THE FIRST BUILDERS

Vitruvius [14] is probably the earliest written reference point for the historical understanding of building materials. His *De Architectura* (Ten Books on Architecture) documented Roman building knowledge in the first century BC. Although Vitruvius did not describe carbonation in the modern chemical sense, his treatise reflects an early stage in which lime materials were understood mainly through practice, empirical material selection and accumulated building experience.

Long before the development of cement chemistry, carbonation controlled the behaviour of air-hardening binders. Lime mortars gained strength through the reaction of calcium hydroxide with atmospheric carbon dioxide, which led to the formation of calcium carbonate [1, 10]. In such conditions, carbonation was not a defect or a durability threat. It was the basis of hardening.

This historical starting point shows that carbonation first appeared in building practice as a useful process. Pre-modern builders did not know the concepts of concrete cover, depassivation or durability design. Their main problem was whether the binder set, hardened and kept cohesion over time. The scientific language later used to describe carbonation did not yet exist, although its visible effects were well known in practice.

This long, pre-theoretical period is not only a simple introduction to concrete technology. It helps to explain why carbonation was later reinterpreted as a durability threat. The reaction itself was not new, but its engineering meaning changed together with the technological context. Figure 1 presents the historical background of

this change, including the move from empirical practice to more systematic knowledge about binders.

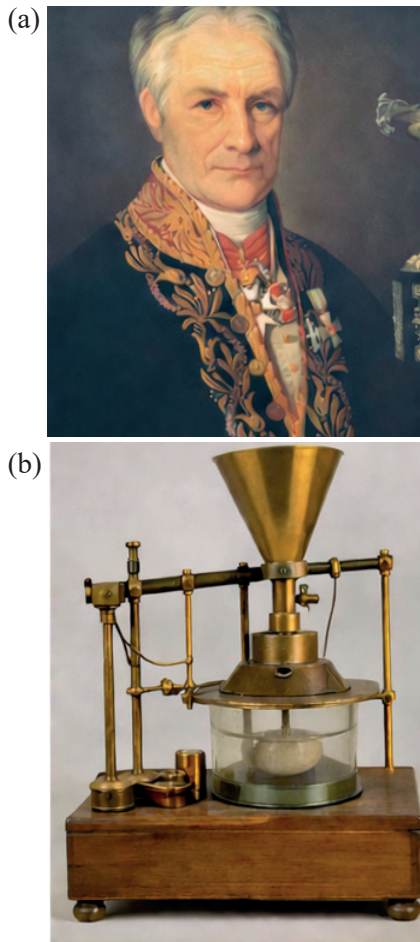


Fig. 1. (a) Louis Vicat (1786-1861), (b) Vicat apparatus. Source: portrait and apparatus image based on Wikipedia materials

4. FROM HYDRAULIC LIME TO PORTLAND CEMENT: LOUIS VICAT AND JOSEPH ASPDIN

Louis Vicat [10] (1786-1861) represents the transition from empirical practice to organized binder science. His work on hydraulic lime and the treatise connected with his name helped to organize knowledge about lime and hydraulic binders. It also created a language in which setting and hardening could be discussed more clearly. From this perspective, Vicat did not discover carbonation as a separate phenomenon, but he helped to create the intellectual basis that later allowed its mechanistic interpretation.

Joseph Aspdin [11] (1778-1855) represents another important change: the industrialization of binder production. His 1824 patent [11] for Portland cement is usually treated as a key starting point of the Portland cement era, as shown in Figure 2. From the viewpoint

of the history of carbonation, Aspdin is important not because his patent solved the carbonation problem, but because it opened the era of cementitious materials in which carbonation was later studied as a change in hydrated cement paste and finally as a durability problem of reinforced concrete.

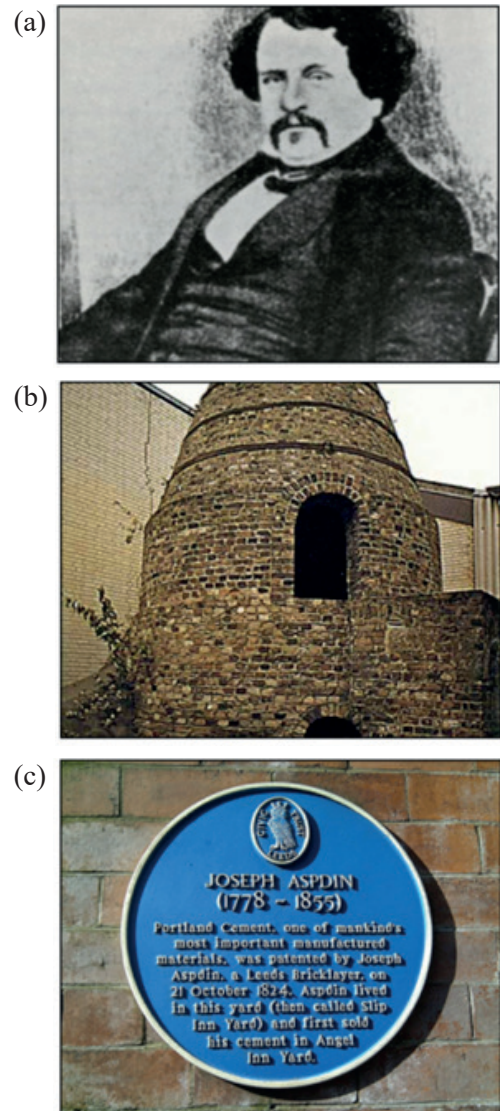


Fig. 2. Joseph Aspdin and selected historical references to the early Portland cement era: a) portrait attributed to Joseph Aspdin, b) preserved 19th-century bottle kiln associated with early cement production, c) Leeds Civic Trust plaque commemorating Aspdin's 1824 Portland cement patent. Source: author's compilation based on Wikimedia Commons and Cement Kilns documentation

Another important milestone of the nineteenth century was the work of Joseph Monier [15], a French gardener and inventor who patented iron-reinforced cement containers in 1867. Although Monier's

contribution was not directly related to the chemistry of carbonation, it marked the beginning of reinforced concrete as a new structural material. When steel was embedded in cementitious materials, carbonation gradually gained a new engineering meaning: it stopped being only a chemical change in the binder and became a process able to reduce alkalinity and influence reinforcement protection.

Together, Vicat and Aspdin mark the beginning of the nineteenth century as a key stage in which the scientific and industrial foundations of modern carbonation research were created. One symbolizes the scientific ordering of binder knowledge, and the other the beginning of an industrial family of materials in which carbonation later became an important long-term problem.

5. CARBONATION BECOMES A SCIENTIFIC AND ENGINEERING ISSUE: VERBECK AND HAMADA

In the twentieth century, carbonation stopped being only a practical observation and became a subject of cement chemistry. A key milestone was the classical work by Verbeck [16] on carbonation of hydrated

Portland cement, which organized the chemical changes connected with carbonation in hardened cement paste. Verbeck's contribution was important because it described carbonation as a transformation of specific hydration phases, not only as an external surface effect. The main chemical changes connected with this approach are shown schematically in Figure 3.

The next important step was to link carbonation with the behavior of reinforced concrete. Hamada's work [17] on neutralization of concrete and corrosion of reinforcing steel explained the engineering meaning of the decrease in alkalinity caused by carbonation. It is also one of the earliest widely cited works describing the use of phenolphthalein to identify the carbonated zone in concrete – a method that later became standard in carbonation assessment. This change connected carbonation with the loss of passive steel protection and helped to establish the classical durability approach in which carbonation is treated as a phenomenon preceding corrosion risk. The engineering meaning of this change, especially for reinforcement protection, is shown in Figure 4.

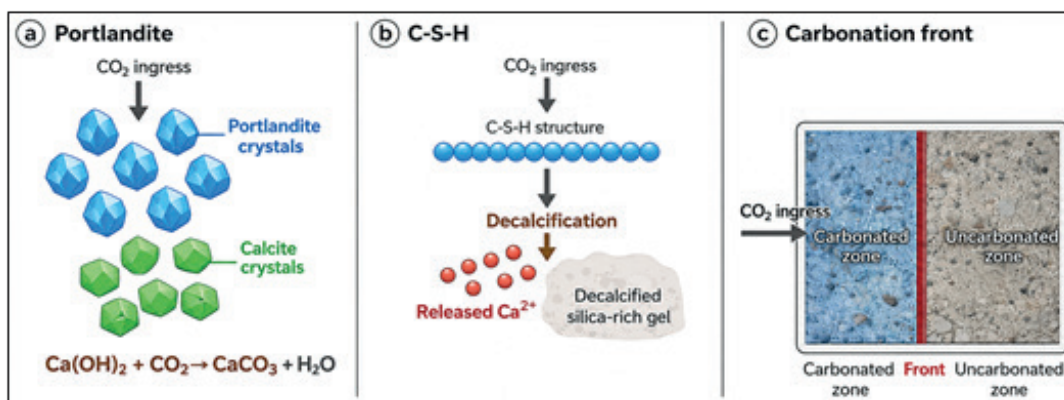


Fig. 3. Author's schematic illustration of carbonation chemistry based on Verbeck [16]: portlandite carbonation, decalcification of C-S-H and an idealised carbonation front in hardened cement paste

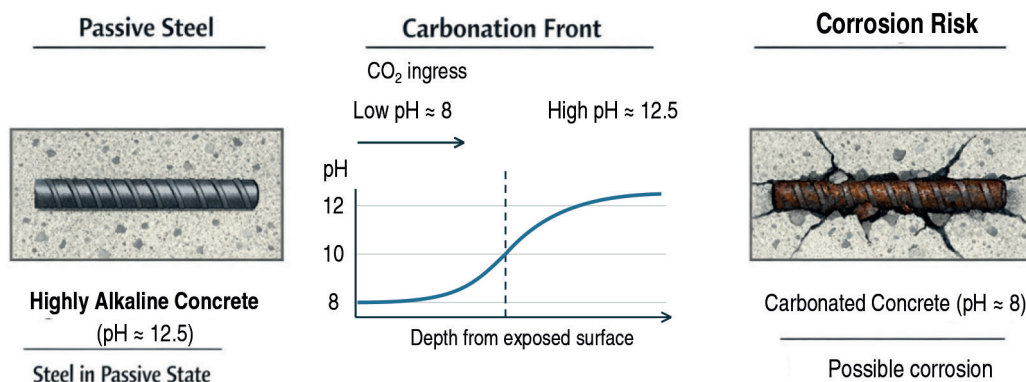


Fig. 4. Author's schematic interpretation based on Hamada [17]: CO_2 ingress, reduced alkalinity and risk of steel depassivation. The pH profile is illustrative; corrosion also depends on exposure conditions

At the same time, current literature clearly shows that the relation between carbonation and corrosion is not strictly deterministic. Carbonation may create a thermodynamic condition for depassivation, but the corrosion rate strongly depends on moisture, degree of pore saturation, oxygen transport and exposure conditions [3, 4]. For a historical review, this distinction is important because it separates the moment when the durability paradigm appeared from its later clarification.

6. CARBONATION AS A DURABILITY

AND DIAGNOSTIC CONCEPT: TUUTTI AND PARROTT

Kyösti Tuutti's 1982 dissertation *Corrosion of Steel in Concrete* [18] is one of the key texts of durability engineering because it included carbonation in the now classical model of initiation and propagation of steel corrosion in concrete. In this approach, carbonation became one of the main mechanisms able to end the initiation phase by lowering alkalinity at the reinforcement depth. The question was no

longer simply whether carbonation occurs, but when the carbonation front will reach the steel. Figure 5 schematically illustrates this durability concept.

Parrott [19, 20] developed this way of thinking towards diagnostics and assessment of real structures. In papers published in 1990 and 1994, he analyzed the relationship between carbonation depth, moisture condition, corrosion and the observed condition of reinforced concrete elements. His contribution is important because it moved carbonation research beyond an abstract mechanism and towards the practical assessment of existing structures. Figure 6 presents this diagnostic interpretation.

The Tuutti-Parrott stage marked the moment when carbonation was fully included in service-life thinking for structures. Modern syntheses also show that carbonated concrete must be assessed with attention to moisture exposure and transport conditions. Without this context, the simple equation $\text{carbonation} = \text{corrosion}$ becomes an excessive simplification [3, 4].

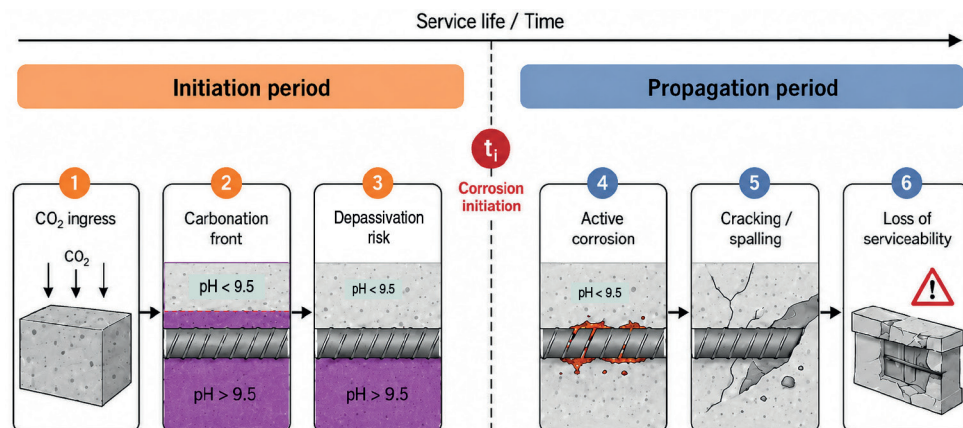


Fig. 5. Author's schematic illustration based on Tuutti's initiation-propagation concept [18]: carbonation, risk of depassivation, corrosion and possible subsequent damage. The pH values are indicative

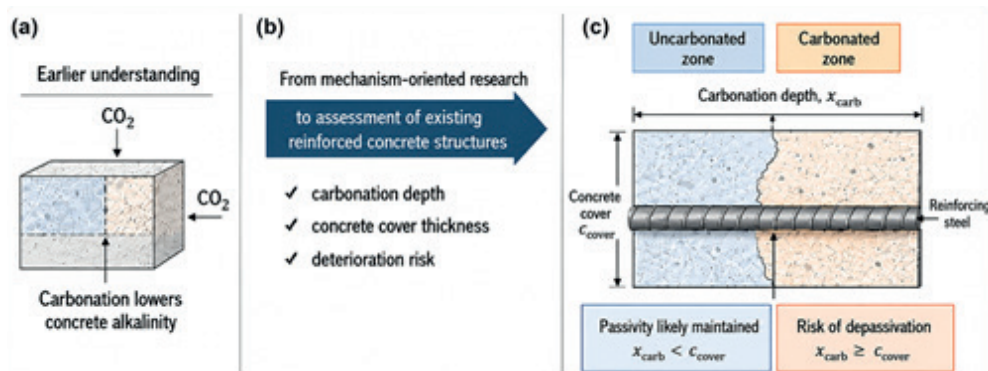


Fig. 6. Author's schematic diagnostic interpretation based on Parrott [19, 20]: carbonation depth relative to concrete cover. Assessment of corrosion risk also requires moisture and exposure conditions

7. FROM OBSERVATION TO MODELLING: PAPADAKIS AND THE MECHANISTIC TURN

Papadakis and co-workers [21, 22] gave carbonation research a mechanistic and predictive dimension by describing carbonation as a coupled transport–reaction process. In this approach, the progress of carbonation depends on CO_2 ingress through the pore system, its dissolution in the pore solution, reactions with portlandite and CSH in the cement paste, and the resulting movement of the carbonation front within a porous cementitious material. The moisture state plays a key role because it controls both CO_2 transport and the availability of pore solution for carbonation reactions. The main elements of this interpretation are presented schematically in Figure 7.

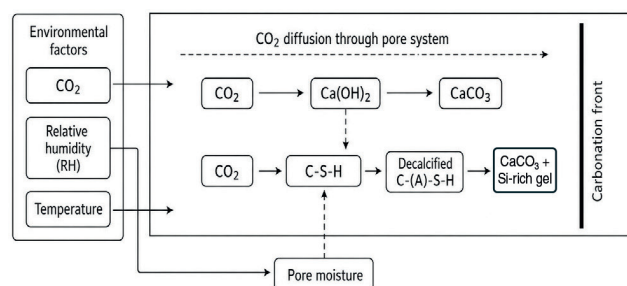


Fig. 7. Author's schematic interpretation based on Papadakis et al. [21, 22]: CO_2 transport, moisture and chemical reactions controlling the advance of an idealised carbonation front

This was an important conceptual advance because carbonation depth could then be treated not only as an empirical observation revealed by phenolphthalein, but also as the result of coupled physical and chemical processes. Binder composition, porosity, moisture content and reactive capacity became explicit variables of the analysis, not only background conditions.

The mechanistic turn remains central for modern modeling. It also explains why accelerated and natural carbonation cannot simply be treated as equivalent phenomena. When carbonation is understood as a coupled transport–reaction process, differences in CO_2 concentration, humidity, pre-conditioning and sample history become scientifically important and not only procedural [5-8, 22].

8. REINTERPRETATION IN THE ERA OF BLENDED AND LOW-EMISSION BINDERS

Modern research on carbonation is largely shaped by changes in cementitious systems themselves. The increasing use of supplementary cementitious materials (SCM) and low-clinker binders means

that conclusions formulated for Portland cement concretes cannot be automatically transferred to blended systems [2, 5-8]. Differences in the hydrate phase assemblage, pore solution chemistry, buffering capacity and pore structure affect both the kinetics of carbonation and the interpretation of test results.

In concretes containing high amounts of SCMs, carbonation may differ from ordinary Portland cement systems in several connected ways. First, the replacement of clinker usually reduces the amount of portlandite and changes the alkaline reserve of the binder. As a result, less CO_2 may be required to lower the pH, even if the measured carbonation depth does not fully describe the risk of corrosion. Second, the main hydrate assemblage changes. In slag, fly ash or other blended systems, C-S-H or C-A-S-H phases may have a lower Ca/Si ratio and may decalcify differently during carbonation. This affects not only the chemical buffering capacity but also the pore structure after carbonation. Third, SCMs often refine the pore structure when curing is sufficient, which may reduce transport, but insufficient curing or high replacement levels may increase carbonation rates because the chemical reserve is lower. Therefore, the carbonation resistance of SCM-rich concretes is controlled by a balance between transport properties and chemical buffering capacity.

This balance is one of the reasons why carbonation depth should not be interpreted in isolation. A larger carbonation depth in an SCM concrete does not always mean the same durability state as the same depth in an OPC concrete. The interpretation should also include moisture exposure, curing history, binder type, replacement level, pore solution chemistry and the presence or absence of reinforcement.

The critical review by RILEM TC 281-CCC [2] showed that carbonation of concretes with SCM is clearly different from carbonation of concretes based only on Portland cement, and that exposure conditions, curing history and SCM characteristics must be interpreted together. Later reports [5, 6] stressed important differences between standardised methods and showed that accelerated and natural carbonation results are not directly interchangeable without proper contextual correction. The 2025 closing letter of the committee [7] and the CPC-18R1 recommendation from 2026 [8] strengthened this conclusion by indicating still existing research needs and updating methodological guidelines for measuring carbonation depth with pH indicators. Figure 8 summarizes the current interpretation context of carbonation in concretes with SCM.

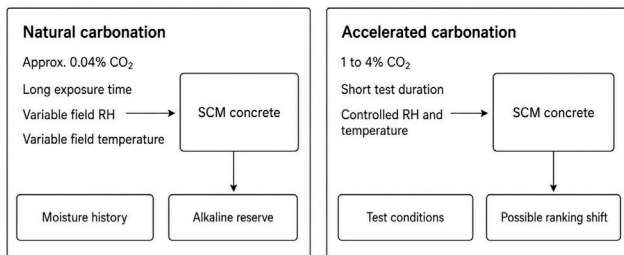


Fig. 8. Current interpretation context of carbonation in concretes containing supplementary cementitious materials (SCM), showing the influence of binder composition and the methodological differences between natural and accelerated exposure.

Source: own elaboration based on [2, 5-8]

The review by Bernal et al. [5] is especially useful here because it stressed the combined role of CO₂ concentration, relative humidity and CO₂ buffering capacity in carbonation tests. This observation helps to explain why the ranking of materials under accelerated exposure may differ from the ranking obtained under natural exposure, especially in blended systems.

8.1. Consequences for carbonation assessment methods

The reinterpretation of carbonation in SCM-rich concretes is closely connected with the method used for its assessment. The phenolphthalein test is simple, inexpensive and useful for determining the apparent carbonation depth. It is therefore suitable for routine comparison and for diagnostics of existing structures. Its limitation is that it identifies a pH-related boundary rather than the complete chemical progress of carbonation. In blended systems, where the pore solution chemistry and buffering capacity differ from OPC concretes, the colour change may not fully represent the degree of carbonation of individual hydrate phases or the actual corrosion risk.

Natural carbonation tests are more representative of field exposure because they use realistic CO₂ concentrations and moisture variations. However, they require long exposure times and their results are strongly affected by local climate, curing history and specimen pre-conditioning. Accelerated carbonation tests are much faster and useful for comparative assessment, but high CO₂ concentrations may change the balance between transport and reaction. This is particularly important for SCM-rich concretes, where the ranking of mixtures under accelerated exposure may differ from the ranking observed under natural conditions.

For this reason, accelerated tests should not be treated as a direct substitute for natural carbonation without validation. They are valuable for screening and

comparative studies, but service-life prediction and durability classification require careful interpretation. In concretes with high SCM contents, carbonation assessment should preferably combine carbonation depth with information on binder composition, curing, moisture state, pore structure and, where reinforcement is present, corrosion-relevant exposure conditions.

9. CARBONATION AS A DURABILITY THREAT AND AS CO₂ UPTAKE

In more recent literature, carbonation is interpreted in two connected but different perspectives. In studies on reinforced concrete, it remains mainly a durability issue because the decrease in alkalinity may lead to steel depassivation and increase the risk of reinforcement corrosion [3, 4]. In environmental studies, the same process is analyzed as a mechanism of CO₂ uptake by cementitious materials during service life, after demolition and during reuse [9, 12].

The importance of this second perspective is confirmed by global assessments. Xi et al. [9] estimated cumulative uptake at 4.5 Gt C in 1930-2013, which corresponds to about 16.5 Gt CO₂ and about 43% of cement process emissions in that period. Updated estimates [12] showed cumulative uptake of 23.89 Gt CO₂ in 1930-2023, equal to about 52.3% of cumulative cement process emissions in that period. Annual uptake reached 0.93 Gt CO₂/year in 2023. Experimental studies [13] also confirmed measurable CO₂ uptake in Portland cement pastes and blended cement pastes under controlled conditions.

It is therefore necessary to separate the scale and aim of assessment. At the structural scale, carbonation may be unfavourable if it leads to the loss of reinforcement protection. At the material, resource and life-cycle scale, it may at the same time be an element of the carbon balance. These perspectives are not contradictory, but they should not be mixed without a clear indication of whether the analysis concerns the durability of a reinforced concrete member, the behaviour of a material, or CO₂ uptake.

10. HISTORICAL SYNTHESIS, PRACTICAL IMPLICATIONS AND FINAL REMARKS

The milestones discussed above show that the history of carbonation is a history of changing research questions. For lime binders, the main questions concerned hardening and cohesion of the material. For Portland cement, the analysis moved towards the chemistry of hydrated paste.

The development of reinforced concrete shifted attention to alkalinity, steel depassivation and corrosion risk, while mechanistic modeling made it possible to describe carbonation as a transport-reaction process depending on binder composition, moisture, pore structure and exposure conditions [1-8, 21, 22]. Modern research on blended and low-clinker binders and on CO₂ uptake does not replace this interpretation, but extends it and requires more careful selection of test methods [2, 5-8, 9, 12, 13]. The main stages in the development of knowledge on carbonation are presented in Table 1.

Three practical consequences for current carbonation research follow from this synthesis. First, conclusions obtained for systems based only on ordinary Portland cement (OPC) should not be directly transferred to concretes with a high content

of supplementary cementitious materials (SCM), because buffering capacity, microstructure and pore solution chemistry change [2, 5, 6]. Second, carbonation depth alone is not a universal durability indicator. Its meaning depends on moisture, exposure class, binder composition, presence of reinforcement and conditions that promote or limit corrosion development [5-8]. Third, the test method should follow the aim of assessment. Service-life prediction, material comparison, diagnostics of existing structures and estimation of CO₂ uptake are all connected with carbonation, but they answer different questions and should not be interpreted by one simplified criterion [5-8, 12].

The ecological transformation of cementitious materials makes this problem more important. Low-emission and blended systems can no longer be

Table 1. Key stages in the development of knowledge on carbonation. Source: own elaboration based on [1-22]

Period / year	Researcher / concept	Main contribution	Importance for the development of knowledge on carbonation
1st century BC	Vitruvius	Early written synthesis of Roman knowledge on building materials, construction practice and durability in <i>De Architectura</i> .	The earliest textual reference point for the practical understanding of lime construction before modern cement chemistry.
Antiquity - 19th century	Lime binder	Carbonation understood in practice as the hardening mechanism of lime mortars exposed to atmospheric CO ₂ .	The earliest stage of understanding: high practical importance, but without formal cement chemistry.
1817-1818	Louis Vicat	Systematisation of knowledge on hydraulic lime and hydraulic binders.	Transition from craft knowledge to scientific description of binders.
1824	Joseph Aspdin	Patent for Portland cement and symbolic beginning of the industrial cement era.	Opening of a family of materials in which carbonation later became a durability problem.
1867	Joseph Monier	Patent for iron-reinforced cement containers and early development of reinforced concrete.	Beginning of the reinforced concrete era, in which carbonation was later directly linked with reinforcement protection and durability assessment.
1958	G.J. Verbeck	Systematic description of the chemistry of carbonation of hydrated Portland cement.	Carbonation became a cement chemistry issue, not only a practical observation.
1968	M. Hamada	Linking concrete neutralisation with loss of alkalinity and risk of reinforcement depassivation.	Carbonation gained a clear engineering meaning for reinforced concrete.
1982	Kyösti Tuutti	Inclusion of carbonation in the model of initiation and propagation of steel corrosion in concrete.	Carbonation became a key durability concept and a basis for service-life thinking.
1989-1991	V.G. Papadakis	Physicochemical modelling of carbonation as a coupled transport-reaction process.	Transition from empirical description to mechanistic and predictive analysis.
1990-1994	L.J. Parrott	Diagnostic interpretation of carbonation depth, moisture influence and damage assessment in existing structures.	Transition from the mechanism itself to diagnostics and condition assessment.
2020	Critical review by RILEM TC 281-CCC	Critical synthesis of carbonation of concrete with supplementary cementitious materials.	It showed that SCM-rich systems need a separate interpretation from OPC systems.
2016-2024	Xi et al.; Wu et al.; Andrade et al.	Quantitative determination of global CO ₂ uptake and experimental confirmation of carbon binding in cementitious systems.	Separation of carbonation as a durability threat from carbonation as a carbon uptake mechanism.
2024	Bernal et al.; Vollpracht et al.	Critical review of standardised test methods and database-based analysis of carbonation factors.	Indication of the limits of direct comparison between natural and accelerated carbonation and of the role of testing methodology.
2025-2026	De Belie and Bernal; CPC-18R1	Synthesis of research needs and updated recommendation for measuring carbonation depth using pH indicators.	Methodological consolidation and modern reinterpretation in low-emission systems.

interpreted only from the durability point of view. They should also be considered in relation to CO₂ uptake, testing methodology and the future development of material strategies based on a circular economy [9, 12, 13, 23-27]. These issues should be treated as an important direction of future carbonation research and not as a separate subject outside the main discussion on durability.

In modern concrete science, carbonation is placed at the intersection of durability, testing methodology and

environmental assessment. It is therefore no longer enough to consider only its influence on the service life of structures. Future research should also explain how carbonation can be interpreted and controlled in low-emission binders and recycled cementitious materials, without excessive simplification of their long-term performance assessment [2, 5-8, 9, 12, 13, 23-27].

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