

CO₂ mineralization of demolished concrete wastes into a supplementary cementitious material – a new CCU approach for the cement industry

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Abstract

This contribution discusses a new carbon capture and utilization (CCU) approach based on CO₂ mineralization of cement paste from recycled concrete that can balance a significant part of the CO₂ emissions associated with cement production. Carbonation of the calcium-bearing phases is achieved in a few hours at ambient temperature and pressure and with a relatively low CO₂ gas concentration (< 10 %). The carbonation of cement paste produces calcite and an amorphous alumina-silica gel, the latter being a pozzolanic material that can be utilized as a supplementary cementitious material. The pozzolanic reaction of the alumina-silica gel is very rapid as a result of its high specific surface and amorphous structure, implying that composite cements containing carbonated cement paste exhibit a rapid strength gain. The successful implementation of this CCU approach relies also on improved concrete recycling techniques and methods to separate out the cement paste fines. Full concrete recycling will further improve concrete's circular utilization by using recycled aggregates instead of natural deposits of aggregates. Although the feasibility of the process has already been demonstrated at the industrial scale, there are still several open questions related to optimum carbonation conditions and performance of carbonated material in novel composite cements.

Keywords: Circular Economy; Carbonation, Supplementary Cementitious Material; Carbon neutrality

1 Introduction

The use of concrete has been growing continuously in the 20th and 21st centuries. Today, concrete is the most widely used man-made material and the most important material of our built environment [1]. The production of concrete consumes substantial amounts of natural resources like limestone, sand and aggregates. The manufacture of cement, the 'glue' in concrete, is associated with significant CO₂ emissions accounting for approx. 7 % of the anthropogenic CO₂ emissions in the World [2]. Therefore, global warming and the significant consumption of natural reserves have pushed the building industry to search for new and sustainable solutions. New breakthrough technologies for lowering the CO₂ footprint include carbon capture in combination with storage (CCS) and utilization (CCU) [2] whereas a circular utilization of demolished concrete and improved waste recycling methods may reduce the use of natural resources [3].

Mineral carbonation is a promising CCU technology [4], which is feasible for applications within the construction industry. When applied to cement, the fundamental concept is that the hydraulic cement components and their hydration products

can react with carbonate ions and thereby CO₂ in a humid/aqueous medium. Mineral carbonation has earlier been proposed as a route to sequester CO₂ for alkaline industrial wastes [4,5]. These materials include steel slags, fly ashes or red mud that can be used for constructions after their reaction with CO₂ [6-8]. In the 1970'ies, it was proposed that carbonation reactions may be used to accelerate concrete and mortar strength evolution [9,10]. However, this type of application has only been rarely used in the past. The interest in carbonation hardening has revived in parallel with the cement industry's attention to the reduction of CO₂ footprint [11], which has resulted in a better understanding of the involved carbonation mechanisms [12,13]. Recently, it has been proposed that carbonation treatment can be applied to improve the properties and applicability of recycled concrete aggregates (RCA) [14]. RCA may have inferior properties compared to fresh aggregates as remains of cement paste may be present on their surface [3]. However, carbonation treatment of RCA results in densification of the layers of hydrates on the RCA surface [15,16] which facilitates the use of RCP for concrete production. The application of the fine fraction from concrete recycling is challenging.

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This fraction, the so-called recycled concrete paste (RCP), is composed of the partially hydrated cement paste, sand and a fine fraction of the aggregates [17]. Application of the fine fraction from concrete recycling to either cement clinker production [17] or composite cement production [18] is currently limited and requires a special treatment of the RCP. When properly treated, RCP is characterized by similar properties as limestone in composite cements [18], e.g. compressive strength evolution. However addition of RCP results in an increased water demand for the composite cements [19]. The enforced carbonation techniques were explored to modify the properties of the RCP, initially by Iizuka et al. [20-24] and Ghacham et al. [25,26]. Fang et al. [27,28] have investigated a two-step wet-carbonation process which allows valorization of recycled cement paste into the added value products of calcite and alumina-silica gel. Lu et al. [29] and Zajac et al. [30-33] have applied CO₂ mineralization to modify the recycled cement paste into a new supplementary cementitious material (SCM). These exploratory studies show that most of the CO₂, originally released by limestone calcination during clinker production, can be sequestered by carbonation of the RCP, provided that the paste is separated from the sand and aggregates. Fully carbonated RCP is composed of calcium carbonate and an alumina-silica gel with pozzolanic properties, implying that the carbonated RCP (cRCP) can be used as an SCM. The application of the cRCP for cement production has two principal advantages: (i) it allows for CO₂ sequestration during the carbonation process and (ii) it can be used in composite cements, thereby further reducing CO₂ emissions of cement production.

This paper discusses the status of current research on carbonation of recycled cement pastes and the applications of the carbonated material in new composite cements.

2 CCU by cement paste carbonation

The method is schematically illustrated in Figure 1, and it relies on improved concrete recycling where the fines rich in cement paste are partly separated out from sand and coarser aggregate.

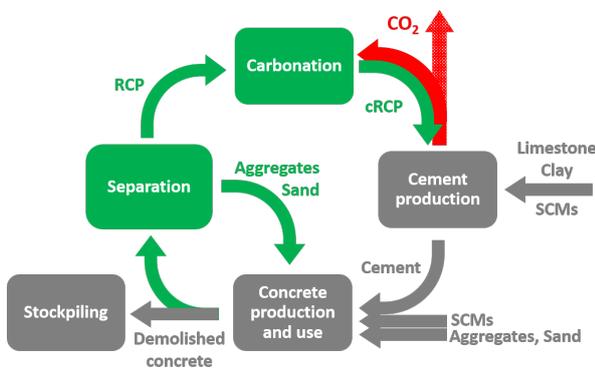


Figure 1. The life cycle of cement and concrete production including concrete recycling and re-carbonation of the concrete fines. The part in gray shows the traditional way of production and use of cement. The green part highlights the improvements of improved concrete recycling and re-carbonation associated with the concept discussed in this contribution. RCP – recycled concrete paste, cRCP – carbonated RCP.

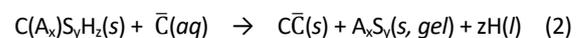
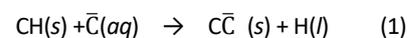
These fines are then wet carbonated, producing a new material that can be utilized in the production of new composite cements [33].

Already today recycled aggregates can be employed in concrete production, despite the disadvantages of higher water demand [34-36]. Still there is a need to improve the properties of these materials [37]. The application of the CCU concept in Figure 1 requires advanced recycling and separation technologies, enabling an appropriate separation of the aggregates, sand and cement paste to produce as pure RCP as possible. This permits the application of high-quality recycled sand for concrete production and the rather pure RCP paste for the re-carbonation processes. To cope with this challenge advanced recycling technologies are being currently developed, including SmartCrusher [38], Advanced Dry Recovery (ADR) [39], vertical rolled soft milling followed by separation [40] among others.

Recent published studies [31,41] reveal that the RCP carbonates readily at ambient pressure and temperature when an aqueous mineralization process is employed, and full carbonation of the materials can be achieved in few hours. The concentration of CO₂ gas in the process includes the levels found in the exhaust gas from industrial productions and combustion plants and has only a limited impact on the carbonation reaction and products. This enables a direct usage of the exhaust gases from cement plants in the wet carbonation process. Thus, this carbonation can be used to sequester CO₂ released from clinker production, reducing further needs for carbon dioxide capturing and/or purification in the production of sustainable cement. The carbonated RCP contains CO₂ bound in the material and can be directly used as an SCM in new composite cements [29,32]. Investigations conducted so far reveal that clinker replacement factors in the range 20 – 40 % may be achieved by this approach, resulting in a significant further reduction in CO₂ emissions. The importance of this application should also be considered in the light of declining availability of conventional SCMs such as slags and fly ashes [42].

2.1 Mechanisms of enforced carbonation

In recent studies [30-33], the wet carbonation process has been investigated in detail as it is more efficient than dry carbonation [43]. The carbonation reaction is a dissolution – precipitation process with some resemblance to those observed in hydrating cementitious systems [44], although it involves an initial dissolution of CO₂ (\bar{C}) from the gas phase forming carbonate ions in solution [31,23]. In the subsequent aqueous carbonation of the partially hydrated cement paste, several reactions occur, which convert the cement clinker phases and hydrates into calcium carbonate ($\bar{C}\bar{C}$) and an amorphous alumina-silica gel (A_xS_y -gel), as schematically represented for the portlandite (CH) and C-S-H phase [30,45]:



In a wet reactor, portlandite (CH) reacts as the first phase followed by the other hydrates along with the anhydrous calcium silicate phases (C₂S) from the cement clinker.

It is noticeable that the minor clinker phases, tricalcium aluminate (C_3A) and ferrite (C_4AF), as well as anhydrous slag in blended cements do not react during the first few hours of wet carbonation [31,32].

Calcium carbonate is the main carbonation product, and in a wet carbonation reactor it precipitates as calcite [30,45] in contrast to carbonation processes conducted at a relative humidity lower than 100 %, where several of the calcium carbonate polymorphs are formed [46,47]. Calcium carbonate precipitates as agglomerates of small and larger crystals that are inter-grown, as illustrated in Figure 2A, and the morphology of calcite is affected by the alkali concentration in the carbonation solution [41]. Calcite precipitates mainly in the space occupied by the solution (Figure 2B), reflecting that calcium is leached from the cement paste grains and combines in the solution with the dissolved CO_2 . Only at the later stages of the carbonation process, a co-precipitation of calcium carbonate with the alumina-silica gel takes place in the space occupied initially by the cement grains [30,31].

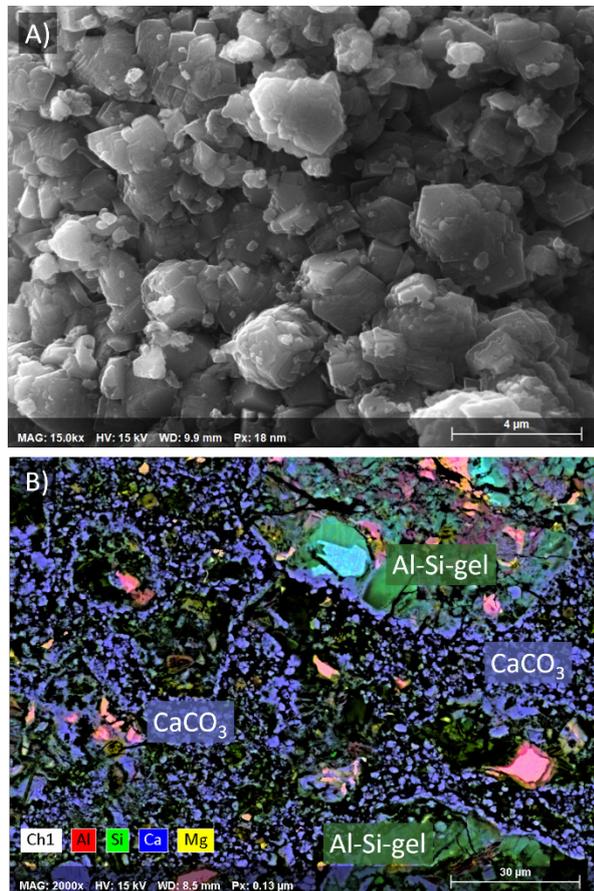


Figure 2. (A) SEM-SE picture showing the calcium carbonate morphology. (B) SEM-BSE image with elemental mapping of the carbonated cement paste. Calcium carbonate and the alumina-silica gel are highlighted.

During carbonation, the principal hydrate phase, C-(A)-S-H phase, initially decalcifies to reach a Ca/Si ratio of approx. 0.67 and then decomposes and form the alumina-silica gel [48]. The complete decomposition of the C-(A)-S-H phase into the alumina-silica gel is clearly seen in the ^{29}Si NMR spectra

(Figure 3) of a hydrated Portland cement paste (CEM-I) carbonated for 360 min. The spectrum of the hydrated cement contains resonances from remains of belite (Q^0 ; -71.3 ppm) and the Q^1 (-79.5 ppm), $Q^2(1Al)$ (-81.4 ppm) and Q^2 (-84.7 ppm) sites of the C-(A)-S-H phase, which is typical for a well hydrated Portland cement pastes. After carbonation these phases have been consumed and transformed into an amorphous alumina-silica phase dominated by Q^3 and Q^4 -type peaks in the range -90 to -110 ppm. The broad and featureless resonance after carbonation reflects the amorphous structure and is characteristic for alumina-silica gels formed by wet carbonation processes at the different conditions [30,46,47]. The decomposition of the C-(A)-S-H phase is also seen by ^{27}Al NMR (Figure 3) as the tetrahedral resonance (Al(IV)) from Al in this phase (~68 ppm) disappears upon carbonation and leads to the formation of a new resonance at approx. 58 ppm, which originates from Al(IV) sites in the alumina-silica gel (*i.e.*, Al(O-Si)₄ sites). Moreover, the resonances from Al in octahedral coordination (Al(VI)) of the AFt and AFm phases decreases also significantly, demonstrating that these phases are also carbonated and contribute with Al to the alumina-silica gel. The small Al(VI) peak after carbonation is attributed to hydrocalcite, which does not decompose up on wet carbonation [30,31]. The ^{27}Al and ^{29}Si NMR spectra clearly reveal the formation of an alumina-silica gel with Al(IV) integrated in the structure and not a pure silica gel as predicted by thermodynamic modelling [30] and proposed in an other study [29]. The formation of a similar gel has also been reported in a study of natural and accelerated carbonation of concrete [47].

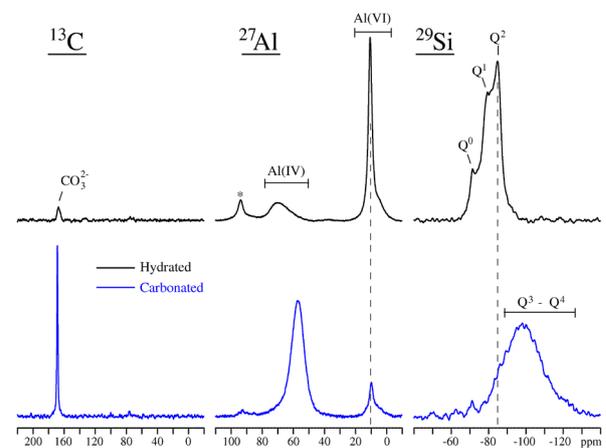


Figure 3. ^{13}C , ^{27}Al and ^{29}Si NMR spectra of a hydrated Portland cement (CEM-I) paste before and after enforced carbonation (360 min.), acquired at 7.05, 14.09 and 7.05 T using spinning frequencies of 7.0, 13.0 and 7.0 kHz, respectively. The ^{13}C NMR spectra show the formation of a significant amount of $CaCO_3$. The ^{27}Al NMR spectra clearly differentiate Al in tetrahedral (Al(IV)) and octahedral (Al(VI)) coordination. Al(IV) is found in the C-(A)-S-H phase (hydrated) and alumina-silica gel (carbonated) whereas Al(VI) is mainly present in the AFt and AFm phases. The ^{29}Si NMR spectrum of the hydrated sample shows resonances from belite (Q^0) and the C-(A)-S-H phase (Q^1 and Q^2 sites) whereas $Q^3 - Q^4$ indicate more condensed SiO_4 sites in the alumina-silica gel of the carbonated RCP. The asterisk indicates a spinning sideband.

Mass balance calculations and ^{23}Na NMR experiments indicate that the alumina-silica gel also contains minor fractions of Ca and alkali ions. Furthermore, it is partly hydrated as it contains hydroxyl groups and chemically bound water [28,30,32,46]. Moreover, the alumina-silica gel exhibits the characteristic pattern for an amorphous phase in the XRD patterns and has a very high surface area in the range of $150 \text{ m}^2/(\text{g gel})$ [32].

The carbonation reaction is rapid under wet conditions provided that the aqueous system is saturated with CO_2 . The content of calcium carbonate is approximately proportional to the amount of bound CO_2 and to the overall carbonation reaction degree following equation 1. The evolution in calcium carbonate with carbonation time (Figure 4) demonstrates that close to full carbonation can be achieved after 2 hours and that more than 90% carbonation is achieved in just 30 minutes. This fast reaction is associated with the strong undersaturation of the hydrates and cement clinker phases under the effect of dissolved CO_2 which favors fast carbonation [31,45]. Furthermore, because of calcium leaching from the cement paste grains, an open microstructure is formed. This contrasts the microstructure formed in dry carbonation experiments or natural carbonation where dense layers of carbonation products may form [49,50].

The aqueous mineral carbonation of the Portland clinker and the hydrates can be divided in to two stages, depending on the rate limiting process [31,45,23], as indicated in the pH evolution with time shown in Figure 4B. The pH increases rapidly in the first stage, which reflects that the hydrates dissolve quickly and in a rapid precipitation of the carbonates and a decrease in the CO_2 concentration of the solution. During this stage of carbonation, the reaction kinetics is limited by the availability of CO_2 . However, recent results [31,45] indicate that the kinetics of calcium carbonate precipitation and nucleation can slow down the reaction as well. This effect can be compensated by addition of seeds for calcium carbonate nucleation and growth. The alkali concentration has a strong impact on the CO_2 solubility in water and thereby on the kinetics of the reaction during the first stage [41]. Dissolved alkalis accelerate the process resulting in a higher CO_2 concentration in the solution. This phenomenon has two consequences. Firstly, there is a larger availability of CO_2 in the solution and secondly, a higher undersaturation with respect to the hydrates is achieved. During the second stage, the dissolution of the hydrates is limiting the carbonation process, resulting in a lowering of pH caused by the increase in CO_2 concentration.

2.2 Pozzolanic properties

Recent studies [29,32] have demonstrated that carbonated recycled concrete paste can be successfully used as an SCM to produce new composite cements. The pozzolanic activity of the alumina-silica gel results in the formation of a phase assemblage which is similar to those found in composite cements containing fly ash and limestone. However, the carbonated alumina-silica gel is characterized by a faster pozzolanic reaction as compared to fly ashes.

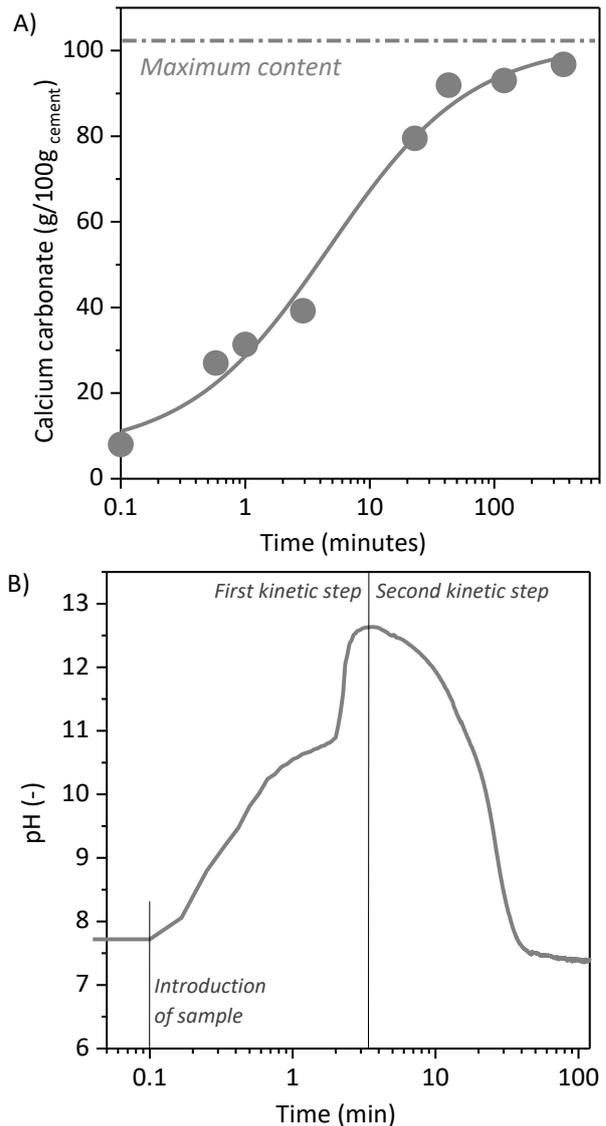
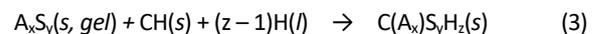


Figure 4. (A) Evolution of the calcium carbonate content during carbonation of the ground Portland cement pastes in a wet reactor with a 10% CO_2 – 90% N_2 gas flow at 10 L/hr/1g cement at $T = 20^\circ\text{C}$ and normal pressure. The line is a guide for eye. (B) Evolution of pH during the carbonation reaction.

Within this contribution, the reactivity of cRCP has been examined in synthesized mixtures containing portlandite and sodium hydroxide in solution to mimic the conditions in a cementitious matrix [32,51]. For these experiments, synthesized cement pastes were prepared from a neat Portland cement and a slag (GGBFS) containing composite cement (1/1 blend). The well hydrated pastes were carbonated and tested as in a recent study [32]. The pozzolanic reaction taking place in the synthesized cements can be summarized as follows:



According to this reaction scheme, the progress of the reaction is proportional to the portlandite consumption, and different samples can be compared by this parameter, assuming similar reaction products.

This is illustrated in Figure 5A and compared to the consumption of portlandite in a blend with fly ash taken from a recent study [32].

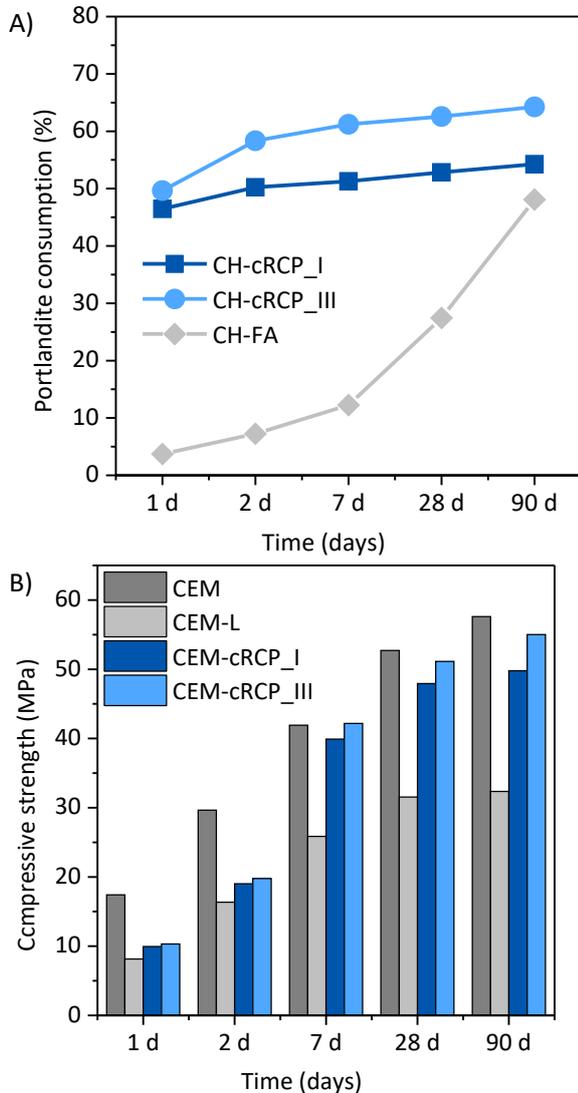


Figure 5. (A) Consumption of portlandite in the synthesized cements. (B) Evolution of the compressive strength in composite cements (see text for further explanation). cRCP_I – carbonated cement paste based on CEM-I Portland cement (acc. to EN 197-1), cRCP_III carbonated cement paste based on a slag containing composite cement (CEM-III acc. to EN 197-1).

The carbonated cement pastes react with portlandite very rapidly when compared to the reference fly ash sample. After one day, the consumption of portlandite in the cRCP containing synthetic cements is the same as in the fly ash analogue after 90 days. However, for the carbonated cement pastes, the consumption of portlandite levels off after 7 days of pozzolanic reaction. It is noticeable that there is a difference between the carbonated pastes based on neat Portland cement and the composite cement, where the latter consumes more portlandite. This finding can be explained by two phenomena. First, the composite cement has a higher content of alumina and silica and may thereby contain more alumina-silica gel after carbonation when compared to the

carbonated paste based on the neat Portland cement. Secondly, the unreacted slag (ca. 50 % of the slag in the original cement) present in the carbonated cement paste [32] can further react and consume a part of the portlandite.

Employing the carbonated cement pastes as an SCM, composite cements were prepared with 60% of cement clinker and 40% of the carbonated cement paste. The evolution of their compressive strengths as compared to reference samples of neat Portland cement and limestone composite cement is shown in Figure 5B. These data clearly reveal that the cement with carbonated cement pastes are characterized by higher compressive strength, as compared to the reference Portland - limestone cement, and slightly lower strength relative to the neat Portland cement. It is important to notice that the compressive strength of the sample with the carbonated composite cement has higher compressive strength in agreement with the Portlandite consumption shown in Figure 5A.

The products of the hydration reactions for the cements containing cRCP are typical for composite cements and include mainly the C-(A)-S-H phase as well as mono- and hemihydrate [29,32]. However, the evolution of the microstructure is not typical for a composite cement as shown in Figure 6. During the hydration of the cements with cRCP, the hydrates precipitate in the space occupied initially by the alumina-silica gel and result in a densification of this space. In other composite cements, hydrates precipitate exclusively in the space occupied initially by the mixing water and the space occupied by the dissolving anhydrous phases.

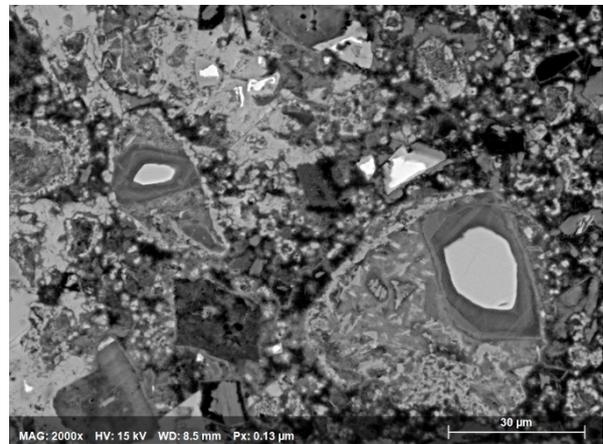


Figure 6. SEM-BSE image of the micro-structure of the composite cement containing cRCP. The grains occupied initially by the alumina-silica gel are filled with the hydrates, mainly C-(A)-S-H phase. Furthermore, the slag grains originating from the cRCP show hydration rims, indicating a post hydration of this phase in the composite cement

2.3 Simplified CO₂ and energy balance

The proposed approach involves several additional steps that are not involved in conventional cement production. These are the advanced recycling and separation technologies to produce the RCP, a recarbonation process and a drying process. To evaluate the overall impact of the proposed approach, a simplified CO₂ and energy balance calculation has

been conducted for the approach as well as for composite cement with 35 % limestone (CEM II/B-LL) and a limestone – calcined clay – cement (LC3) composition with 20 % limestone and 30 % calcined clay.

In the balance, only process emissions and approximate energy consumption are considered. For clinker production, process emissions of 500 kg CO₂ per ton clinker is assumed and energy of 1000 kWh/t corresponding to 3.4 GJ/t for clinker production and about 60 kWh/t for grinding and other processes. Limestone production as well as clay calcination are assumed free of process emissions. For cRCP production, a CO₂ uptake of 250 kg CO₂ per ton cRCP is considered, corresponding to 90 % of the theoretical uptake of cement composed of 50 % clinker and 50 % of a typical slag containing 30 % CaO. No emissions from fuels and energy are considered as all new processes involved can be electrified, and electricity is expected to be provided by sustainable sources such as wind mills and solar cells in the next decade [52]. Thus, the impacts from energy consumption are therefore expressed in kWh/t of material.

For the energy balance, it is assumed that clay drying plus calcination consumes 750 kWh/t, which corresponds to 2.7 GJ/t. For cRCP, 0.75 GJ/t is assumed for drying, 10 kWh/t for advanced recycling and 20 kWh/t for the re-carbonation, all these values being a conservative estimate [38,40]. Remaining processes are considered identical to the CEM II/B-LL production. All these assumptions are summarized in Table 1.

Table 1. Assumed process emissions at material level and approximate energy consumption for the cement constituents

	Process emissions (kg CO ₂ /t)	Additional energy consumption (kWh/t)
clinker	500	1000
limestone	-	10
calcined clay	-	750
cRCP	-250	238

Table 2 shows the considered cement compositions and the resulting process emissions and approximate energy consumptions of the cements compared.

Table 2. Assumed cement compositions and resulting process CO₂ emissions and approximate energy consumptions

	CEM II/B-LL	LC3	40 % cRCP
clinker	65 %	50 %	60 %
limestone	35 %	20 %	-
calcined clay	-	30 %	-
cRCP	-	-	40 %
process CO ₂ emissions (kg CO ₂ /t cement)	325	250	200
Approximate energy consumption (kWh/t cement)	654	727	695

As can be seen, the presented approach results in the lowest process CO₂ emissions for the three cement compositions and in energy demands that do not exceed other solutions such as

limestone – calcined clay – cements with 50 % clinker replacement.

3 Conclusions

The construction sector needs to reduce its CO₂ emissions and consumption of natural raw materials to decrease its environmental footprint. Traditional approaches such as use of conventional SCMs to reduce the clinker content in cements are nowadays mainly limited by the availability of suitable replacement materials. Hence, industrial and academic research laboratories are exploring several other solutions to further reduce the environmental footprint. In this work, a solution based on carbonation of the cement paste from old demolished concrete and its application as an SCM has been described as a novel carbon capture and utilization process. Recent studies indicate that the process of enforced carbonation of cement paste can be conducted under mild conditions and may easily be upscaled to an industrial scale. The wet carbonation is conducted at normal temperature and pressure, where full carbonation can be achieved after only two hours. The carbonated cement paste is composed of two carbonation products: calcium carbonate and an amorphous alumina-silica gel. The calcium carbonate precipitates mainly as calcite. The alumina-silica gel is amorphous in the view of powder XRD and exhibits a high surface area. Solid-state ²⁷Al and ²⁹Si NMR reveal that it is composed mainly of condensed Q³ and Q⁴ SiO₄ tetrahedra with integrated AlO₄ units in the structure. The carbonated cement paste can be successfully applied as a supplementary cementitious material to produce new composite cements. This reflects a high pozzolanic reactivity of carbonated cement paste and the formation of hydrate phase assemblages similar to those in composite cements containing fly ash and limestone. As a result of the high reactivity, the carbonated cement paste has a positive effect on the compressive strength evolution even at early times.

The recarbonation of cement paste contributes to the circular economy and preserves natural resources as all materials derived from demolished concrete can be recycled and used in new concrete production. The principal feasibility of the new CCU approach has been demonstrated in the available literature. However, there are still several open questions which require further research in order to upscale the solution presented in this work to the industrial scale. This includes optimization of the chemical and physical carbonation conditions. Furthermore, investigations are needed to unravel the impact of elements like alkalis, magnesium and sulfate that are present in the system in significant amounts. The composition range, structure and physical properties of the alumina-silica gel requires further research as these properties may explain the impact on the performance of the alumina-silica gel in composite cements. Finally, there is only little information available about the development of microstructure and performance of cements containing carbonated cement paste.

Authorship statement (CRediT)

Maciej Zajac: Conceptualization, Methodology, Investigation, Formal analysis, Writing – original draft, Supervision, Project administration.

Jan Skocek: Conceptualization, Investigation, Writing – original draft, Project administration.

Jørgen Skibsted: Investigation, Formal analysis, Writing – original draft.

Mohsen Ben Haha: Writing – original draft, Supervision.

Declaration of interest

We wish to confirm that there are no known conflicts of interest associated with this publication and there has been no significant financial support for this work that could have influenced its outcome.

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