

## Full length article

# Thermal and carbonation activation of recycled cement paste: A comparative data analysis and future research directions<sup>☆</sup>

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## ABSTRACT

Concrete is the predominant construction material worldwide, but cement production generates significant CO<sub>2</sub> emissions. Furthermore, large amounts of concrete waste are difficult to recycle and are therefore mostly landfilled. Using recycled concrete fines (RCF) as a supplementary cementitious material (SCM) through activation offers a promising strategy to reduce clinker consumption while valorizing this waste stream. This study provides comprehensive insights into the effectiveness of two activation methods, thermal activation and carbonation activation, through systematic quantitative evaluation based on published literature. Thermal activation dehydrates the material, forming a modified calcium silicate phase (C<sub>2</sub>S) with hydraulic reactivity. Carbonation activation produces CaCO<sub>3</sub> and an amorphous alumina-silica gel with pozzolanic activity. Both methods substantially improve the performance of recycled cement paste (RCP) relative to untreated RCP, although neither matches OPC performance. Thermal activation demonstrates greater potential than carbonation activation. However, limited data and substantial variations in starting material, replacement level, particle size, and activation conditions impede direct comparison and prevent their individual effects from being reliably isolated.

## 1. Introduction

Cement production is a major source, contributing around 8% of the global total CO<sub>2</sub> emissions. This substantial impact is primarily driven by the enormous volume of cement consumed worldwide, even though the specific emissions associated with concrete itself are comparatively lower. This significant environmental impact underscores the need for sustainable practices in the construction industry [1–5]. Furthermore, the amount of construction and demolition waste (C&DW) is steadily growing. Recycling C&DW converts a waste product into an important resource [6]. Recycled concrete waste (RCW) can be broadly categorized into coarse (4 mm to 40 mm) and fine (< 4 mm) aggregates [3,7–9]. Typically, separation of concrete waste yields approximately 60 wt.% to 80 wt.% coarse and 20 wt.% to 40 wt.% fine recycled aggregates, depending on processing objectives and parameters [10,11].

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**Table 1**  
Definition of RCP, RCF and RCW.

| Abbreviation | Name                    | Mixture                          | Production                    |
|--------------|-------------------------|----------------------------------|-------------------------------|
| RCP          | Recycled Cement Paste   | Binder + Water                   | Laboratory Mixtures           |
| RCF          | Recycled Concrete Fines | Binder + Water + Sand            | Laboratory Mixtures           |
| RCW          | Recycled Concrete Waste | Binder + Water + Fine Aggregates | Crushed/Milled Concrete Waste |

Furthermore, processes are currently being developed that increasingly focus on separating sands and aggregates from the fine particles in order to increase the proportion of cement paste in the fine fractions [12]. Activating these fine particles, including high volume of cement paste for use as a supplementary cementitious material (SCM), offers an effective way to reduce both the consumption of natural resources and the associated CO<sub>2</sub> emissions. Based on the current literature, this paper provides a comparative quantitative analysis of two promising activation methods for recycled concrete fines (RCFs). Since the largest data set is based on laboratory-prepared RCFs and the sand fraction is generally considered inert, this study focuses on the cementitious portion, referred to as recycled cement paste (RCP), which is also the most frequently examined material in the compiled data set. The activation methods discussed include thermal and carbonation activation, both of which are often combined with mechanical processing, an approach that is also addressed in this comparison. The examined key aspects include the influence of these activation methods on the hydration behaviour, microstructure, and hardened concrete properties.

Despite the fact that a considerable number of experimental studies exist on these activation methods, they are mostly conducted on individual materials under specific boundary conditions, making it difficult to determine which method is more effective and for which practical applications these approaches are suitable. Therefore, the novelty of this paper lies in the comparative approach that has been adopted. It synthesizes literature on two distinct activation methods for RCP, evaluates trends, and identifies research gaps. In addition, the study examines the extent to which findings obtained from research on model materials (RCPs/RCFs) can be applied to the practical use of the fine fraction of RCW, guiding both experimental and industrial applications.

## 2. Methodology

In this work, RCW refers to concrete debris obtained from demolished structures, while RCF and RCP are prepared as controlled laboratory mixtures. RCF contains both fine aggregate, namely sand, and residual cement paste, whereas RCP consists exclusively of hydrated cement paste without aggregates (Table 1).

The collected papers were screened based on three criteria:

1. Activation method employed (only carbonation and thermal activation are accepted).
2. Type of precursor material (only RCP is accepted).
3. Reference system used (only binary cement paste or mortar systems with OPC are accepted).

For the literature search in Scopus and Google Scholar, the following keywords were used: Activation Method (Concrete), Thermal Activation, Carbonation Activation, Comparison of Activation Methods (Concrete), Recycled Concrete, Concrete Waste, Concrete Fines, and Concrete Paste.

For inclusion, studies had to explicitly define activation parameters and report results in graphical or tabular form. The evaluation of the literature was performed qualitatively as well as quantitatively through a defined methodology presented in the following subsections.

A total of 200 studies were considered for this work, of which 57 met the three criteria for inclusion in the quantitative analysis. The qualitative comparison presented in this paper is based on 57 studies on unactivated material, 15 studies on thermal activation, 21 studies on carbonation activation, and two studies that directly compare both activation methods. Only studies employing RCP were included as data points in the dataset used for the subsequent analysis. The remaining studies were primarily used to support the quantitative analysis and to provide a broader understanding of transferability to RCW.

### 2.1. Quantitative comparison

The methodology for quantitative comparison was formalized based on approaches presented in some key literature examples [1, 3,9,13–23] to derive specific trends or insights. Performance is assessed using two key indices, which are described in Sections 2.1.1 and 2.1.2. Therefore, each study had to include at least one of the following reference measurements:

- A test without substituting OPC with activated RCP, or
- A test with substitution performed using unactivated RCP.

#### 2.1.1. Activity Index (AI)

The Activity Index (AI) compares a selected property  $P$  of a mixture containing activated RCP with a pure OPC reference:

$$AI = \frac{P_{\text{sample}}}{P_{\text{ref}}} \quad (1)$$

AI is most commonly used for compressive strength, but requires identical substitution rates to ensure a valid comparison.

**Table 2**  
Quantitative comparison using AI and II methods.

| Property                                                        | P <sub>sample</sub> | P <sub>ref.</sub> | P <sub>RCP</sub> | AI   | II              |
|-----------------------------------------------------------------|---------------------|-------------------|------------------|------|-----------------|
| Mass loss $\Delta m$                                            | $\Delta m_{t/cRCP}$ | –                 | $\Delta m_{RCP}$ | –    | $\Delta m - II$ |
| $^{29}Si$ MAS NMR (all $Q^n$ )                                  | $NMR_{t/cRCP}$      | –                 | $NMR_{RCP}$      | –    | $NMR - II$      |
| $N_2$ -BET                                                      | $BET_{t/cRCP}$      | –                 | $BET_{RCP}$      | –    | $BET - II$      |
| True Density $\rho$                                             | $\rho_{t/cRCP}$     | –                 | $\rho_{RCP}$     | –    | $\rho - II$     |
| Particle size $d_{50}$                                          | $d_{50,t/cRCP}$     | –                 | $d_{50,RCP}$     | –    | $d_{50} - II$   |
| Cumulative heat release (at approx. 20 °C after 3 d) $Q_{h,3d}$ | $Q_{h,3d,t/cRCP}$   | $Q_{h,3d,ref.}$   | $Q_{h,3d,RCP}$   | H-AI | H-II            |
| Heat flow – second peak $W_{h,3d}$                              | $W_{h,3d,t/cRCP}$   | $W_{h,3d,ref.}$   | $W_{h,3d,RCP}$   | P-AI | P-II            |
| Mortar strength (28 d) $f_{28d}$                                | $f_{28d,t/cRCP}$    | $f_{28d,ref.}$    | $f_{28d,RCP}$    | S-AI | S-II            |

**Table 3**  
Thermal activation of RCP and resulting phase transformations.

| Temp. range                    | Mineral phases                             | Reaction process                      | Effect on reactivity                                 | Source           |
|--------------------------------|--------------------------------------------|---------------------------------------|------------------------------------------------------|------------------|
| $\leq 110^\circ C$             | Free water                                 | Physical evaporation of pore water    | –                                                    | [3,25]           |
| $\leq 200^\circ C$             | Gypsum, Ettringite (AFt)                   | Dehydration of sulphate phases        | –                                                    | [30–32]          |
| $\leq 250^\circ C$             | Monosulfate (AFm) phases                   | Gradual dehydration                   | –                                                    | [33,34]          |
| $260^\circ C$ to $400^\circ C$ | Tricalcium aluminate hydrate ( $C_3AH_6$ ) | Dehydration and partial decomposition | Increased surface area; reactive product             | [35]             |
| $110^\circ C$ to $500^\circ C$ | Calcium silicate hydrates (C–S–H)          | Dehydration and structural breakdown  | Increased surface area; reactive product             | [3,25,33,36]     |
| $400^\circ C$ to $500^\circ C$ | Portlandite (CH)                           | Dehydration and transformation to CaO | Enhanced reactivity due to CaO formation             | [3,25,33,36–38]  |
| $600^\circ C$ to $900^\circ C$ | $CaCO_3$                                   | Decarbonation, $CO_2$ release         | Formation of reactive CaO; high activation potential | [22,25,28,36,39] |

### 2.1.2. Impact Index (II)

Impact Index (II) replaces the OPC reference in Eq. (1) with a reference using the analogous substitution of unactivated RCP, isolating the effect of activation:

$$II [-] = \frac{P_{\text{sample}}}{P_{\text{RCP}}} \quad (2)$$

II can also evaluate the effects of “pure” materials, such as particle size, specific surface area, or reactivity. All properties used are listed in Table 2.

## 2.2. Qualitative comparison

For the qualitative comparison, all collected data are visualized in figures, allowing an overview of trends across the entire dataset. When data are insufficient to apply AI and/or II for detailed comparison, the evaluation is based on absolute values from the general dataset rather than a direct comparison of materials from the same precursor.

## 3. Activation methods for RCPs

Activated RCP can be used as SCM to reduce clinker content. Two main approaches, namely thermal and carbonation activation, have been extensively studied so far in the literature [3]. Mechanical activation (grinding/milling) might improve reactivity. However, it is often combined with thermal or carbonation activation. Hence, in this study, it is considered primarily as a preconditioning for thermal and carbonation activation. Recently, the feasibility of mechano-chemical activation of RCF has been demonstrated [24]. However, due to a lack of extensive data, this activation technique has been further excluded from the study.

### 3.1. Thermal activation

Thermal activation results in the disintegration of various mineral phases in the RCP depending on the temperature range. The dehydration and decomposition of these phases lead to the formation of partially hydraulic products, thereby activating the RCP and enabling its use as a latent-hydraulic SCM [18,22,25–27]. The first paper to establish the basis for the reactivation of hardened cement paste was published by Schneider in 1982 [28]. The concept of thermal activation was later introduced and explained, firstly by the dissertation of Dora in 2001 [29]. Table 3 shows the different dehydration, decomposition, and activation processes occurring at various temperature ranges.

At around 110 °C, both physically and chemically bound water started to be released due to the decomposition of hydrated phases such as calcium silicate hydrate (C–S–H), ettringite, and AFm phases. At temperatures above approximately 400 °C, portlandite (CH) decomposes, followed by the thermal breakdown of calcium carbonate (CaCO<sub>3</sub>) at 600 °C. These processes promote the formation of reactive solid phases, which can significantly enhance the overall reactivity of the thermally activated material, provided that appropriate thermal activation conditions are maintained [3,22].

At temperatures between 390 °C and 490 °C, the chemical composition of dehydrated C–S–H gradually leads to formation of belite-like phase (C<sub>2</sub>S). Depending on the specific thermal regime, this transformation can lead to the formation of reactive polymorphs similar to  $\alpha'_H$ -C<sub>2</sub>S or  $\alpha'_L$ -C<sub>2</sub>S, both of which exhibit hydraulic reactivity [15,16,18,22,40–43]. However, when the temperature exceeds roughly 800 °C, these reactive forms tend to convert into the thermodynamically stable but less reactive  $\beta$ -C<sub>2</sub>S phase [16,18,44]. This transition is also associated with the release of CO<sub>2</sub>, resulting from the decarbonation of carbonate-containing phases [18,40,41,45]. Even at activation temperatures below 800 °C, a rapid cooling process can induce the transformation of  $\alpha'_L$ -C<sub>2</sub>S to  $\beta$ -C<sub>2</sub>S, since  $\alpha'_L$ -C<sub>2</sub>S is fully stable only down to about 600 °C [44]. As a result of the formation of reactive phases (mixture of  $\alpha$ -C<sub>2</sub>S and  $\beta$ -C<sub>2</sub>S), thermal activation of RCP (tRCP) leads to significant changes in its chemical composition and phase assemblage. These transformations contribute not only to the development of hydraulic reactivity but also to improving the mechanical strength properties compared to mixtures containing unactivated RCP.

### 3.2. Carbonation activation

Carbonation activation can be performed under dry, moist, or wet carbonation conditions [19,46]. The extent of carbonation can be quantified, for example, using thermogravimetric analysis (TGA) [47], by comparing carbonate contents determined before and after the reaction. This paper focuses primarily on wet carbonation, as it enables a high degree of carbonation (DoC) within a few hours [20,21,23,48–50]. This efficiency is attributed to the enhanced accessibility of dissolved CO<sub>2</sub> within the pore structure to the reactants. Notably, such high carbonation levels can be achieved under ambient temperature and pressure, whereas dry carbonation typically requires elevated temperatures and/or pressures to reach comparable results, and moist carbonation is difficult to perform on a laboratory scale. The reactions involved have been described in detail [1,19,51–55]. However, to broaden the dataset for analysing governing effects, the specific carbonation pathway is largely disregarded. Instead, the emphasis is placed on achieving a high DoC. Consequently, RCPs exhibiting high carbonation extents are considered.

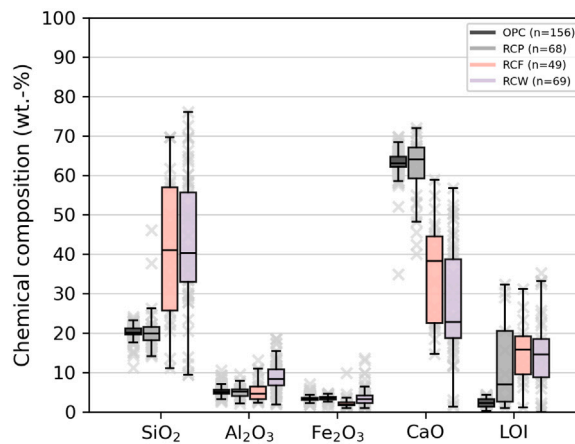
Wet carbonation of RCP involves the continuous injection of CO<sub>2</sub> as fine bubbles into a reactor containing an aqueous suspension of the material, thereby enhancing gas–liquid mass transfer and enabling efficient contact between dissolved CO<sub>2</sub> and the reactive phases. The dissolution of CO<sub>2</sub> in the aqueous phase initiates a sequence of dissolution–precipitation reactions that transform phases in RCP, such as portlandite (CH), C–(A)–S–H, AFm phases, ettringite (AFt), and residual clinker phases, into CaCO<sub>3</sub> and an amorphous Al–Si gel [23,56–60].

Aluminate is incorporated into the gel via carbonation of C–(A)–S–H and aluminate-bearing hydrates such as ettringite [56,61]. The formation of the phase assemblage present in carbonated activated RCP (cRCP) can be conceptually simplified by the following Eqs. (3)–(5):



Several studies have shown that under current wet carbonation conditions, where DoC exceeding 90% can be achieved, not all phases present in RCP carbonate or carbonate completely. Examples include hydration phases that carbonate only slowly under the given conditions, such as hydrogarnets and AFm phases. Some phases are essentially inert to carbonation, such as quartz, and unreacted clinker phases such as C<sub>3</sub>A, C<sub>4</sub>AF, or unreacted SCM [19,56,61–64]. Therefore, incomplete carbonation reduces the achievable CO<sub>2</sub> uptake relative to the theoretical maximum.

The Al–Si gel present in the cRCP is characterized by a high specific surface area, and is able to react with portlandite when used as an SCM, forming additional cementitious hydrates in a manner analogous to conventional pozzolanic SCMs [59]. Compared to traditional SCMs, the pozzolanic reaction of cRCP generally proceeds at a faster rate, as evidenced by the rapid early consumption of portlandite and the associated accelerated early-age strength development reported in several studies [19,49,62,65]. While not considerably reactive itself, the CaCO<sub>3</sub> fraction present in cRCP can still affect hydration in cRCP-OPC blends. Physically, CaCO<sub>3</sub> can act as a filler, exerting effects comparable to those observed in limestone-OPC systems, such as accelerating early hydration of cement clinker phases [66–68]. Chemically, it can react with C<sub>3</sub>A (or in blended cements, also other sources of aluminium, such as other SCMs or Al from Al–Si gel) to form hemi- or monocarbonate, thereby stabilizing ettringite and inhibiting its conversion to monosulfate, which causes a slight increase in overall compressive strength [69–71].



**Fig. 1.** Chemical composition of OPC [3,7,9,13,15–18,20–22,25,26,36,40,41,45,48,50–56,62,65,74–154], RCP [1,16–18,20–23,25,26,40,41,45,50–57,61,62,72,73,75,83,90,93,95–97,102,105,109,112,116,117,123,126,129,150,155–165], RCF [9,14,17,26,48,49,79,86,89,101,104,108,110,116,129,131,134,142,147,148,156,160,162,166,167] and RCW [7,11,13,15,20,36,72,81,82,88,94,106,107,109,113–115,118–121,124,125,127–130,133,135,137–141,143,144,149,151–154,161,168–177].

#### 4. Comparison of the thermal and carbonation activation method

##### 4.1. Phase composition

The chemical phase composition of anhydrous OPC differs from that of RCP, as shown in Fig. 1. Both thermal and carbonation activation modify the phase composition, depending on the temperature or the degree of carbonation, thereby enhancing the reactivity of the recycled material. Replacing OPC with RCP notably alters the spread of the SiO<sub>2</sub> and CaO content. Thus, a SiO<sub>2</sub> content ranging from 15 wt.% to 38 wt.% and a CaO content ranging from 40 wt.% to 72 wt.% is now possible, as illustrated in Fig. 1, which limits the potential for forming new C–S–H phases and portlandite [9,13].

During thermal activation of RCP, several chemical transformations occur (Section 3.1). A notable result is the formation of both  $\beta$ -C<sub>2</sub>S and  $\alpha$ -C<sub>2</sub>S, which typically emerge at activation temperatures between 600 °C to 900 °C [15,16,18,43,44,72]. X-ray diffraction analysis also confirms the presence of C<sub>2</sub>S, as well as other calcium silicates, such as Ca<sub>3</sub>SiO<sub>5</sub> and Ca<sub>2</sub>SiO<sub>4</sub>, in RCP activated at temperatures between 400 °C to 600 °C. At approximately 800 °C, CaCO<sub>3</sub> in the RCP decomposes into CaO and CO<sub>2</sub>.

For the carbonation activation of RCP, Zhao et al. [21] investigated microstructural changes, confirming the phase transformations reported by Zajac et al. [19]. TG analysis shows a first mass loss peak around 100 °C due to the dehydroxylation of C–(A)–S–H, AFt, silica gels, and AFm. Peaks at 450 °C and 600 °C to 900 °C correspond to the decomposition of portlandite and CaCO<sub>3</sub>, respectively [73]. Calcite, aragonite, vaterite, and amorphous CaCO<sub>3</sub> start to decompose at temperatures around 600 °C to 700 °C [21].

The mass loss impact index ( $\Delta m - II = \Delta m_{t/cRCP} / \Delta m_{RCP}$ , Section 2.1.2) provides a measure of phase changes in RCP during activation, reflecting variations in portlandite, C–S–H, and carbonation products such as CaCO<sub>3</sub>. Based on a data analysis of the literature, cRCP consistently exhibits very high CaCO<sub>3</sub> content, with reported  $\Delta m - II$  values ranging from 3.7 to 12.1, indicating extensive conversion of portlandite and C–S–H into stable CaCO<sub>3</sub> (see Fig. 2(a)). In contrast, thermal activation of RCP at temperatures between 550 °C to 650 °C decreases the portlandite content relative to unactivated RCP, with  $\Delta m - II$  values approximately between 0.3 and 0.5, while promoting the formation of calcium silicates and shifting the chemical composition towards that of OPC. These results reveal a clear distinction between the two activation methods. Carbonation leads to the formation of CaCO<sub>3</sub>, whereas thermal activation decomposes portlandite and C–S–H, thereby enhancing the similarity of RCP to conventional cement.

<sup>29</sup>Si MAS NMR spectroscopy classifies silicate species as  $Q^n$  units according to the number of bridging oxygens:  $Q^0$  represents isolated silicates in unreacted clinker,  $Q^1$  corresponds to chain ends,  $Q^2$  to middle-chain units in C–S–H,  $Q^3$  to branching sites, and  $Q^4$  to fully polymerized silicates. Al-induced sub-environments (e.g.,  $Q^3(1Al)$ ) are not resolved and are included within the corresponding  $Q^n$  signals. In tRCP, <sup>29</sup>Si MAS NMR analyses show a progressive reduction of the  $Q^1$  and  $Q^2$  signals with increasing activation temperature, reflecting the decomposition of silicate dimers and middle-chain C–S–H units. This trend indicates the breakdown of the primary silicate network and the loss of gel-like C–S–H phases [22].

In cRCP, the microstructure, particularly the distribution of silica gel and CaCO<sub>3</sub>, strongly influences chemical reactivity [1]. Reactions between portlandite and silica gel generate additional C–S–H, enhancing the mechanical properties of the material [20, 23]. <sup>29</sup>Si MAS NMR shows a pronounced increase in the  $Q^3$  and  $Q^4$  populations, indicating the formation of a more polymerized silica network as C–S–H decalcifies and converts into silica gel.

The <sup>29</sup>Si MAS NMR Impact Index (Section 2.1.2,  $NMR-II = NMR_{t/cRCP} / NMR_{RCP}$ ) provides a relative measure of silicate network polymerization ( $Q^0$ – $Q^4$  distribution) in activated RCP. For tRCP at an activation temperature of 750 °C,  $Q^1$  and  $Q^2$  signals,

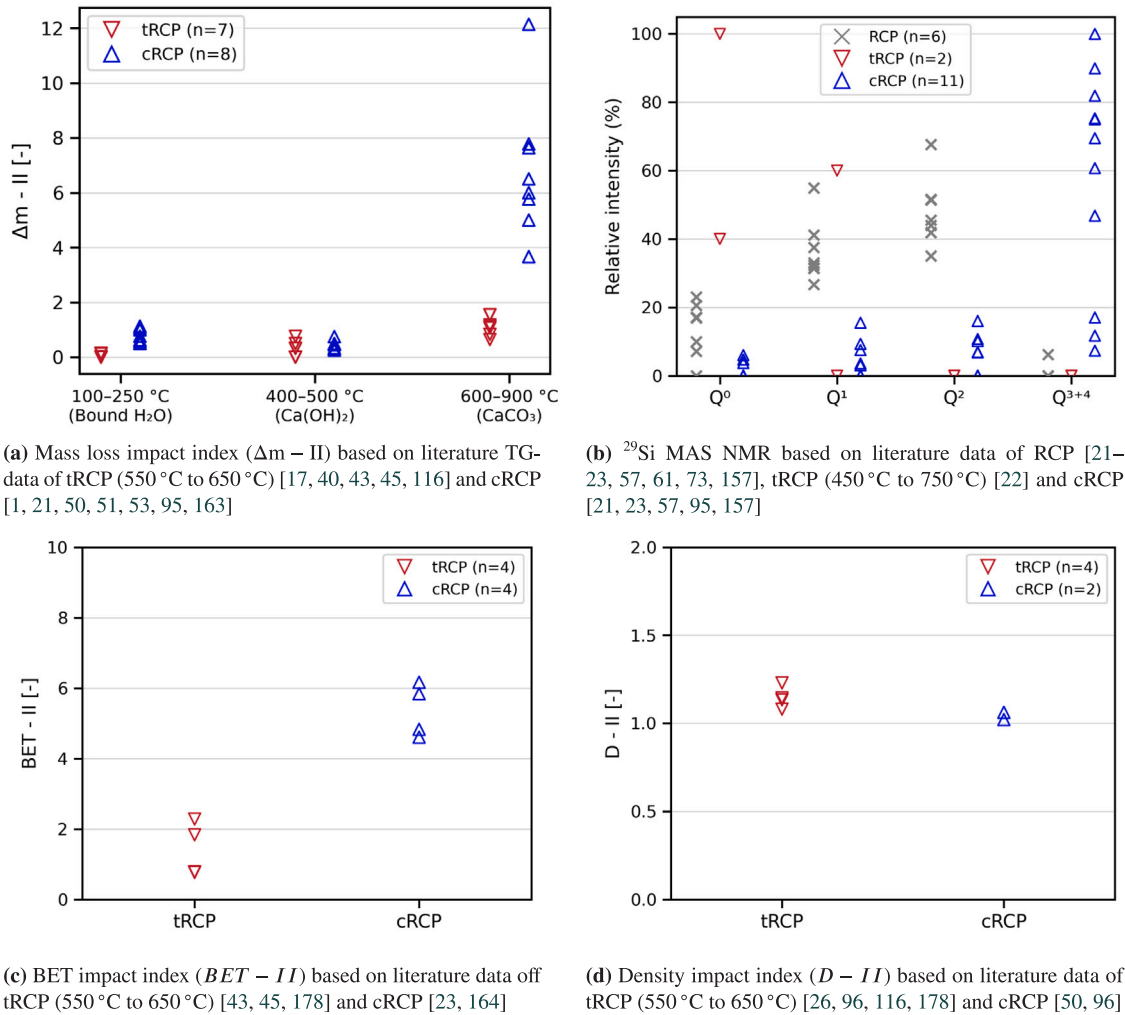


Fig. 2. Investigation of the phase composition based on literature data.

representing chain ends and middle-chain C–S–H units, are reduced, while  $Q^0$ , corresponding to a modified form of  $C_2S$ , is increased. Calculated from the available literature,  $Q^1$  and  $Q^2$  decrease to approximately 0.0, and  $Q^0$  increases to an NMR-II of 10 [22]. In cRCP,  $Q^3$  and  $Q^4$  signals, corresponding to branching and fully polymerized silicates, are substantially higher, with NMR-II for  $Q^{3+4}$  reaching 16.1 [157]. It should be noted that the deconvolution and quantification of  $^{29}Si$  MAS NMR spectra can vary significantly between studies due to differences in fitting procedures, including peak positions, Full Width at Half Maximum (FWHM), and the assignment of  $Q^n$  species. Consequently,  $Q^n$  distributions are method-dependent and should be compared with caution, particularly across materials with different compositions and measurement conditions. Although the data are insufficient to establish continuous trends or to construct a complete NMR-II diagram, a summary figure of the calculated values highlights the relative silicate abundances in tRCP and cRCP. It clearly indicates that thermal activation primarily leads to the depolymerization of C–S–H chains, whereas carbonation promotes the formation of highly polymerized silica networks (Fig. 2(b)).

#### 4.2. Microstructural characterization

Thermal activation significantly modifies the pore structure of RCP. In its untreated state, RCP exhibits a larger number of pores, ranging from 50 nm to 1  $\mu m$ , compared to OPC. After thermal activation, the pore size distribution narrows, with most pores in the 50 nm to 300 nm range [18,25]. The microstructure also evolves with temperature. Unactivated RCP consists of irregularly shaped particles, whereas treatment at 400 °C produces a denser material (Fig. 2(d)), despite the absence of C–S–H gel phases and the slightly higher density of the product phase, a modified form of  $C_2S$  [178–181]. At the same time, the BET surface area increases (Fig. 2(c)), indicating a greater accessible internal surface [9,13,43]. This apparent contradiction could be explained by the intrinsic microporosity of the modified  $C_2S$  phase. The presence of numerous small internal pores increases the accessible surface

area measured by BET, while having only a limited influence on the measured density, which is primarily affected by larger pores and the overall packing of the particles. But this explanation requires further investigation.

Carbonation activation alters the pore structure of RCP differently, primarily through the formation of silica gel and  $\text{CaCO}_3$ , which influence chemical reactivity [20,51]. In cRCP, silica gel generates extensive gel pores while reducing macropores relative to unactivated RCP. Consequently, the BET surface area increases, while the overall material density remains similar to that of untreated RCP. Nevertheless, the porosity of cRCP remains higher than that of OPC [20,89,97].

Based on reported literature data, BET and density impact indices were calculated to compare activated and untreated RCP. For tRCP, the calculated BET Impact Index (BET-II) decreases from 2.3 at 550 °C to 0.8 at 600 °C [45], and the corresponding Density Impact Index ( $D - II = \rho_{t/cRCP} / \rho_{RCP}$ ; see Section 2.1.2) ranges from 1.1 to 1.2. For cRCP, calculated indices indicate that both material density ( $D - II$  approx. 1.0) and cumulative porosity (Pore Volume II approx. 0.9, data based on [1,20,23,43]) remain similar to untreated RCP (Fig. 2(d)). In contrast to that, the BET-II increased significantly with a BET-II of approx. 5.0 (Figs. 2(c)).

#### 4.3. Particle size

The particle size of RCP is more variable and relatively large [170]. While the average particle size  $d_{50}$  of an OPC is approximately  $d_{50} = 17.3 \mu\text{m}$  with a standard deviation of  $7.2 \mu\text{m}$ , the mean value of RCP is approximately  $d_{50} = 18.5 \mu\text{m}$  with a standard deviation of  $11.1 \mu\text{m}$ , both based on the following literature data [1,9,13,17,21,23,25,40,41,45,48,50,79,81,88,92,95–97,99,100,109,110,112,114,115,118,122–124,127–129,140,142,150,161,162,171]. Given that particle size significantly impacts reactivity, both activation methods are preceded by a mechanical activation step to reduce particle size through crushing. In the following, the effect on the particle size of thermal and carbonation activation is examined.

Thermal activation significantly influences the particle size distribution of RCP. Heat treatment, particularly at elevated temperatures, alters particle size, though the effect is initially minor. Zhang et al. [13] report that RCP shows at room temperature (20 °C,  $d_{50} = 9.3 \mu\text{m}$ ) only slight increases after treatment at 400 °C and 600 °C ( $d_{50} = 10.5 \mu\text{m}$  and  $10.2 \mu\text{m}$ , respectively), indicating minimal agglomeration. In contrast, activation at 800 °C leads to a pronounced increase in average particle size ( $d_{50} = 22.6 \mu\text{m}$ ) [13,14,40]. Similarly, Sui et al. [15] and Baggio et al. [36] observe nonlinear behaviour, with  $d_{90}$  ranging from  $3.1 \mu\text{m}$  at 700 °C to  $17.9 \mu\text{m}$  at 600 °C. Considering  $d_{50}$ , both studies report an initial increase with temperature followed by a decrease at 900 °C, consistent with coarsening and subsequent fragmentation of particles.

During the carbonation activation, portlandite (CH) is converted to  $\text{CaCO}_3$ , causing a volume expansion of approximately 11 % to 14 % that refines pores and densifies the matrix. Carbonation of C–S–H gel can result in shrinkage due to its lower density compared to its carbonation products. The particle size of  $\text{CaCO}_3$  in cRCP typically ranges from  $0.1 \mu\text{m}$  to  $2.0 \mu\text{m}$ , which corresponds to capillary pore sizes in RCP, indicating that these fine particles may detach from the matrix, migrate within the cementitious system, and form agglomerates [20,21,182]. However, the final particle size largely depends on the particle size distribution of the starting material.

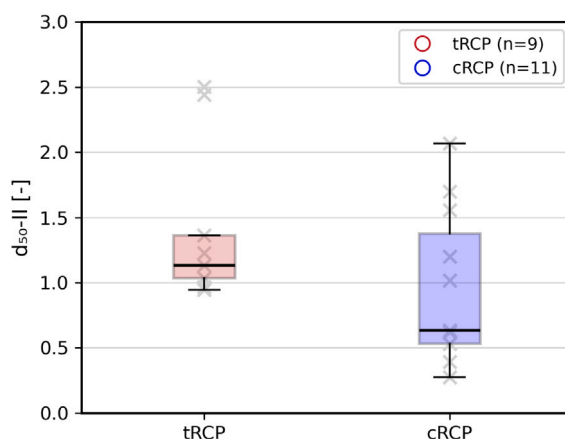
To quantitatively compare the impact of activation on particle size distribution, the Impact Index (II) method for the median particle size  $d_{50}$  ( $d_{50-II} = d_{50,t/cRCP} / d_{50,RCP}$ ) is used. Calculated  $d_{50-II}$  values from literature data show that thermal treatment of RCP at 550 °C to 650 °C increases the median particle size by approximately 1.5, with reported ranges from 1.0 to 2.5 depending on the source (Fig. 3). These changes are primarily attributed to structural transformations of calcium silicate hydrate (C–S–H) and concurrent modifications in surface area and porosity (Section 4.1). Carbonation, in contrast, can either refine the RCP through the formation of fine  $\text{CaCO}_3$  and silica gel crystals, reducing the median particle size to a  $d_{50-II}$  of 0.3 to 0.6 [23,95,164], or increase it up to 1.8 [1,21,54] due to crystal growth and partial agglomeration [3,52,82,89]. These differing results indicate a research gap that should be investigated further. Possible influencing factors could include the carbonation procedure and the initial particle size. Overall, these observations indicate that thermal activation leads to relatively consistent changes in particle size distribution, whereas carbonation activation effects remain less well established, with inconsistent trends reported across studies.

#### 4.4. Reactivity

##### 4.4.1. Heat flow calorimetry

The incorporation of RCP modifies the early hydration behaviour of cement by shortening the induction period and reducing both the heat release rate and the cumulative heat within the first 72 h compared to ordinary Portland cement [3,54,170]. Increasing RCP content shifts the secondary hydration peak to earlier times, indicating accelerated hydration kinetics, particularly for fine particles that act as nucleation sites [170]. Although RCP can intensify early hydration despite lower cumulative heat release [100], its overall effect depends strongly on dosage, particle fineness, and pretreatment [3,170]. In addition, residual phases may remain reactive after activation and contribute to the overall hydration behaviour when RCP is used as a supplementary cementitious material, similar to partially unreacted slag [19,184–187].

Thermally activated RCP enhances heat release during the first 4 to 6 h of hydration compared to ordinary Portland cement, while the subsequent heat evolution decreases more rapidly, indicating a reduced long-term hydration rate that depends on activation temperature [14,27,90]. Although the total cumulative heat remains lower than that of pure cement, it exceeds that of untreated RCP, demonstrating the beneficial effect of activation [3,101]. The highest initial heat flow peak and cumulative heat release are observed for RCP activated at 600 °C, whereas at 800 °C, the heat flow peak occurs earlier and is reduced, reflecting the formation of less reactive crystalline phases [14,16,18,40,43]. Systems composed entirely of tRCP retain distinct hydration stages but exhibit an extended acceleration period and lower peak heat flow. These behaviours are primarily attributed to modifications of the phase



**Fig. 3.** Particle size  $d_{50}$  impact index ( $d_{50}$ -II) based on literature data of tRCP (550 °C to 650 °C) [40,41,45,96,183] and cRCP [1,21,23,50,95–97,164,165].

composition induced by thermal activation (see Section 4.1), including full depolymerization of the C–S–H phase and maximal  $C_2S$  content at 600 °C, and the formation of polymorphs such as  $\beta$ - $C_2S$  and  $\alpha$ - $C_2S$  at higher temperatures. In addition, when thermal activation does not exceed high temperatures (over approx. 1000 °C), the number of nucleation sites remains comparable to that of unactivated RCP, promoting early formation of hydration products and accelerating initial hydration [3,101].

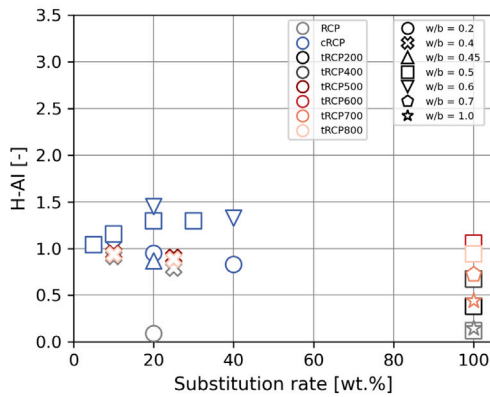
Carbonation activation leads to a different hydration behaviour, characterized by increased early heat release within the first 12 h and reduced cumulative heat over longer periods [97]. The heat evolution resembles that of limestone blended cements and is strongly influenced by the amount of carbonated material [49,51,62]. The initial heat peak increases due to rapid interactions between  $CaCO_3$  and clinker phases such as  $C_3A$  and  $C_3S$ , forming monocarbonate and calcium silicate hydrates, while  $CaCO_3$  particles also act as nucleation sites [19,51,89,164]. This so-called filler effect accelerates cement hydration and can generally be divided into dilution, nucleation, and accelerated dissolution effects [187,188]. During the carbonation activation, however, the nucleation effect appears to be the predominant mechanism governing the accelerated hydration kinetics. The early heat signal associated with  $C_3A$  hydration shows an earlier onset, suggesting this catalytic effect of the alumina silica gel formed during carbonation. Despite this acceleration, the cumulative heat release remains lower after 48 h [51]. The hydration behaviour is further governed by the surface chemistry of the products due to wet carbonation activation, as silica-rich and highly polymerized phases formed during carbonation exhibit rapid early reactivity but contribute to a lower overall heat release [23,49,102].

In the following, a quantitative comparison based on literature data is conducted using calorimetric indicators. This quantitative comparison uses the AI as well as the II for the cumulative heat release after 3 d and the second peak of the heat flow (Table 2).

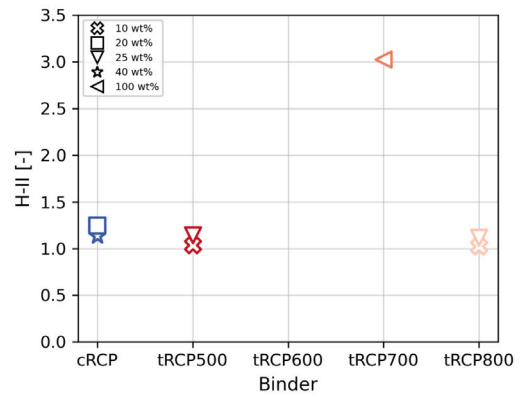
The hydration behaviour of thermally activated RCP is strongly influenced by the activation temperature and the water-to-binder (w/b) ratio, as reflected in the calorimetric results shown in Figs. 4(a) and 4(b). The cumulative heat release of systems incorporating thermally activated RCP remains lower than that of OPC and higher than that of untreated RCP. This trend is expressed by the H-AI, which is approximately 0.6 at a full replacement level of 100 wt.% and increases to about 0.9 at a substitution level of 10 wt.%, depending on the w/b ratio. In comparison with untreated RCP, the H-II reaches values around 1.2 and increases up to 3.0 at higher w/b ratios and an activation temperature of 700 °C, corresponding to a threefold increase in cumulative heat release after 3 d [16].

The second heat flow peak is generally reduced by using thermally activated RCP in comparison with ordinary Portland cement, with P-AI values ranging from 0.7 to 0.2 depending on w/b ratio and particle size for full replacement levels and activation temperatures between 500 °C to 800 °C (Fig. 4(c)). In contrast, when compared to untreated RCP, the P-II is higher, with values ranging from approximately 0.2 at an activation temperature of 800 °C [25] to 2.7 at 600 °C [40], and around 1.7 at 700 °C [16] (Fig. 4(d)). These results indicate that thermal activation substantially enhances hydration compared to untreated RCP, although the reactivity remains below that of ordinary Portland cement. Consequently, complete replacement of cement with untreated RCP results in very low cumulative heat release, while thermally activated RCP in the temperature range between 550 °C to 650 °C shows markedly higher reactivity, as illustrated in Fig. 4(a).

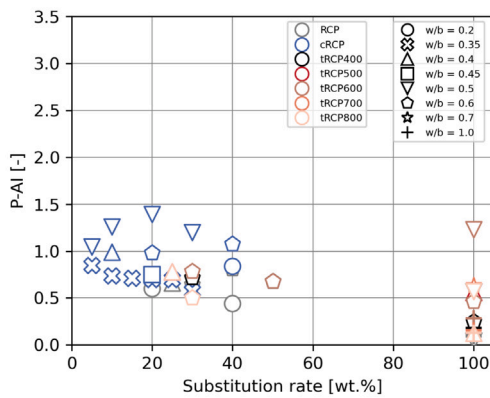
Carbonation activation, similar to thermal activation, can significantly enhance the reactivity (H-II  $\approx$  1.2 [50]) of RCP materials, leading to a marked increase in the early evolution of the heat of hydration, which is shown in Fig. 4(a). If, instead, the cumulative heat release after 3 d is compared with an OPC (H-AI), very different results are obtained at a substitution rate of, for example, 20 wt.%, ranging from 1.5 [50] down to 0.8 [97], with the H-AI visibly increasing as the w/b ratio rises (shown in Fig. 4(a)). A similar picture emerges when comparing the second heat peak from the literature when compared to OPC (P-AI) (Fig. 4(c)) and, in comparison with untreated RCP (P-II) (Fig. 4(d)). However, since cRCP exhibits pozzolanic reactivity, the presence of  $Ca(OH)_2$  is required to sustain the reaction. As shown in Fig. 4(a) (cumulative heat release) and Fig. 4(c) (second peak of the heat flow), the literature does not report data for a 100 wt.% substitution of OPC with cRCP, as in the absence of any additional binder component, there is no available  $Ca(OH)_2$  to drive the reaction.



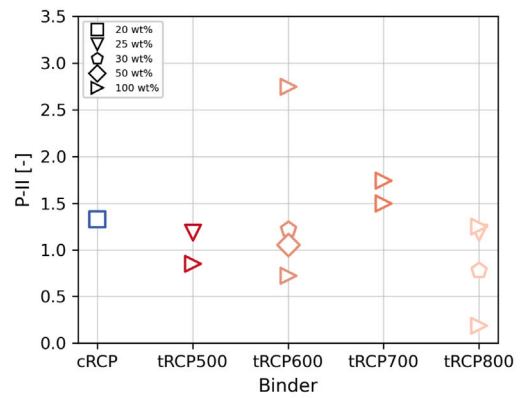
(a) H-AI of Cumulative heat release after 3 d based on literature data of RCP [16, 25, 162, 165], cRCP [21, 50, 95, 97, 136] and tRCP [25, 43] with different w/b (water/binder) ratios over the substitution rate



(b) H-II of Cumulative heat release after 3 d based on literature data of cRCP [50, 97] and tRCP [16, 25] with different w/b ratios



(c) P-AI of second heat peak based on literature data of RCP [16, 25, 40, 50, 97, 156, 162], cRCP [21, 50, 51, 95, 97, 136, 165] and tRCP [16, 18, 25, 40, 43, 90, 156] with different w/b ratios over the substitution rate

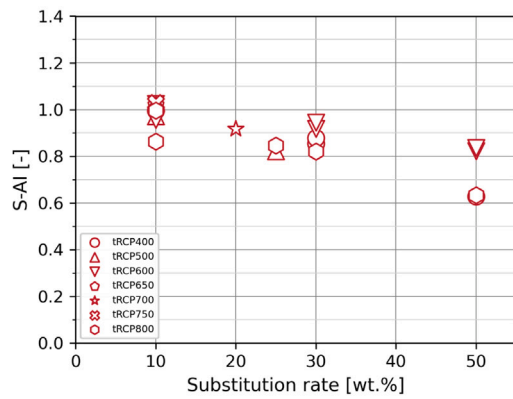


(d) P-II of second heat peak based on literature data of cRCP [97] and tRCP [16, 25, 40, 156] with different w/b ratios

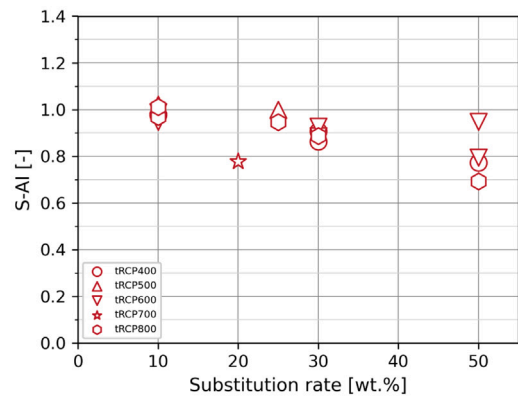
**Fig. 4.** H/P-AI and H/P-II of the heat of hydration as a function of clinker substitution, based on literature data for RCP, cRCP, and tRCP across varying w/b ratios.

#### 4.4.2. $R^3$ test

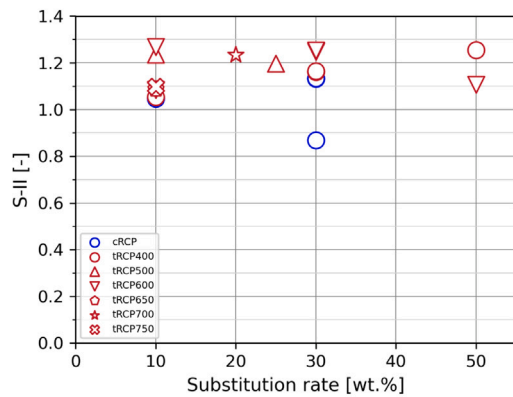
Another test method for assessing the reactivity, especially of pozzolanic or latent-hydraulic SCMs, is the  $R^3$  test [189–192]. This test can also be applied to evaluate the reactivity of tRCP and cRCP [72,96,165,184,193,194]. Two ways can be used to determine the reactivity of an SCM in the  $R^3$  test: on the one hand, the heat release and on the other hand the bound water content [191]. Several promising studies have explored the potential for chemically bound water as an indicator of hydraulic reactivity of activated RCPs, which correlated well with the compressive strength of the corresponding mortar samples. Ilki et al. [193] reported that RCP activated at 750 °C binds significantly more water than material activated at 800 °C, indicating higher hydraulic activity. Comparative studies [96,165,194] support this trend. The precursor materials differ, with Hashmi et al. [194] using RCF and Ye et al. and Shah [96,165] using RCP. Nevertheless, the water uptake of the unactivated materials is similar, around 3 g/100 g of dried starting material. Thermal activation at 750 °C to 800 °C increases water uptake to approximately 5 g/100 g, approaching OPC values of 6 g/100 g and indicating comparable hydraulic reactivity. The effect of carbonation activation has not been consistent among different studies. Hashmi et al. [194] investigate a value of about 7 g/100 g, whereas Ye et al. and Shah observe approx. 4.5 g/100 g. These differences are likely caused by both the type of precursor and the carbonation method, with Ye et al. [96] employing wet carbonation and Hashmi et al. [194] a moist process. In the case of wet activation, the material must subsequently be dried before further processing. As a result, the initial water content can vary significantly between the samples. An alternative determination parameter is the heat evolution in a modified  $R^3$  test. Although cRCP exhibits rapid early reaction kinetics, cumulative heat release values after 3 d of only approximately 140 J g<sup>-1</sup> SCM have been reported, which are lower than those of conventional SCMs such as fly ash or slag [184]. This relatively low overall reactivity is attributed to the high CaCO<sub>3</sub> content of cRCP, which can reach



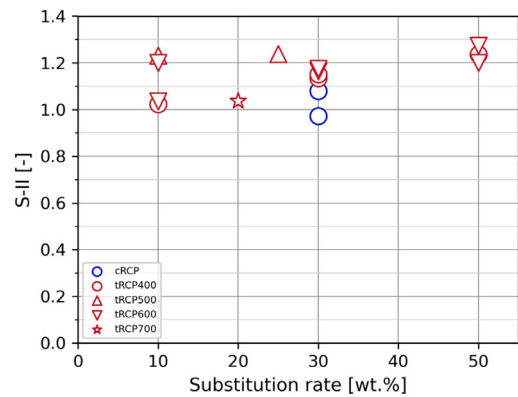
(a) S-AI of the compressive strength of mortar at 28 d depending on the clinker substitution rate of tRCP, depending on the temperature [18, 25, 96, 117, 178] at a w/b ratio of 0.50



(b) S-AI of the flexural strength of mortar at 28 d depending on the clinker substitution rate of tRCP, depending on the temperature [18, 25, 117, 178] at a w/b ratio of 0.50



(c) S-II of the compressive strength of mortar at 28 d depending on the clinker substitution rate of tRCP [18, 25, 96, 117, 178] and cRCP [52, 54, 96] at a w/b ratio of 0.50



(d) S-II of the flexural strength of mortar at 28 d depending on the clinker substitution rate of tRCP [18, 25, 117, 178] and cRCP [52] at a w/b ratio of 0.50

**Fig. 5.** S-AI and S-II of mortar at 28 d as a function of clinker substitution by tRCP at varying activation temperatures, compared with cRCP, all evaluated at a w/b ratio of 0.50.

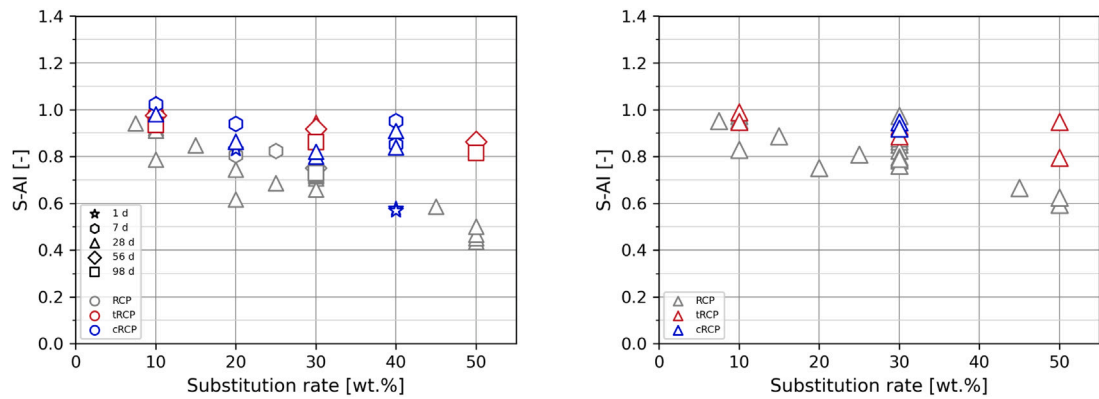
up to 65 wt.% [184]. According to Biscotti et al. [72], thermally activated RCP at 600 °C exhibited a cumulative heat release of approximately  $157.4 \text{ J g}^{-1}$  after 3 d, whereas OPC reached  $324.6 \text{ J g}^{-1}$  over the same period.

It should be emphasized that the available literature provides only limited data on the reactivity of both thermally and carbonated RCP. This lack of comprehensive datasets restricts the possibility of performing a detailed quantitative comparison and prevents the consistent application of the Activity Index (AI) and Impact Index (II) methods introduced in Sections 2.1.1 and 2.1.2.

#### 4.5. Hardened mortar properties

The replacement of clinker with RCP in mortar has a significant impact on mechanical performance. As the proportion of RCP increases, mechanical strength generally decreases, particularly at higher levels of substitution [9]. Mortars containing RCP typically exhibit reduced 28 d compressive strengths compared with standard mortars, primarily due to the limited availability of reactive phases acting mainly as filler material, leading to detrimental effects on the pore structure at higher substitution rates (Section 4.1) [25,48,195].

Thermal activation improves the mechanical performance of mortars containing RCP and mitigates strength loss [9]. Increasing the activation temperatures up to 600 °C improves the mechanical properties [3,13]. The hydraulic reactivity is enhanced (Section 4.4), and the porosity is reduced (Section 4.2), resulting in an improvement in mechanical properties compared with untreated RCP [3,13]. In addition to chemical effects, tRCP activated at 700 °C increases packing density and accelerates hydration through its finer particle size (Section 4.3), which provides additional nucleation sites on cement surfaces [15,92]. This combination of physical and chemical mechanisms enhances mortar performance. Strength declines when activation exceeds 750 °C, as shown in Figs. 5(a) and 5(b). At 1200 °C, tRCP softens, and hydration products disappear (Section 4.4), indicating a loss of hydraulic reactivity [3].



(a) S-AI of the compressive strength of mortar at different ages depending on the clinker substitution rate of RCP [18, 25, 52, 96, 109, 110, 117, 160], tRCP (600 °C to 650 °C) [96, 117, 178] and cRCP [19, 52, 65, 96] at a w/b ratio of 0.50

(b) S-AI of the flexural strength of mortar at 28 d depending on the clinker substitution rate of RCP [25, 26, 52, 109, 110, 117], tRCP (600 °C to 650 °C) [26, 117, 178] and cRCP [52] at a w/b ratio of 0.50

**Fig. 6.** S-AI of mortar strength at various ages as a function of clinker substitution by RCP, tRCP (600 °C to 650 °C), and cRCP, evaluated at a w/b ratio of 0.50.

Wu et al. [9] reported that the strength activity index reaches its maximum around 600 °C, with a value of 0.88 after 28 d at 30 wt.% replacement, while Sui et al. [15] observed 0.86 at 700 °C under comparable conditions, indicating only a minor difference. Furthermore, Höffgen et al. [17,27] report a k-value of 1.0, which corresponds to an S-AI of 1.0 and is therefore comparable to OPC at an activation temperature of 600 °C with a replacement rate of 30 wt.%. From 600 °C onward,  $\text{CaCO}_3$  begins to decompose, releasing  $\text{CO}_2$ , which suggests that 600 °C represents a balance between mechanical performance and environmental impact [14,17,27].

Additionally, Bogas et al. [90] demonstrated that the w/b ratio strongly influences strength development in cement paste and mortars containing RCP. When 100 wt.% of CEM I is replaced with tRCP activated at 700 °C and the w/b ratio is increased to maintain equivalent workability, compressive strength decreases by approximately 75 %. Maintaining a constant w/b ratio limits the reduction to 32 %, reflecting the combined effects of higher particle porosity, lower inherent strength of tRCP, and reduced formation of dense hydration products at particle interfaces.

Carbonation activation can significantly enhance the mechanical performance of cementitious systems compared with unactivated alternatives. The pozzolanic reaction of Al–Si gel in cRCP improves the strength of blended systems [1]. At low substitution rates (10 wt.%), the filler effect of cRCP further contributes to a denser cement matrix [182] (Section 4.2), enhancing the mechanical development of the composite [3]. The early hydration (Section 4.4) leads to rapid increases in compressive strength, particularly compared with limestone-containing cements [19,62]. After 7d, once the Al–Si gel is fully consumed, strength gain plateaus, emphasizing the importance of gel content and reactivity. A high DoC is essential for reactive gel formation, which enables pozzolanic behaviour. The concentration of the carbonation solution also affects gel properties and, in turn, the mechanical performance of the cementitious system [19,48,62]. Optimizing carbonation conditions can therefore significantly enhance performance. For example, Zhu et al. [54] report a strength activity index of 0.76, while Kaliyavaradhan et al. [82] observed an index exceeding 0.75 at 28 d in cement mortar containing 20 wt.% cRCP.

In the following quantitative comparison, the activation index (AI, Section 2.1.1) for mechanical properties is quantified using the strength activity index (S-AI). For unactivated RCP, literature data indicate that S-AI generally decreases with increasing substitution rate. Nevertheless, both strength values and the resulting S-AI display considerable variability, as illustrated in Figs. 6(a) and 6(b).

Thermal activation of RCP mitigates strength loss, resulting in improved mechanical performance compared with unactivated RCP. Consequently, the S-II is approximately 1.3 for several substitution rates, as illustrated in Figs. 5(c) and 5(d). Relative to untreated RCP, thermally activated RCP exhibits reactivity, which enhances hydraulic activity and promotes more refined microstructural development [9,16,156]. At lower replacement ratios (10 wt.%), the addition of thermally activated RCP can even produce a slight increase in compressive strength, with S-AI values exceeding 1.0, as shown in Figs. 5(a) and 5(b), compared with the reference mortar containing only OPC. This enhancement can be attributed to the nucleation and filler effects of finer particles generated by thermal activation [9,18]. With increasing substitution rate, a slight decrease in strength can be observed. However, at a replacement rate of 50 wt.% and an activation temperature of 600 °C, an S-AI greater than 0.8 can be achieved (see Fig. 6(a)). Nevertheless, there are too few comparable experimental data available to make a definitive statement. Furthermore, the benefits of thermal activation are not uniform over time due to the rapid reactivity of the material (Section 4.4) [13]. Thermal activation substantially increases the hydraulic reactivity of RCP, particularly within the temperature range of 600 °C to 800 °C. Figs. 6(a) and 6(b) demonstrate that activation within 600 °C to 650 °C allows reaching an S-AI exceeding the 0.8 threshold, indicating good hydraulic reactivity [13].

Carbonation activation of RCP enhances the compressive strength of cement pastes compared with those containing unactivated RCP, with S-II around 1.1, as shown in Figs. 5(c) and 5(d). This improvement is primarily due to the pozzolanic reactivity of the

Al–Si gel formed during carbonation (see Sections 3.2 and 4.1) [21,49,51,182]. Flexural strength generally evolves in parallel with compressive strength over the curing period [97]. Despite these gains, the compressive and flexural strengths of cRCP cements remain lower than those of OPC [97]. Cement pastes containing either unactivated RCP or cRCP consistently exhibit reduced strength relative to pure OPC at all curing ages, as illustrated in Figs. 6(a) and 6(b). Depending on the literature data, S-AI values range from 1.0 [96] to 0.6 [65]. A trend with increasing substitution rate at the same age is not discernible due to the limited data available (see Fig. 6(a)).

## 5. RCP-to-RCW transferability: Challenges and limitations

Numerous studies have demonstrated the feasibility of using both activated and unactivated RCP as SCMs at laboratory scale (see Sections 3 and 4). However, significant challenges remain for their transfer to real-world RCW and large-scale applications.

A key limitation arises from the heterogeneous nature of RCW. Complete separation of concrete constituents, including cement paste, fine aggregates, and coarse aggregates, is difficult to achieve during recycling. As a result, RCW is not composed solely of RCP but also contains residual aggregates that significantly influence its chemical and mineralogical composition. In addition to hydration products such as calcium silicate hydrate (C-S-H), calcium hydroxide (CH), AFm, and Aft phases, and unhydrated cement, RCW contains phases originating from aggregates, including quartz ( $\text{SiO}_2$ ), calcite ( $\text{CaCO}_3$ ), gismondine ( $\text{Ca}(\text{Al}_2\text{Si}_2\text{O}_8) \cdot 4\text{H}_2\text{O}$ ), and dolomite ( $\text{CaMg}(\text{CO}_3)_2$ ). Further  $\text{CaCO}_3$  is formed through natural carbonation processes of CH [3,13,156,196].

As illustrated in Fig. 1, RCW typically exhibits higher  $\text{SiO}_2$  and lower CaO contents than laboratory-produced RCP, particularly when derived from concretes containing siliceous aggregates [170]. Recycled concrete fines (RCF) produced under laboratory conditions are generally more comparable to RCW due to the presence of residual aggregates. The relative proportions of CaO and  $\text{SiO}_2$  are primarily governed by the origin of the material and the aggregate type [3]. Since chemical and mineralogical composition strongly control material performance [7], these variations must be carefully considered when assessing suitability for construction applications [196].

To enhance performance, additional processing steps are required. Mechanical separation techniques can be applied to concentrate the cementitious fraction, thereby shifting the composition of RCW closer to that of RCP and ultimately OPC or the initial SCM. In combination, activation methods such as carbonation or thermal treatment can be employed for the finer fraction, especially for the pure cement paste. The available data indicate a preliminary potential for improved performance. However, the presence of largely inert aggregate phases as well as natural carbonated phases reduces the overall reactivity of both tRCP and cRCP when applied as SCMs. This effect has been demonstrated in both laboratory-produced and industrial materials, including comparisons with synthetic cement pastes subjected to identical treatments [49,86,197,198]. Furthermore, there is already a study by Höffgen et al. [17,27], which allows predictions as to whether RCW material is suitable for thermal activation, based on mass loss during thermal treatment. However, regarding the potential for carbonation activation, there are currently no studies available, and so further investigations are needed.

Beyond material-related aspects, the integration of activation technologies into existing recycling systems poses additional challenges. These include plant integration, infrastructure requirements, process scalability, throughput, and transportation logistics [199]. In addition, the facilities must be economically viable, meaning that the capital expenditures (CapEx) and operating expenditures (OpEx) associated with the respective activation method must not exceed the generated revenue or overall profit. Nevertheless, a limited data set of pilot-scale implementations exists for carbonation-based processes, including wet and moist approaches, which demonstrate the technical feasibility of producing cRCP for SCM applications [200–204]. In contrast, thermal activation has so far been investigated primarily at the laboratory scale. Existing studies suggest that preliminary trends observed for tRCP can be qualitatively transferred to RCW, although performance is generally reduced due to dilution by inert constituents [15, 17,36,81,145]. The absence of pilot-scale implementations currently limits the validation of these findings under realistic conditions. However, the industrial-scale application of thermal activation has already been established for other kinds of cementitious materials, such as calcined clays, and can be adapted [205].

More generally, the comparability of activation methods across studies remains limited. The available literature is fragmented and typically based on differing raw materials and experimental conditions. While mechanical performance exhibits some degree of consistency, microstructural characteristics are strongly affected by the variability of the original concrete sources, thereby hindering direct comparison. Consequently, the transferability of laboratory findings to practical applications remains uncertain. Only a limited number of studies address real-world implementation, and these do not yet provide robust or generalizable conclusions regarding the relative effectiveness of different activation methods. Current evidence suggests that carbonation activation results in less pronounced strength improvements compared to thermal activation, indicating a higher effectiveness of the latter in terms of mechanical performance. Nevertheless, carbonation activation provides the additional benefit of  $\text{CO}_2$  sequestration, which may enhance its overall environmental performance. Future research should therefore focus on systematic investigations using identical source materials to enable direct comparison of activation methods. In addition, the influence of raw material variability on activation efficiency should be explicitly quantified. A comprehensive evaluation framework is required that integrates both performance-related metrics and environmental impacts, particularly process-related emissions. Such an approach is essential to assess whether activation yields a net benefit or whether the direct use of unactivated RCW represents the more sustainable alternative. Finally, further studies addressing durability and resistance to degradation mechanisms are necessary to evaluate long-term performance.

However, a full life cycle assessment (LCA) of the entire carbonation activation process is not yet available, as the process itself has not been fully optimized or clearly defined. With regard to thermal activation, there is already a study by Jing et al. [206]. In

this work, an activation temperature of 600 °C for 2 h is assumed, using RCW material. At a substitution rate of 10 wt.%, thermally activated RCW (tRCW) shows a CO<sub>2</sub> reduction of nearly 10 % compared to ordinary Portland cement (OPC). This represents a better performance than that of limestone powder (about 5 %) or fly ash (about 3.6 %), mainly due to the advantage of local availability. For reference, pure OPC accounts for roughly 1000 kg CO<sub>2eq</sub>/m<sup>3</sup>. However, this calculation should be regarded as an initial indication rather than being directly transferable to real-scale applications. Consequently, there remains a general research gap for both activation methods with regard to comprehensive LCAs. It is therefore advisable to first address the previously identified gaps, particularly the need for a systematic investigation of performance.

## 6. Conclusion

This paper compares the two most prominent activation methods for real recycled concrete waste (RCW). Since most studies to date have been conducted on cement pastes, mortars, or concretes prepared in the laboratory, and the activation primarily affects the cement paste rather than the inert components, the data collection is therefore limited to laboratory-prepared cement pastes (RCP). Each activation method was introduced and evaluated in terms of its impact on microstructural and micromechanical properties, hydration behaviour, and strength development, drawing on the current body of literature. The key findings can be summarized as follows:

- **Phase Composition:** RCP differs from OPC in both chemical composition and hydration state, which limits its direct applicability as an SCM; however, this can be partially compensated by activation. Thermal activation at approximately 600 °C represents the optimal activation temperature, as it induces dehydration without direct material-related CO<sub>2</sub> emissions and leads to the decomposition of portlandite, aluminate and C–S–H phases. This results in the formation of reactive aluminate and calcium-silicate phases, such as a modified Al<sub>2</sub>O<sub>3</sub> and modified polymorph of C<sub>2</sub>S, which imparts latent-hydraulic reactivity while refining the pore structure towards smaller pore sizes and maintaining a relatively high BET surface area. In contrast, carbonation activation relies on the reaction of RCP with CO<sub>2</sub>, resulting in the formation of CaCO<sub>3</sub> and, more importantly, an amorphous alumina-silica gel that governs pozzolanic reactivity, alongside a densification of the microstructure through reduced macroporosity and increased gel pore content, while simultaneously enabling CO<sub>2</sub> binding. Overall, both methods enhance reactivity and densify the microstructure through different mechanisms, latent-hydraulic in the case of thermal activation and pozzolanic in the case of carbonation, with current data suggesting a stronger increase in BET surface area for carbonation and slightly higher densities for thermal activation, although these trends remain uncertain due to the limited and partly inconsistent data basis.
- **Particle Size:** Thermal activation generally increases particle size due to agglomeration and recrystallization, although partial decomposition of these structures occurs at higher temperatures (700 °C to 800 °C). In contrast, carbonation activation appears to have a comparatively minor effect on particle size. However, direct comparison between the two activation methods remains challenging, as reported particle size distributions and decomposition behaviour vary significantly across studies due to differences in material characteristics and experimental conditions. In particular, the type of RCP, prior processing history, and initial particle size strongly influence the observed effects, making it difficult to draw general conclusions regarding the impact of either activation method on particle size.
- **Reactivity:** Thermal activation in the range of 400 °C to 600 °C generally enhances the heat of hydration, whereas higher temperatures (e.g., 800 °C) lead to a reduction due to structural degradation. Nevertheless, all thermally activated RCP samples exhibit improved early hydration kinetics compared to untreated material, primarily due to increased surface area, the presence of reactive dehydrated phases, higher calcium aluminate content, and the exothermic hydration of CaO. Carbonation activation similarly enhances RCP reactivity, as reflected by increased initial and cumulative heat release, although the overall performance remains below that of OPC. The initial heat evolution of carbonated RCP is largely governed by the rapid interaction between C<sub>3</sub>A and CaCO<sub>3</sub>, resulting in behaviour comparable to limestone composite cements. A comparison of literature data using the AI and II approach reveals considerable variability depending on material origin, processing, and testing conditions. While cumulative heat release generally decreases with increasing OPC substitution (H-AI < 1.0), both thermal and carbonation activation lead to higher reactivity compared to untreated RCP (H-II > 1.0). For thermal activation, maximum reactivity is typically observed around 600 °C. However, reported values for carbonation activation vary widely, with H-AI ranging from approximately 0.8 to 1.5 at 20 wt.% substitution, influenced in particular by the w/b ratio. A general trend in the reactivity of both activated RCPs as a function of the substitution rate is currently difficult to determine, since increasing the substitution rate also raises the w/b ratio, thereby introducing another influencing factor. Overall, despite the variability and limited comparability of existing studies, both activation methods consistently increase hydration heat relative to unactivated RCP, indicating enhanced reactivity. However, standardized testing conditions are required for a more reliable assessment. Another test for assessing reactivity, particularly of pozzolanic and latent-hydraulic SCMs, is the R<sup>3</sup> test. This method evaluates the potential for the uptake of chemically bound water as well as heat release, from which reactivity is inferred. However, the current database is very limited and, especially regarding the reactivity of cRCP, shows inconsistent results.
- **Strength development:** Thermal and carbonation activation of RCP both enhance mechanical performance (S-II > 1.0) by generating reactive phases and refining the microstructure, mitigating the typical strength loss associated with its use. Carbonation activation increases strength through the formation of pozzolanic alumina-silica gel, although overall performance remains below OPC (S-AI < 1.0). Thermal activation at approximately 600 °C is generally optimal, producing reactive calcium-silicate phases that improve hydraulic reactivity while avoiding significant CO<sub>2</sub> emissions, resulting in higher S-AI values

compared to cRCP and untreated RCP, particularly at higher replacement levels. Strength development is influenced by multiple factors, including replacement rate, precursor composition, particle size, activation parameters, w/b ratio, and mix design. At low replacement levels, filler effects can slightly enhance strength, whereas at higher substitution rates, activation helps mitigate strength loss, with tRCP at 600 °C typically outperforming cRCP. Despite these observed trends, variability across studies and interdependent influencing factors make it difficult to definitively compare the effectiveness of thermal versus carbonation activation, highlighting the need for standardized testing and controlled comparative investigations.

For most of the studies considered in this work, several factors such as type of raw material, replacement level, particle size, experimental boundary conditions, mortar composition, and water-to-binder (w/b) ratio used to produce RCP, which directly influence the performance of RCP, are not consistent. Hence, a more detailed quantitative comparison of the two activation methods at present remains difficult.

## 7. Future research directions

The present study demonstrates that it is neither possible to fully compare the two different activation methods and understand their differences nor to scale up the laboratory investigations towards industrial application using real RCW and industrial-scale dimensions. Therefore, further research is required. The identified research gaps, as well as the order in which they should be addressed, are listed below:

- Currently, there is insufficient data for a direct comparison of cRCP and tRCP systems, for example regarding the cRCP values for SII and HII properties. Therefore, a side-by-side experimental investigation using the same starting materials and experimental conditions is necessary for both activation methods, together with a reference material for comparison. In particular, the reactivity, strength, and workability should be the main focus.
- Regarding the need to reduce the clinker content to a minimum, the use of other SCMs, such as calcined clay, in combination with activated RCP should be investigated, as recent studies have primarily focused on their individual application rather than their combined use.
- To increase the industrial relevance, the starting material should be changed from RCP to real-world RCW, in which the RCP is not fully separated from the fine aggregate fractions. This would allow the transferability of the results to be assessed and facilitate the application of findings from other investigations.
- One of the biggest challenges in reusing RCW is its inhomogeneity. Therefore, it is necessary to establish a larger database to estimate the scatter and variability in the performance of the different activated materials. In this case, two requirements must be met: a consistent activation process and consistent performance of the activated material.
- Durability has to be investigated comprehensively, as it is an essential prerequisite for assessing the long-term sustainability of the material. Without sufficient durability and long-term performance, the potential sustainability benefits cannot be reliably realized in practical applications.
- Larger batch sizes should be investigated for both the activation process and the subsequent application of the activated material in concrete to evaluate process consistency, variability, and the transferability of laboratory-scale results to larger scales.

Beyond the technical performance and scalability, the environmental and economic feasibility of constructing and operating an activation plant should be assessed.

- The environmental impact and sustainability of the activation process should be evaluated through a comprehensive life-cycle assessment (LCA), with particular focus on energy consumption, CO<sub>2</sub> savings, resource consumption, and the overall environmental benefits of reusing RCW.
- The economic feasibility of the activation process should be investigated, including the investment and operating costs (CAPEX, OPEX) of an activation plant, energy requirements, material costs, and the overall costs associated with the production and application of the activated material.

## CRediT authorship contribution statement

**Maureen Denu:** Writing – original draft, Visualization, Methodology, Formal analysis, Data curation, Conceptualization. **Jan P. Höffgen:** Writing – review & editing, Validation, Supervision, Methodology, Formal analysis, Conceptualization. **Fabian Niewöhner:** Writing – original draft, Formal analysis. **Maciej Zajac:** Writing – review & editing, Validation. **Ravi A. Patel:** Writing – review & editing, Validation, Supervision, Methodology, Formal analysis, Conceptualization. **Alisa Machner:** Writing – review & editing, Validation, Supervision, Funding acquisition. **Frank Dehn:** Writing – review & editing, Validation, Supervision, Funding acquisition.

## Declaration of competing interest

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

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## Appendix A. Supplementary data

Supplementary material related to this article can be found online at <https://doi.org/10.1016/j.cscm.2026.e06611>.

## Data availability

Data will be made available on request.

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