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Thermoactivated Recycled Cement

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Abstract

The cement industry is currently faced by the great challenge of reducing its vast carbon footprint, due to being the second highest industrial greenhouse gases (GHG) emitter. This value is expected to further increase, since cement production is foreseen to rise by about 20% until 2050. Therefore, more eco-efficient alternatives to ordinary Portland cement have been developed towards a sustainable concrete industry. This chapter presents some of the latest advances in low-carbon thermoactivated recycled cements (RC) obtained from old waste concrete, leading to a significant reduction of the GHG emissions, while also encouraging the valorization reuse of waste materials and the reduction of natural resource depletion. The manufacture and general performance of RC, including the main production issues, rehydration behavior and phase and microstructure development, as well as its incorporation in cement-based materials are discussed. Some of the most recent research, main challenges and future perspective of RC are addressed.

Keywords: recycled cement, thermoactivation, low-carbon binder, sustainable concrete, waste concrete

1. Introduction

The construction industry is one of the most relevant sectors contributing to global warming, involving the extensive consumption of raw materials, depletion of non-renewable resources, extensive greenhouse gas (GHG) emissions and significant construction and demolition waste (CDW) disposal. Therefore, stringent environmental measures and relevant international agreements have been established to reduce the environmental impact of this industry. In this context, large companies worldwide have been investing in more sustainable practices, promoting the development of alternative more eco-efficient building materials towards a truly circular economy supported on recycling, resource efficiency and low carbon emissions [1, 2]. Two major goals of the European council are the reduction of GHG emissions to 60–80% by 2050, on a 1990 base year and the reuse of at least 70% of CDW, excluding backfilling operations [3, 4].

In particular, concrete, as the most used building material in the world and involving substantial extraction of raw materials, significant GHG emissions and extensive CDW generation, becomes a serious source of environmental concern [5, 6]. Cement is the main concrete constituent responsible for the significant carbon footprint of concrete, accounting with over 80% of the total CO₂ emissions of concrete production [7]. In fact, over 5% of the world's anthropogenic CO₂ emissions are attributed to the cement industry [8, 9]. This alarming value is expected

to further increase due to the ever-growing demand for this product, which is expected to rise by over 20% in 2050 [8, 9]. Therefore, the concrete industry and the scientific community have been focused on the urgent development of more sustainable eco-efficient concrete. The use of recycled aggregates in concrete production has been largely studied [10], but its effective acceptance in the construction sector is still a way off. On the one hand, the available technology is not efficient in providing high quality recycled aggregates with minor contamination with adhered paste that reduces their physical, mechanical and durability properties. On the other hand, the simple substitution of natural aggregates with recycled aggregates fails to significantly reduce the GHG emissions, which, as mentioned, are essentially related to cement production. Therefore, various strategies have been implemented concerning the efficient reduction of cement's environmental impact [8, 11, 12], namely the development of carbon capture solutions, new and more efficient production technologies, alternative fuels, alternative cements and the reduction of the clinker to cement ratio. Among these carbon reduction levers, the most promising and effective solution lies in carbon capture, but its viable implementation has yet to overcome some challenges [13]. The use of supplementary materials as partial clinker substitution has been considered for many years, but further GHG emissions reduction with currently available mineral additions is hard to explore. Moreover, the availability of some of these additions, namely those that are by-products of pollutant industries, such as fly ash, is becoming scarce.

Alternative low-carbon cements, such as calcium aluminate and alkali-activated cements have also been the object of intense research, but their implementation in the construction market is still far from being economically viable [13, 14]. More recently, a very promising approach relies on the production of low-carbon recycled cements (RC) from dehydrated waste hardened cement. The idea is to recover the binding properties of waste cement through its thermal activation at low temperatures, reducing the thermal energy of the clinker manufacture and avoiding the limestone decarbonation phase, which represents about 60% of the carbon emissions during the sintering process [13, 15].

The rehydration capacity of concrete subjected to high temperatures has long been shown from post-fire studies of concrete behavior [16–18]. This recovery was found to be related to the regeneration of new hydration products, despite the eventual existence of unreacted cement left in old concrete [19]. The possible reactivation of the hardened cement was a relevant finding and many authors started to explore this idea regarding the production of an innovative recycled binder. As mentioned, if thermoactivation temperatures are set to be under the decarbonation stage, the CO₂ emissions may be significantly reduced, and a low-carbon binder is obtained [20]. Moreover, retrieving the waste concrete highly encourages the valorization reuse of CDW, the reduction of natural resources depletion and the relevant decrease of landfill disposal. However, recycled cement is still a very young domain of research and various aspects related to its production process and its behavior when incorporated in cement-based materials must be further explored before the implementation of this very promising eco-efficient solution in the construction industry.

The objective of this chapter is to review some of the most relevant research and main contributions achieved in the domain of thermoactivated recycled cements. First, a general overview of the recycled cement manufacture is presented. Then, Sections 3 to 5 are dedicated to the phase development in anhydrous RC and subsequent rehydration, Sections 6 to 8 discuss the main physical, microstructural and mechanical properties of cement-based materials. Section 8 also covers the use of RC as partial cement replacement, showing the higher potential of this new recycled binder compared to current mineral additions used by the concrete industry.

2. Overview of recycled cement production

The first published work concerning the specific recovery of cementitious materials regarding cement recycling was probably presented by Splittgerber and Mueller [21]. The authors suggested exploring the inversion of cement hydration based on previous studies concerning the rehydration of hardened concrete subjected to fire temperatures. In fact, as mentioned, various studies have for long been carried out in this domain [16–18]. Basically, the thermal activation turns the cement hydration into a reversible process, obtaining dehydrated compounds with similar characteristics to those of the original clinker phases [21]. As better discussed in Section 3, the thermal activation explores the chemical transformations of the cement paste that occur at different temperatures, namely involving dehydration, dehydroxylation and decarbonation stages [22–26]. The production of recycled cement essentially involves three relevant steps, namely: the separation of the cement fraction from the other constituents of waste concrete; the comminution of the waste cement to an average particle size in the range of common ordinary Portland cements (OPC); thermoactivation of waste cement into RC. The closed circular economy involved in RC manufacture is illustrated in **Figure 1**.

One main obstacle that has hindered the production of recycled cement at an industrial scale is related to the individualization and separation of waste concrete constituents. This is not only a challenge for the production of RC, but also for obtaining high quality recycled aggregates. In fact, the contamination of aggregates by adhered cement paste increases their porosity and waste absorption [27], with repercussions on the durability, shrinkage and control of fresh concrete workability [28, 29]. For this reason, current recycled concrete waste is essentially reused as low-quality recycled aggregates for low grade concretes or as backfilling in road base layers and landscape recovery. Therefore, in order to enhance the CDW valorization and encourage a closed circular economy for concrete

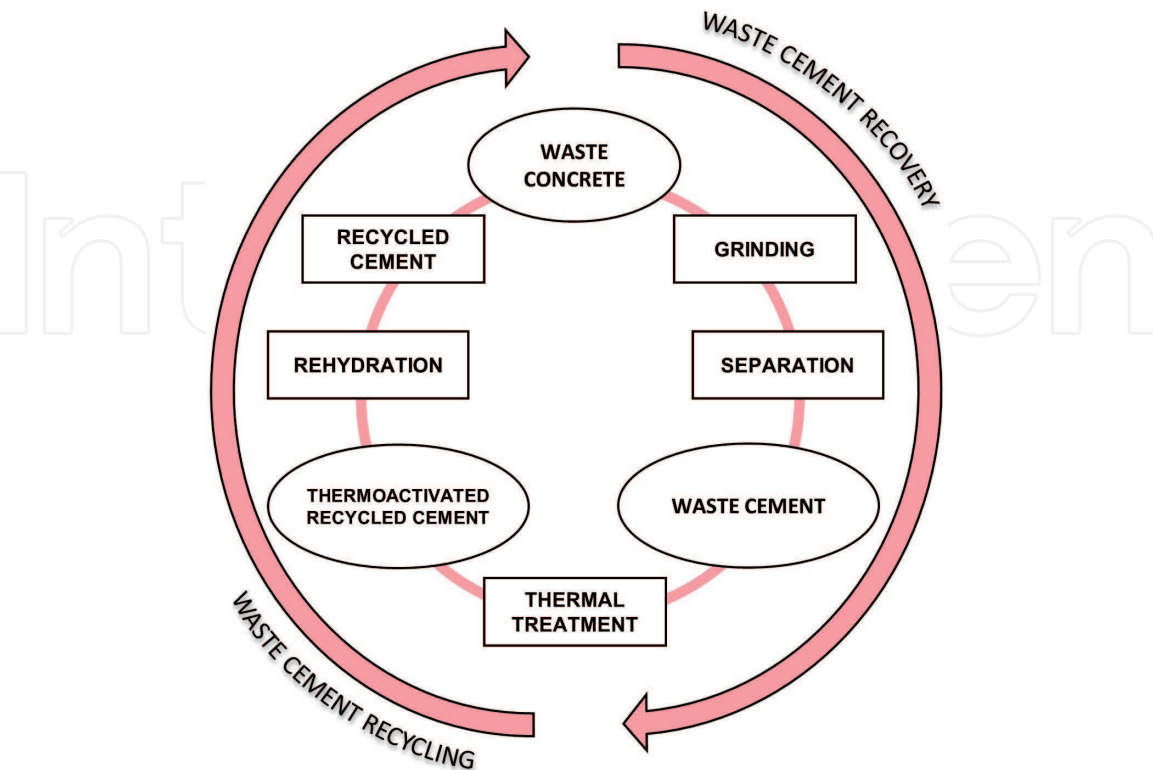


Figure 1.
Closed circular economy of thermoactivated recycled cement retrieved from hardened waste cement-based materials (adaptation from Carriço et al. [15]).

production, the development of an efficient separation process is a priority goal. However, despite various attempts, essentially focused on the recovery of cleaner recycled aggregates [30–32], no effective solution has been achieved yet. Most suggested separation methods essentially include mechanical [33] and thermal processes [31]. These methods take advantage of the different physical properties of concrete constituents, such as the crushing strength and thermal expansion, to promote separation. However, these processes are usually high-energy intensive and high contamination levels are difficult to avoid [34]. Nevertheless, some studies have thermoactivated recycled concrete fines (RCF), up to 5 mm, obtained from these processes [33, 35, 36], but the level of contamination may be high, especially when non-siliceous aggregates are considered. Other methods based on microwave heating [37, 38] or high-voltage electrical pulse discharge [39] fail to be easily implementable at an industrial scale. The lack of an effective method for waste concrete separation explains why most studies regarding the characterization of RC have involved the consideration of laboratory produced cement pastes, avoiding the challenging stage of concrete separation. Recently Bogas et al. [40] have patented a novel easily implementable and cost-effective separation process that is reported to yield waste cement with less than 12% aggregate contamination, by volume, and high quality recycled sand with as low as 3% of adhered cement paste [41].

In a second stage of RC production, waste cement is subjected to gridding, usually by means of ball milling as done in the cement industry [42, 43]. Some authors opt to previously oven dry the waste cement before gridding, since it reduces the baling phenomenon and wall mill adhesion [36, 44, 45]. In other studies, the RC grinding was performed after thermal activation [42, 46–49], however this turns the thermal process less effective and may lead to less homogeneous RC.

The increase of RC fineness enhances its rehydration reactivity, which leads to denser microstructures [50, 51]. Moreover, reducing the particle size of porous RC particles decreases their absorption properties. Therefore, it is recommended to produce RC with at least the same fineness range of OPC. However, this goal is not easily achieved in laboratory mills, especially when large amounts of RC are required. For this reason, most studies have considered particle sizes up to 150 μm . Zhang et al. [43] suggested the intergrinding of waste cement with slag (of higher hardness) in order to prevent the waste cement agglomeration and improve the fineness level.

The RC production conditions adopted by different authors are summarized in **Table 1**. The thermal treatment typically follows a thermal curve initiated by a heating rate of 5–10°C/min, followed by a residence time at maximum temperature and the respective cooling. It is expected that a lower heating rate will favor a more effective dehydration, but optimization of this parameter has never been reported [26]. However, the maximum treatment temperature and respective residence time are the main factors affecting the complete dehydration process at a given stage [59].

The first studies in this domain considered a wide range of thermoactivation temperatures, from as low as 200°C to over 900°C [46, 49, 60]. Later research has been focused on a narrower range, between 500 and 800°C [19, 33, 35, 43, 47, 61, 62], in order to comprise the phases of C-S-H dehydration and CH dehydroxylation, without relevant decarbonation. Optimal treatment temperatures have been reported to be in the range of 600–700°C, ensuring high rehydration ability and low thermal energy consumption [2]. The residence time has ranged from 1 to 8 hours in literature, although 2–3 hours is most often adopted [44, 46, 48, 49, 54, 55, 60]. The influence of the residence time and treated temperature on the mechanical strength of mortars produced with 25% of RC from the cement fraction of waste

Thermal treatment conditions				Refs.
Temperature (°C)	Residence time (hours)	Heating rate (°C/min)	Cooling	
200, 500, 800	1	10	cooling to room temperature	[35]
400, 650, 900	1	ND	accelerated cooling	[52]
750	1	ND	accelerated cooling	[48]
700, 750, 800	1.5	10	cooling to room temperature	[19]
650	4	10	accelerated cooling	[53]
600, 700, 800, 900	2	ND	ND	[44]
200, 400, 600, 800	2	5	ND	[46]
300, 500, 650	2	10	accelerated cooling	[54]
660–940	2.5	10	cooling to room temperature	[55]
700, 800, 900	1.5	10	600°C/min	[56]
120, 450, 750	8	7	cooling to room temperature	[42]
600	4	ND	cooling to room temperature	[42]
500	2	ND	ND	[57]
400, 500, 600, 700, 800, 900	5	20	cooling to room temperature	[58]
400, 450, 500, 600, 650, 700, 750, 800, 900	3	10	cooling to room temperature	[2]

ND, not disclosed.

Table 1.
Thermoactivation procedure of RC according to various authors.

mortar was analyzed by Kalinowska-Wichrowska et al. [63]. The maximum compressive strength was attained for RC treated at 650°C for 80 minutes.

The cooling procedure may occur inside the kiln at low cooling rates or may be accelerated by means of cooling devices, following the same philosophy adopted in OPC production [45, 48, 54, 61]. Serpell and Zunino [56] did not find significant changes when the cooling rate was accelerated.

3. Dehydrated thermoactivated recycled cement

After thermoactivation, the dehydrated RC particles are characterized by a porous structure and rough surface with high surface area, which is up to 15 times higher than that of OPC particles [21, 57]. In opposition, common OPC particles are non-porous and with smooth surface [42]. The helium particle density of RC tends to be slightly lower than that of OPC, ranging between about 2400–3200 kg/m³ depending on the treatment temperature [2, 21, 33].

Dehydrated waste cement is usually characterized by the absence of tricalcium silicate (C₃S), as well as the significant presence of free lime (CaO) or calcite (**Figure 2**) [1]. In fact, considering the main hydration reaction of calcium silicate

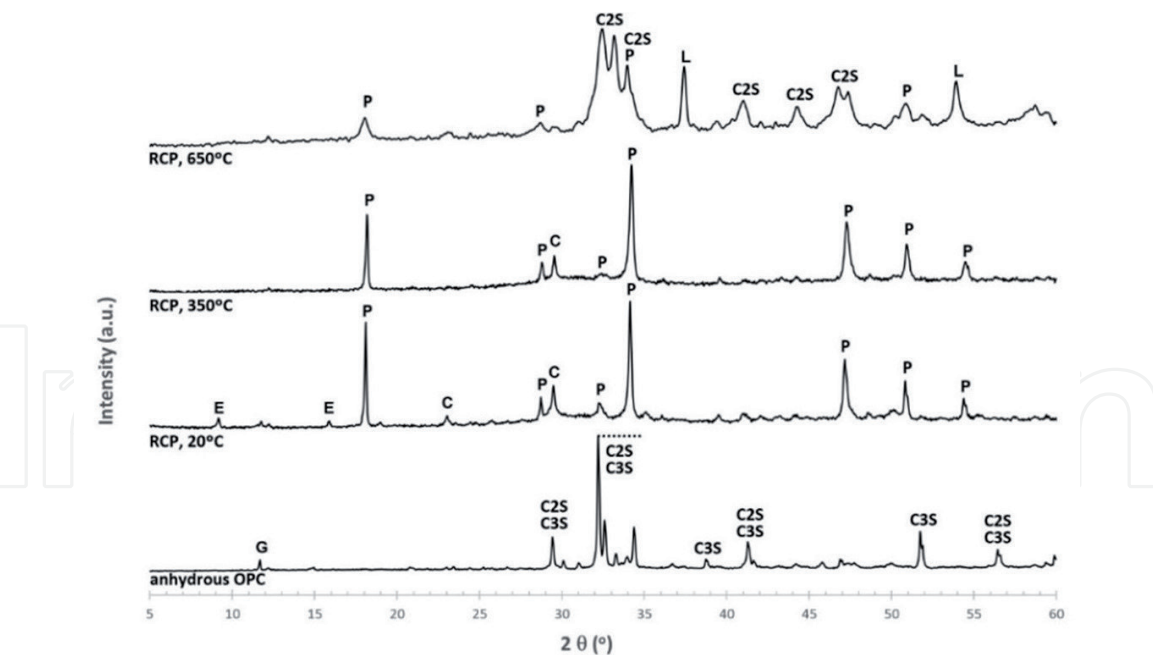
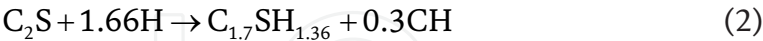
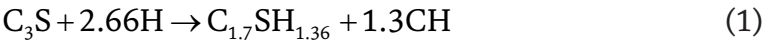


Figure 2. XRD of anhydrous OPC, hydrated cement paste (RCP, 20°C) and RC treated at 350 and 650°C (RCP, 350°C or RCP, 650°C) [1].

compounds of OPC Eqs. (1) and (2), the obtained C-S-H may be assumed to present an average C/S ratio (CaO/SiO_2) of about 1.7 [64]. Considering the reverse dehydration of the obtained hydration products, a C/S ratio lower than 2 would be expected for the new calcium silicate as well as the presence of free clime (CaO). Based on chemical and ^{29}Si NMR analysis of RC treated at 750°C, Alonso and Fernandez [24] estimated the value of 1.78 for the C/S ratio of the new nesosilicate, concluding that the dehydrated phase coefficients pertained to a structure close to C_2S .



The RC is also composed by other phases, such as dehydrated calcium aluminates and carbonated compounds, besides unreacted original anhydrous OPC grains and eventual residual hydration products [42, 49, 54, 58]. **Table 2** summarizes the main dehydrated phases identified by XRD in the literature.

Shui et al. [35] analyzed the dehydrated phases developed in RC treated at 200°C, 500°C and 800°C, by means of thermogravimetry (TG) and X-ray diffraction (XRD) analysis. Up to 500°C, RC was essentially composed of partially dehydrated C-S-H and CH, as well as CaCO_3 and C_2S , besides other amorphous dehydrated phases. Over 500°C free lime was progressively formed from the CH dehydroxylation and at 800°C the CaCO_3 was also decomposed into free lime. Similar conclusions regarding these major transformations with increasing temperature were also documented by other authors [1, 2, 42]. However, Wang et al. [42] reported a higher content of CaCO_3 after treatment at 450°C than in the source cement paste. The increase of carbonated products after thermoactivation was also documented by other authors [24, 46, 52, 65], attributing this phenomenon to the

Temperature (°C)	Precursor	Dehydrated phases	Refs.
200	RCF	C-S-H, CH, C ₂ S, CaCO ₃ , 4CaO.Al ₂ O ₃ Fe ₂ O ₃	[35]
500		CH, CaO, C ₂ S, CaCO ₃ , 4CaO.Al ₂ O ₃ Fe ₂ O ₃	
800		CaO, C ₂ S, CaCO ₃ , 4CaO.Al ₂ O ₃ Fe ₂ O ₃	
400	CP	β-C ₂ S, CH	[52]
650		β-C ₂ S, CaO	
900		β-C ₂ S, CaO	
600	AAC	quartz, tobermorite, CaO, β-C ₂ S, CaAl ₂ Si ₂ O ₈	[44]
700		quartz, tobermorite, β-C ₂ S, CaAl ₂ Si ₂ O ₈ , CaO, CS	
800		quartz, β-C ₂ S, CaAl ₂ Si ₂ O ₈ , CaO, CS, α-C ₂ S	
900		quartz, β-C ₂ S, CaAl ₂ Si ₂ O ₈ , CaO, CS, α-C ₂ S	
200	CP	CH, CaCO ₃ , C ₂ S, C ₄ AF	[46]
400		CH, CaCO ₃ , C ₂ S, C ₄ AF	
600		CH, CaCO ₃ , anhydrite II, C ₂ S, C ₄ AF	
800		CH, CaCO ₃ , anhydrite II, C ₂ S, C ₄ AF	
660–940	CP	α'-C ₂ S, β-C ₂ S, CaCO ₃ , C ₄ AF, CaO	[55]
700	CP	α' _H -C ₂ S, α-C ₂ S, CH, C ₁₂ A ₇ , C ₄ AF, CaSO ₄	[56]
800		α' _H -C ₂ S, β-C ₂ S, CaO, C ₁₂ A ₇ , C ₄ AF	
900		β-C ₂ S, α' _H -C ₂ S, CaO, Ca ₅ (SiO ₄) ₂ SO ₄ , C ₄ AF	
120	CP	CH, 1.2 nm tobermorite, jennite, CaCO ₃	[42]
450		0.96 nm tobermorite, disordered jennite, CaCO ₃	
750		C ₂ S, CaO, CS	
500	CP	quartz, CaMg(CO ₃) ₂ , CaCO ₃ , C ₂ S, CH	[57]
400	CP	CH, tobermorite	[2]
450		CH, CaCO ₃	
500		CH, CaCO ₃	
600		CH, C ₃ A, α' _L -C ₂ S, CaO	
650		CH, C ₃ A, C ₄ AF, α' _L -C ₂ S	
700		CH, C ₄ AF, α' _L -C ₂ S, CaCO ₃	
750		CH, C ₄ AF, α' _L -C ₂ S, CaO	
800		CH, C ₄ AF, α' _L -C ₂ S, β-C ₂ S, CaO	
900		C ₄ AF, α' _L -C ₂ S, β-C ₂ S, CaO	

RCF, recycled concrete fines; CP, cement paste; AAC, autoclave aerated concrete.

Table 2.
Dehydrated phases of RC identified through XRD according to various authors.

partial carbonation of dehydrated CH at intermediate temperatures. In addition, a higher content of carbonation products in RC is expected, because old concrete is prone to carbonate [2, 58].

Naturally, the free lime content tends to increase with the treatment temperature, especially after the decarbonation stage [25, 66]. The free lime must be taken into account during RC rehydration, since it contributes to the increase of heat release, consumption of mixing water and false setting [2, 55, 67]. Due to the

high hydration susceptibility of free lime, its previous partial hydration has been reported during the stages of cooling or storage [2, 58]. It was also suggested that these newly formed CH presented lower bonding energy than the original CH in waste cement.

Wang et al. [42] reported the presence of tobermorite and jennite within the temperature range 120–450°C. The former gradually dehydrated with increasing temperature, reducing the C-S-H layer spacing from 1.2 nm to 0.96 nm up to 450°C. Above 450°C, the diffraction peaks of tobermorite and jennite disappeared, suggesting the full depolymerization of C-S-H. At 750°C poorly crystallized peaks of wollastonite (CS) and larnite (C₂S) were identified, both presenting lower reactivity than partly dehydrated C-S-H phases [42].

In another study, Lü et al. [52] documented the full dehydration of C-S-H at 400°C. This was confirmed through ²⁹Si NMR analysis, in which incipient Q⁰ peaks started to replace the disilicates and chain silicates (Q¹, Q²) of the C-S-H structures. Over 650°C, Q⁰ peaks were predominant, indicating a relevant decomposition of C-S-H into what was termed as poorly crystallized β-C₂S. Similar findings were obtained by Alonso and Fernandez [24] in RC treated at 750°C, but the Q⁰ peaks were attributed to a new nesosilicate of higher reactivity than β-C₂S. Finally, at 900°C, only sharp Q⁰ peaks were identified by Lü et al. [52], suggesting the formation C₂S of low reactivity. The identification of β-C₂S peaks from XRD at 900°C was also reported by Serpell and Lopez [55], which become progressively sharper up to 940°C. However, at lower temperatures in the range 660–800°C, the presence of C₂S was attributed to the high temperature polymorph α'-C₂S, which is usually unstable at room temperature. The formation of this polymorph of higher reactivity than β-C₂S allowed to explain the better performance of RC treated at this temperature range than over 900°C, as also found by Shui et al. [49]. Based on XRD Rietveld analysis, Serpell and Zunino [56] later explored the development of different C₂S polymorphs in thermoactivated RC, for the range 600–800°C. According to the authors, the fraction of β-C₂S increased with the treatment temperature, substituting the polymorphs α'-C₂S and, with less relevance, γ-C₂S. The low temperature formation of α'-C₂S was attributed to the direct decomposition of C-S-H into this polymorph. These new polymorphs of higher surface area and lower crystallite size presented higher reactivity [56].

Considering a wide range of RC treated between 400°C and 900°C, Real et al. [2] confirmed the partial dehydration of C-S-H to tobermorite 9 Å up to 500°C, as well as the absence of ettringite. Up to this temperature, only incipient C₂S peaks were detected by XRD, indicating the onset of C-S-H transformation into C₂S crystalline polymorphs. After 600°C, the intensity of these peaks was increased and the C-S-H depolymerization was confirmed through ²⁹Si NMR analysis. The new nesosilicate form was identified as possible α'-C₂S or α'-C₂S. Finally, above 800°C, the C₂S peaks were more intense and sharper, indicating a higher crystallinity and β-C₂S was progressively formed, suggesting the generation of a less reactive product as also reported by other authors [52, 55].

The influence of the residence time and cooling rate in the RC thermoactivation has barely been studied. Based on a surface response model, the influence of these parameters on the formation of C₂S polymorphs was analyzed by Serpell and Zunino [56]. For cooling rates ranging 187°C/h to 1925°C/h only a slight reduction of strength development was found at higher rates. The formation of dehydrated phases was significantly affected by varying the residence time between 40 and 130 minutes, increasing the fraction of α'-C₂S for RC treated at nearly 700°C. However, for higher temperatures, this factor assumed less relevance.

The phase composition of dehydrated RC is also affected by the composition of the precursor source material. From XRD analysis, Vyšvařil et al. [46] analyzed

the relative amount of crystalline phases in different cement pastes (only OPC; 20% ground granulated blast furnace slag (GGBFS); 20% fly ash (FA)) subjected to dehydration temperatures between 200°C and 1200°C. In all samples C₂S was identified and mayenite appeared in GGBFS and FA pastes treated over 800°C. Calcium sulfoaluminate AFt and AFm phases were not identified at any temperature and treated material. However, these compounds are expected to be present in an amorphous form. This was observed by Baldusco et al. [57] from SEM images.

In sum, these studies suggest the development of a more reactive calcium silicate polymorph at intermediate temperatures, which becomes less reactive for temperatures higher than about 800°C. Overall, optimal thermoactivation temperatures are defined for the range 600–800°C. The final complex composition of RC depends on the thermal activation curve and the precursor source waste material [46, 49]. Other factors, such as the cooling rate and residence time may also affect the phase composition of RC, but further research is needed [56]. There is still some uncertainty in the characterization of the phase composition of anhydrous RC, which is affected by the different thermoactivation procedures and the limitation of the available techniques in identifying and quantifying the formed compounds (Table 2).

4. Rehydration of RC

The hydration mechanism of RC has yet to be fully understood. As discussed in Section 3, the dehydrated phase composition depends on the treatment temperature of RC, thus affecting the phase composition after rehydration. From XRD analysis, Xuan and Shui [60] characterized the crystalline phase composition of hydrated RC treated up to 800°C. The main phases identified in common hydrated OPC were also confirmed in RC, namely CH, CaCO₃ and residual C₂S and C₄AF, but associated to broader and less intense peaks. From SEM analysis, the authors found that at 400°C the RC pastes presented a looser microstructure with fine bundles of C-S-H intermixed with visible AFt phases of ettringite. However, for high temperatures up to 800°C more rehydration products were developed, and the microstructure was densified, but also with a rough and irregular morphology.

In rehydrated pastes treated up to 600°C, Vyšvařil et al. [46] also reported the presence of CH, CaCO₃, ettringite, C₂S and C₄AF. However, at 800°C ettringite peaks were absent and gismondine (CA₂S₂H₄) was identified above 600°C. Wang et al. [42] found that CH was almost absent in rehydrated RC treated at 450°C, confirmed by TG and XRD analysis. This was compensated by the increase of CaCO₃ content when compared to the source hydrated paste. From SEM/EDS analysis, RC presented a distinct morphology, characterized by massive nanosized clusters of C-S-H gel and CaCO₃, also evidencing the presence of calcium carboaluminates resulting from the reaction between calcite and aluminates. Baldusco et al. [57], for paste thermoactivated at 500°C, also reported the development of carboaluminates. In a recent work of the authors of this chapter [67], it was also confirmed the possible development of carboaluminates in regions where the calcium/carbon ratio measured by SEM/EDS was nearly 4.

According to Zhang et al. [68] the rehydration mechanism of RC implies the C-S-H formation, attributed to the repolymerization of partly dehydrated C-S-H in the presence of Ca²⁺ and water, as well as the hydration of β-C₂S. In addition, the formation of portlandite from CaO and calcium aluminates (possible C₂AH₈) from dehydrated aluminate phases was identified by the authors.

Real et al. [2] analyzed the phase composition of RC pastes after testing different temperatures, between 400°C and 900°C, through TG, XRD and ²⁹Si NMR analysis.

In general, the authors found that the rehydration was effective for any treatment temperature, presenting a similar amount of binding water as reference OPC. However, incomplete depolymerized RC, treated up to 500°C, failed to provide significant cohesive bonding between anhydrous particles. Above 600°C, the weakly formed CH presented less binding energy than the original ones in source OPC pastes. The authors found slight differences between RC and reference OPC from TG analysis, namely related to the formation of higher amounts of carbonate/sulfate Afm phases, a lower amount of CH and a higher amount of CaCO₃ in RC pastes. XRD analysis also confirmed the formation of similar crystalline phases in RC and OPC pastes, supporting the adequate rehydration ability of thermoactivated waste cement, especially above 600°C. The ²⁹Si NMR analysis clearly evidenced the effective depolymerization and rehydration of pastes treated above 600°C. Rehydrated RC treated between 600 and 900°C presented mean silicate chain lengths (MCL) between 3 and 6, which indicates the formation of C-S-H with a C/S ratio over 1.2 [51, 69]. Moreover, the MCL and Q²/Q¹ ratio were of the same order in OPC and RC pastes treated at 700–800°C, suggesting the development of the same type of C-S-H in both materials. However, the estimated coefficient of hydration was higher in RC pastes, indicating the formation of a higher volume of C-S-H, at least up to 28 days. This was attributed to the higher surface area of RC and the development of interparticle products as discussed later in Section 7. Noteworthy was the slightly higher MCL and Q²/Q¹ ratio reported for RC treated at 600°C, indicating a higher reactivity of this product with α'-C₂S in its constitution, which was able to form a greater amount of C-S-H of longer chain length. On the other hand, above 800°C an opposite trend was found in the MCL and Q²/Q¹ ratio, confirming the slower rehydration capacity of RC associated with the formation of less reactive C₂S polymorph.

From the above studies, it is concluded that the rehydration of RC involves the generation of C-S-H, as found in OPC, and AFt or Afm phases associated with sulfoaluminate and/or carboaluminate compounds. The type and morphology of hydrated phases depend on the treatment temperature of RC. The carbonated products tend to be more abundant in RC pastes, due to residual carbonation products in source waste cement paste and the possible carbonation that occurs after thermoactivation.

5. Kinetics of RC hydration

The hydration mechanism of RC has yet to be fully understood. As discussed in Section 3, the dehydrated phase composition depends on the treatment.

The mechanism and kinetics behind the hydration process of RC are still barely understood and subject of current research. Based on isothermal calorimetry (IC) tests, Wang et al. [42] analyzed the rehydration kinetics of RC heated at 200–1010°C. A significant initial heat release rate immediately upon contact with water was observed, followed by an induction period of a few hours and an acceleration stage with a maximum peak at about 10 hours. After the acceleration stage, the heat release rate was lower in rehydrated RC than in Ref. OPC. Among all tested temperatures, RC treated at 450°C showed the highest peak of heat release, as well as the highest cumulative hydration heat. The high reactivity for 450°C was attributed to the faster rehydration of partly dehydrated tobermorite and jennite (Section 3). This seems to have been triggered by the high calcite content that may have also provided nucleation and filler effects. Similar findings were reported by Angulo et al. [54] for RC treated at 500°C, then presenting less reactivity at 650°C.

As discussed in Section 4, other studies have shown higher RC reactivity for treatment temperatures above 600°C.

Carriço et al. [58] also found that similarly to OPC, the hydration mechanism of RC treated at 700°C involves four main stages corresponding to the initial hydrolysis, induction, acceleration and deceleration. However, RC shows a remarkably higher initial heat rate, especially up to 90 minutes, in which the heat release was nearly three times higher than that of reference OPC. This phenomenon was in line with the observation of a lower setting time in RC. However, the long-term heat release of RC tended to be lower than that of OPC, suggesting the formation of less hydrated products over time.

Real et al. [2] analyzed the hydration heat evolution of RC treated between 400 and 900°C through IC analysis. The maximum heat release and acceleration peak were attained for RC treated at 600°C. The authors also observed that RC required longer periods to initiate the acceleration stage and reach the maximum heat peak than OPC, especially those cements treated above 800°C. Moreover, the RC heat release during the acceleration stage was significantly lower than that of OPC, regardless the treatment temperature. RC treated at 400°C had no significant long-term reactivity, explained by the low depolymerization level of this product. The low reactivity of RC treated at 900°C was related to the low reactivity of the C₂S polymorph obtained at this temperature. Overall, it was concluded that RC pastes tended to present longer induction periods and less intense acceleration stages.

In all these studies [2, 42, 54, 58] the IC analysis were carried out after external mixing, preventing the determination of the heat released during the first few minutes of hydration. This initial higher hydration heat release of RC was attributed by various authors to the exothermic reaction of CaO [36, 42, 54]. However, according to Baldusco et al. [57] the high initial hydration cannot be solely attributed to the hydration of CaO, because high hydration rates are observed even in RC treated at as low as 500°C, in which the amount of free lime is not as significant. The authors suggest that as stated by Shui et al. [49], this phenomenon is better explained by the high surface area and instable nature of dehydrated RC phases, which are ready to repolymerize, despite the high calcium aluminate content of RC.

Taking into account the thermoactivation at 500°C of a waste binder composed by OPC and GGBFS, Balsusco et al. [57] documented a different shape for the IC curve. In this case, the curve was characterized by a high initial peak of heat release followed by a continuous deceleration, without the identification of typical induction and acceleration stages. According to the authors, instead of a dissolution-precipitation mechanism, the rehydration followed an instantaneous re-adsorption of water. These apparently contradictory results may be explained by the partly dehydrated state of RC anhydrous phases at 500°C. Indeed, the same behavior was found by Real et al. [2] for RC treated at 400°C. As the IC analyses were prepared with internal mixing, allowing to record the heat release upon contact with water, the cumulative heat was higher in RC than in OPC. Zhang et al. [43] also recorded the initial heat release of RC treated at 600°C, reporting that this was 10 times higher than in Ref. OPC paste. In this case, the addition of gypsum was not successful in reducing the initial reactivity of RC. In another study, Zhang et al. [68] tested other set retarders in order to control the hydration evolution of RC. For RC treated at 500°C the authors found that 1–1.5% of sodium borate was able to reduce the initial peak of heat release and extend the initial setting time.

In sum, although it is generally accepted that RC is responsible for a significant initial heat release upon contact with water, the kinetic mechanisms and involved rehydration reactions are still under debate.

6. Fresh properties of recycled cement

Two main obstacles that have been hindering the application of RC are its high water demand [43, 46, 48, 53] and fast setting time [49, 58, 67]. The high water demand is essentially attributed to the high surface area [57] and porous nature [2, 43, 48, 58] of RC, and in a second plan to the free lime content [49, 53, 55, 60] and particle agglomeration [53]. Therefore, for a given workability the RC pastes usually require a high water/binder (w/b) ratio.

Shui et al. [49] found that the amount of water required for normal consistency increased from 0.48 to 0.68 with increasing treatment temperature between 300°C and 900°C. These values were about 1.8 to 2.5 times higher than those obtained for OPC. The free lime and high surface area of RC were the main reasons attributed to this phenomenon. Both factors increased with increasing temperature. Xuan and Shui [60] studied the influence of the source paste composition (w/c ratio) on the water demand of RC treated at 200–900°C. The authors found that increasing the w/c of the source paste from 0.3 to 0.5 led to a 10–20% increase of the RC water demand, regardless of the treatment temperature. This may be explained by the higher porosity and higher amount of dehydrated products, with subsequent increase of the specific surface area, in RC treated from high w/c waste paste.

Yu and Shui [53] confirmed the great propensity for RC particles to agglomerate, which is encouraged by their high surface area. The particles are agglomerated by capillary action, trapping the mixing water between them. Therefore, part of the mixing water becomes unavailable and the water demand is increased for a given workability. After sonication, the authors found a slight increase of workability due to the partial dispersion of RC particles.

However, the authors found that the water demand was not significantly affected up to 50% replacement of OPC with RC treated at 700°C. The reported increase in water demand was 1.4 and 2.5 times higher for 50% and 100% RC incorporation, respectively. According to the authors, the lubrication effect provided by the fine OPC particles aided the reduction of the friction between coarser particles, partly compensating the adverse effect of RC. In a later study, Real et al. [2] found that RC treated between 400°C and 900°C may present 2 to 3 times higher water demand than OPC pastes of equal normal consistency, increasing with the treatment temperature. A more significant increase was found above 800°C, which was attributed to the higher free lime content, after decarbonation.

In order to reduce the effect of free lime on the water demand, Serpell and Lopez [55] suggested a two-stage mixing procedure. First, water is added in order to slake the free lime and the additional water is added to compensate the water loss by evaporation, which is measured by weight difference. Nevertheless, the reported increase for RC heated between 650 and 850°C was lower than 2%, which is not significant. Similar values were reported by Carriço et al. [58]. It was concluded that the surface roughness and porosity of RC were the most influential parameters.

Most studies report lower initial and final setting time in RC than in OPC [1, 35, 43, 48, 49, 55, 58, 60, 62, 68]. The initial setting time may be as low as less than 20 minutes, which makes it less viable for building applications [49, 58]. The low setting times in RC are associated to the fast rehydration of RC, owed to their higher surface area and higher reactivity [43, 49, 57].

The low setting time is also attributed to the free lime content [24, 46]. According to Vyšvaril et al. [46] this may explain the decrease of setting time with increasing dehydration temperature. In addition, Serpell and Lopez [55] suggested that the apparent rapid setting of RC is related to a “false setting” phenomenon that

may disappear after a while. A similar phenomenon is confirmed by Carriço et al. [58] for RC treated over 700°C.

The reduction of the setting time with the treatment temperature up to 800°C has been reported by various authors [43, 49, 57, 60]. Shui et al. [49] found that the initial setting time decreased from 42 to 17 minutes when the treatment temperature was increased from 300–800°C. This phenomenon was attributed to the quick repolymerization of new C-S-H, which does not involve the dissolution-precipitation process of OPC. Carriço et al. [58] found that the potential setting time of RC treated at 900°C was 2 times longer than that treated at 600°C, confirming the lower reactivity of RC for high dehydration temperatures. Similar findings were reported by Real et al. [2]. The authors also found that the potential replacement of OPC with up to 50% RC only slightly affected the setting time (less than 10%). However, Yu and Shui [53] reported a progressive reduction of the setting time with the incorporation of up to 30% RC. The same trend was documented by Yu et al. [61], for replacement percentages between 5 and 25%.

However, less expected high setting times in RC than in OPC were found by some authors [1, 2, 46]. The authors attributed this phenomenon to the possible agglomeration of RC, as well as the eventual pre-hydration of RC during cooling and storage. Setting times over 2 times higher in RC than in OPC are reported by Real et al. [2].

Taking into account source cement pastes of distinct w/c, Xuan and Shui [60] found lower setting times for high w/c cement pastes, attributed to the greater amount of dehydrated phases.

As well known, the setting time of OPC pastes is governed by the dissolution rate of C₃A and its combination with added calcium sulphate [51, 70]. However, in RC the lack of knowledge regarding the morphology of the dehydrated aluminate phases at different temperatures and their reaction mechanism does not allow to consider the same reasoning. As mentioned in Section 5, Zhang et al. [68] explored the use of different set retarders in RC treated at 600°C. The authors found that the addition of 4% gypsum only slightly increased the setting time. It seems that the dissolution of gypsum in RC can be slower than the reaction involved in the rehydration of dehydrated compounds. Contrary to OPC, in which the gypsum dissolution prevents the formation of CAH from C₃A, its addition showed to be ineffective in RC [15, 68]. Nevertheless, Sun et al. [62] documented a 6-fold increase of the setting time when 8% gypsum was added to RC thermoactivated at 550°C.

The above studies underline the challenging task of controlling the fresh behavior of RC pastes, still without an efficient solution to overcome their high water demand and non-standard setting time. Further research is needed in this domain, contributing to higher confidence in using RC.

7. Microstructure of RC pastes

Few works have been published concerning the microstructure characterization of RC cement-based materials. Most studies only involved simple qualitative scanning electron microscopy (SEM) analysis [55, 57, 68], where the phase morphology and global porosity were poorly assessed and only for a limited range of temperatures.

From SEM analysis, Shui et al. [49] documented the morphology of rehydrated phases of RC treated at 600°C, as fine bundles of C-S-H intermixed with honeycomb shape structures over the surface of dehydrated RC particles. In opposition, C-S-H in OPC presented a typical foil and fiber morphology. Overall, the RC pastes presented a looser structure than reference OPC pastes.

Zhang et al. [43], for RC treated at 600°C, also reported a highly porous hydrated paste with a weak bonding between phases. In a later study, Baldusco et al. [57] determined the total porosity of pastes produced with RC treated at 500°C from blended OPC and GGBFS. Based on water saturation-vacuum tests it was found that RC pastes developed higher total porosity at 7 days than reference OPC pastes. This was essentially explained by the higher intrinsic porosity of dehydrated cement particles. Noteworthy are two recent studies developed by Bogas et al. [67] and Real et al. [2], where the microstructure of RC pastes was deeply characterized over time and for different dehydrated temperatures, respectively.

Bogas et al. [67] analyzed the microstructure and phase evolution of cement pastes with RC thermoactivated at 700°C, by means of TG, XRD, SEM, quantitative backscattering electron (BSE), mercury intrusion porosity (MIP) and nitrogen adsorption (NA) analysis. Tests were performed in pastes with 8, 14 and 24 hours, as well as after 3, 7 and 28 days. The RC showed effective rehydration with the formation of C-S-H, AFm, Aft and other carbonation phases since early ages. Contrary to common OPC, AFm phases were developed, at least since 8 hours, associated with the formation of sulfoaluminates and carboaluminates. It was confirmed the progressive increase of CH and C-S-H over time, which shows that the hydration mechanism extends in time. The authors described the RC paste microstructure as a “dual structure”, where the intraparticle porosity is surrounded by a bulk interparticle matrix, in which the available space is lower. Basically, part of the mixing water is retained by inner hydration products, reducing the amount of water and w/b ratio between RC particles. This leads to a greater proximity between RC particles, increasing the packing density and paste cohesion, and consequently the early mechanical strength. Compared to OPC pastes, RC pastes showed higher reactivity up to 3 days, higher volume of hydrated products and more refined porosity, especially at early age. However, at 28 days the compressive strength was 32% lower in RC pastes, with less development of long-term interparticle hydration products. The total porosity was only slightly lower in RC and OPC pastes of equal w/b. The higher final setting time of RC was attributed to the very early formation of interparticle hydration products that do not contribute to the cement paste cohesion.

Following this work, a comprehensive study was carried out by Real et al. [2], where the influence of the dehydration temperature, between 400°C and 900°C, on the hydration and microstructure of RC pastes was analyzed by means of TG, XRD, ²⁹Si NMR, MIP and SEM analysis. Due to the above mentioned “dual microstructure” of RC pastes, a denser microstructure was found for RC pastes tested over 600°C than in Ref. OPC pastes. However, due to the insufficient repolymerization, pastes tested up to 500°C developed very loose microstructures associated with coarse porosity and low mechanical strength. On the other hand, RC treated above 800°C was less reactive, resulting in a less dense C-S-H network. It was thus concluded that the optimal treatment temperature was within the 600–650°C range. Similar to the previous work, the compressive strength was higher in RC pastes than in OPC pastes until 3 days and 27% lower at 28 days.

8. Mechanical properties of cement-based materials with RC

Various studies have already been carried out concerning the mechanical characterization of pastes or mortars produced with incorporation of 100% RC. For different dehydration temperatures and a wide range of w/b ratios the reported compressive strength varied as much as between about 4 and 30 MPa (**Table 3**). Besides these factors, the compressive strength is also affected by other parameters, such as the characteristics of the precursor material, grinding fineness and agglomeration

Thermal treatment conditions			Cementitious material				Refs.
Precursor material	Temperature (°C)	Duration (min)	Type	w/c	$f_{cm,28d}$ (MPa)	$f_{c,Rc}/f_{c,OPC}$	
CP	500	60	paste	0.40	8.0	ND	[35]
CP	800	150	paste	0.64	19.5	ND	[49]
CP	800	150	paste	0.50	20.0	ND	[60]
AAC	700	120	paste	0.45	4.2	ND	[44]
CP	800	120	paste	0.75	16.5	ND	[46]
CP	755	140	paste	0.70	31.2	ND	[56]
CP	500	120	paste	0.60	18.0	68%	[45]
CP	450	480	paste	0.55	32.3	85%	[42]
CP	650	180	paste	0.72	19.2	72%	[2]
RCP	500	60	mortar	0.26	4.7	ND	[35]
C	750	60	mortar	0.50	12.3	21%	[48]
CP	800	150	mortar	0.93	11.6	ND	[55]
CP	500	120	mortar	0.48	7.6	18%	[54]
CP	600	180	mortar	0.50	30.3	62%	[43]
CP	650	180	mortar	0.68	8.3	19%	[1]
C	650	180	mortar	0.81	0.9	2%	[1]
CP	700	300	mortar	0.58	21.6	54%	[58]
CP	650	180	concrete	0.65	33.2	83%	[2]

ND, not disclosed; RCF, recycled concrete fines; CP, cement paste; AAC, autoclave aerated concrete; C, concrete.

Table 3.
Maximum values for the 28-day compressive strength of 100% thermoactivated recycled cement pastes, mortars and concrete reported in the literature.

level of RC. When available, the relative compressive strength of RC versus that of reference OPC of equal w/b ($f_{c,Rc}/f_{c,OPC}$) is also indicated in **Table 3**. In all cases, the 28 days compressive strength of RC tended to be lower than that of OPC, confirming the development of a less dense net of interparticle hydration products, as discussed in Section 7.

When RC was directly thermoactivated from recycled concrete fines (RCF), the 28 days compressive strength was under about only 10 MPa (**Table 3**). This is related to the lack of effective separation methods allowing the increase of rehydratable compounds in the precursor material. Therefore, the development of new methods for the efficient individualization of waste concrete constituents is a priority goal in waste cement recycling. Following the new patented method of Bogas et al. [40], Real et al. [2] could retrieve cement paste from waste cement with only up to 12% aggregate contamination by volume. In this case, concrete produced with up to 30% incorporation of RC from waste concrete showed similar behavior to RC directly obtained from pure laboratory waste cement paste of equal composition.

As mentioned, most authors have opted to conduct their works using lab-made well-hydrated paste as precursor materials, in order to avoid the complex stage of concrete separation. Moreover, the full potential of RC is better accessed through non-contaminated waste precursors.

Xuan and Shui [60] showed that the mechanical strength of RC pastes is also affected by the w/c of the waste material. It was found that higher strength would

be obtained using waste cement pastes with lower w/c, of 0.3 instead of 0.5. This was attributed to the higher amount of unhydrated cement particles in less hydrated low w/c pastes, which may contribute to the subsequent development of more hydrated products. Another reason may also be related to the possible production of more interparticle hydration products in these systems with less porous RC obtained from low w/c pastes.

Different trends may be found in the literature regarding the evolution of the compressive strength as a function of the treatment temperature. In some studies, the maximum mechanical strength was attained for dehydration temperatures around 500°C [42, 45, 54]. For an optimal temperature of 450°C, Wang et al. [42] explained the obtained highest strength by the quick rehydration of partially dehydrated tobermorite and disordered jennite. For higher temperatures, the presence of wollastonite and crystalline larnite reduced the subsequent reactivity of RC. However, in most cases the optimal mechanical strength has been reported to be attained within the range of 600–800°C [19, 44, 49, 56, 58, 60].

A maximum compressive strength of as high as 32 MPa was achieved by Serpell and Zunino [56] for RC treated at 750°C. As discussed in Section 3, at this temperature range the authors identified the formation of a more reactive polymorph form of dicalcium silicate ($\alpha'_H\text{-C}_2\text{S}$), which progressively turned into the less reactive $\alpha\text{-C}_2\text{S}$ at higher temperatures. From various studies, including those exploring the use of ^{29}Si NMR analysis to characterize the structure of C-S-H in RC [2, 52], it seems reasonable to conclude that below 600°C the phase dehydration is incomplete and at higher levels the reactivity of the anhydrous Q^1 phases are highly dependent on their morphology and crystallinity. It also seems evident that over a maximum optimal treatment temperature the compressive strength is reduced [2, 42, 46, 49, 54, 56]. In fact, various studies in the literature suggest that above about 800°C the RC dehydrated phases react slowly and compressive strength is reduced [2, 42, 49, 55].

Lü et al. [52] first demonstrated that at 900°C the dehydrated SiO_2 tetrahedrons of $\alpha\text{-C}_2\text{S}$ were not significantly repolymerized upon water contact. Similar findings were obtained by Real et al. [2].

Bogas et al. [1] showed that the compressive strength of RC mortars might be increased when the maximum particle size of RC is reduced from 250 μm to 63 μm . Similar findings were obtained by Letelier et al. [36], comparing the performance of mortars with RC of 150 μm and 300 μm . However, taking into account RC with up to 75 μm or 150 μm , the authors did not find significant differences in the compressive strength. One reason for these differences is attributed to the agglomeration state of RC particles in the upper particle size range.

The agglomeration issue related to fine RC particles is documented by various authors [1, 2, 43]. From SEM analysis, Shui et al. [49] confirmed the poor dispersion of fine RC. Then, the same authors [53] demonstrated that the compressive strength could be almost doubled when RC was previously dispersed through sonication.

As discussed in Section 6, the increase of the treatment temperature increases the w/c ratio needed for a given workability. Naturally, this leads to a reduction of the compressive strength when compared to reference OPC of equal workability. In fact, Real et al. [41] reported a reduction of about 15% in the compressive strength when only 15% OPC was replaced with RC treated at 650°C. To compensate this, high dosages of superplasticizer have been considered in RC cement-based materials [2, 33, 61, 68]. According to Real et al. [41], the superplasticizers (SP) are also effective in reducing the mixing water in RC concrete, but the SP saturation point tends to be higher due to the porous nature and high surface area of RC. Nevertheless, the authors report that when SP is adopted, a better dispersion is

attained, and a high percentage of RC may be incorporated without significantly affecting the mechanical strength.

Regarding the strength evolution, there is an almost general consensus that the hardened properties are developed faster in RC than in OPC [41, 43, 57, 67]. Shui et al. [49] found that pastes with RC treated between 400°C and 800°C showed hydration degrees at 1, 3 and 28 days of 70, 80 and 90%, respectively. Moreover, the compressive strength of RC pastes at 3 days could be higher than that of reference OPC pastes. After this point, the relative strength of RC progressively decreased to 60% of that of OPC, at 28 days. Similar findings were obtained by Bogas et al. [67] and Real et al. [2]. As mentioned in Section 7, the compressive strength at 3 days was similar to higher in RC pastes than in OPC pastes but was about 30% lower at 28 days.

On the other hand, Baldusco et al. [45] reported that the 3 days compressive strength of RC paste could be about twice that of OPC. This was explained by the hydration of residual unhydrated calcium silicate grains, as well as the fast rehydration properties of RC. After 3 days, the strength evolution was not significant, being only 6% higher at 28 days. As discussed in Section 7, the authors suggested the formation of a long-term looser microstructure in RC pastes than in OPC pastes. According to Zhang et al. [43] in RC pastes, the compressive strength at 28 days is limited by the weaker RC particles of porous nature and low hardness. However, Bogas et al. [1] suggested that the strength evolution may be severely affected by the size and agglomeration state of RC particles, explaining the delayed strength evolution beyond 7 days found in RC mortars produced by the authors.

In order to access the potential use of RC as cement addition, various authors have explored various partial substitution percentages of OPC with RC. In a first study, Bogas et al. [1] analyzed the mechanical strength of mortars produced with 20–100% RC treated at 650°C. A progressive reduction of the compressive strength was observed, but up to 20% replacement the strength was similar to that of reference OPC mortars.

In mortar with 10 to 30% replacement of OPC with RCF treated at 500°C and 800°C, Florea et al. [33] confirmed the decrease of the compressive strength with increasing RCF content. Up to 10% replacement of OPC with RCF treated at 800°C, strength was little affected, but for 30% replacement, the reduction was as high as 35%. However, Xinwei et al. [48] reported more optimistic results for pastes with 40% replacement of OPC with RC treated at 750°C, leading to only a slight reduction of 12% in the compressive strength compared to reference OPC pastes. However, for 60% replacement, the compressive strength was 36% lower. Similar replacement ratios were studied by Araújo et al. [71] for RC treated at 700°C. The authors confirmed a slight reduction of the 28 days compressive strength for 40% replacement (of about 14%) but a significant decrease for 60% replacement. In another study involving pastes produced with 5–15% RC treated at 650°C, Yu and Shui [53] found maximum compressive strengths for 5% replacement, being 30% higher than that of OPC paste. The strength was further increased by about 15%, when the RC dispersion was improved by the addition of ethanol and a sodium-based dispersant followed by sonication.

So far, only a few studies have been published regarding the mechanical strength of concrete with RC. Letelier et al. [36] analyzed the mechanical strength of concrete produced with 5–15% of RCF treated at 400°C, 500°C and 900°C and 20 to 40% recycled aggregates. No significant compressive strength reduction, below 1%, was observed when OPC was replaced with up to 15% RCF. The strength reduction was more affected by the substitution of natural aggregates with recycled aggregates than by the OPC replacement with RC.

Carriço et al. [58] studied the influence of the incorporation percentage and dehydration temperature on the physical and mechanical behavior of RC mortars.

For replacement percentages with up to 20% RC the workability and compressive strength were not significantly affected. Mortars with RC treated at 600–800°C showed the best mechanical performance. For up to 50% incorporation, the mechanical strength was only 12 to 23% lower than that of reference OPC mortars of equal w/b. Moreover, even for a high w/b of about 0.6, mortars with 100% RC were able to attain as high as 27 MPa at 28 days. It was found that RC can be comparable to the low grade OPC of class 32.5.

Qian et al. [72] analyzed the production of ultra-high performance concrete produced with lime powder, silica fume and different replacement percentages of OPC with RC treated at 650°C. The authors found that up to 25% OPC replacement the concrete workability was only minorly affected. Regarding the compressive strength, it generally decreased with the incorporation of RC. The increased air content caused by the loss of workability was the main reason attributed to this reduction, especially for incorporation percentages over 25%. Nevertheless, at 7 days the compressive strength of concrete with up to 12.5% RC was higher than that of OPC concrete.

In a more recent study, Real et al. [41] investigated the mechanical strength behavior of concrete produced with RC treated at 650°C, obtained from waste cement paste or waste concrete. Concretes were produced with total or up to 40% RC incorporation. Up to 15% RC, workability was not significantly affected. Over this level, SP had to be incorporated in RC concrete production. Overall, the mechanical strength was not significantly affected or even increased by the incorporation of up to 40% RC. Even for 100% RC, the strength reduction was only 17%. It was thus concluded that the eco-efficient RC might have great potential as a supplementary cementitious material. Moreover, the RC concrete showed higher 3 days compressive strength than OPC concrete, regardless of the RC content. As expected, the modulus of elasticity decreased with the RC content due to its lower hardness and stiffness than OPC.

9. Conclusions

In this chapter, some of the most relevant research and main challenges concerning the production of low-carbon eco-efficient thermoactivated recycled cement were addressed. It is shown that recycled cement has a great potential for the efficient reuse of the large amount of waste concrete generated worldwide and the production for the very first time of fully recycled concrete, towards a truly circular economy. So far, recycled cement comparable to the low-grade Portland cement class 32.5 can be achieved and 28-day mortar compressive strengths over 20 MPa may be easily obtained, even considering their high w/b ratios. The main challenges that must be overcome are the high-water demand and non-standard setting times of RC. However, these issues are less relevant if RC is used as supplementary material for up to 40% replacement. In addition, other issues related to the optimization of the thermal activation process, effective separation of concrete constituents, and a deeper understanding of the dehydration and hydration process, as well as the physical, mechanical and durability behavior of recycled cement based materials need to be further addressed. However, although further research is needed, a significant step forward has been already achieved with the development of a more eco-efficient recycled binder able to respond to the very demanding environmental goals of the cement industry's road map. The aim is to supply the concrete industry with a very promising low-carbon binder, addressing the more efficient use of resources, the waste disposal issue and the decrease of the carbon footprint and associated quota.

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