

Improving cement waste recycling by carbonation: The feasibility of a Short Carbonation Process

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Extended Abstract

Concrete is responsible for an estimated total annual CO₂ emissions of 5-8% globally, derived from the high consumption of this versatile construction material world-wide [1]. Furthermore, out of the different stages in the production process of concrete, the cement manufacturing is responsible for about 80% to 95% by mass of the CO₂ emissions [2]. Thus, a promising prospect for reducing the environmental impact of concrete should be based on this critical concrete's component.

In 2009, the Cement Technology Roadmap highlighted four technology levers with the higher potential for an effective CO₂ emission reduction [5]. More recently, two of the identified technologies by the Roadmap have gathered more recognition and awareness by the academy, given its potential for carbon emission mitigation in the cement and concrete industry: the carbon capture utilization and storage and clinker substitution. Studies targeting both these technologies have applied a strategy specifically aimed to uncover the carbonation potential of end life recycled mature concrete prior to its reutilization in new concrete batches. In these studies, a by-product of mature concrete recycling, composed by a hydrated cement paste powder (CPP) and some aggregate residue, is carbonated and then reused as a supplementary cementitious material. Thus, this cementitious material works as a carbon sink, clinker substitution material and promotes a circular economy in the concrete industry [4].

Even though these studies expose a feasible strategy for carbon mitigation in the concrete industry, the novelty associated with this line of investigation still requires further research to obtain an industrial viable application process. One important stage in the viability of this strategy is the carbonation process of CPP, which still presents a long duration from three to twenty-eight days, depending on the study, to ensure a maximum carbonation degree [4]. A reason usually attributed to this long duration carbonation process is the different carbonation rate of different cementitious compounds present in CPP. While calcium hydroxide (CH) is able to fully carbonate in one day, provided proper humidity conditions, calcium silicate hydrate (C-S-H) takes a longer time, being the rate-determining carbonating compound towards the fully carbonation of CPP. Moreover, both these main calcium bearing cementitious compounds present different mechanisms during the carbonation reaction: CH is characterized by a dissolution-precipitation process, whereas C-S-H is associated to the decalcification and polymerization process [5].

The microstructural analysis of cementitious materials, aiming at accurately characterize their chemical composition, requires special attention because of the diverse set of compounds in presence, with also different chemical structures. Also, the carbonation reaction further contributes to complicate the analysis, considering the aforementioned different carbonation mechanisms that occur in cementitious materials. Furthermore, studies have demonstrated that calcium carbonate (CC), the main carbonate product, precipitates simultaneously in different polymorphs, with different crystallinity degrees and thermodynamic stability, with the following energetic downhill sequence: amorphous calcium carbonate (ACC), vaterite, aragonite and calcite [6].

The intricacy and complexity of characterizing cementitious materials, before and after a carbonation process, usually motivate the utilization of complementary analytical techniques, e.g. thermogravimetric analysis (TGA) or X-ray diffraction (XRD), which complement each other [14]. However, spectroscopy based techniques have demonstrated potential to characterize both amorphous and crystalline phases of cementitious materials, before and after a carbonation process, permitting both a qualitative analysis of the sample composition and a structural characterization of the main compounds (e.g. C-S-H and CC) [7, 8, 9]. In this study two analytical techniques based on spectroscopy are explored, Raman Spectroscopy and Fourier Transform Infrared Spectroscopy, to analyse the cementitious samples, intending also to evaluate the capacity of these techniques to characterize these materials. Nevertheless, the main objective of this paper is to propose and analyse a short duration (two hours) carbonation process for CPP with a specific carbonation condition setting, aiming at reducing the long duration of the carbonation process usually applied in prior studies, thus, increasing the industrial viability of this strategy for CO₂ uptake.

For this, a synthetic CPP was produced in the laboratory, simulating the CPP obtained in recycling facilities, but with an elevated level of purity and with a controlled and monitored composition. Thus, a cement paste with a water/binder ratio of 0.45 was produced and cured in a moist chamber for one year. After this, the hardened cement paste was dried, crushed, grinded and sieved through a 250 µm mesh, ensuing the CPP material used in the study. Water content was increase to values between 15% and 20% (by mass) to ensure a proper carbonation reaction. The short carbonation process was performed using an airtight chamber with an initial CO₂ concentration of 80% at the atmosphere pressure (1 atm), with a duration of two hours. The composition of CPP and the chemical

structure of both CC and C-S-H, before and after the short carbonation process, were investigated through Raman spectroscopy and Fourier-transform infrared spectroscopy.

Results revealed firstly that both spectroscopy based techniques were able to independently characterize both the composition of CPP and the chemical structure of C-S-H and CC, the two main compounds in CPP. Nevertheless, the redundancy of results obtained by using both techniques permitted an increase in the confidence of CPP characterization. Furthermore, these techniques do not provide a quantitative analysis of CPP, opposed to XRD and TGA techniques; yet, the expeditious results obtained by these spectroscopy techniques further emphasises the potential for a proper preliminary characterization of cementitious materials [14].

Concerning the main objective of this paper, the results revealed that CH, C-S-H and ettringite (AFt) contributed to the carbonation reaction. Results suggest that CH was consumed during the two hour carbonation process, and AFt was at least significantly consumed, although the full consumption of these calcium bearing compounds could not be entirely demonstrated. Regarding C-S-H, results demonstrated that these compounds reacted with CO₂ during the carbonation process originating a polymerization reaction, associated with the decalcification of C-S-H; yet, silica-gel was not obtained. The main carbonation product obtained was CC, and even though calcite was the main polymorph achieved, FTIR point out ACC as the second most probable polymorphs after calcite.

In sum, this study demonstrated that the main difference between the CPP carbonated using the short carbonation process proposed and the previous longer ones, is the absence of silica-gel. A result that increases the feasibility of this carbon capture technology by reducing the duration of a production process from a few days to a few hours, even more considering the positive results reported by literature regarding the utilization of C-S-H seeding in cementitious materials.

Acknowledgements

This work was supported by the Polish National Agency for Academic Exchange under Grant No. PPI/APM/2019/1/00042/U/00001, by the CERIS Research Centre, Instituto Superior Técnico, Universidade de Lisboa and by FCT through scholarship SFRH/BD/147856/2019. The authors would also like to acknowledge Professor Luís Santos for the experimental support with Raman Spectroscopy analysis.

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