Microstructural characterization of pastes produced with recycled cement

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The European commission have defined stringent targets regarding the reduction of greenhouse (GHG) gas emissions and the responsible generation, reuse and recycling of solid wastes (EC 2009). In this particular, concrete as the most used building material in the world, is the cause of significant GHG emissions, consumption of natural resources and generation of construction and demolition waste (CDW) (Schneider et al 2011). On the one hand, concrete waste represents over 30% of the CDW, which comprises the largest waste stream in Europe (Wang et al 2021). On the other hand, concrete production represents over 7% of the world man-made CO_2 emissions, essentially attributed to clinker production (MPA 2021, Wang et al 2021). For this reason, the concrete industry and research community have been intensively exploring the development of more eco-efficient alternative binders and the reincorporation of CDW in building products, towards a more sustainable and circular construction practice (CEMBUREAU 2020, Carriço et al 2020). Bearing this in mind, relevant research has been carried out regarding the production of recycled low-carbon binder from the hardened cementitious fraction of concrete waste. The idea is to recover the binding properties of cement waste through mechanical and thermal processing (Real et al 2021, Carriço et al 2021). Basically, the process involves one step of comminution (grinding and milling) followed by thermoactivation at low temperatures, in the range of 600-700ºC (Yu and Shui 2013, Carriço et al 2020). The manufacture of the obtained recycled cement (RC) may involves over 60% lower carbon emissions than conventional clinker, besides contributing to the lower depletion of natural resources and the greater valorization and reuse of CDW (Sousa et al 2021).

However, studies on this recent research domain are still limited. Most advances in this field have been essentially devoted to the production and mechanical characterization of cement-based materials produced with RC (Carriço et al 2020, Wang et al 2018). It has been demonstrated that the performance of RC may be greatly affected by the thermoactivated temperature (Yu and Shui 2013, Real et al 2020), and less by other factors such as the cooling rate and type of precursor concrete waste (Serpell and Zunino 2017, Carriço et al 2020). Depending on these factors and paste composition, the compressive strength of RC pastes may vary from as low as less than 10 MPa to as high as over 30 MPa (Wang et al 2018, Bogas et al 2019). So far, previous work of the authors have demonstrated that RC cam be comparable to low grade common Portland cement (PC) of class 32.5 (Carriço et al 2020b). Today, two major drawbacks on the industrial application of RC are its high water demand and difficult retrievement of the cementitious fraction from old concrete [Carriço et al 2020]. Regarding the later issue, the present authors recently developed an innovative separation method able to individualize the cement fraction from other concrete constituents, with only up to 15% aggregate contamination, by volume (Carriço et al 2021b). A comprehensive review on the last findings regarding the production and application of RC is presented in Carriço et al (2020).

Despite the significant research that have been carried out in this domain during the last 5 years, knowledge is still scarce, namely in relevant aspects, such as the hydration evolution, microstructure development, durability and volume changes of RC cement-based materials. Regarding the morphology and microstructure characterization of RC pastes, published studies are essentially limited to qualitative analysis based on scanning electron microscopy (Carriço et al 2020). Those studies have pointed out the apparent looser structure and long-term higher porosity of RC pastes compared to those with PC of equal composition. However, no comprehensive studies have been conducted regarding the detailed microstructural characterization of RC pastes.

In this context, the present study discusses the microstructure of pastes with incorporation of RC retrieved from cement paste waste. For comparison purposes, reference pastes with common PC of the same workability were also analysed. All pastes were characterized in terms of mechanical strength, isothermal calorimetry, X-ray diffraction, thermogravimetry and microstructure (qualitative scanning electron microscopy, quantitative backscattering electron imaging, mercury intrusion porosimetry and nitrogen adsorption). The microstructure development was followed since early age, between 8 hours and 28 days. The combination of all aforementioned techniques allowed a more reasoned microstructure characterization.

In this study it is shown the rehydration capacity of RC, evidencing the development of hydration heat and the generation of common hydration products since early age, namely calcium silicate and calcium aluminate hydrates. Contrary to PC, Afm phases are identified in RC pastes since 8 hours. However, Aft phases were less evident in RC pastes. The amount of carbonated phases tended to be higher in RC than in PC pastes. Hydration of

RC progressed overtime, although at lower rate than PC. In fact, the volume of external hydration products at 28 days tended to be higher in PC pastes.

RC pastes showed more refined porosity than OPC pastes of equal w/b, especially at early ages, until 3 days. However, the total porosity was similar. This is attributed to the biphasic microstructure of RC pastes, composed by the intraparticle porous structure and the interparticle space built by the outer hydration products. This led to a reduction of the water/binder ratio and a consequent densification of the outer space between particles. Although the mechanical strength of RC and PC pastes was similar up to 3 days, the 28 days compressive strength of RC paste was about 30% lower than that of PC paste.

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