INFLUENCE OF NANO-TiO_2 ADDITION ON THE ENVIRONMENTAL PERFORMANCE OF CEMENTITIOUS COMPOSITES: A HOLISTIC APPROACH

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Cement is one of the main compounds of concrete, the most consumed construction material globally. Cement manufacturing is responsible for around 8% of the total carbon dioxide (CO_2) emissions each year globally. Besides cement, concrete production requires a significant volume of aggregates and water. Over 8 billion m^3 of aggregates are used to produce 30 billion tons of concrete and other cementitious composites such as mortar every year. Thus, these cementitious composites have a significant environmental impact, and a small step in improving their environmental performance could lead to a giant leap in terms of global sustainability. It is known that the TiO_2 nanoparticles (nano-TiO_2) addition provides the cementitious composites the ability to reduce different pollutants concentrations (e.g., nitrogen oxides (NO_x) or volatile organic compounds (VOCs)) due to its photocatalytic activity. A preliminary study from Velay's Research group suggested that TiO_2 nanoparticles may also promote the carbonation of mortars. This preliminary data has served as the initial evidence of the potential use of nano-TiO_2 addition to promoting CO_2 capture. Besides, previous research showed that nano-TiO_2 addition may increase the compressive strength of cementitious composites. However, there is no agreement on what the optimum nano-TiO_2 percentage in terms of compressive strength enhancement is. Researchers also observed that an excess of nano-TiO_2 may reduce the strength of the composite. Understanding the mechanism and factors that may define the optimum percentage of nano-TiO_2 in terms of compressive strength would be vital in designing sustainable cementitious composites containing TiO_2 nanoparticles.

This dissertation aims to study the effect of nano-TiO_2 addition on the environmental performance of cementitious composites based on a holistic approach with three main pillars: (i) the composite performance, (ii) the impact of material production, and (iii) the effects during the service life.

Regarding material performance, this study analyzes the effect of the water-to-binder ratio (w/b) in the optimum percentage of nano-TiO_2 in terms of compressive strength. This part of the research concludes that the higher the w/b, the higher the optimum percentage of nano-TiO_2. In terms of age, the lower the age, the higher the optimum percentage of nano-TiO_2. Mixtures with high initial porosity present a dual beneficial effect of nano-TiO_2 addition due to the promotion of hydration (nucleation effect) and the reduction of porosity (filling effect). On the other hand, an excessive nano-TiO_2 may cause a lack of space for calcium hydroxide (CH) growth, thus strength reduction.

Concerning the environmental performance of material production, this research aims to assess the combined effect of nano-TiO_2 and recycled aggregate on mortars' life cycle assessment (LCA), using cradle-to-gate as the system boundary. Results indicate that nano-TiO_2 addition on mortar mixtures enhances the sustainability of mortars with recycled concrete aggregates (RCA) to a greater extent than on mortars with natural aggregates. Indeed, a 0.5% nano-TiO_2 addition highly decreased the global warming potential in mortars with RCAs, particularly with 100% RCA replacement.

Furthermore, this dissertation focused on unveiling new mechanisms to increase the sustainability of cementitious composites during their service life. This research concludes that using nanoparticles to change the microstructure and modify the pore structure of the concrete's matrix can make concrete to capture greater amounts of CO_2 through both active (CO_2 curing) and passive (weathering) carbonation. In terms of passive or weathering carbonation, results show that the addition of nanoparticles to cementitious composites produce two simultaneous competing mechanism in terms of CO_2 uptake: (i) a beneficial effect due to the reduction of the CH crystals' size that makes CH more prone to be carbonated, and (ii) a detrimental effect due to the reduction of porosity, that reduces the CO_2 penetration. These two competing mechanisms may explain why nano-TiO_2 can be beneficial or detrimental for the CO_2 uptake as a function of the water-to-cement ratio (w/c). The higher the w/c, the higher the maximum level of nanoparticles that increases the CO_2 capture of cement paste. Regarding active carbonation, cement pastes with nano-TiO_2 addition increase both CO_2 uptake and compressive strength after 12-hour CO_2 curing compared to mixtures without nanoparticles cured under the same conditions. These effects may produce a significant improvement in terms of the environmental performance of cementitious composites. For instance, the combination of 0.5% nano-TiO_2 addition and CO_2 curing reduces net CO_2 emissions of over 20% in a cement paste with a 0.55 w/c. Another study of this dissertation assesses how CO_2 curing may influence the self-cleaning activity of cementitious composites. Results suggest that the lower porosity of cement pastes containing slag produced higher and more concentrated surface carbonation during CO_2 curing. Thus, CO_2 curing produced a higher reduction of the superficial porosity in samples containing slag than in samples without slag. This higher reduction of the superficial porosity resulted in a higher self-cleaning enhancement after CO_2 curing; the lower penetration of pollutants makes them more exposed to the UV light, translating into higher self-cleaning activity. Results indicate that the combination of using slag cement and CO_2 curing has a synergistic effect that enables the production of photocatalytic cementitious composites with lower percentages of nanoparticles.