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Natural Low-Kaolinite Clays and Volcanic Materials as SCMs: Activation, Hydration and Evolution of Pore Structure.

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ABSTRACT

This thesis investigates the viability of naturally heterogeneous low-kaolinite clays and volcanic materials as supplementary cementitious materials (SCMs) for low-carbon binders. The work is motivated by the need to reduce clinker content in cement under climate constraints and in view of the declining availability of conventional SCMs such as fly ash and slag. In contrast to the kaolinite-rich clays that dominate much of the literature, the materials studied here are mineralogically complex and more representative of abundant natural aluminosilicate resources. The thesis therefore examines how activation modifies their structure and reactivity, and how these changes govern hydration, aluminium partitioning, strength development, and pore structure evolution in blended cement systems.

The results show that the two pozzolan groups respond differently to activation routes. For the clays, a combined thermo-mechanochemical activation (TA-MCA) was effective in improving pozzolanic reactivity. Compared to conventional thermal activation (TA), the TA-MCA improved reactivity by up to 353% and by 127% relative to mechanochemical activation alone (MCA). This was accompanied by increases in amorphization of up to 124%, specific surface areas increases up to 374%, and pronounced atomic-scale Al-Si network disruption. TA‑MCA produced the greatest structural disruption, together with high atomic-scale Al–Si network disruption. TA-MCA caused reduction of crystalline Al(VI) by 88% compared with TA and by 94% compared with MCA and the strongest broadening of $^{27}Al/^{29}Si$ resonances. For the volcanic materials, MCA improved reactivity across mineralogically diverse precursors, increasing MR3 reactivity from about 40-60 to 400 J/g SCM. In blended cements, the activated clays produced dense microstructures and strong later-age performance, with binary blends at 20-40 wt.% replacement reaching up to 125% of ordinary Portland cement (OPC) strength at 56 days and the 30 wt.% blend showing a 42% reduction in total porosity. Aluminium was found to be incorporated preferentially into C-(A)-S-H rather than predominantly into AFm phases, which promoted pore refinement but limited limestone synergy. The volcanic blends hydrated through a more distributed assemblage involving C-(A)-S-H, AFt, possibly strätlingite, and Mg-Al layered double hydroxide-type phases. Although their strength development was slower, 30 wt.% replacement reached OPC-comparable strength at 56 days, while 40 wt.% replacement still achieved about 92% of OPC strength. Overall, the thesis highlights the potential of complex heterogeneous natural pozzolans to be viable SCMs if mechanisms of their activation and hydration are adequately understood.

Keywords: Supplementary cementitious materials; low-kaolinite clays; volcanic materials; thermo-mechanochemical activation; pozzolanic reactivity; hydration; aluminium partitioning; pore structure; low-carbon binders.