



Orthogonal design optimization of low-grade limestone calcined clay cement: Workability, strength, and microstructural insights

Ying Liu^a, U. Johnson Alengaram^{a,*}, Shaliza Binti Ibrahim^b,
Gokulanathan Venkatesan^a, Zikai Xu^c

^a Centre for Innovative Construction Technology, Department of Civil Engineering, Faculty of Engineering, Universiti Malaya, Kuala Lumpur 50603, Malaysia

^b Institute of Ocean and Earth Sciences, Universiti Malaya, Kuala Lumpur 50603, Malaysia

^c School of Highway, Chang'an University, Xi'an 710064, China

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ABSTRACT

To explore the potential application of low-grade limestone (LGL) and illite clay (IC) in limestone calcined clay cement (LC³) systems, this study investigates the compressive strength, flowability and microstructure of mortar incorporating LGL and IC. An orthogonal experimental design (OED) was used to optimize the mix design by evaluating OPC replacement levels (30 %, 40 %, 50 %, 60 %) and the influence of IC and LGL calcination temperatures (300, 500, 800 °C). The highest 28-day strength (39.05 MPa) was achieved with both IC and LGL calcined at 800 °C. The optimized LC³-50 mix reduced CO₂ emissions by 33 % and energy use by 15 %. Response surface methodology (RSM) was applied to refine mix proportions. From an engineering perspective, LGL with incomplete calcination should be strictly avoided, as it negatively affects both flowability and compressive strength. Microstructural analyses (XRD, SEM/EDS, FTIR) revealed carboaluminate formation, denser C-A-S-H gels, and portlandite consumption. Pearson correlation analyses showed delayed hydration. The findings support optimizing locally available IC and LGL in sustainable LC³ formulations.

1. Introduction

Limestone calcined clay cement (LC³) is promising low-carbon binder system that combines reduced clinker content with enhanced durability and mechanical performance compared to ordinary Portland cement (OPC) [1–3]. The cement sector remains the second-largest industrial contributor to anthropogenic CO₂ emissions and the third-largest industrial energy consumer, responsible for nearly 8 % of total direct emissions globally. To address environmental challenges, supplementary cementitious materials (SCMs) such as fly ash, slag, and limestone have been widely adopted as partial clinker replacements, effectively reducing emissions while enhancing hydration and durability. However, their applications are often limited: fly ash suffers from low reactivity, restricting replacement levels [4]; slag allows higher substitution but is constrained by steel production [5]; and limestone primarily functions as a filler, and in many studies its replacement as clinker or cement is typically capped at around 15 %, beyond which performance losses often emerge [6,7]. In contrast, LC³ has gained broad research attention as a more versatile and sustainable system. Owing to the synergistic interaction between calcined clays, limestone, and clinker, LC³ not only achieves higher clinker substitution levels but also

* Corresponding author.

E-mail address: johnson@um.edu.my (U.J. Alengaram).