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Comparison of the effect of mix proportion parameters on behaviour of geopolymer and Portland cement mortars

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HIGHLIGHTS

- Lithomarge-based geopolymer mortars behaved in a similar way as cement mixes.
- w/s ratio had greater effect on workability of geopolymer mixes than paste content.
- Setting times were increased while strength decreased with increase in w/s ratio.
- Workable geopolymer mixes could be made with less free water than cement mixes.
- Geopolymer mortars had shorter setting times and a very rapid strength development.

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ABSTRACT

This work focuses on low-purity kaolin, widely accessible throughout the globe. Room temperature cured geopolymer mortars (GPMs) were formulated using an aluminosilicate precursor based on calcined lithomarge and potassium silicate activator. The effect of mix proportion parameters on the engineering properties of GPMs was investigated. The behaviour of GPMs was compared with that of Portland cement-based mortars (PCMs). Statistically designed experiments revealed that an increase in water-to-solid (w/s) ratio had a dominant effect on increasing the workability and setting time while decreasing the compressive strength of GPMs. In contrast to PCMs, GPMs proportioned with a constant water content showed a non-linear relationship between the w/s ratio and workability, which could be associated with changes to paste/sand proportions and/or water/alkali proportions. Like-for-like comparison of GPMs and PCMs showed that GPMs require lower free water content, and can offer shorter setting times and a rapid strength development.

1. Introduction

Geopolymer-based concretes are a new class of construction materials, where the cementitious binder is replaced with geopolymer alternatives, typically of low carbon footprint. Geopolymer binders are produced by reacting an alumino-silicate precursor, often a waste or a by-product material, with an alkali-silicate solution, also called chemical activator [1]. An inorganic polymerisation reaction results in the formation of hardened material with a three-dimensional and amorphous microstructure. Thanks to the unique, ceramic-like microstructure, geopolymer-based materials have been reported to have potentially equivalent, or even superior, physical and durability properties when compared to conventional materials made with Portland cement [2]. Geopolymers are most frequently renowned for a fast rate of strength development, fast setting time, resistance to chemical attack and improved fire resistance [3]. However, where the concrete/construction industry is concerned, geopolymer concrete still has to be proven to be more user-friendly and cost-effective, and to comply with specific engineering properties in order to gain more popularity.

Various alumino-silicate source types can be used as precursors for geopolymerisation. Among the most common are metakaolin (*i.e.* high purity kaolin) [4,5] and different types of calcined clays [6–9], slags [2,10,11] and ashes [2,12–14]. However, due to geographical or industrial diversity across the globe, precursors containing metakaolin or some industrial by-products (such as fly

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ash or ground granulated blast furnace slag) may not be locally available. Economy and sustainability of geopolymer technology are hindered by the need to source the precursor elsewhere and transport it to the place of further processing or intended use. Therefore, it is important to investigate the possibility of using locally available, naturally occurring, low purity materials, such as clays. These clays, being abandoned by industry, have the advantage of being cheaper than the high purity alternatives (*e.g.* metakaolin) or materials which are difficult/expensive to get access to. It has been recently shown that low purity kaolinitic clays can be calcined and used to produce geopolymer binders with compressive strengths exceeding 50 MPa [15–21].

Large deposits of kaolin-containing soft rock, called lithomarge, exist in Northern Ireland as part of the Interbasaltic Formation (IBF) [22]. Cooper [23] reported that lithomarge primarily contains kaolinite ($Al_2Si_2O_5(OH)_4$), gibbsite ($Al(OH)_3$), goethite ($FeO(OH)$), hematite (Fe_2O_3) and various smectite minerals. IBF material is typically seen as a nuisance by quarry owners. However, because of its kaolinite content, IBF could be used as an aluminosilicate source for the commercially viable formation of geopolymer binders, hence providing a large resource for future commercial production. Since the mineralogy of lithomarge varies, it is important to have an appropriate methodology in place to be able to identify the most appropriate precursor material for the production of geopolymer binder. McIntosh et al. [19] developed a protocol, not geographically limited to Northern Ireland, for refining the selection process of lithomarge suitable for calcination. It was shown that to produce binders with minimum compressive strength of 50 MPa, the kaolinite content should exceed 60% by weight of the original rock [19].

After decades of research evidence, it has been well established that the water-to-cement (w/c) ratio (or water-to-binder ratio for concrete made with additions, also called supplementary cementitious materials) is the dominant factor influencing most properties of conventional Portland cement-based concrete [24]. For a given set of concrete ingredients, selection of the w/c ratio and binder content are required at the mix design stage to produce concretes that meet specific strength and durability requirements. On the other hand, to achieve a desired workability at a given w/c ratio, a suitable content of free water in the mix or, more specifically, a suitable content of paste filling spaces between the aggregate particles, is needed. In the upcoming years, geopolymer binder concretes formulated using low purity kaolinitic clays will likely gain wider construction market access. Therefore, it is of importance to understand their behavior and compare it to that of conventional concretes. Recognising these needs, the overall aim of this work was to characterise the behaviour of lithomarge-based geopolymer mortars (GPMs), paving the way for the future development of a mix design of geopolymer concrete. GPM mixes were compared to Portland cement mortars (PCMs) to demonstrate whether the GPMs can be used by the industry in a similar way to a Portland cement system. Therefore, the primary objective of this research was to assess the effect of mix proportion parameters, *i.e.* water-to-solid (w/s) ratio, paste volume and free water content, on workability, setting times and compressive strengths of room temperature cured geopolymer mortars formulated using an aluminosilicate precursor based on calcined lithomarge and a potassium silicate activator. Design of experiments approach (DoE) was used to simultaneously investigate the effect of w/s ratio and paste volume on the properties of GPMs. In addition, the effect of a wide range of w/s ratios was studied on GPM mixes made with either fixed paste volume (varied free water content) or with a fixed free water content (varied paste volume). The behaviour of these two groups of GPM mixes was compared with that of Portland cement counterparts made with varied w/c ratios. The secondary objective was to directly compare the performance of selected GPMs with that of Portland cement alternatives in the same strength class (normal and high strength) and formulated with the same paste volume.

2. Research significance

In recent years there has been tremendous research effort into development and characterisation of cement free binders and concretes, to overcome shortcomings and lower the overall environmental impact of Portland cement concrete. However, most of the effort has been dedicated towards usage of slags, ashes or pure metakaolin. This paper provides data regarding the effect of variation in selected mix proportion parameters (w/s ratio, paste volume and free water content) on workability, setting time and compressive strength of geopolymer mortars formulated with a lithomarge based precursor, *i.e.* a low purity kaolin. An essential part of this work was devoted to benchmarking the behaviour of mortars made with the new binder against that of conventional Portland cement mortars, to find similarities and differences between these two binder systems. This data should lay strong foundations towards the development of methodologies for the mix design of concrete made with low purity kaolin geopolymer binders, encouraging their popularisation and industrial acceptance. Such data can be of interest to the wider scientific community, designers and producers of concrete, as well as contractors, to better understand key mix proportion parameters affecting fundamental properties of geopolymer concrete formulated using a lithomarge based binder.

3. Experimental programme

The research methodology is first outlined, followed by a short overview of the design of experiments technique (*i.e.* central composite design) which was adopted in the opening part of this work. Afterwards, the description of materials and mix proportions used is shown. Mortar mixing and sample preparation are then described, followed by the presentation of testing procedures.

3.1. Methodology

To satisfy the first objective, seven families of mortars, five GPMs and two PCMs, were tested. Their mix proportion parameters are reported in Table 1.

Design of experiments (DoE) [25] approach was used to simultaneously investigate the influence of w/s ratio (factor A) and paste volume (factor B) on workability, setting times and compressive strengths of geopolymer mortars (GPMs) – mixes called GPM-0. The DoE approach has been chosen because it allows identification of the most influential factor(s) or factor interaction(s) affecting the investigated properties. Taking into account that the investigated properties were not expected to change linearly, the GPM-0 group of mortars was proportioned with a wide range of w/s ratios and paste volumes according to 2^2 full central composite design (CCD) plan, to obtain quadratic mathematical response models. A summary of the investigated levels of factors, in terms of actual and coded values (*i.e.* transformed actual values), is given in Table 2, while an overview of the CCD is presented in the subsequent section.

In addition to the DoE work, workability and compressive strengths of GPMs were studied using a wider range of w/s ratios, *i.e.* from 0.275 to 0.6, either by keeping a constant paste volume or a free water content. For this range of w/s ratios, ten mortars were made with a constant paste volume of 500 L/m^3 (GPM-1) and another ten were made with a constant water content of 235 L/m^3 (GPM-2). To verify workability findings for GPM-2 mixes,