

Geopolymer Binders

Subjects: Others

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Based on numerous studies conducted in recent years, geopolymer binders are considered to be sustainable and environmentally friendly cementitious materials and are attractive in terms of their good cost–performance ratio due to low energy consuming production process and use of secondary raw materials instead of virgin ones [1][2].

Keywords: geopolymer ; workability ; compressive strength ; initial temperature

1. Introduction

Millions of tons of industrial wastes are being stored and disposed in landfills annually creating a pollution risk. The environmental aspect is a great driver to initiate as human society strives for cleaner production by finding opportunities for re-cycling of waste materials instead of landfilling. Disposal of industrial waste also requires the use of land, which can be potentially utilized for agricultural purposes or other needs. Reuse of secondary raw materials in production of alkali-activated aluminosilicate materials could not only expand the raw material base of the construction industry without increasing the use of natural mineral resources, but also reduce the effect of greenhouse gases associated with ordinary Portland cement and concrete production. Production of geopolymers requires significantly less energy than the production of Portland cement [3][4][5][6]. In addition, Portland cement-based concrete is subject to certain durability problems, including aggregate–alkali reactions, chloride-induced corrosion in reinforcing steel, retarded ettringite and thaumasite formation [7], which are not characteristic for geopolymers.

The proportion of the raw materials, viscosity of the fresh mixes and molarity of the alkaline activation solutions for geopolymer synthesis can be varied over a wide range, depending on the chemical and mineralogical composition of the precursors, which can also alter the reaction kinetics, chemical properties and microstructure of the geopolymers [8]. Hydroxide solutions with different molarities, as well as mixes of hydroxides and sodium silicates, are most frequently used as alkaline activation solutions to generate a high pH and activate source materials [9]. In cases where the molarity of the alkaline activation solution is insufficient (for example, the molarity of NaOH is less than 1 M), incomplete ion dissociation can occur, which results in an insufficient amount of intermediate from which new compounds can be formed. Alternatively, the opposite—if the molarity of the alkaline activation solution is too high (greater than 15 M), dissolved products can form a layer around unreacted particles restricting or significantly slowing ion dissociation. Even if an undesirable layer is established around the particles, it has been found that the geopolymerization process does not happen completely, which results in a lower mechanical strength of the material [10][11].

2. Studies on Rheology of Geopolymer Paste

The acceptance of alkali-activated materials by the construction industry is needed at industrial scale. To do so, it is important to determine key parameters affecting workability and fluidity of the fresh paste of geopolymers. Especially it is important for construction sites where temperature can rapidly change during day time or to be extremely different depending from the part of year. Initial temperature of the geopolymer paste and extra water addition to the composition can change workability of the fresh mix which will resulting into different properties of the hardened materials compared with planned ones.

Some studies show an important effect of the alkaline activation solution on the rheology of geopolymer paste [9][12] that plays a key role in its practical applications [13]. In particular, NaOH-activated slag pastes behave as Bingham fluid [9][14], while slag pastes activated by using a sodium silicate solution alone-fit the Herschel–Bulkley model [9][12]. In most cases, a higher water/alkaline ratio of the activation solution improves the flowability and workability of the paste [15]. The size, shape and other geometrical properties of the precursor's particles effect the rheological properties of the fresh geopolymer paste [16]. Mixing time and mixing speed have an influence on the rheology of geopolymer pastes as well. According to the research of Kaur et al. [16], the active mixing of raw materials results in increased viscosity of the paste and improved mechanical properties of the hardened samples. This is explained by the input of mixing energy, which can partially break down the size of the raw material “aggregates” which can occur due to agglomeration of the powdered raw

materials during milling to allow the complete reaction to continue. Mixing is also a temperature-dependent process, which is influenced by the activation energy of raw materials used for geopolymer production. The viscosity and geopolymerisation reaction rate depends on the paste temperature because Brownian motion is slowing down and the dissolution rate of aluminum decreases at lower temperatures [9][16].

It is reported that the higher molarity of activation solution made of sodium hydroxide increases the viscosity of the mixture, which results in the increased strength of the hardened samples [15][17]. Moreover, regardless of the alkaline activation solutions used (NaOH, Na₂CO₃ or sodium silicate solution), an increased concentration of Na₂O in the activation solution increases the viscosity of the mixture [18]. Furthermore, when sodium silicate solution is used as the alkaline activation solution, a rise in the SiO₂-Na₂O ratio decreases the workability of the geopolymer paste [9][18][19]. There is an opinion that the water in a geopolymer mixture does not play any role in the chemical reaction and merely provides for the workability of the paste [8], but other studies discuss the positive effects of water retention on the geopolymerisation process and on the final properties of metakaolin-based and fly ash-based geopolymers [20][21][22].

However, a higher water/alkaline ratio in the activation solution can negatively affect the formation of the geopolymer structure, pore structure, mechanical properties and durability, as well as the modulus of elasticity, due to the increased pore volume and pore size [23][24].

The water in the micro-structure of the geopolymers accumulates in the pores, providing only alkaline transfer functions; it is not directly involved in the geopolymerisation process [25] but has an effect on the rheological properties of the material [26][27]. The decrease in workability of the geopolymer paste is associated with an increase in the molarity of the activation solution [17][28]. Koutnik et al. [24] conclude that the viscosity of the geopolymer paste, and later compressive strength of the specimens, strongly depends on the amount of filler (sand) and additional water used in the composition: increasing the amount of water results in a decrease in viscosity and compressive strength. On the contrary, when the water content of the composition is decreased from 45% to 30%, the viscosity increases from 0.68 Pa·s to 8.1 Pa·s—that is, 12 times higher—and the compressive strength increases from 11 MPa to 85 MPa, which is eight times higher.

Only a few studies discuss the influence of decreasing the alkaline solution molarity by diluting it with water to obtain better workability of the mixture or the properties of the hardened geopolymers after the initial curing at an elevated temperature and prolonged curing time at room temperature [29]. A higher water/alkaline ratio in the alkaline activation solution can negatively affect the structure of the geopolymers by increasing the size of the dominant pores and total porosity, which decreases its mechanical properties and reduces long-term durability [30][31].

Although viscosity is an important property affecting fluidity, workability and applicability of geopolymer paste, only a few detailed studies have been published clarifying this complex issue [9][19][20].

The rheological properties of geopolymer pastes depend on the ratio of water to alkaline in the activation solutions and can strongly influence durability of the hardened geopolymers. This is important for adapting geopolymer compositions for practical use in construction sites in ambient temperatures, which depend on the weather conditions and period of year.

The paste's initial temperature and extra water in activation solution, reflected in an increase in the water/10 M NaOH ratio, significantly affect the electrical conductivity, viscosity and spread values of geopolymer pastes and, therefore, the structural development, density, porosity and compressive strength of the samples after curing.

The formation and compaction of the geopolymer structure is accelerated by increasing the initial temperature of the paste and decreasing the extra water in the activation solution. This is confirmed by the UPV measurements: the highest UPV values were detected for the specimens made using an alkali activation solution with a lower water/10 M NaOH ratio, meaning an accelerated formation of structure during the curing process compared to other compositions. The basic formation of the structure for the samples with a lower water/10M NaOH ratio in the activation solution with initial paste temperatures of 35°C, 15°C and 5°C occurred during first 4, 8 and 24 hours. The structure formation was delayed until 22–24 hours for samples with the highest water/10 M NaOH ratio in the activation solution. The UPV measurements also depended on the water/10 M NaOH ratio of the activation solution used for geopolymers: a lower water/10M NaOH ratio of the activation solution led to an increase in the UPV values for the samples after seven to 56 days of curing. Increases to the initial temperature of the pastes used to prepare the samples led to the UPV increment becoming slower for the samples during their curing period.

During the whole curing period of up to 56 days, the compressive strength of the samples prepared from paste with an initial temperature of 5°C increased more than 1.65–2 times; for samples prepared from pastes with higher temperatures (15°C and 35°C), the increase was 1.3–1.7 times. After 56 days of curing, the strength and density of the samples

prepared at different temperatures with the same water/10 M NaOH ratio in the activation solution had practically levelled off.

The XRD confirms the compressive strength test results and reveals that a lower water/10 M NaOH ratio in the activation solution and a higher initial paste temperature leads to a more intensive geopolymerisation reaction and formation of the reaction by-product hydroxy sodalite.

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