MECHANICAL CHARACTERISATION OF ALKALI ACTIVATED CLAY-BASED GEOPOLYMER BINDER MADE OUT OF GRAVEL WASH MUD

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Introduction

One of the most promising cement alternatives are geopolymer cements. The term "geopolymer" was mentioned for the first time by Davidovits[1] and classifies all forms of inorganic polymeric material synthesised by chemical reaction of aluminosilicates and an alkaline activating solution[2-3]. The production of geopolymer binder comprises in two main procedures: calcination and geopolymerisation. The synthesised geopolymer shows interesting characteristics like good mechanical properties, high strength and good durability[4-5].

Most of previous investigations used metakaolinite with high content of reactive SiO_2 and Al_2O_3. In this study, a waste product from gravel mining, namely, gravel wash mud (GWM) was examined and processed into a powder to synthesise an alkali-activated binder. The chemical composition and the mineralogical composition was examined using XRF analysis. The GWM powder is calcined at different temperatures and varying calcination times. After calcination, the mineralogy of the calcined material is analysed using the XRD-Rietveld method. In the following step, the geopolymerisation process is initiated by alkaline activation of the calcined clay with an 8M NaOH solution. Finally, the performance of the geopolymer is examined for mixtures with different liquid/ solid ratios after different curing times. This study allows to identify the suitability of the gravel wash mud as a performing geopolymer binder and provides early indications about its performance without addition of further strengthening components.

Material properties & Experimental procedures

The raw material is a gravel wash mud (GWM) which occurs as a waste product from gravel mining. The mud was collected from a sludge reservoir, located in the North

West of Luxembourg and stored into sealed buckets. The raw material showed no impurities beside some gravel stones, which were picked up during the extraction at the edge of reservoir. In the laboratory, the GWM was dried in an oven at 105°C for 2 days. After cooling down to room temperature (RT), the dried GWM was finely crushed into a powder using a laboratory jaw crusher. Additionally, the particle size distribution was determined by laser granulometry (HELOS & RODOS) and resulted in particle sizes within the range of 0.9 - 175 µm with an average of 6.35 µm. Furthermore, Table 1 shows the chemical composition of the dried GWM powder (Bruker S4 Explorer). The results show a high content of SiO_2 and Al_2O_3, whereas compared to kaolinite (SiO_2: 44.2% and Al_2O_3: 40.2%)[12], the SiO_2 content of the GWM powder is higher and the Al_2O_3 content is lower.

Table 1: Chemical composition of the dried GWM powder

Element	SiO_2	Al_2O_3	Fe_2O_3	K_2O	MgO	TiO_2	CaO	Na_2O	P_2O_5	SO_3	MnO
Percentage (%)	65.77	18.92	9.04	3.31	1.30	0.74	0.41	0.17	0.14	0.11	0.09

The mineralogical spectroscopic analysis was carried out for dried GWM powder, which was calcined in a laboratory oven at 550°C, 650°C and 750°C for seven different calcination times from 15 to 120 minutes in intervals of 15 minutes (Bruker D8 Discover).

For the preparation of the alkali-activated binder, the GWM powder was calcined at 650°C for 1 h. The alkaline activating solution of 8M NaOH was prepared by mixing weighted proportions of NaOH pellets of 99% purity and distilled water in a laboratory mortar mixer. Mixtures with liquid/solid ratios of 0.30, 0.35, 0.40, 0.45, 0.50 and 0.55 were prepared to assess the workability as well as the influence of water content to the performance of the geopolymer binder. After mixture, the binder is put in prism moulds (40x40x160 mm^3) and stored in a ventilated oven at 50°C for 6 days, respectively 49 days for the second compression test. After demoulding, the specimens were stored for 24 h, respectively 4 days for the second set, at RT in plastic foil until the compression strength test.

Results

X-ray powder diffraction (XRD) analysis

The mineral composition of the material has been obtained from a quantitative analysis of the X-ray diffraction pattern data of calcined GWM powder samples using the Rietveld refinement technique. Figure 1 shows the quantitative analysis of the mineral composition of uncalcined GWM and calcined GWM at 750°C for different calcination times. The phase identification of the XRD pattern of the raw and calcined GWM detected the presence of quartz, muscovite, illite, kaolinite, hematite and potassium aluminium silicate. Furthermore, compared to uncalcined GWM, with increasing calcination time, the amount of quartz, muscovite and kaolinite phases

decreases, the content of hematite remains constant and the portion of illite, potassium aluminium silicate increases. The amorphous portion is higher for the calcined GWM compared to the uncalcined GWM, but with increasing calcination time its content is decreasing with the development of other phases. Similar evolutions of the mineral phases were observed for the calcination temperatures of 550°C and 650°C.

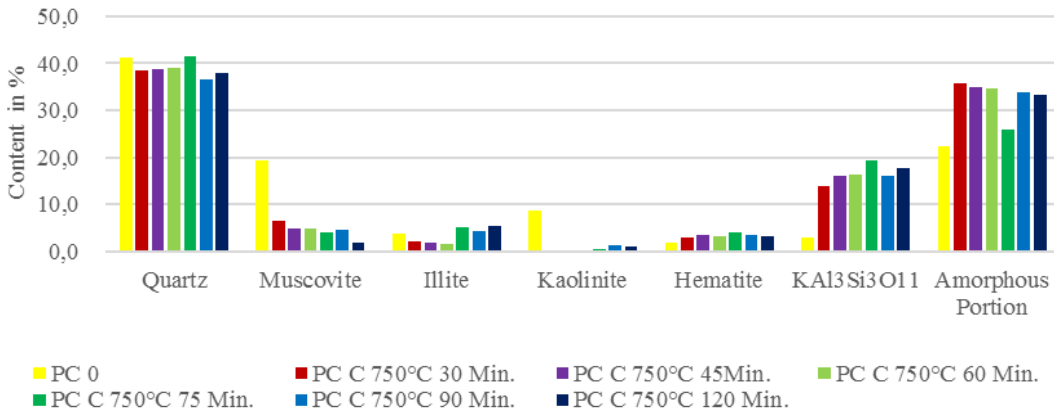

Figure 1: XRD - Mineral composition of uncalcined GWM and calcined GWM at 750°C for different calcination times

The quantitative mineralogical analysis of calcined GWM at different temperatures after calcination time of 60 min and 120 min showed a decrease of muscovite, illite and kaolinite phases with increasing calcination temperature. Moreover, higher thermal treatment result in an increase of hematite, potassium aluminium silicate and amorphous portion contents. However, it has been observed that the calcination process of the clay minerals is completed after a calcination time of 60 min. Moreover, it has to be remarked that further XRD analysis have to be performed for the geopolymer binder after alkaline reaction to identify new mineral phases due to geopolymerisation and analyse the changes compared to the unreacted material.

Compression strength tests

The results of the compression test after 7 days and after 53 days in relation with the different liquid/solid ratios are plotted in Figure 2. For the compression test after 7 days, the prisms with a liquid/solid ratio of 0.50 and 0.55 did not build a strong compound and therefore broke during the demoulding process. The hardened geopolymers reached low strength for the analysed mixture with a maximal value of 0.96 MPa. However, this experiment allowed to assess the workability of the mixture regarding the liquid/solid ratio. It was observed that the optimum L/S ratio in terms of workability amounts between 0.35 and 0.40. For the compression test after 53 days, all prisms remained unbroken after demoulding and in general performed better compared to the prisms compressed after 7 days. However, the achieved strengths are low, but better than those from the 7-days compression test with a maximal value of 2.76 MPa. However, it can be considered that longer curing times under thermal

treatment have a positive effect on the strength of the geopolymer binder made of calcined GWM powder.

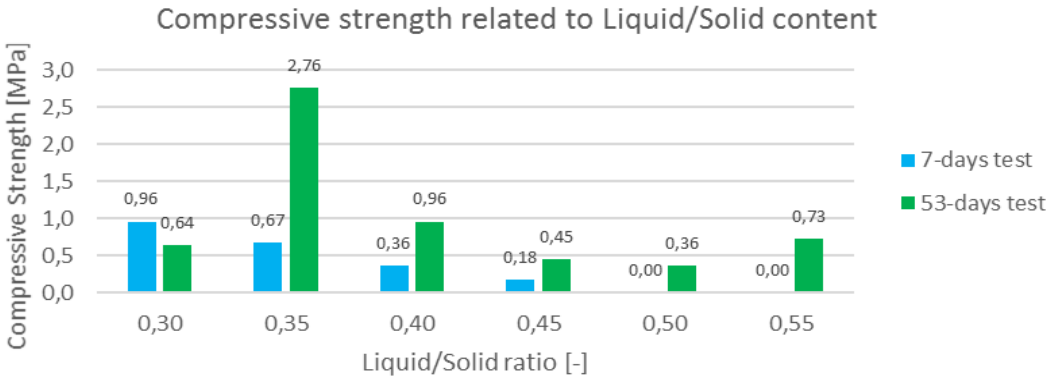

Figure 2: Compressive strength related to liquid/solid content - 7 days test

Summary and Outlook

GWM powder is used as raw material for making a geopolymer. The mineralogical analysis shows presence of kaolinite, which is beneficial for the strengthening of the geopolymer. A liquid/solid ratio between 0.35 and 0.40 provides the best workability of the geopolymer paste. Furthermore, higher curing times with thermal treatment lead to higher strengths. However, only low strengths were achieved with current mixtures. Therefore, further studies have to be carried out in order to optimise the process of producing geopolymer binders and to improve the strength of geopolymer binder made of calcined GWM powder. In further investigations, two paths need to be analysed: The influence of higher concentrations of alkaline solution on the strength of geopolymer binder and the development of strength by substitution of portions of cement with calcined GWM powder.

References

1. J. Davidovits, “Geopolymers: Inorganic polymeric new materials”, *J Therm Anal*, **37** (8) 1633-1656 (1991).
2. C. Ferone, F. Colangelo, G. Roviello, D. Asprone, C. Menna, A. Balsamo, A. Prota, R. Cioffi and G. Manfredi, “Application-oriented chemical optimization of a metakaolin based geopolymer”, *Materials*, **6** (5) 1920-1939 (2013).
3. F. Slaty, H. Khoury, J. Wastiels and H. Rahier, “Characterization of alkali activated kaolinitic clay”, *Appl Clay Sci*, **75** 120-125 (2013).
4. M. Schmücker and K. J. MacKenzie, “Microstructure of sodium polysialate siloxo geopolymer”, *Cer Int*, **31** (3) 433-437 (2005).
5. P. Duxson, G. C. Lukey and J. S. J. van Deventer, “Physical evolution of Na-geopolymer derived from metakaolin up to 1000°C”, *J Mater Sci*, **42** (9) 3044-3054 (2007).