Article

Development of Geopolymers Formulations Based on Low-Grade Kaolins as a Potential Construction Material

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Abstract: Sustainability, particularly in construction materials, has been a subject of growing interest. Civil construction is one of the industries where more materials are consumed, which leads to high energy consumption and CO₂ emissions. The production of cement, especially clinker is largely responsible for these problems. As a solution, new materials emerge, which do not require much energy for their production, which are the alkaline cements, specifically the geopolymers. Geopolymers are inorganic polymers obtained by the alkaline activation of aluminosilicate precursors. In the present study geopolymers were developed with low grade kaolin (as a precursor) from a Portuguese company. The development of these geopolymers will be, due to their properties, a good solution for rehabilitation of earth buildings, especially in adobe. The development of these geopolymers is also a contribution to the sustainability of kaolin exploitations as it opens new markets for the low grade kaolins, presently not easily commercialized. As mechanical strength of adobe materials ranges in literature from 0.6 to 8.3 MPa, the values obtained for the developed geopolymers (between ~2 to 10 MPa) can be considered as totally adequate.

Keywords: geopolymers; kaolins; diatomite; applications

1. Introduction

In construction industry, the growing concerns about the exhausting natural resources and consequent energy consumption are increasing during the last decade, mainly due to the production of cement that, in addition to consuming different materials, entails high energy expenditure with a release of CO₂ corresponding to at least 5 to 7% of global emissions [1]. In order to minimize these effects, other materials arise trying to substitute OPC cements and also to fill shortages in supply. Previous investigations for new more sustainable building materials aiming cement substitution by geomaterials submitted to alkaline activation or geopolymers [2].

A geopolymer is an inorganic polymer obtained by the alkaline activation of an aluminosilicate under certain temperature and pressure conditions [3]. According to several authors [2, 4, 5, 6], the geopolymers can be a great solution to use as construction material, especially in earth construction to rehabilitation and restoration works.

Their properties depend mainly on the chemical structure. In addition to replacing the cements, these materials can also be applied to other products, such as composites with fire-resistant fibers, anti-fire protection, in the ceramic industry (bricks and others), encapsulation of toxic and radioactive waste, among others [7].

The main advance is that sludge can be also used in the composition with fly ash [4]. In general, all aluminosilicates in which the atomic ratio Si: Al varies between 1 and 3 are considered the best fit

[26].The geopolymers can be produced from naturally high-strength natural phyllosilicates cured at room temperature; thus, they have good physic and chemical characteristics, such as: high workability, quick hardening, surface finishing, rapid development of mechanical strength, surface hardness, good resistance to chemical attack, heat and fire [3].

As interest in these products increases, research is starting in Portugal aiming to produce geopolymers from abundant Portuguese low-grade kaolin deposits.

2. Materials and Methods

Geopolymers were prepared using two Portuguese low grade kaolins (MIB-A and MIB-C), provided by a Portuguese company (Mibal – Minas de Barqueiros SA; location at Figure 1) with a grain size less than 30 μm.

Figure 1. Kaolin deposits location, and geological map 1:50 000 adapted, 1-C sheet - Barcelos [8].

The main constituent of kaolin is kaolinite ($Al_2Si_2O_5(OH)_4$), which undergoes successive structural and microstructural transformations during its firing [9]. The decomposition of kaolinite in metakaolinite occurs between 400 and 630°C. The calcination (up to 400°C) breaks down the structure of kaolin (alumina and silica layers become puckered and disorder) resulting in a metakaolin. Metakaolin is a highly reactive transition phase, an amorphous material with pozzolanic and latent hydraulic reactivity, suitable for use in cementing applications [10]. Both kaolins were used in their natural state and calcined at temperatures of 300, 500 and 750°C during 4 hours, to evaluate the effect of the temperature calcination on final products (Figure 2).

Figure 2. Portuguese kaolins MIB-C A and MIB-A, as natural and in different calcination temperatures.

Hydrated sodium silicate (Merck, Germany; 8.5 wt.% Na₂O, 28.5wt.%, SiO₂, 63wt.%, %H₂O) and sodium hydroxide (6M) (ACS AR Analytical Reagent Grade Pellets) were used as alkaline activators. Deionized water was the reaction medium.

The following 8 compositions (Figure 3) were prepared: GeoMIB-A and C (natural kaolin), GeoMIB-A and C 300 (kaolin calcined at 300°C), GeoMIB-A and C 500 (kaolin calcined at 500°C) and GeoMIB-A and C 750 (kaolin calcined at 750°C).

Figure 3. Geopolymer performed specimens.

The mixing of the blends was carried out by a laboratory stirrer (200 rpm for 2 min) to ensure their homogeneity and avoid bubbles and agglomeration into the sample. The pastes were immediately placed in cubical molds (20mm x20mm x 20mm) and put on oven for 24h. During the curing time, the geopolymers specimens were stored in sealed plastic bags at room temperature [11]. The assays were performed in 3 specimens of each formulation at 7, 14 and 28 days of cure.

Chemical composition of natural and calcined kaolins was assessed by X-ray fluorescence (XRF) using an XRF Panalytical AXIOS PW4400/40. Loss-on-Ignition (LOI) was determined by heating 1 g of the sample at 1000°C for 1 h in furnace.

Mineralogical composition was determined both on un-oriented (random) powder mounts for < 30 µm fraction analyses and on oriented aggregates for the clay fraction ones, for kaolin, and in the bulk samples of geopolymers specimens. The clay fractions were obtained by sedimentation according to Stokes law, using 1% sodium hexametaphosphate solution to avoid flocculation. For the preparation of preferentially oriented clay mounts, the suspension was placed on a thin glass plate and air-dried. XRD measurements were performed using Philips PW 3050 and X' Pert PW 3040/60 equipment using Cu Kα radiation. Scans were run between 2° and 60° (unoriented powder mounts) and between 2° and 20° 2θ (oriented clay mounts) in air-dry state, after a previous glycerol saturation, and heat treatment (500°C). Qualitative mineralogical analyses followed the criteria recommended by [12, 13, 14]. The degree of crystallinity was estimated from XRD determination of the Hinckley crystallinity index. The Hinckley crystallinity index consists in the ratio of the height above background of the (110) and (111) peaks above the band of overlapping peaks occurring between 20 to 23° 2θ compared to the total height of the (110) above background [15].

Microstructural characterization was carried out by scanning electron microscopy (SEM – Hitachi, SU 70) and energy dispersive X-ray spectrometry (EDS – EDAX with detector Bruker AXS, software: Quantax) operated at 3–30 kV.

Thermogravimetric analysis (TGA) and differential thermal analysis (DTA) were carried out as simultaneous thermal analysis (STA) on a Setaram TGA 92 balance, under argon between ~20°C and 1000°C, with a heating rate of 10°C min⁻¹.

The cubical specimens were submitted to compressive strength tests, using a universal mechanical compression testing machine (Shimadzu Autograph AG 25 TA). The testing procedure was carried out with a maximum force of 5 kN at the speed of 50 N/s as per the standard EN 1015-11 [16].

Water absorption of the immersed specimens was calculated for each series according to Eq. (1), where W% is the adsorption of water expressed as percentage; Ww and Wd are the weights of the immersed specimen and of specimen after drying (g), respectively [11, 17].

$$W \% = \frac{W_w - W_d}{W_d} * 100$$

(1)

3. Results and Discussion

3.1. Kaolin characterization

The textural behavior of both kaolins is presented in Table 1. Through these results it's verified that MIB-C kaolin is finer, which means that it has better reactivity. However, both kaolins are very fine, presenting more than 85% of particles less than 25µm.

Table 1. Texture of MIB-A and MIB-C kaolins.

	Texture (µm)									
	<100	<75	<50	<25	<10	<5	<2	<1.5	<1	<0.5
MIB-A	98.9%	98.8%	97.1%	84.1%	70.9%	51.5%	44.1%	33.2%	13.7%	3.4%
MIB-C	98.2%	98.7%	96.1%	85.8%	75.9%	68.8%	55.7%	48.5%	32.4%	7.7%

XRD patterns of studied kaolin samples, natural or after calcination, are presented in Figures 4 and 5.

Figure 4. XRD patterns of MIB-A samples (1 – quartz, 2 – kaolinite, 3 – illite).

Figure 5. XRD patterns of MIB-C samples (1 – quartz, 2 – kaolinite, 3 – illite, 4 - gibbsite).

Both kaolins present similar mineralogical composition, being essentially composed by quartz and kaolinite, with illite in smaller amounts. Above 500°C, kaolinite was destructed and a broad reflection centered at $2\theta = 20^\circ$ to 25° attributable to the amorphous metakaolinite appeared. MIB-C kaolin shows the presence of gibbsite in small amounts.

XRD patterns of fractions $<2\ \mu\text{m}$ show that Hinckley crystallinity index of kaolinite increases with calcination at 300°C; at 500 and 750°C calcination temperatures these kaolins lose their crystallinity (Figures 6 and 7), becoming amorphous.

Figure 6. XRD patterns of MIB-A samples (fraction $<2\ \mu\text{m}$).

Figure 7. XRD patterns of MIB-C samples (fraction <2μm).

Thermal behavior of kaolin samples was assessed by DTA/TG. Both samples MIB-A and MIB-C have a similar thermal behavior, showing no significant differences between them (Figure 8).

MIB-A

MIB-C

Figure 8. ATG/TG of kaolins MIB-A and MIB-C.

DTA/TG curves of both kaolins show the presence of two endothermic peaks. The first one is less intense located around 80°C, reflecting the losing of the surface water. The second one appeared around 560°C, being related to the losing of structural water (dehydration of kaolin to form

metakaolin) and associated to a mass loss of (close to 13% for both kaolins), between 450°C and 600°C. Furthermore, DTA curves of MIB-C present an endothermic peak located between 280°C and 300°C associated to a mass loss around 1%; corresponding to the dehydroxylation of gibbsite and formation of alumina [18].

Chemical composition (major elements) of kaolins used on geopolymers design is presented on Table 2.

Table 2. Chemical analyses of studied kaolins.

Sample	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	Na ₂ O	K ₂ O	TiO ₂	LOI (%)
MIB-A	47.34	35.81	2.11	0.13	0.26	0.06	0.89	0.50	12.51
MIB-A300	47.52	35.21	2.13	0.05	0.27	0.06	0.94	0.50	13.05
MIB-A500	52.10	40.39	2.39	0.08	0.29	0.07	1.01	0.55	2.81
MIB-A750	53.21	41.08	2.39	0.06	0.31	0.07	0.96	0.56	0.94
MIB-C	44.92	36.11	3.42	0.03	0.24	0.06	0.90	0.47	13.54
MIB-C300	45.90	36.71	3.45	0.03	0.26	0.06	0.96	0.49	11.85
MIB-C500	50.02	41.00	3.84	0.03	0.28	0.07	0.97	0.54	2.88
MIB-C750	50.43	42.15	4.02	0.05	0.28	0.06	1.03	0.57	1.01

On kaolin calcined at low temperatures (300°C) Al₂O₃ content is maintained, whereas on kaolins calcined at higher temperatures (500°C and 750°C) the aluminum content increases slightly due to the changes on the structure of these materials. Other important aspect to mention is the loss-on-ignition (LOI), decreasing with temperature, due to the dihydroxylation related to the transition from crystalline to amorphous state. This aspect is very important, showing that with the increases in temperature we can obtain a more resistant material, in particular, to fire.

3.2. Geopolymers characterization

3.2.1. Si:Al, Na/Al, Na:Si and water:Si ratios

The Si:Al ratio determines the degree of polymerization [19] and was computed according to the chemical analysis of precursors and the alkaline solutions used (Table 3).

Table 3. Computed ratios of geopolymers produced formulations.

Geopolymers	SiO ₂ :Al ₂ O ₃	Na ₂ O:Al ₂ O ₃	Na ₂ O:SiO ₂	H ₂ O:SiO ₂
GEOMIB-A	1.63	0.35	0.22	1.31
GEOMIB-A300	1.66	0.36	0.22	1.31
GEOMIB-A500	1.56	0.31	0.20	1.21
GEOMIB-A750	1.56	0.31	0.20	1.19
GEOMIB-C	1.55	0.35	0.23	1.37
GEOMIB-C300	1.55	0.34	0.22	1.34
GEOMIB-C500	1.49	0.31	0.21	1.25
GEOMIB-C750	1.46	0.30	0.21	1.24

According to [20], higher mechanical forces are obtained for higher SiO₂: Al₂O₃ ratios, and higher porosities for lower SiO₂: Al₂O₃ ratios; they also found that for ratios between 1.4 and 1.65 there is a change of structure, changing from a porous structure to a more homogeneous one. The same authors also conclude that the porosity increases on higher H₂O: SiO₂ ratios. Accordingly, the formulations

are all within the range between 1.4 to 1.65. Developed geopolymers have very similar chemical compositions and as such, the differences in values between the results of either mechanical strength or water absorption are mainly attributed to the presence of amorphous material.

3.2.2. Mineralogical composition and SEM analysis

Mineralogical characterization of the studied geopolymers was carried out on the specimens after 28 days of cure. XRD patterns are closely similar to the used kaolins. However, a slight shift in the amorphous phase broad band is observed. The amorphous material undergoes a clear evolution from 2θ between 20° to 27° (Figures 9, 10 and 11), at the raw material, to 23° to 30° on geopolymers [21]; these changes allow us to conclude that the polymerization process is occurring in all geopolymers developed.

Figure 9. XRD patterns of geopolymer GEOMIB-A and kaolin MIB-A (1 – quartz, 2 – kaolinite, 3 – illite).

Figure 10. XRD patterns of geopolymer GEOMIB-A 500°C and kaolin calcined at 500°C (1 – quartz, 2 – kaolinite, 3 – illite, 4 amorphous phases).

Figure 11. XRD patterns of geopolymer GEOMIB-A and kaolin MIB-A (1 – quartz, 2 – kaolinite, 3 – illite).

Produced geopolymers are composed essentially by quartz, kaolinite, illite and metakaolinite (Figure 12).

Figure 12. XRD patterns of all geopolymeric formulations (1 – quartz, 2 – kaolinite, 3 – illite).

Figure 13 shows SEM images for geopolymers made with kaolins calcined at higher temperatures, displaying the amorphous structure obtained after 28 days of curing time.

Figure 13. SEM images of GEOMIB-A 500°C (left) and GEOMIB-C 750°C (right) formulations.

3.2.3. Compressive strength

The compressive strength studied for all geopolymers after different curing times is presented in Figure 14.

Figure 14. Compressive strength of geopolymers at different curing times (7, 14 and 28 days).

The GEOMIB-C 300°C geopolymers presents higher values in term of mechanical resistance(~10MPa). This result can be related with the presence of gibbsite and its dihydroxylation.

When kaolins A and C were calcined at higher temperatures, the compressive strength decreased considerably in comparison of raw kaolin and calcined at 300°C. Moreover, it is observed that with larger curing period the strength decreased. This means that the consolidated material is losing its cohesion in function of time. For the GEOMIB-A and GEOMIB-A 300°C, the opposite behavior it is verified as mechanical strength increases during the curing time. The decrease of mechanical strength on raw materials calcined at higher temperatures means that the reactivity of kaolins decreased with calcination. Thus, the desired pozzolanic effect, which confers resistance to geopolymers, is compromised. This may be related to the crystallinity of the original kaolin. According to [22], the degree of crystallinity of the kaolinite precursor can affect pozzolanic reactivity of metakaolinite. In this case, the pozzolanic effect of kaolinite could improve the mechanical strength of these consolidated materials, instead of metakaolinite [23].

The low Na/Al ratios (Table 1) are compromising the mechanical strength of the geopolymers. According to [24], Na/Al ratios <1 mean that the alkaline activator used was insufficient. This means that higher concentrations of NaOH solution should be used. These authors also report that Na/Al ratios <1.2 can also lead to a decrease in mechanical resistance. The best ratios Na / Al should be between 1 and 1.2.

3.2.4. Water absorption

The results obtained in the compressive strength are reflected in the water absorption capacity analysis results (Figure 15).

Figure 15. Water absorption (%) of geopolymers in different curing times (7, 14 and 28 days).

The lower the value of the mechanical resistance higher is the absorption. It means that in the case of better cohesion of the consolidated products, the porosity decrease. Also, the materials obtained with raw and calcined at 300°C kaolins presented values around 5 to 10%, 3 to 4 times lower than the other ones. Geopolymers with kaolin without calcination and with calcination at 300°C exhibit similar behavior; after around 20 minutes they are saturated (Figure 16).

Figure 16. Water absorption (%) of geopolymers with kaolins without temperature and calcined at 300°C.

4. Conclusions

Mineralogically, the geopolymers are essentially composed by quartz, kaolinite, and aluminum and silicon rich amorphous material. The increase of amorphous material in all geopolymers comparing to precursors reveals that polymerization is occurring.

The mechanical strength shows that natural kaolins (MIB-A and MIB-C) produce geopolymers with good results for compressive strength. These results are related to the reduced particle size of the kaolins, and the reduction of the reactivity of the calcination at higher temperatures. The development of these geopolymers is also a contribution to the sustainability of kaolin exploitations as it opens new markets for the low grade kaolins, presently not being easily commercialized.

According to [2], after studying several adobe construction buildings it was found that there is a need to develop sustainable materials. The developed materials need to be compatible with the old adobe materials to carry out intervention, conservation and rehabilitation works. The development of these geopolymers will be, due to their properties, a good solution for rehabilitation of earth buildings, especially in adobe. The better geopolymers developed for these propose are: GEOMIB-A, GEOMIB-C, GEOMIB-A300°C, GEOMIB-C300°C and GEOMIB-A750°C; taking into account that the mechanical strength of adobe materials ranges in literature from 0.6 to 8.3 MPa [25], the values obtained for these geopolymers (between ~2 to 10 MPa) can be considered as totally adequate. However, it is suggested that preference must be given to the geopolymers not demanding heating in the process, in order to obtain a more sustainable product, that is, more environmentally friendly, as it will cause much less CO₂ emissions.

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