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Thermal behavior and characteristics of fired geopolymers produced from local Cameroonian metakaolin

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Abstract

The thermal behavior along with certain characteristics of geopolymers produced from local Cameroonian metakaolin and heated up to 1000 °C were examined. Geopolymers fired up to 900 °C had the same physical aspect as initial ones and those fired at 1000 °C warped, were glazed and blistered. The TG showed elimination of water according to two stages. The dilatometric curves of preheated samples showed shrinkage between 90 and 250 °C followed by expansion and sintering. The samples heated up to 700 °C were amorphous and new crystalline phases appeared around 900 °C. The microstructure of geopolymers heated between 300 and 900 °C showed progressive disruption and the linear shrinkage increased. The water absorption of the samples fired up to 700 °C increased slightly and tremendously around 900 °C. A drastic decrease of compressive strength was observed with the samples fired between 300 and 900 °C. Hence, the characteristics of geopolymers lessened with elimination of the water which forms hydration spheres around the compensating cations (Na^+) opposed to tetrahedral groups AlO_4^- along with transformation of amorphous phase.

Keywords: Metakaolin; Geopolymers; Characteristics; Thermal behavior; Amorphous phase

1. Introduction

Geopolymers are materials that consist of polymeric silicon–oxygen–aluminum framework with silicon and aluminum tetrahedral alternately linked together in three directions by sharing all the oxygen atoms. The negative charge of aluminum in IV-fold coordination is compensated by cations such as Na^+, K^+ [1]. These materials can be considered as being made up of non-crystalline network which is close to that of zeolites [2]. According to certain authors [3,4] geopolymers are generally endowed with thermal stability which depends on characteristics such as the charge of compensating cation, the ratio of Si/Al and the phases which they are found [5,6]. When geopolymers are heated at high temperature, there are certain modifications such as change of dimension due to thermal shrinkage and sintering, formation of new T–O–T bonds or phases whose thermal stability is more or less important. Several studies on heating of metakaolin-based geopolymers, fly ash-based-geopolymers or certain by-products-based geopolymers at high temperatures have been carried out including determination of new crystalline phases [4,7,8], change of physical and mechanical properties [6,9–11], modification of microstructure [12,13], etc. It has been observed that the highest heating temperature experimented is more or less variable and depends on the nature of the raw aluminosilicate material and composition of alkaline medium used for the synthesis of geopolymers [14]. As for metakaolin-based geopolymers, serious damage is noticeable around 800–850 °C [8,14].

The objective of this work is to investigate the thermal behavior and to determine certain chararacteristics of fired products of geopolymers produced by using local Cameroonian metakaolin as aluminosilicate. This study focuses particularly on the fate of water and amorphous phase of geopolymers under elevated temperatures. With this in mind, the samples were exposed between ambient temperature and

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1000 °C. The resulted products were characterized via visual appearance, thermal analysis (DSC, TG and dilatometry), X-ray diffractometry, Fourier transform infrared spectrometry, scanning electron microscopy as well as determination of certain physical and mechanical properties (thermal shrinkage, water absorption and compressive strength).

2. Materials and methods

2.1. Materials

The metakaolin was obtained from kaolin provided by the group Nubru Holding which is involved in the valorization of certain local raw materials in Cameroon. Before to be used, the kaolin was enriched with kaolinite via the sedimentometry process based on Stockes′ law [15]. The clay fraction obtained was dried at 105 °C until its weight became constant then it was ground and sifted through a sieve of mesh 80 µm followed by calcination at 700 °C [16] for 10 h at a heating rate of 5 °C/min in a programmable electric furnace (Nabertherm, Mod. LH 60/14). The diffractograms of both the clay fraction and the metakaolin are given in Fig. 1. The chemical composition (%wt) of the metakaolin was as follows: SiO_2 (51.46), Al_2O_3 (41.71), Fe_2O_3 (2.66), CaO (0.10), Na_2O (0.39), K_2O (0.70), TiO_2 (0.95) and LOI, 2.03. The specific surface area of metakaolin determined according to BET method was $20.88 \pm 0.06\ m^2/g$ and the particle size distribution was as follows: $d_{10}=5.65\ \mu m$, $d_{50}=30.75\ \mu m$ and $d_{90}=70.01\ \mu m$. The alkaline solution was obtained via mixing of a solution of sodium hydroxide (12 M) and sodium silicate made up of 28.7 wt% of SiO_2, 8.9 wt% of Na_2O, 62.4 wt% of H_2O and SiO_2/Na_2O molar ratio of 3.3.

2.2. Paste samples preparation and analytical techniques

To get the geopolymer paste samples, metakaolin powder and alkaline solution were mixed according to the following molar ratios: $SiO_2/Al_2O_3=3.4$, $Na_2O/Al_2O_3=0.8$ and $H_2O/Na_2O=15.1$. The mixture was homogenized in a Hobart mixer (M & O, model N50-G) for 10 min and the fresh paste poured in PVC molds. Once molded, the pastes were vibrated for 10 min on an electrical vibrating table (M & O, type 202, N°106) to remove entrapped air bubbles. Three types of samples were made i.e. large cylinders (diameter: 30 mm and height: 60 mm), small cylinders (diameter: 18 mm and height: 20 mm) and parallelepipeds ($7 \times 7 \times 65\ mm^3$) and were covered with polyethylene film and stored at room temperature (25 ± 3 °C) for 24 h before demoulding. The large cylinder samples were used for visual observation and determination of thermal shrinkage and compressive strength. The water absorption and scanning electron microscopy were performed using the small cylinder samples while the dilatometric analysis was done on the parallelepiped ones. After 28 days of storage at ambient temperature (25 ± 3 °C) of the laboratory, the samples were either cured at 90 °C (24 h in an oven) or heated in a programmable electric furnace (Nabertherm, Mod. LH 60/14) at heating rate of 5 °C/min and soaking time of 2 h at the following temperatures: 300, 500, 700, 900 and 1000 °C. The determination of thermal shrinkage was performed using a vernier calliper and the compressive strength with the help of a hydro-electric press (M & O, type 11.50, N°21) which operates at an average rate of 3 mm/min. The determination of water absorption was carried out according to the ASTM C 373-88 standard [17]. The thermal analysis (DSC and TG) were done using a NETZSCH STA-429 at heating rate of 20 °C/min up to 1200 °C in air. The dilatometric analysis was performed with a mechanic dilatometer (Adamel–Lomargy, model DM-15). Fourier transform infrared spectroscopy was carried out on powders of geopolymer using a Bruker Alpha-P in absorbance mode (interval of wave number ranging was 4000–400 cm^{-1}). Scanning Electron Microscopy (SEM) was carried out using a Philip XL 30 EM. The crystalline phases were determined using X-ray diffractometry with the aid of a Philip PW 3050/60 diffractometer, operating by reflexion of $K_{\alpha 1}$ radiation of Cu.

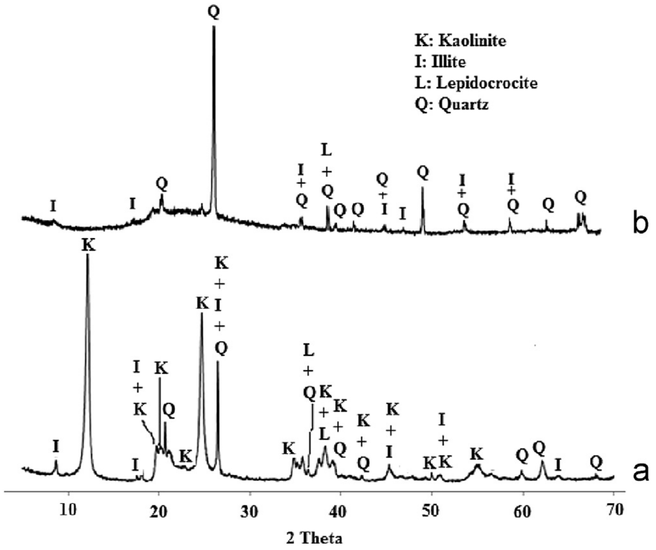

Fig. 1. X-ray diffractograms of kaolin (a) and metakaolin (b).

3. Results and discussion

3.1. Appearance, phase composition and microstructure before and after thermal treatment of geopolymers

After demoulding, the geopolymer cylinders maintained at room temperature, at 90 °C or heated at 900 °C had the same physical appearance (Fig. 2a). On the contrary, large and small cylinder geopolymers heated at 1000 °C warped and were glazed, blistered and presented cracks (Fig. 2b). FTIR spectra (Fig. 3) showed that unlike the metakaolin and the geopolymer heated at 900 °C, the samples cured at room temperature or heated at 90, 300 and 500 °C showed absorption bands at 3400–3450 cm^{-1} and at 1640–1650 cm^{-1} and their intensities decreased with increase in temperature. These bands correspond respectively to deformation vibration of H–O–H bonds and elongation of O–H groups of molecules of water [18–21]. Hence, though heated at 500 °C, not all the water involved in