Construction and Building Materials 24 (2010) 1176–1183

Contents lists available at ScienceDirect

Construction and Building Materials

journal homepage: www.elsevier.com/locate/conbuildmat

Effect of curing temperature on the development of hard structure of metakaolin-based geopolymer

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ARTICLE INFO

Article history:
Received 8 June 2009
Received in revised form 11 December 2009
Accepted 16 December 2009
Available online 15 January 2010

Keywords:
Geopolymer
Metakaolin
Mechanical properties
Curing temperature
Porosimetry

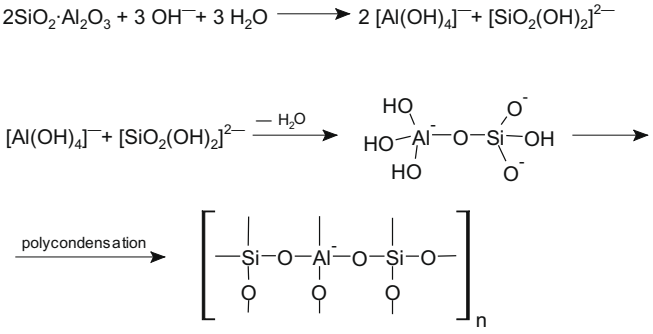
ABSTRACT

The properties of metakaolin-based geopolymer are directly impacted not only by the specific surface and composition of initial metakaolin and the type, composition and relative amount of alkali activator used but they also depend on the conditions during the initial period of geopolymerization reaction. This study aimed to analyze the effect of curing temperature (10, 20, 40, 60 and 80 °C) and time on the compressive and flexural strengths, pore distribution and microstructure of alkali activated metakaolin material. The results have shown that the treatment of fresh mixture at elevated temperatures accelerates the strengths development but the 28 days mechanical properties are deteriorated in comparison with results obtained for mixtures that were treated at an ambient or slightly decreased temperature. The influence of curing temperature on microstructure of geopolymer matrix was verified in terms of pore distribution studied by means of mercury intrusion porosimetry. The study revealed a tendency to increase pore size and cumulative pore volume with rising temperature, which is reflected in mechanical properties. It has been also shown the possibility of monitoring the geopolymerization reaction by means of Infrared Spectroscopy.

1. Introduction

Since the late seventies of the 20th century geopolymers have been considered for replacing traditional structural materials by reason of their excellent properties and high performance [1,2]. During the last decade, increased research efforts have been directed to this area due to the wide range of potential applications of these materials besides civil engineering in many other branches of industry. Geopolymers belong to the group of strong and durable cementitious materials that harden at temperatures below 100 °C [3–5]. It is three-dimensional CaO-free aluminosilicate binder, which is usually prepared by alkali activation of metakaolin or other inorganic material having pozzolanic properties such as fly ash and some of the aluminosilicate-based natural minerals [6–8]. The reason for using mainly metakaolin to produce geopolymers could be the fact that it is common industrial mineral which can be obtained in a large quantity with homogeneous properties. Metakaolin is also environmentally friendly compared to Portland cement: its production requires much lower calcining temperature and emits 80–90% less CO_2 than Portland cement [9].

Alkali activation of metakaolin can be performed by solution of alkali hydroxide [10] or by alkali salt that gives after hydrolysis strongly basic solution, e.g. alkali silicate [11]. The process comprises dissolution of primary aluminosilicate framework from metakaolin followed by condensation of free silicate and aluminate species to form three-dimensional structure. This structure consists of cross-linked SiO_4 and AlO_4^- tetrahedra where the negative charge on Al^{3+} in IV-fold coordination is balanced with positive charge of alkali ion (Na^+, K^+) [9]. The geopolymerization reaction can be expressed according to the following scheme:



This structure is very closed to the structure of zeolites but without regular ordering to longer distance – it has amorphous character. Long range ordering of tetrahedra is affected by temperature at which the polycondensation reaction occurs. During long term hydrothermal conditions at temperatures above 85 °C the

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doi:10.1016/j.conbuildmat.2009.12.023

formation of crystalline structure is preferred [3,12,13]. The reaction between metakaolin and alkaline solution has been studied by several analytical methods, among others by calorimetric measurements [14,15], thermal analysis (DTA, TGA and DSC) [16–18], FTIR and NMR [19–22] and X-ray diffraction analysis [13,14,23]. It has been proved that metakaolin-based geopolymer is highly amorphous material and the only crystalline phases that can be found in XRD patterns were assigned to quartz traces that were already present in metakaolin precursor.

The mechanical properties and microstructure of geopolymer strongly depends on the initial Si/Al ratio. Better strength properties have been reported for mixtures with SiO_2/Al_2O_3 ratios in the range of 3.0–3.8 with an Na_2O/Al_2O_3 ratio of about one that can be easily achieved when sodium silicate with appropriate SiO_2/Na_2O ratio is used. Changes in the SiO_2/Al_2O_3 ratio beyond this range usually result in low strength systems [24]. However, several results that have been published are not easy to compare because of different conditions, such as temperature, curing time, relative humidity, at which geopolymerization was carried out. Unfortunately, to date little is known about the influence of curing temperature on the mechanical properties and microstructure of geopolymer binder, which is supposed to be one of the important factors affecting the rate of formation and quality of the hard structure. Such information would be worth for instance in production of prefabricated elements made of composite that would use geopolymer as a binder.

This paper reports on the mechanical properties of metakaolin-based geopolymer mortars that were synthesized in different curing conditions concerning temperatures from 10 to 80 °C and different time of curing. The results of compressive and flexural strengths were explained from the viewpoint of microstructure changes that were determined by means of mercury intrusion porosimetry and FTIR spectroscopy during hardening process. The composition of the investigated geopolymer had been chosen on the basis of previous experience with several types of geopolymer materials prepared at an ambient temperature as the mixture having the best mechanical properties after 28 days.

Table 1
Chemical composition of starting materials.

Component (%)	Metakaolin	Alkaline silicate solution
Al_2O_3	40.94	–
SiO_2	55.01	24.9
K_2O	0.60	–
Na_2O	0.09	18.5
CaO	0.14	–
MgO	0.34	–
Fe_2O_3	0.55	–
TiO_2	0.55	–
LOI	1.54	–
H_2O	–	56.6

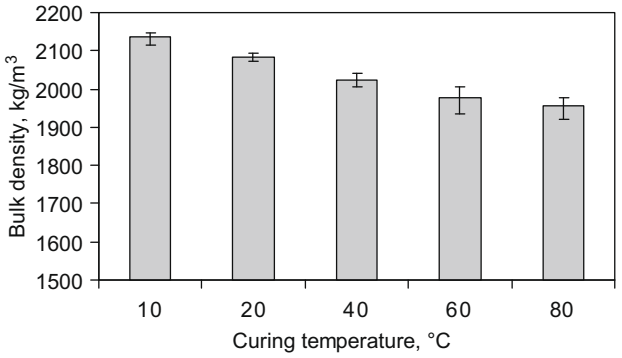

Fig. 1. Bulk density of geopolymer material prepared at different curing temperatures (10, 20, 40, 60 and 80 °C) at the age of 28 days. Heated mixtures were treated at elevated temperatures for initial 4 h. Error bars indicate the minimum and maximum value in the measurement series.

2. Experimental methods

2.1. Materials

Metakaolin was purchased from ČLUZ (CZ) under the brand name of Mefisto K05 that was produced by calcination of kaolin at 750 °C in rotary kiln. The molar composition of metakaolin determined by X-ray fluorescence is presented in Table 1. The Brunauer–Emmett–Teller (BET) surface area [25] of metakaolin determined by nitrogen absorption is 13.1 m^2/kg, and the mean particle size (d_{50}) is 4.82 μm.

Alkaline silicate solution with silicate modulus (SiO_2/Na_2O) 1.39 was prepared by dissolving of solid sodium hydroxide (Lachema, 98.0%) in commercial sodium water glass (Zaklady chemiczne Rudniki, S.A., $SiO_2/Na_2O = 3.26$ and $H_2O/Na_2O = 10.40$) until clear. Standard quartz sand was added as an aggregate for the preparation of geopolymer mortars that were used for testing of mechanical properties. The maximum grain size of quartz was 2.5 mm.

2.2. Sample preparation

Geopolymer samples were prepared by mechanically mixing metakaolin and activator solution in planetary mixer for 5 min. Then quartz sand was mixed into this geopolymer paste with some additional water in order to obtain better workability of the mortar. The geopolymer mortar composed of 450 g metakaolin, 372 g

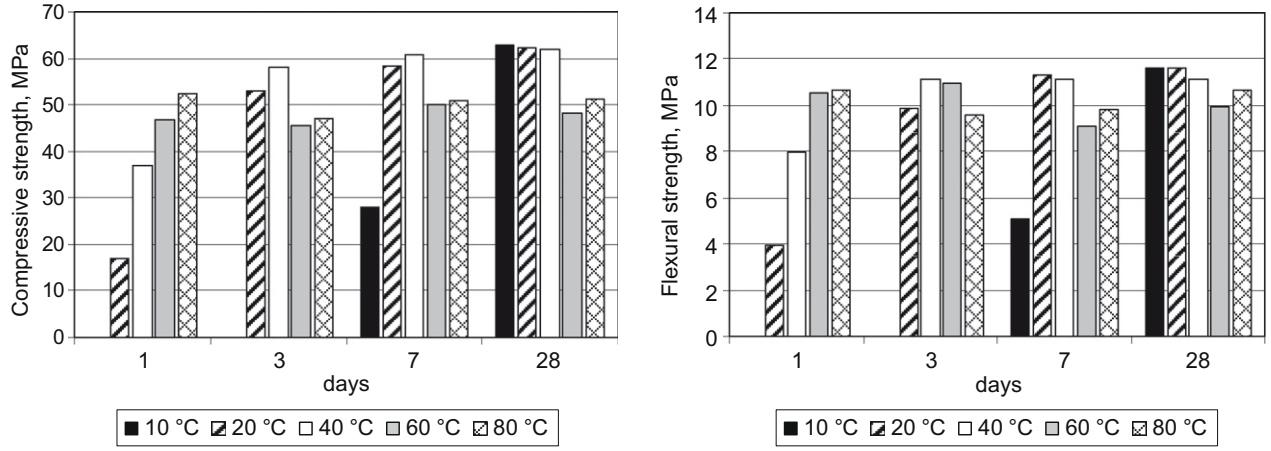

Fig. 2. Development of compressive and flexural strength of geopolymer material cured at 10, 20, 40, 60 and 80 °C over time (1, 3, 7, and 28 days). Curing at 40, 60 and 80 °C was carried out for initial 4 h.