Resilient Structures and Sustainable Construction
Edited by Pellicer, E., Adam, J. M., Yepes, V., Singh, A., and Yazdani, S.

ISBN: 978-0-9960437-4-8

MICROSTRUCTURE, TEXTURE, AND MECHANICAL PROPERTIES OF GEOPOLYMERS PREPARED USING INDUSTRIAL WASTE

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Geopolymers prepared by processing industrial waste present an environmental friendly alternative to the traditional binders which are characterized by very energy-demanding production. Due to an appropriate composition, fine-ground ceramic powder represents a good candidate for the production of geopolymers. In this paper, microstructure, texture, basic physical properties and compressive strength of three geopolymers with different silicate modulus (SM) are studied. Experimental results showed that the densest microstructure had the material with the lowest SM = 0.8, which also did not have any visible cracks. The open porosity and matrix density were found to increase with the increasing SM. The highest compressive strength was reached for the highest SM = 1.2. Having the compressive strength of ~38 MPa, such a geopolymer can be considered as comparable to cement based materials.

Keywords: Industrial waste, Alkali activation, Pore size distribution, Basic physical properties.

1 INTRODUCTION

The manufacturing process of clay masonry units usually faces a problem of extensive waste generation. Besides a high volume of scrapped ceramics, the total amount of waste is increased also by powder as a product of grinding of brick blocks to have a precise shape and dimensions. A utilization of coarse fragments of ceramics as aggregates in concrete is generally known, but also fine particles can be employed in concrete production because of their pozzolanic activity. The ceramic powder is, therefore, predisposed to be used as a supplementary cementitious material, having according to Gonçalves *et al.* (2009) or Gonzalez-Corominas and Etxeberria (2014), a positive influence on mechanical properties or durability of final products. Additionally, since the production of cement is very energy demanding, its partial substitution by waste materials may bring economical, as well as environmental benefits. This has been reported by several researchers before (Bektas *et al.* 2007, Lavat *et al.* 2009, Matias *et al.* 2014, O'Farrell *et al.* 2006).

However, utilization of waste ceramic powder may be even friendlier to the environment when used in geopolymers after alkali activation. On the other hand, due to technological

problems that might occur during the manufacture, only low attention has been paid to alkali activation of such a material so far, while metakaolin, fly ash or blast furnace slag have been rather preferred (Junaid *et al*. 2017). Nevertheless, a good potential of ceramic waste materials for alkali activation has been pointed out by Puertas *et al.* (2006), Zedan *et al*. (2015) or Rieg *et al.* (2013), in spite of the mentioned manufacture issues. That makes waste ceramic powder a good source of raw material for the geopolymer production.

In this paper, several geopolymer mixtures are prepared using waste ceramic powder as the main component and a combination of water glass and sodium hydroxide as the alkali activator. After hardening, the mixtures are subjected to detailed characterization including microstructural and textural studies using scanning electron microscopy and mercury intrusion porosimetry, respectively. Determining the basic physical and mechanical parameters, the functional characteristics of designed geopolymers are assessed as well. The main objective of the experimental work is to prepare such geopolymer mixtures which could be considered as good candidates for environmental friendly alternatives to cement-based materials.

2 MATERIALS

Three different mixtures were prepared, based on alkali activated ceramic powder, of which chemical composition is given in Table 1. It has been supplied by the Heluz company in the form of waste material from a production of perforated brick blocks. Having different silicate modulus calculated as the molar ratio of SiO_2/Na_2O, the dosage of components for mixtures preparation is summarized in Table 2. In order to reach the desired silicate modulus, the mass of sodium hydroxide was calculated, weighed and dissolved in water glass. The purpose of NaOH addition is to increase the alkalinity of the environment and to gain sodium cations, which have the vital stabilization function for the product structure. After the addition of water, the solution was mixed with the prepared ceramic powder.

Table 1. Chemical composition of ceramic powder.

Component	**SiO_2**	**Al_2O_3**	**Fe_2O_3**	**CaO**	**MgO**	**SO_3**	**Na_2O**	**K_2O**	**TiO_2**	**MnO**	**P_2O_5**
(% by mass)	66.400	13.390	4.660	3.660	1.670	1.090	0.920	2.440	0.700	0.073	0.130

Table 2. The dosage of components for the mixtures preparation.

	Ceramic powder (g)	**Water glass (g)**	**NaOH (g)**	**Water (ml)**	**Silicate modulus (-)**
K-120	200	70	15.57	50	0.8
K-121	200	70	9.50	50	1.0
K-122	200	70	5.50	50	1.2

3 EXPERIMENTAL METHODS

3.1 Study of Microstructure

Microscopic studies were performed using a scanning electron microscope (SEM) Zeiss Merlin, inspecting the samples without coating. The following conditions were adhered to: accelerating voltage (HT) 4 kV, probe current 17 pA – 20 pA and work distances between 5.9 mm and 3.9 mm.

3.2 Pore Size Distribution

Using the mercury intrusion porosimetry (MIP) the pore size distribution was determined. For this purpose, Pascal 140 and 440 devices (Thermo) were used. The contact angle of mercury and a sample was assumed 130°, the surface tension of mercury 480 mN m^{-1} and density of mercury 13.534 g cm^{-3}. The samples were dried before testing, having the size of ~1 mm and weight of ~1 g.

3.3 Basic Physical Properties

Basic physical properties were determined by helium pycnometry (Pycnomatic ATC, Thermo Scientific) and the conventional gravimetric method. Measuring the dimensions and mass of the samples, the bulk density ρ_b [kg m^{-3}] was found. The matrix density ρ_{mat} [kg m^{-3}] was determined using the helium pycnometer. The open porosity ψ_0 [%] was then calculated in Eq. (1).

$$\psi_0 = 100\left(1 - \frac{\rho_b}{\rho_{mat}}\right) \tag{1}$$

3.4 Mechanical Properties

Compressive strength was determined as the main representative of mechanical properties. The testing procedure was conducted according to ČSN EN 12390-3 (2002). The age of the samples was 28 days.

4 RESULTS AND DISCUSSION

4.1 Study of Microstructure

The microscope images of samples microstructure are shown in Figure 1. Comparing the particular samples, the K-120 (see Figure 1a) differs from the others as it does not contain ceramic grains smaller than 10 μm, making the matrix very homogeneous. The reaction processes involved in geopolymerization were probably more intensive in K-120. In K-121 and K-122 the grains are much bigger, which can be clearly seen in Figures 1b and 1c. Additionally, there are small cracks present in K-121 and K-122. It is apparent that in K-121 the crack follows grains' boundaries which can be interpreted as an evidence of poor adhesion of the remaining ceramic particles to the activated products. On the other hand, the crack in K-122 goes through the grains indicating that the adhesion forces are higher than the strength of ceramic.

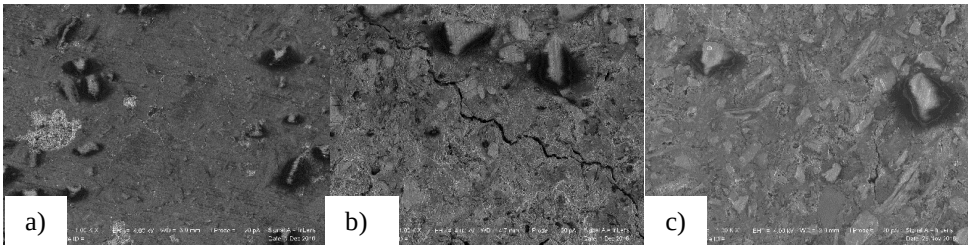

Figure 1. Microstructure of studied samples: a) K-120, b) K-121, c) K-122.

4.2 Pore Size Distribution

The pore size distribution curves and cumulative pore volume of the studied samples are shown in Figure 2 and 3, respectively. According to the results presented in Figure 2, there are notable peaks in the case of K-121 and K-122 that correspond to the pore size of about 1 μm. Nothing similar can be observed in K-120. There is only a slightly higher volume of pores in the range of 0.01 to 0.1 μm. The presence of cracks in K-121 and K-122 can explain these differences. The cumulative curves, presented in Figure 3, show that K-122 had approximately four times higher pore volume than K-120. The porosity thus increased fast with the increasing silicate modulus.

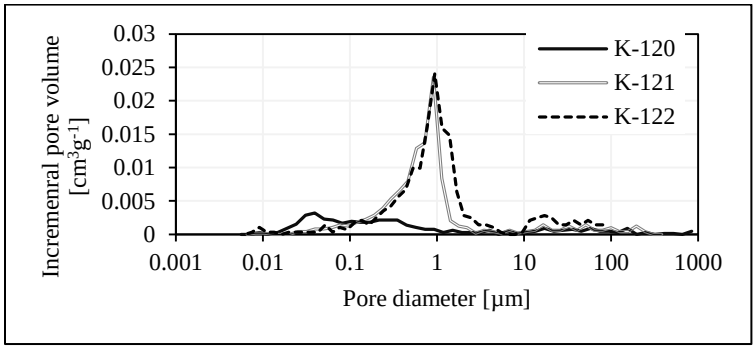

Figure 2. Pore size distribution curves of studied samples.

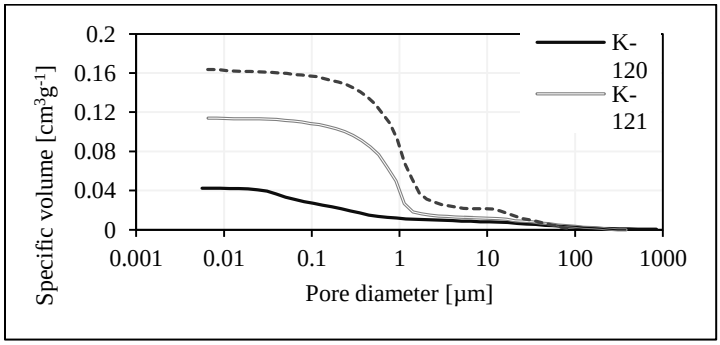

Figure 3. Cumulative pore volume of studied samples.

4.3 Basic Physical Properties

The values of basic physical properties are given in Table 3. The bulk density slightly decreases with increasing silicate modulus, but the differences are within 4 %, which can be considered as insignificant. On the other hand, an increase of matrix density is notable and corresponds to the values of silicate modulus. Compared to K-120, the matrix densities of K-121 and K-122 are by 5 and 15 % higher, respectively. The same can be stated about the open porosities that are higher by 50 % in the case of K-121 and by 158 % for K-122.

4.4 Mechanical Properties

The results of compressive strength tests are shown in Figure 4. It has been found that the mechanical performance of the studied samples is better with increasing silicate modulus even if the open porosity is increasing and there are visible cracks on the specimens. While the

compressive strength of K-121, as compared to K-120, is higher by 37 %, the difference between K-122 and K-121 is only 9 %.

Table 3. Basic physical properties of studied materials.

	Bulk density (kg m^{-3})	Matrix density (kg m^{-3})	Open porosity (%)
K-120	2080	2291	9.2
K-121	2072	2402	13.8
K-122	2006	2630	23.7

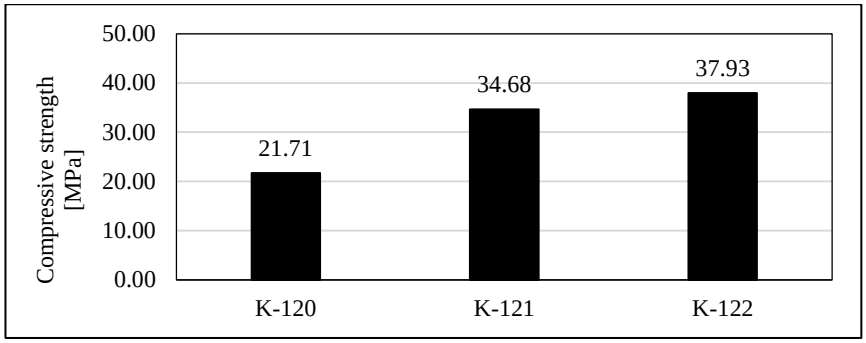

Figure 4. Compressive strength of studied samples.

The obtained results can be compared to those reported by Robayo *et al.* (2016) who investigated broad scale of alkali-activated red clay brick waste as alternative binders. Considering their mixtures without addition of Portland cement, they reported 28-day compressive strengths at room temperature between 7.49 and 54.38 MPa, depending on several factors such as Na_2O content or SiO_2/Al_2O_3 and Na_2O/SiO_2 molar ratios of the activator. The results obtained in this paper are therefore in compliance, especially when different mineralogical composition of ceramic powder is taken into account.

5 CONCLUSIONS

Three different geopolymer mixtures made of alkali activated waste ceramic powder had been prepared and analyzed from the point of view of their microstructure, texture, basic physical and mechanical properties. Since a different amount of sodium hydroxide had been used, each mixture had different silicate modulus that ranged from 0.8 to 1.2.

The varying silicate modulus caused, of course, significant changes in the structure of the resulting materials and thus affected their properties. Higher amount of sodium hydroxide in the mixture composition led to dissolution of fine ceramic grains, creating a very compact structure with the lowest porosity among all the investigated samples. However, such a sample evinced the lowest compressive strength. For the silicate modulus equal to 1.0, the adhesion of activated products to the grains surface was low which resulted in crack formation on the grain boundaries. The formation of these cracks was avoided when silicate modulus of 1.2 had been assumed. After hardening, this mixture formed the sample with the highest compressive strength that was comparable to traditional Portland-cement based materials.

The results presented in this paper can be considered as very promising from the point of view of a utilization of industrial waste as the designed geopolymers had similar mechanical properties to traditional binders of which production is usually very energy-demanding. Anyway, further detailed investigation aiming at other aspects must be still performed before any practical application could be started.

Acknowledgments

This research has been supported by the Czech Science Foundation, under project No 16-02862S.

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