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EFFECT OF THE ENCAPSULATION ON CHEMICAL DURABILITY OF GEOPOLYMERS

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ABSTRACT: This work deals with chemical durability of encapsulated geopolymers against distilled water at temperature of 85 °C. Goal of the work is to find out and interpret the influence of individual encapsulations on chemical durability of geopolymers. Experimental part covers dynamic flow experiments and a method of specification of leached constituents. Time dependence of leaching of Si, Na and Al was obtained. We found out, that the encapsulations of geopolymers which are the subject of our testing in this work, increases the leaching of Si and Al. On the contrary, leaching of Na has got opposite tendency.

KEY WORDS: chemical durability, geopolymers, leaching process

1. INTRODUCTION

Geopolymers are inorganic polymeric materials, which are prepared by polycondensative reaction in basic surroundings at normal temperature and pressure [1]. By this reaction, amorfic alkalic geopolymers with three-dimensional structure resembling to the one of zeolitic precursors come to exist [2].

Term "geopolymer" was first used by Davidovitson [3] in order to describe group of mineral cements resembling to unnaturally created zeolits. Structure of these materials is made of polymeric system Si-O-Al, which is identical with the structure of zeolits. Exact character of the structure is not fully quantificated yet. According to this, geopolymers can be used as amorfic equivalents of zeolites, having the same ratio of Al/Si as zeolites, but without crystallic structure. These differences in structure give zeolites priorities in comparison with common cements.

Chemical durability is defined as ability to resist the influence of external chemical influences, especially atmosphere, water, acids, bases and different chemical agents. Methods of chemical durability take advantage of a fact, that surroundings influencing examined material for a long time at low temperature can have the same effect as the surroundings influencing this material for shorter time at higher temperature [3].

Ceramics [4] have extraordinary resistance to effects of chemical agents even at higher temperatures. This fact is connected to high thermodynamic stability of separated phases of ceramic materials. During effect of some chemical agents on the ceramic material, chemical reactions with separated solid phases can take place. By these reactions new compounds come to exist. Reactions' products are often created at connection of separated surroundings and mostly they are dissolved in liquid phase. Corrosion of ceramics can happen: a) by chemical reaction connected to creation of new compound, b) by dissolving of one or more phases.

Observation of influence of lasting corrosion on decisive qualities of ceramics, for example mechanic firmness is also important. It enables us to predict length of applicability of ceramic components in given corrosive surroundings. When corrosion is taking place, with decrease of body's mass the loaded cross-section is being reduced and that leads to decrease of its mechanic firmness.

Ceramics' durability against effects of chemical agent can be influenced by changing chemical and phase composition, and also by the microstructure of ceramics. For example, when amount of Al_2O_3 in ceramic material is being increased, its durability against effects of hydrofluoric acid is increasing, but durability against effects of sulphuric acid is decreasing.

Increase of earthenware's chemical durability can be achieved for example by addition of $BaCO_3$ into production mixture. During burning barium silicates originated in splinter come into being. These have high durability against errects of alkalic agents. Decrease of ceramic splinter's open porosity also helps it to increase its chemical durability.

During influence of chemical agents on glazing surfaces of ceramic bodies', similar processes as during the glass corrosion take place. By gradual leaching of glass-grid modificators their chemical durability is increasing. Glazing layers are not always homogeneous; they contain bubbles very often and also different crystalic phases.

One of the possible applications of geopolymers is immobilization of radioactive waste with different use; due to catching radioactive waste by absorption of them on the surface of pores particles SiO_2, Al_2O_3, eventually of combinated systems. Another possibility of application of geopolymeric systems is the vitrification of radioactive waste, and eventually encapsulation of radioactive wastes' particles and concretion of these encapsulated particles [5, 6]. Encapsulation is one of possible ways how to increase chemical durability of geopolymeric matrix.

2. EXPERIMENT

Geopolymeric matrix was prepared from water glass, in which NaOH and water had been added while stirring. Then metacaolin was added while stirring. Mixture was divided into shut off-able polypropylene containers and these were dried while closed at laboratory temperature for 21 days. After this term were samples put out of containers and dried free for 7 days. Samples were crushed on shredder Hounsfield H20K-W. Firmnesses in pressure were measured and from the rest the crushed material was prepared.

Crush was encapsulated by tetraethoxysilicate: water mixture (ratios and labelling are shown in table 1). Amounts of raw materials were being mixed for 10 minutes, and then 6 grams of crush were added. Mixture was being mixed for another 10 minutes. After its homogenization, mixture was filtrated and dried at 80 °C in dryer. Finally, mixture was put into ceramic container and burned in oven at 180 - 200 °C for 1 hour.

Tab. 1: Molar ratio of separated components of encapsulated samples

Gels	Tetraethoxysilicate	Isopropyl alcohol	Distilled water
G1	1	16	0
G2	1	15.4	4
G3	1	14	10

Dynamic flow tests were done on crush from samples of geopolymeric matrix without encapsulation (G0) and with different ratio of encapsulation (G1 - G3) at temperature of 85 °C in distilled water. Four parallel determinations of one sample simultaneously took place. 1 gram of crush with grain fraction of 0.35 - 0.5 mm was put into each container for samples. Takings of leaches were done in 0.5; 1; 2; 3; 4; 5; 6; 7; 8 hours.

3. DISCUSSION AND RESULTS

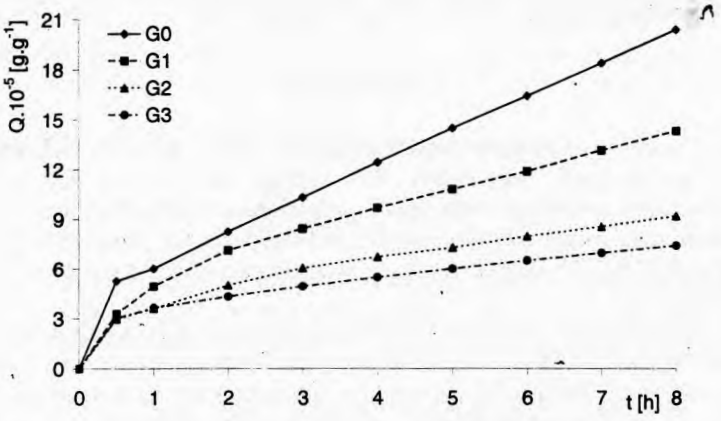
Work dealt with time dependence of leaching substances separated from geopolymeric matrix. Calibration of machine was done and it was found out, that its optimal setting for wanted flow is 1 $ml.min^{-1}$ through 1 g of crush at the temperature of 85 °C.

Composition of geopolymeric matrix in molar ratios was determined after backward calculating from raw materials' weights, which had been used for preparation of geopolymeric matrix. Al_2O_3 : SiO_2 : Na_2O : H_2O are in ratio1 : 3.51 : 1.32 : 19.86.

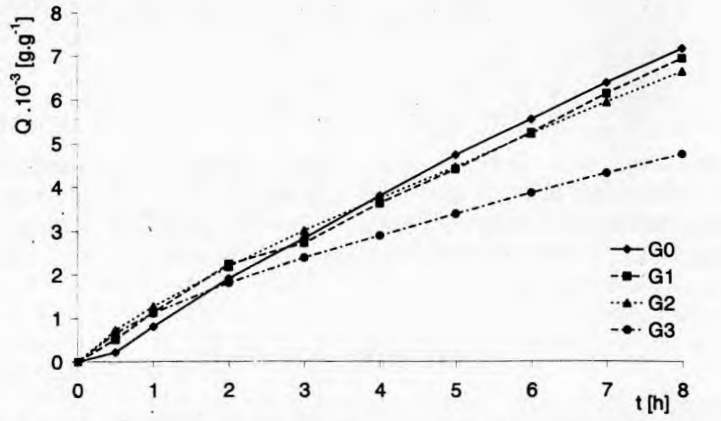
Analysis of leaches was oriented at determination of amount of Si, Na and Al. Amount of Si was determined spectrophotometrically by using yellow form of silicomolybdate acid [7]. Representation of Na and Al was determined spectrophotometrically by using atomic absorption of spectrophotometer AAS 3 Carl Zeiss Jena in emission and absorption mode.

Amount of separated substances in geopolymeric matrix was determined for each parallel determination by magnitude Q, which stands for extracted amount of material, which was backward, counted according to crush weight. From average values of Q, graphic dependences on time were made for Si, Na and Al.

Graph 1 shows the amount of extracted Si for G0 - G3, which was prepared by appropriate procedure. It is obvious from the graph, that encapsulation by individual gels has marked influence on the course of leached Si, because the amount of leached Si decreases. The most noticeable change in comparison to non-wrapped crush is set during encapsulation by G3; for preparation of this gel highest molar ratio of H_2O was used.



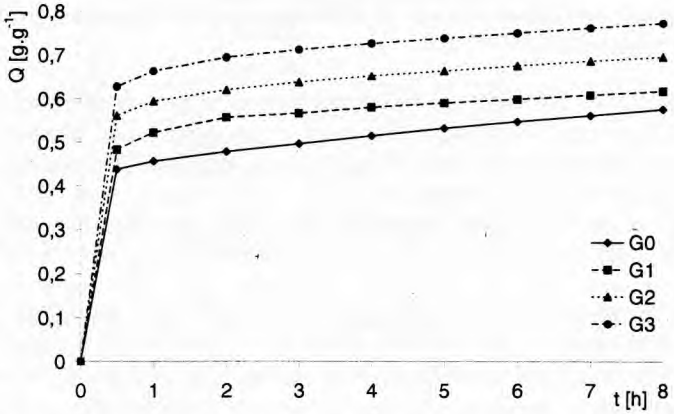
Graph 1: Dynamic flow test of geopolymers G0 - G3 in distilled water at the temperature of 85 °C – time dependence of amount of leached Si (experimental points, average values)



Graph 2: Dynamic flow test of geopolymers G0 - G3 in distilled water at the temperature of 85 °C – time dependence of amount of leached Al (experimental points, average values)

Also it is obvious from graph 2, where the amount of extracted Al for G0 - G3 is shown, that amount of extracted Al has decreasing tendency with most noticeable changes for G3. Graphic dependence grows with time of test almost linearly and from this fact it is obvious, that after 8 hours of experiment running there was no saturated state of Al in the solution.

From graph 3 it follows, that influence of geopolymers' encapsulation on the course of Na leaching has opposite effect than on the course of Al and Si leaching. In first 30 minutes is increase of leaching maximal, but with the time of experiment going on this state is stabilizing and leaching of Na gradually becomes minimal.



Graph 3: Dynamic flow test of geopolymers G0 - G3 in distilled water at the temperature of 85 °C – time dependence of amount of leached Na (experimental points, average values)

4. CONCLUSION

We used 3 gels for the encapsulation of a crush made of geopolymeric matrix. Dynamic flow tests on the experimental equipment, set on the optimal conditions, were done. Amounts of individual extracted substances were determined in the leaches values for each parallel determination of Si, Na and Al obtained by measuring were processed into graphs, where dependence of amounts of leaching substances (calculated according to weight of crush) on time of experiment was shown. The most important step in this work was evaluation of influences of individual encapsulations on the chemical durability of the geopolymers. From graphic dependences 1-3 it follows, that encapsulation by individual gels has an influence on chemical durability of geopolymers as follows: chemical durability of Si and Al is increased by encapsulation; chemical durability of Na is decreased.

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