DE GRUYTER OPEN
CIVIL AND ENVIRONMENTAL ENGINEERING REPORTS
ISSN 2080-5187 CEER 2015; 16 (1): 25-40
DOI: 10.1515/ceer-2015-0002

GEOPOLYMERS IN CONSTRUCTION

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Abstract

Within the framework of quests of supplementary and „healthier" binders to the production of concrete followed the development of geopolymers in construction. However the practical application of these materials is still very limited. The production of each ton of cement introduces one ton of CO_2 into the atmosphere. According to various estimations, the synthesis of geopolymers absorbs 2-3 times less energy than the Portland cement and causes a generation of 4-8 times less of CO_2. Geopolymeric concretes possess a high compressive strength, very small shrinkage and small creep, and they possess a high resistance to acid and sulphate corrosion. These concretes are also resistant to carbonate corrosion and possess a very high fire resistance and also a high resistance to UV radiation.

Keywords: geopolimers, geopolymeric concretes, durability, CO_2 emission

1. INTRODUCTION

The term of geopolymer consists of two elements: geo and polymer. The *geo* prefix specifies that the material is of a natural origin, yet in the case of geopolymers this is not the case. This prefix was given to geopolymers in connection with the fact that their structure is analogical to the structure of natural minerals. The *polymer* particle means that this material is constructed from many repeated particles known as unit cells. Today, the geopolymer is

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considered to be a modern material, yet the beginnings of its application unofficially date back to as early as 25,000 years ago [1]. The oldest artefact for which the use of a geopolymer material was needed is exactly estimated to be this age. This is a terracotta statuette of Venus. A modern theory reports that even ancient Egyptians used a technology connected with geopolymers. They used them when building pyramids and, strictly speaking, to form huge blocks of rocks at the building site [1]. This information replaces the theory about huge blocks of rock being transferred over large distances by hundreds or even thousands of slaves. French scientists proved it that geopolymers were used on a large scale when building channels and water reservoirs that would provide water to human habitats in the ancient Egypt and when building aqueducts in the ancient Rome [1]. The return to this forgotten technology took place in the 1950s [2]. A lot of interest in this technology was shown in numerous branches of industry, yet the greatest possibilities of its quick implementation appeared in the construction industry.
The production of each ton of cement introduces one ton of CO_2 into the atmosphere. In the beginning of XXI century the global production of Portland cement introduces ca. 1.6 billion tons of CO_2 into the atmosphere [3 - 5]. In the year 2013, the global production of Portland cement exceeded 3.5 bn. tons annually, which is ca. 8% of the annual global quantity of CO_2 that is emitted into the atmosphere (Fig. 1) [6-8].

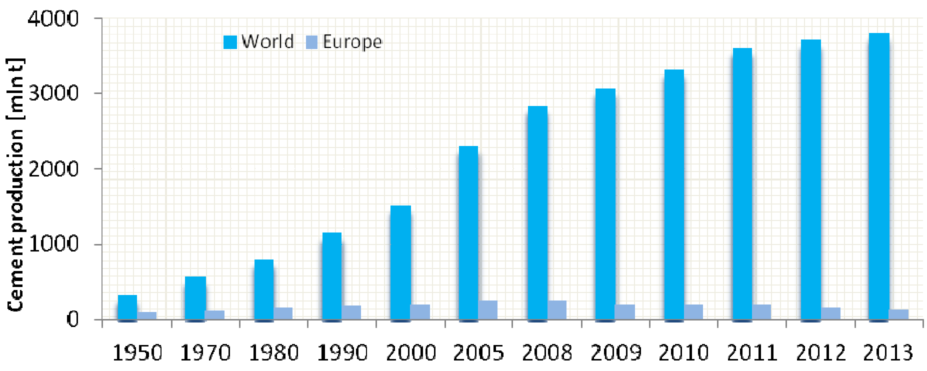

Fig. 1. Proportion of world cement production compared with European cement production [8]

According to various estimations, the synthesis of geopolymers absorbs 2-3 times less energy than the Portland cement and causes a generation of 4-8 times less of CO_2. In connection with this, those who propose the application of geopolymer cements perceive in them a way of an essential reduction of the environmental burden [9]. Owing to these properties, this material has been given the name of a green concrete.

Professor Joseph Davidovits, who established the Institute of Geopolymers in Saint-Quentin in France, is the creator of geopolymers. In the year 1978, Davidovits introduced the term of “geopolymer” to denote the mineral polymer which is produced on the base of geochemistry [10]. The majority of the synthesis methods of geopolymers come down to one process: a fragmented and dried off pozzolanic material (metakaolin or fly ash) is mixed with a water solution of an appropriate silicate (e.g. sodium or potassium silicate) with an addition of a strong base: usually, this is concentrated sodium or potassium hydrate). The paste that is created behaves similarly as the cement: it sets within several hours to become a hard mass. An alternative method to prepare geopolymers is burning of a pozzolanic material with a metal hydroxide to obtain a homogenous powder which fixes water very well, similarly to the Portland cement. This method, however, is problematic due to much worse mechanical properties of the material produced [11]. Yet another method that has recently been proposed is similar to the traditional synthesis with the use of metakaolin, a silicate and hydroxide solution, but colloidal silica is additionally used. It allows to reduce the consumption of the pozzolanic material and to increase the content of silicon in the geopolymer to exceed the maximum value achieved with the use of traditional preparative methods [12].

2. WHAT IS GEOPOLIMER

The geopolymer is the polymer of aluminosilicate which is synthetically produced by means of a synthesis of silicon (S_i) and aluminium (Al), which are geologically acquired from minerals. The chemical composition of the geopolymer is similar to the composition of zeolite but it reveals an amorphous microstructure [13]. Geopolymers consist of long chains: copolymers of alsifer and aluminium as well as the metals cations which stabilize them, most frequently of sodium, potassium, lithium or calcium and also bounded water. Apart from well-defined polymeric chains, various mixed phases occur in them as a rule: silicon oxide, non-reacted aluminosilicate substrate and sometimes crystallized aluminosilicates of a zeolite type (Fig. 2).
Two main categories of the division of geopolymers are accepted. The first one takes into account the elementary units of polymeric chains:

- PSDS Si-O-Al-O-Si-O-Si-O - poly(sialate-disiloxo),
- PSS Si-O-Al-O-Si-O - poly(sialate-siloxo),
- PS Si-O-Al-O - polysialate.

The second category takes into account a division concerning the origin of geopolymers and, strictly speaking, their pozzolanic aluminosilicate material.

Due to this criterion, we distinguish geopolymers that are formed on the base of the following:
- fly ash,
- metakaolin,
- various types of rocks,
- volcanic agglomerates,
- silicas,
- fossil materials.

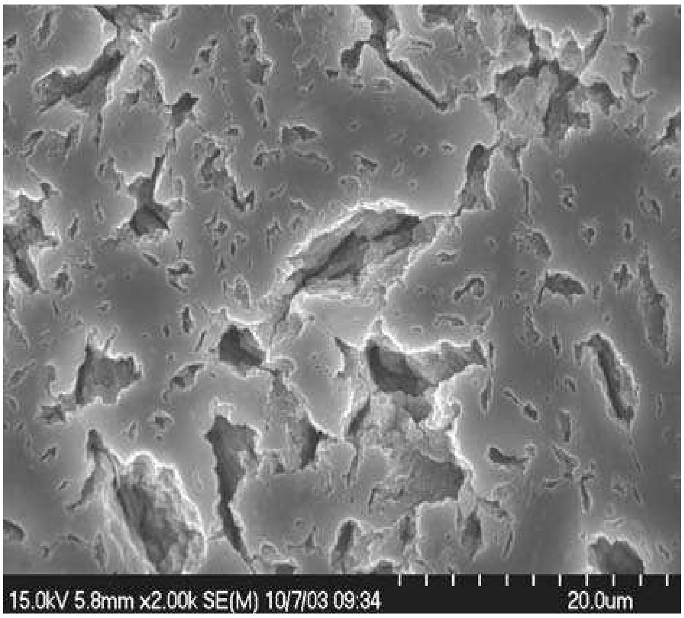

Fig. 2. Typical microstructure of polished geopolymer (100% KOH) [14]

In practice, the use of geopolymeric concretes is still very limited due to their higher price. The decisive influence on this price is related to the use of relatively large quantities of sodium hydroxide and water solutions of silicates. The first applications of the geopolymeric material date back the 1990s; specifically, this was the year 1991. At that time the technologies of concrete on the base of geopolymers that had been developed by the laboratories of the US army were implemented in the construction of airports [15]. Nevertheless, this was not owing to a reduced emission of one of greenhouse gases but in consideration of another property, i.e. the increment time of the strength, which declassed the competitive material that is the cement concrete. After 4 hours, the airstrip obtained the strengths that were sufficient for planes of Boeing 747 size to land on it. The technology of geopolymers is currently used by the Airbus company in the production of composite cables in the structures of jet engines, where geopolymeric cables are able to withstand temperatures in the range of 400-800 ^{0}C [16]. There are also solutions which permit the production of window and door profiles with the use of this material.

3. GEOPOLYMER AS A STRUCTURAL MATERIAL

The application of geopolymers in the building industry mainly consists in the production of concrete from them, which in its composition would include instead of the classical cement a binder produced on the base of aluminosilicates. The type of the chemical reaction, which causes their hardening and a conversion of a plastic concrete blend in a solid body, is the main difference between both binders. Let us compare the difference in the composition of the classical cement and the most popular geopolymer that is used in the production of the concrete blend, i.e. the fly ash. Table 1 presents the average chemical contents in fly ashes that are produced in the combustion of hard bituminous coal in one thermal power station.

Table 1. Chemical composition of geopolymeric cement [17]

Mineral composition	SiO_2	Al_2O_3	Fe_2O_3	TiO_2	CaO	K_2O	MgO	SO_3	Na_2O	P_2O_5
Percentage content	53.7	32.9	5.5	2.1	1.84	1.76	0.92	0.46	0.37	0.15

Table 2 presents the average content of minerals in Portland cement produced as a result of burning of limestones, cement rocks, clay and gypsum.

Table 2. Mineral composition of Portland cement [18]

Mineral composition	$3CaO{\bullet}SiO_2$	$2CaO{\bullet}SiO_2$	$3CaO{\bullet}Al_2O_3$	$4CaO{\bullet}Al_2O_3{\bullet}Fe_2O_3$	$CaSO_4{\bullet}2H_2O$
Percentage content	50	25	10	10	5

As it can be seen in Table 1 and 2, the mineral compositions of the materials compared are considerably different. The oxides of silicon and aluminium constitute the bases of the composition of the geopolymer. The additives of metal cations such as sodium or potassium constitute the stabilizing material here. In the case of the Portland cement, the situation is different. The main element is tricalcium and dicalcium silicates known as alite and belite and tricalcium aluminate, which depending from the composition of the clinker mass, constitute jointly even up to 90% of its volume [18]. The pure Portland cement is free from any additives of other oxides due to the fact that is burnt from those materials that are appropriately segregated and selected. The geopolymer is formed on the base of fly ashes, which constitute merely a by-product of the coal combustion process.

The classical cements set owing to the special phenomenon of solvation, i.e. hydration. It is a compound process due to overlapping and a mutual influence of individual clinker phases that react with water. The total hydration process consists of three basic stages. The dissolution of soluble compounds in water, that is the proper hydration, which consists in the creation of the primary phase in a colloidal state (the formation of the plastic mass) and the crystallization of the hydration products (hardening of the plastic mass). The initial stage of the proper hydration of cement is connected first of all with the C_3A phase [18]. As a result of a fast reaction of this phase, large crystals of hydrated calcium aluminates are produced (Fig. 3a). All the stages of hydration, as compared with the setting of the polymeric blend, are presented in Fig. 3.

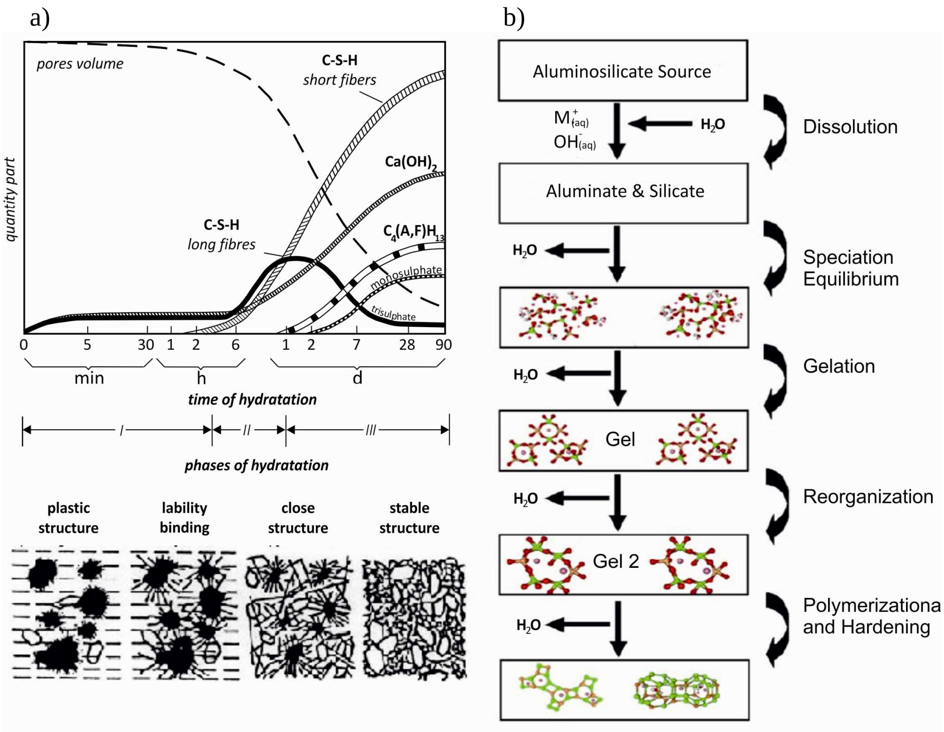

Fig. 3. Comparison of phases: a) hydration of the Portland cement, b) polymerization [19]

As it is evident in Fig. 3, in the formation process of concrete on the base of geopolymers, the situation looks different than in the case of concrete on the base of the Portland cement. In the case of concrete on the base of geopolymers, the whole process commonly known as polymerization consists of a number of reactions which do not interpenetrate one another but follow one another. In each of the phases, we first observed a gradual release of added water, which is the characteristic phenomenon of the polymerization process [20]. The substance, which initially constitutes a powder, enters the gel phase to become a solid body after the start of the proper polymerization [21]. A detailed diagram is presented in Fig. 3b. A different process of the setting of the two binders presented makes the materials examined different with respect to their strength parameters. The polymerization process is a significantly more violent process, which translates into the geopolymeric concrete obtaining nominal strengths faster than in the case of the cement concrete. What is more, polymeric bindings ensure obtaining strength for the concrete blend at a level being 2-3 times higher as compared with the strength of the classical concrete on the base of the Portland cement [18, 21]. Below, the structure is presented of the polymeric slurry at the moment of setting (Fig. 4) and after 28 days starting from the moment of its preparation (Fig. 5), and concrete on the cement binder (Fig. 6).

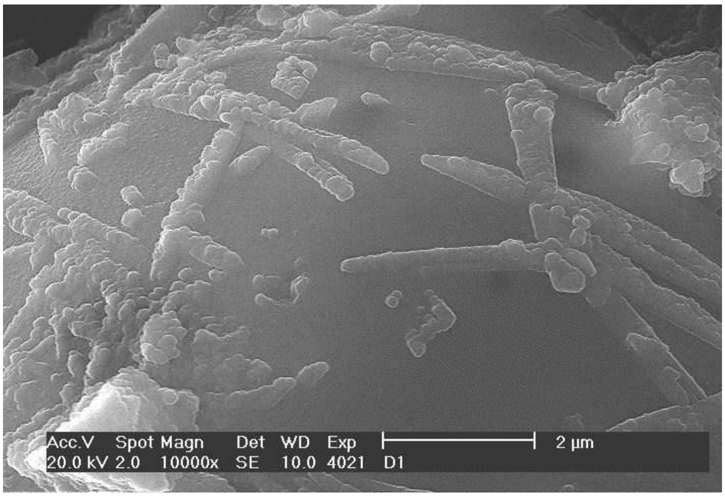

Fig. 4. Initial phase of the polymeric slurry [17]

The first and most simple method to obtain the geopolymeric binder for the production of concrete is to use fly ashes for this purpose, which constitutes a by-product in the combustion process of coal in coal power plants. Compositions of blends have already been developed which allow obtaining of a high strength concrete. Detailed investigations were carried out among others in the Czech Republic, in the course of which the properties of concrete were analyzed that was produced on the base of a geopolymeric binder obtained through the reactions of an alkaline activator (in the form of sodium hydroxide and sodium silicate), that was acting on the by-product of the combustion of hard bituminous coal. These investigations demonstrated that concerning properties, this concrete diverges from the one produced on the base of the Portland cement [17].

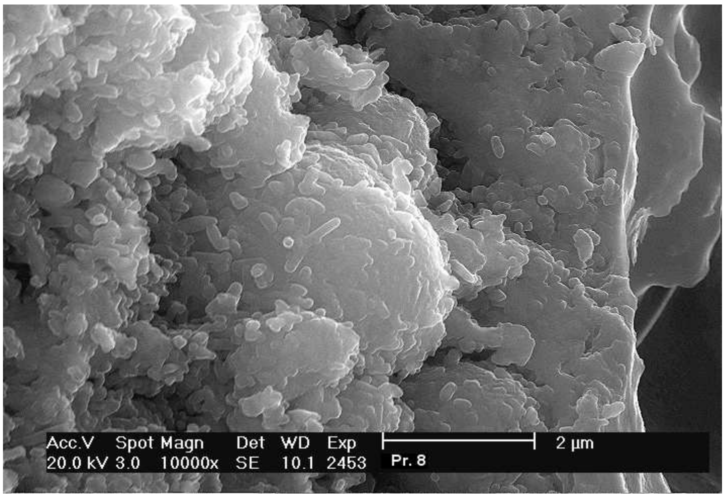

Fig. 5. Final phase of the polymeric slurry [17]

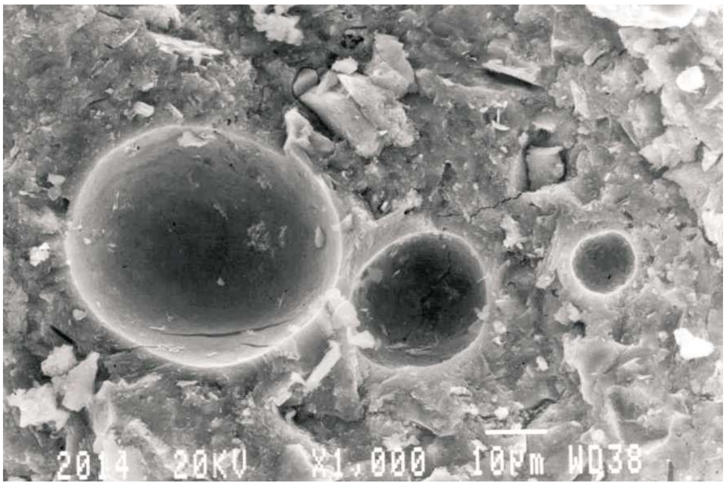

Fig. 6. Microstructure of concrete on the base of the Portland cement [19]

Below, the main differences are listed concerning the properties that were observed during a number of investigations in relation to the geopolymeric concrete:

- The structure of the geopolymers obtained from fly ashes consists chiefly of AlQ4(4Si), SiQ4(4Al) and SiQ4(4Al).
- The geopolymer obtained on the base of fly ashes is the strongly porous, which determines its strength. Additions in the form of those materials that

include Ca, such as gypsum, significantly reduce the porosity of this material.

- The concrete produced on the base of the geopolymeric binder does not undergo the phenomenon of the occurrence of shrinkage cracks; furthermore, the relation of compressive strength to the value of tensile strength is within the limits of 10:5.5, whereas in the case of the classical cement concrete this relation is at 10:1 -10:1.5 [22].
- The rheological features are different from those which occur in concretes on the base of the Portland cement. An increase was observed of the resistance of the concrete to chemical corrosion and to the activity of low temperatures.
- No change is observed of the structure of the contact layer on the boundary between the binder and reinforcement, as it is the case when using the classical Portland cement.

As mentioned previously, the use of fly ashes is the cheapest method to obtain geopolymers. Additionally, this is also an environment-friendly approach, which makes a practical utilization of this material possible. However, one needs to remember that not all fly ashes may be used for the production of the green concrete. Some of those fly ashes which are produced in the reaction of coal combustion in power plants and thermal power stations may be radioactive. As reported by the Scientific American Magazine, those American scientists who have been conducting research in connection with coal as an energy fuel and its environmental impact for a dozen or so years now have obtained surprising results which contradict the stereotyped approach. It appears that coal wastes that are released by power plants in the course of its exploitation can be 100 times more radioactive than waste generated by nuclear power stations, for the purpose of the production of the same quantity of energy [16]. The composition of coal that possesses trace amounts of uranium and thorium (i.e. radioactive elements) is the cause of this phenomenon. Their quantity is harmless for the environment provided that coal is not burnt. In the course of the combustion reaction, the concentration of both elements increases ten times. In connection with this fact, the name of “green concrete” cannot be granted to blends on the base of fly ash with higher radioactivity. Consequently, it is not recommended to use such concrete in housing industry. Nevertheless, it can be used in industrial and transport structures.

The application of a binder produced from kaolin burnt in high temperatures constitutes the second technology of the production of the geopolymeric concrete [16, 23]. This mineral has already undergone numerous examinations as an addition to the Portland cement in the production process of concretes. By adding it to cement, a concrete with an increased strength can be obtained. However, it is not only kaolin and kaolinite that may replace fly ashes. In the German Bauhaus University in Weimar, a number of examinations were carried out to demonstrate the possibility of the use of clay for the production of the geopolymeric binder [23]. Two different burning methods of the materials presented are compared below aimed at the production of the geopolymeric cement. Fig. 7 presents a simplified diagram of the production of the geopolymeric cement from kaolin or kaolinite (Fig. 7a) and clay (Fig. 7b).

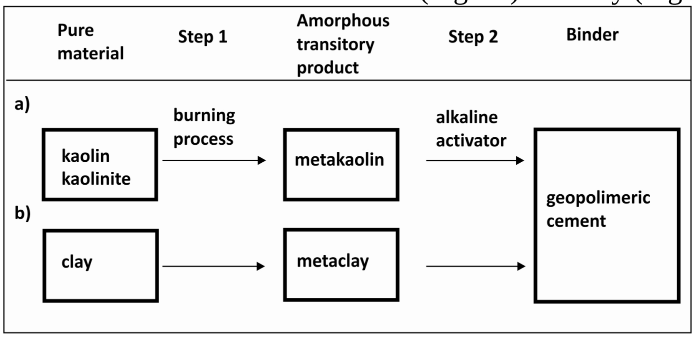

Fig. 7. Production process of the geopolymeric cement from kaolin and kaolinite or clay [23]

Table 3 presents the chemical composition of the three most popular and most easily available materials which the geopolymeric cement can be produced from. As it is evident, the contents of individual oxides are similar. All the three ones are built on the base of the oxides of silicon and aluminium. They also possess metal oxides that are responsible for the stabilization of the whole material. No occurrence of sulphur and phosphorus oxides in the case of metakaolin and meta-clay demonstrates that these materials are more pure than fly ash and their composition does not include any pollutants.

Table 3. Chemical composition of the geopolymeric cement [17, 23]

Mineral composition	SiO_2	Al_2O_3	Fe_2O_3	TiO_2	CaO	K_2O	MgO	SO_3	Na_2O	P_2O_5
Fly ash	53.70	32.90	5.50	2.10	1.84	1.76	0.92	0.46	0.37	0.15
Metakaolin	52.00	45.00	0.50	1.50	0.40	0.05	0.20	0	0.15	0
Meta-clay	64.80	13.70	4.90	0	1.00	4.00	2.60	0	0.20	0

The burning of meta-clay was carried out in two different technological conditions. The first one was direct combustion, which took place on an open furnace with a direct access to the atmosphere. The second one was a burning process in a closed environment (indirect burning), which took place in a hermetic combustion chamber. The mineral was burnt for a period of one hour in

the temperatures of 650 ^{0}C, 750 ^{0}C, 850 ^{0}C and 950 ^{0}C respectively. The creation phases of meta-alumina are shown in Fig. 8.
The data presented above indicate the advantage of the burning process of clay in the closed chamber. Smaller inputs of energy are required to obtain similar physicochemical parameters of the material. The situation is quite the same after putting the material to Chapell's test, i.e. the test for the determination of pozzolanity [20]. The highest ability of the meta-clay produced concerning bonding of calcium occurs at the temperature of 750^{0}C (for burning in a closed chamber) and 850^{0}C (for burning in free atmosphere) respectively. This test also demonstrated that the blend produced as a result of burning possesses the best aluminate and silica solubility in the alkaline solution in the same temperatures where it achieves the highest degree of pozzolanity. Additionally, the most desirable relation of the moles of silicon and aluminium which is favourable for the formation of polymeric networks occurred with these values of temperatures, too. It was in the range of $2.2 < Si/Al < 3.0$ for the temperature of 750^{0}C (when burning in a hermetic chamber) and in the range of $2.3 < Si/Al < 3.5$ for the temperature of 850^{0}C (burning with an air inflow). Apart from a smaller energy input for the preparation the binder blend, indirect burning possesses one more essential asset. After adding the alkaline activator, the blend forms a binder with considerably higher strength values (Fig. 9). These values are even 20% higher in comparison with those obtained by the binder that is prepared in the course of direct burning.

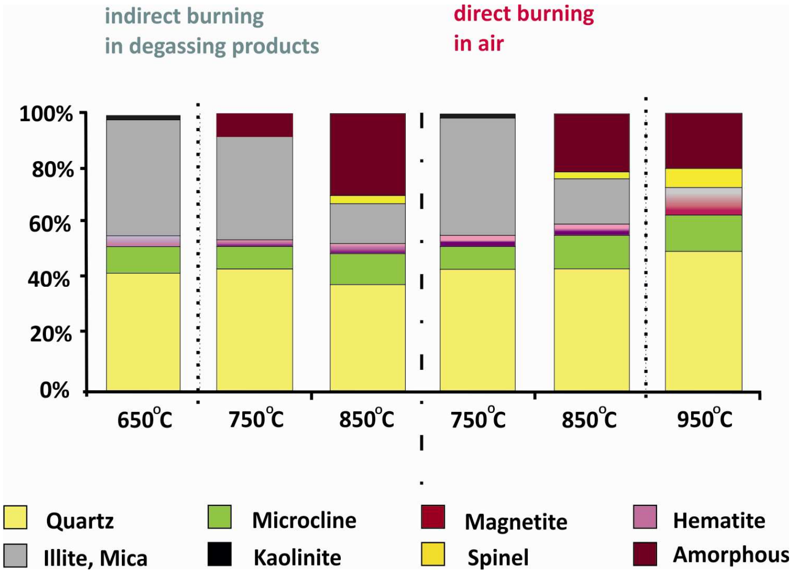

Fig. 8. Creation phases of meta-clay in the burning process [23]

The greatest strengths of the ready geopolymeric concrete produced from the samples burnt at the different temperatures were obtained with the use of burning temperatures ranging from 750 to 850^{0}C (for burning in the closed chamber). Any further increase of the temperature caused a reduction of the strength of the subsequent samples.

Geopolymeric concretes possess a high compressive strength, very small shrinkage and small creep, and they possess a high resistance to acid and sulphate corrosion [24, 25]. Some researchers also found that this concrete is also resistant to carbonate corrosion and possesses a very high fire resistance [26] and also a high resistance to UV radiation [27].

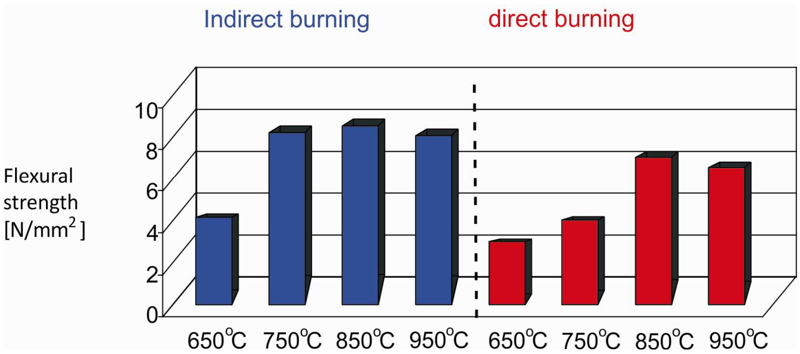

Fig. 9. Influence of burning temperature on the strength of the geopolymeric material (90 days, 75% of humidity, temperature of 20^{0}C) [23]

The widest and most interesting investigations concerning reinforced concrete elements (beams and pillars) from the geopolymeric concrete were carried out by the Curtin University of Technology in Australia in the year 2006 [28]. The results were obtained both for beams and pillars which were similar in behaviour to reinforced concrete elements from a concrete on the Portland cement (Fig. 10, 11).

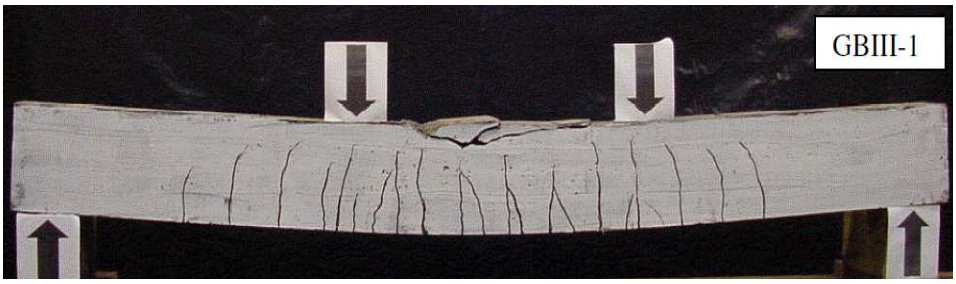

Fig. 10. Example of reinforced concrete beams from geopolymeric concrete damage [28]

Fig. 11. Example of reinforced concrete pillars from geopolymeric concrete after the performed durability tests [28]

4. CONCLUSIONS

A low emission of CO_2, a fast increment of resistance and high values of resistance constitute evident advantages of the geopolymeric binder over the Portland cement. Most probably, these features will not be used very soon. It is only legal regulations that put limits to carbon dioxide emission that will contribute to the geopolymeric concrete technology being implemented on a wide scale in the production of concrete. The European Commission is not intending to impose any penalties in relation to CO_2 emissions by the year 2020. There is no information on what new legal regulations will be like after that date. It is certain that the cement industry that has invested huge amounts of money to the existing production lines of standard cement is not interested in any rapid changes in relation to the new binder. As of the present day, the use of the green concrete will be implemented in special situations only, i.e. when better properties of geopolymers are sought. What is meant here is chiefly the setting time and very good fireproof properties of the new material. Other industry branches, i.e. mainly foundry engineering or the production of composites, will be keener on using the new technology. The aeronautic industry is already making use of the properties of geopolymer composites on a wide scale. Owing to research into this binder, geopolymers may soon become the leading material in many industry branches including maintenance of monuments. The potential of geopolymers which has already been discovered will undoubtedly contribute to this; geopolymers are used to restore damaged works of art produced from ceramics or stone.

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ZASTOSOWANIE GEOPOLIMERÓW W BUDOWNICTWIE

S t r e s z c z e n i e

W ramach poszukiwania zastępczych i „zdrowszych" spoiw do produkcji betonu nastąpił rozwój geopolimerów w budownictwie. Jednakże praktyczne zastosowanie tych materiałów jest jeszcze nadal bardzo ograniczone. Produkcja każdej tony cementu wprowadza do atmosfery tonę CO_2. Według różnych szacunków, synteza geopolimerów pochłania 2-3 razy mniej energii, niż cementu portlandzkiego oraz powoduje wydzielenie 4-8 razy mniejszej ilości CO_2. Do tego betony geopolimerowe posiadają wysoką wytrzymałość na ściskanie, bardzo mały skurcz i małe pełzanie oraz dają wysoką odporność na korozję kwasową i siarczanową. Betony te są także odporne na korozję węglanową i posiadają bardzo wysoką odporność ogniową, a także wysoką odporność na promieniowanie UV.

Słowa kluczowe: trwałość, geopolimery, betony geopolimerowe, emisja CO_2

Editor received the manuscript: 27.12.2014