

Structural changes of fly ash-based geopolymers induced by a large amount of lead

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Abstract: Geopolymers has emerged as possible technological and environmentally solution for the effective immobilization of heavy metals and other toxic materials. In this paper, structural changes of geopolymers induced by large amount of lead (up to 30%) were examined. Geopolymers were synthesized by alkali activation of fly ash using sodium silicate with modulus 1.5 as alkali activator. Lead was added in the form of water-soluble salt - lead-nitrate, during the synthesis of geopolymers. Structural changes of geopolymers due to Pb addition were assessed by means of XRD and ^{29}Si MAS NMR analysis. XRD analysis confirmed that the Pb addition led to the formation of new Pb-phases, which indicated that Pb formed chemical bonds with the geopolymers. ^{29}Si MAS NMR analysis showed the increase of less condensed Q^1 and Q^2 structural units of geopolymers with Pb addition.

Keywords: geopolymer, fly ash, immobilization, lead

1. Introduction

Geopolymers are the hotspot in research of new green cementitious materials. They are produced by alkaline activation of amorphous aluminosilicates, including industrial waste such as fly ash (FA) (Shi et al., 2011). Fly ash is a by-product produced from combustion of coal and collected by electrostatic separators from the fuel gasses of the thermal power plants (Komljenović et al., 2010). Accumulation quantities of fly ash are increasing yearly and estimated to be around 780 million tons annually (Toniolo & Boccaccini, 2017). There is a considerable interest in utilizing such by-product material in the synthesis and processing of high performance cementitious materials. These inorganic binder materials are very attractive environmentally for a number of reasons (Duxon et al., 2007). First, this type of binder represents a considerable benefit in terms of reducing global CO_2 emissions. High emission of CO_2 during Portland cement production (1 t of CO_2 per 1 t of Portland cement) is a reason enough to develop and use geopolymers. Further, the use of industrial by-product is consistent with the growing trend towards waste re-utilization (van Deventer et al., 2007). Geopolymers are also an alternative to Portland cement due to excellent mechanical properties, chemical resistance and

durability in the long-term period (Provis & van Deventer, 2009). Some of these materials cross the boundaries defined by traditional cementitious materials (Fernandez-Jimenez & Palomo, 2003).

Geopolymers are the amorphous equivalent of certain synthetic zeolites, although the distinctive zeolite structure is absent (van Jaarsveld et al., 1997). The main product of fly ash geopolymerization is the three-dimensional hydrated sodium aluminosilicate gel "N-A-S-H", in which Si and Al atoms are bridged through oxygen atoms and tetrahedrally coordinated, while alkali metal ions play an important role in geopolymers structure, as they balance the negative charge of aluminum in tetrahedral coordination (Provis & van Deventer, 2009).

Geopolymer properties are influenced by numerous factors, such as the characteristics of the starting material (fly ash), the type and concentration of the alkali activator, as well as the synthesis and curing conditions of geopolymers (temperature, reaction time and the relative humidity) (Nikolić et al., 2015).

On the other hand, with the development of industry, there is an increasing amount of heavy metal wastes generated from metalwork industry, chemical industry, electrical equipment manufacturing and other fields, which brings great damage to

the environment (Wang et al., 2018). Unfortunately, landfilling is still the most widely used technic for processing of waste from the mining industry causing the environmental problems (van Jaarsveld et al., 1997). It is clear that a need exists for a technology that can handle large amount of fly ash, mine tailings, contaminated soil, municipal waste containing heavy metals (Shi & Fernández-Jiménez, 2006). Heavy metals pollution has become a very serious global environmental problem. In addition, heavy metals in soil, water and air may cause bioaccumulation of heavy metals in animals and humans. Lead (Pb) is one of the most common hazardous heavy metals due to the increasing human activity and industrial progress. Lead is a pollutant, which has long-term persistence in the environment and has detrimental effects on the human body, especially on the nervous, immune and reproductive system (Landrigan et al., 2000).

Geopolymers are a possible solution in the immobilization of heavy metals (Xu et al., 2006; Guo et al., 2017) for several reasons. Firstly, geopolymer has a three-dimensional network structure, and heavy metals can be “lock” in its structure. Secondly, the permeability of the geopolymers is very low, which can prevent leaching of heavy metals. Thirdly, Al (III) is a fourfold-coordinated atom, so $[\text{AlO}_4]^{5-}$ has ability to attract positive charge. In addition, geopolymers are durable and chemical resistant (van Jaarsveld et al., 1997).

The mechanism immobilization can be physical or chemical nature. Although it is well known that the geopolymers are suitable material for immobilization of lead (Guo et al., 2017), the mechanism of Pb immobilization is still unclear (Nikolić et al., 2014). Some authors (van Jaarsveld et al., 1998) indicated that Pb immobilization proceeds by a combination of physical encapsulation and chemical bonding, including adsorption, while other authors (Phair et al., 2004) concluded that the Pb immobilization is a chemical process, rather than the sorption or physical encapsulation. Perera et al. (2005) suggested that Pb is encapsulated in the amorphous aluminosilicate structure of geopolymers while no new crystalline phases were observed. On the other hand, Palomo & Palacios (2003) reported that the addition of $\text{Pb}(\text{NO}_3)_2$ in the fly ash-based geopolymer led to the formation of vaguely identifiable insoluble crystalline compound Pb_3SiO_5 . Zhang et al. (2008) tentatively supported the identification of Pb_3SiO_5 . In addition, Lee et al. (2016) suggested that lead might contribute to the balancing charge of geopolymers network by replacing Na in the structure.

Regardless of the fact that there is no overall agreement regarding the mechanism of Pb immobilization, there is a general agreement that the process of Pb immobilization in geopolymers is successful (Nikolić et al., 2014; Palomo & Palacios, 2003). However, relatively low concentrations of Pb compound (up to 1% Pb in relation to the FA mass) in the Pb immobilization were mainly used (Nikolić et al., 2014; Perera et al., 2005), which made the mechanism of Pb immobilization still indefinable. In the previous studies, Nikolić et al. (2018) investigated the influence of curing conditions on the lead immobilization and confirmed that 4% of lead was successfully immobilized. Guo et al. (2017) were conducted research in order to immobilize higher concentration of Pb (up to 8%) in geopolymer matrix and to establish the

immobilization mechanism of Pb. Obviously, higher concentrations of Pb are required to clarify the mechanism of Pb immobilization.

Structural changes of geopolymer induced by Pb addition were usually studied by XRD, FTIR, and SEM or TEM coupled with EDS analysis. However, these methods usually provide limited information, particularly XRD analysis, since the major product of alkali-activation - the aluminosilicate gel is amorphous. In order to obtain additional useful information regarding the mechanism of Pb immobilization, more appropriate method, such as ^{29}Si MAS NMR (Nikolić et al., 2018) was also used.

The aim of this paper is to determine how a large amount of lead (up to 30%) affects the structure of geopolymers, using XRD and ^{29}Si MAS NMR analysis and possibly clarify the mechanism of Pb immobilization.

2. Materials and Methods

2.1. Materials

In this study, FA Morava from thermal power plant “Morava” Svilajnac, Serbia was used. Chemical composition and classification according to ASTM C 618-03 of the initial FA sample are shown in Table 1.

Table 1. Chemical composition and classification according to ASTM C 618-03 of the initial FA sample.

Component	FA
SiO ₂	55.23
Al ₂ O ₃	21.43
Fe ₂ O ₃	7.42
CaO	7.94
MgO	2.61
SO ₃	0.81
Na ₂ O	0.64
K ₂ O	1.35
Loss on ignition at 1000 °C	1.66
<i>Total</i>	<i>99.09</i>
Classification according to the ASTM C 618-03 standard	class F

Sodium silicate solution (“Galenika–Magmasil”, Serbia, 13.60% Na₂O, 26.25% SiO₂, 60.15% H₂O) was used as an alkaline activator. Starting sodium silicate modulus n (SiO₂/Na₂O mass ratio) was 1.93. This modulus was further adjusted by adding NaOH pellets (NaOH p.a. ASC reagent, (min. 98%), “Sigma Aldrich”, Sweden). Lead nitrate $\text{Pb}(\text{NO}_3)_2$ was used as a heavy metal contained source ($\text{Pb}(\text{NO}_3)_2$ p.a., “Centrohem”, Serbia).

2.2. Synthesis of geopolymers

The fly ash-based geopolymers was synthesized according to previously optimized procedure (Nikolić et al., 2014; 2018). Sodium silicate solution with modulus 1.5 was used as an alkali activator. The concentration of the activator was 10% Na₂O with respect to the FA mass.

Geopolymer paste samples were prepared by mixing FA with alkali activator and water. Water/binder ratio (water represents the total amount of water in the system including water from the activator, while binder represents the total FA

mass and solid part of the activator) was 0.41. Method preparation of geopolymers containing lead was similar to the method preparation geopolymers without lead. Pre-calculated amount of lead nitrate was dissolved in the amount of distilled water required to achieve appropriate consistency. The order of geopolymers preparing was as follows: the activator was added in the FA and then lead nitrate solution. The content of Pb accounting for the total mass of FA were 0%, 10% and 30%. Pastes were poured into plastic moulds on a vibrating table and then cured for 24 h at 55 °C, based on the previous research (Nikolić et al., 2018). After curing, pastes were demolded, crushed and pulverized in isopropyl alcohol for 1 h in order to stop further reaction, filtered, rinsed with acetone and dried at 50 °C for 2 h.

2.3. Methods of characterization

Mineralogical characterization of geopolymers was conducted by X-ray powder diffraction analysis (RIGAKU X-ray spectrometer KG-3). The diffraction patterns were recorded using Cu K α radiation ($\lambda = 1.54178 \text{ \AA}$) within 10 – 50 ° 2 θ range, with a rate 1 °/min. For the identification of crystalline phases, the “PCPDFWIN” software (based on JCPDS-ICDD database) was used.

²⁹Si MAS NMR spectra of geopolymers were obtained at a Larmor frequency of 79.49 MHz using Bruker MSL 400 system, Apollo console upgraded (Tecmag). Chemical shifts $\delta(^{29}\text{Si})$ were externally referenced to 2,2-dimethyl-2-silapentane-5-sulfonate (DSS) standard. Gaussian peak deconvolution of the obtained spectra was performed using DMFIT application (Massiot et al., 2002), by employing common routine for analyzing peak distribution. Prior to ²⁹Si MAS NMR analysis, iron content in the geopolymer samples was minimized by exposing the samples to a strong magnetic field (Nikolić et al., 2018).

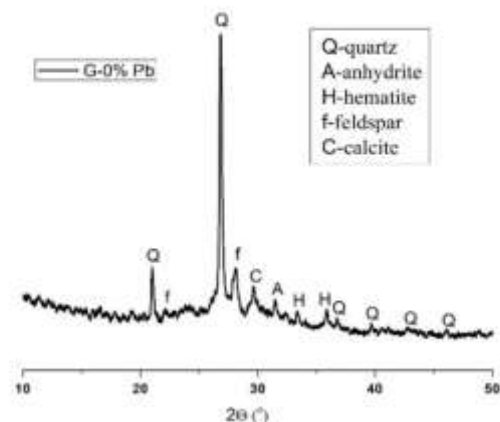
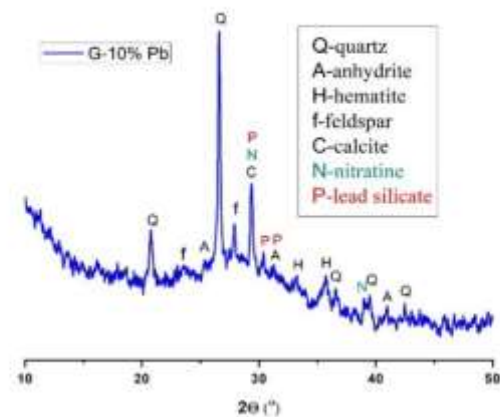
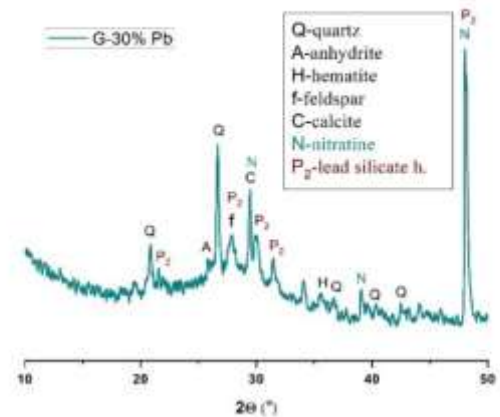
3. Results and Discussion

3.1. XRD analysis of geopolymers

X-ray powder diffraction analysis of geopolymers with 0, 10 and 30% of Pb added is shown in Figure 1. XRD analysis of geopolymers samples indicated a wide and diffusive reflection in an interval from 25° to 35° 2 θ , so-called “geopolymer hump”. In addition, XRD analysis confirmed the presence of crystalline phases typical for the initial FA: quartz (PDF#33-1161) as the most prominent, anhydrite (PDF#06-0226), feldspar (PDF#20-0528), hematite (PDF#24-0072), as well as secondary calcite (PDF#47-1743) formed due to geopolymer carbonation.

Apart from the mentioned phases, in the geopolymer samples with 10% and 30% Pb added, nitratine (sodium nitrate NaNO₃, PDF#07-0271) was detected. The nitrate formation was not surprising given that Pb was added in the form of highly soluble nitrate salt, while sodium came from the activator. In previous study (Nikolić et al., 2018), the presence of nitratine in the sample with 4% Pb added was also confirmed.

In the geopolymer samples containing lead, the appearance



of new Pb-peaks was observed. In the sample with 10% Pb added, lead silicate Pb₅SiO₇ (PDF# 35-0274 marked with P) was noticed, although a main peak for this phase overlapped with the peaks for the calcite and nitratine. On the other hand, in the sample with 30% Pb added the presence of one more phase, probably lead silicate hydroxide (PDF# 44-0276 marked with P₂) was detected. However, because of the fact that some peaks of this phase overlap with the peaks of other phases it cannot be reliably verified. In addition, the possibility of the formation of other lead silicate phases should not be excluded. In the previous study (Nikolić et al., 2018), XRD analyses did not revealed formation of any new crystalline Pb

Figure 1. XRD analysis of geopolymers depending on concentration of Pb added

phase, although the formation of amorphous lead phases, either amorphous lead silicate or amorphous lead-incorporated aluminosilicate gel, was not excluded.

Palomo & Palacios (2003) have already indicated the occurrence of lead silicate in the Pb immobilization by fly ash-based geopolymers. The occurrence of new Pb-phases could indicate that Pb formed chemical bonds with the geopolymers. Furthermore, this points to the fact that the mechanism of Pb immobilization is the chemical nature.

3.2. NMR analysis of geopolymers

^{29}Si MAS NMR spectra of geopolymer samples characterized symmetric resonance lines, which were centered around -93 ppm indicating poorly ordered aluminosilicate structures (Figure 2).

The spectra consist of overlapping resonances attributed to the silicon sites present in aluminosilicate gel, unreacted crystalline phases and unreacted glassy phase present in the initial FA (Nikolić et al., 2018). It is assumed that ^{29}Si MAS NMR spectrum of geopolymers consists of all five $\text{Q}^4(m\text{Al})$, $m = 4, 3, 2, 1$ or 0) silicon species, with $\text{Q}^4(4\text{Al})$, $\text{Q}^4(3\text{Al})$, $\text{Q}^4(2\text{Al})$, $\text{Q}^4(1\text{Al})$ and $\text{Q}^4(0\text{Al})$ resonating at approximately -84, -89, -94, -99 and -107 ppm, respectively (Engelhardt & Michel, 1987; Duxon et al., 2005).

The deconvolution of broad ^{29}Si MAS NMR spectra of geopolymers consisting of individual $\text{Q}^n(m\text{Al})$ structural units of silicon, are shown in Table 2. In addition to $\text{Q}^4(m\text{Al})$ silicon peaks, small resonances at approximately -73, -79, -107 (sharp peak) and -112 ppm were observed in the spectra of all investigated geopolymer samples. Peaks at approximately -73 and -79 can be ascribed to the less condensed silicon species Q^1 and Q^2 (Criado et al., 2008; Ruiz-Santaquiteria et al., 2012). Sharp peak at approximately -107 ppm indicated an ordered crystalline structure and can be attributed to quartz originating from the initial FA (Nikolić et al., 2018). Quartz content in geopolymer samples was almost constant, indicating its inertness during alkali activation. The resonance at -112 ppm is usually attributed to different SiO_2 polymorphs (Fernández-Jiménez, 2006), i.e. unreacted fractions from the initial FA or $\text{Q}^4(0\text{Al})$ units in the products of alkali activation reaction (Ruiz-Santaquiteria et al., 2012).

By comparing the results of the spectra deconvolution of geopolymers with 0% and geopolymers with 10% and 30% Pb added, it could be concluded that the Pb addition led to an increase in the fraction of less condensed Q^1 and Q^2 structural units.

In addition to an increase in the fraction of less condensed Q^1 and Q^2 structural units, previous research of Nikolić et al. (2018) has shown that the Pb addition leads to changes in the structure of Q^4 geopolymer units. The authors pointed that addition of 4% Pb led to the reduction of the fraction of $\text{Q}^4(m\text{Al})$ structural units richer in aluminum ($m = 3, 4$) with respect to a fraction of the structural units richer in silicon ($m = 0, 1, 2$). This trend is also noticeable in the case of addition of a significantly higher amount of lead. However, more

pronounce changes are the increase of less condensed Q^1 and Q^2 structural units (Figure 3), which means that Pb addition led to depolymerization of N-A-S-H gel.

The increase in the fraction of less condensed structural units, as well as reducing the fraction of higher condensed $\text{Q}^4(m\text{Al})$ structural units, with an addition of Pb has been also observed in lead silicate glasses (Fayon et al., 1998). This leads to the conclusion that geopolymers containing a considerable amount of Pb are similar to lead-silicate glasses.

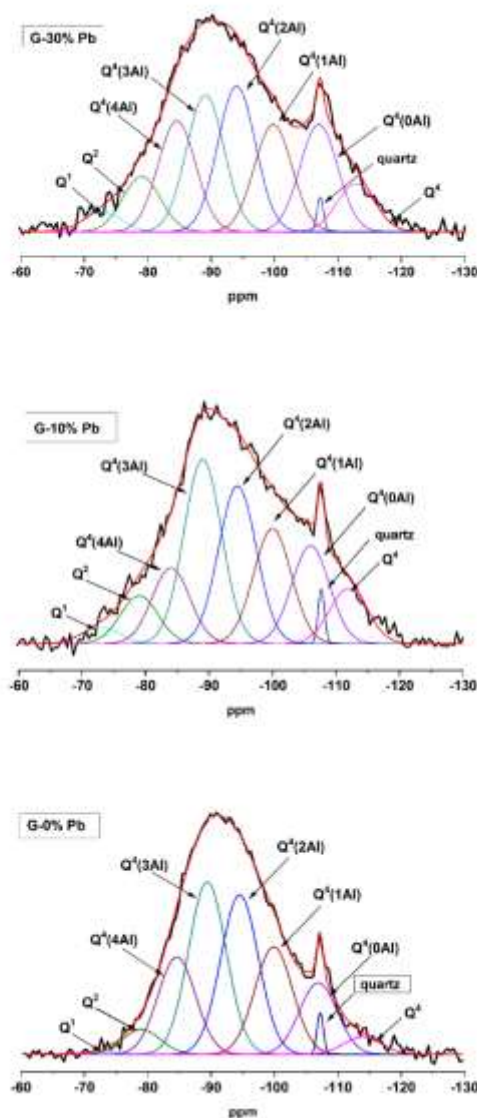


Figure 2. ^{29}Si MAS NMR spectra deconvolution of geopolymers depending on concentration of Pb added

4. Conclusion

In this paper, structural changes of fly ash-based geopoly-

mers induced by a large amount of lead (up to 30%) were examined by means of XRD and ^{29}Si MAS NMR analysis.

XRD analysis confirmed that crystalline phases, from the initial fly ash, as well as secondary calcite, were present in the fly-ash based geopolymers.

In the geopolymer samples containing lead, nitratine was detected. In addition, the appearance of new peaks, attributed

to Pb-phases was observed. In the sample with 10% Pb added, lead silicate Pb_5SiO_7 was noticed, while in the sample with 30% Pb added the presence of one more phase, probably lead silicate hydroxide was observed. The occurrence of new phases indicated that Pb formed chemical bonds with the geopolymers. Furthermore, this points to the fact that the mechanism of Pb immobilization is the chemical nature.

Table 2. ^{29}Si MAS NMR spectra deconvolution of geopolymers depending on the concentration of Pb added (Figure 2)

Pb concentration		Q ^{1*}	Q ^{2*}	Q ^{4(4Al)}	Q ^{4(3Al)}	Q ^{4(2Al)}	Q ^{4(1Al)}	Q ^{4(0Al)}	quartz**	Q ^{4**}
0 %	δ (ppm)	-72.95	-79.00	-84.50	-89.40	-94.00	-99.30	-106.4	-107.0	-112.7
	w (ppm)	7	7	7	7	7	7	7	1.2	7
	area (%)	0.4	3.78	14.72	26.1	24.13	16.26	10.83	1.08	2.67
10 %	δ (ppm)	-72.90	-79.00	-84.58	-89.40	-94.50	-99.87	-106.9	-107.2	-113.9
	w (ppm)	7	7	7	7	7	7	7	1.2	7
	area (%)	2.12	6.3	9.97	24.27	20.78	15.14	12.88	1.24	7.28
30 %	δ (ppm)	-72.9	-79.00	-84.74	-89.98	-94.96	-99.97	-106.8	-107.2	-113.9
	w (ppm)	7	7	7	7	7	7	7	1.2	7
	area (%)	2.66	7.5	15.04	18.58	19.76	14.57	14.57	0.8	6.52

* Signals associated with less condensed silicon species, Q¹ and Q²

** quartz and other Q⁴ silicon unit

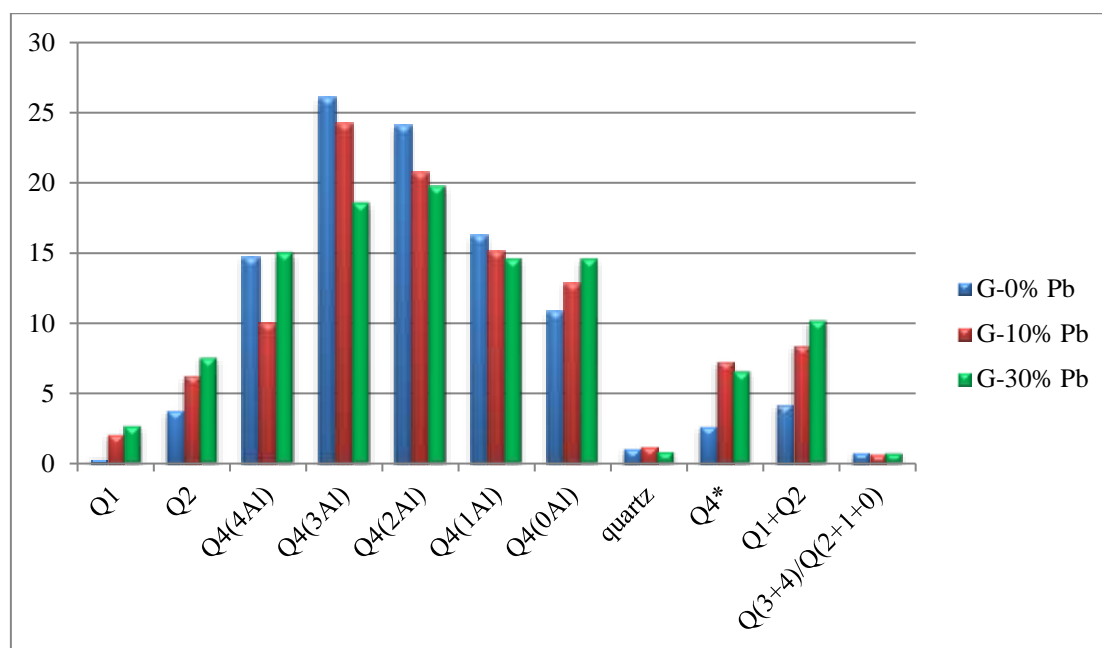


Figure 3. Qⁿ distribution of fly ash-based geopolymers depending on concentration of Pb added

The results of ^{29}Si MAS NMR spectra deconvolution of geopolymers with and without Pb added showed that the Pb addition led to an increase in the fraction of less condensed Q¹ and Q² structural units.

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Strukturne promene geopolimera na bazi elektrofilterskog pepela izazvane velikom količinom olova

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Apstrakt: Geopolimeri se smatraju mogućim tehnološkim i ekološkim rešenjem za efikasnu imobilizaciju teških metala i drugih toksičnih materijala. U ovom radu su ispitivane strukturne promene geopolimera izazvane velikom količinom olova (do 30%). Geopolimeri su sintetisani alkalnom aktivacijom elektrofilterskog pepela korišćenjem natrijum silikata modula 1,5 kao alkalnog aktivatora. Olovo je dodato u obliku soli rastvorljive u vodi – olovo-nitrata, tokom sinteze geopolimera. Promene strukture geopolimera usled dodavanja Pb određivane su XRD i ^{29}Si MAS NMR analizom. XRD analiza je potvrdila da je dodavanje Pb dovelo do formiranja novih Pb-faza, što je pokazalo da Pb formira hemijske veze sa geopolimerima. ^{29}Si MAS NMR analiza je pokazala porast slabije umreženih Q^1 and Q^2 strukturnih jedinica geopolimera sa dodatkom Pb.

Ključne reči: geopolimer, elektrofilterski pepeo, imobilizacija, olovo
