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Influence of Na_2O/Al_2O_3 and SiO_2/Al_2O_3 ratios on the immobilization of Pb from electric arc furnace into the fly ash based geopolymers

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Abstract. Electric arc furnace dust (EAFD) is by-product of iron and steel scrap remelting in electric arc furnace. This waste is classified as a hazardous material due to the leaching of heavy metals into the environment. Geopolymeryzation of fly ash into construction materials called - geopolymer is a considering as a possible way of immobilization of heavy metals from EAFD. Mechanical properties and chemical stability of geopolymers are mostly determined by the ratio between main oxides, Na_2O, Al_2O_3 and SiO_2. In this paper we have investigated possibility of Pb immobilization from EAFD into the fly ash based geopolymer. Geopolymers are synthesized at different Na_2O/Al_2O_3 and SiO_2/Al_2O_3 ratios. Compressive strength and chemical stability of geopolymers with respect to the leaching of Pb in water according to the EN 12457 regulation were tested. The results have shown that Na_2O/Al_2O_3 and SiO_2/Al_2O_3 ratios have influence on the compressive strength of geopolymers and Pb release in the water environment.

Key words: geopolymerization, lead, fly ash, electric arc furnace

Introduction

Geopolymers (inorganic polymer) are environmentally friendly materials possessing very good mechanical properties. Besides, they exhibit a high resistance when are exposed to the elevated temperature and durability in a different aquatic conditions. Geopolymers are formed as results of reaction of alumosilicate material with high alkali solutions. The solid alumosilicate material for geopolymerisation reaction may be Al-Si reach natural or waste material, (Jaarsveld and Deventer 1999; Xu 2002; Xu et al. 2002; Maragkos et al.,2009). Due to the high content of amorphous silica and alumina, fly ash, granulated blast furnace slag and metakaolin were widely used as a binder for geopolymer synthesis. Depending on the chemical composition of source materials, geoplymers are generally classified in the two groups:

- Geopolymers synthesized using a source materials rich in Ca like blast furnace slag and
- Geopolymers synthesized using source materials rich in Al and Si with a low Ca contents like metakaolin and fly ash, (Khale and Chaudhary 2007).

Besides binder, alkali solution is necessary for geopolymerizations process. Alkali solution is mostly the mixture of concentrated MOH solution and water glass i.e. M_2SiO_3 solution ((where, M is Na^+ or K^+ ion). Geopolymers properties depend on the ratio of main oxides in a geopolymer mixture, M_2O/Al_2O_3, SiO_2/Al_2O_3 and H_2O/M_2O, solid to liquid ratio, temperature and time of curing. Source of M_2O in geopolymer mixture are MOH and M_2SiO_3 solutions, while SiO_2 come from M_2SiO_3 solution and solid raw material (fly ash, metakaolin, granulated blast furnace slag). Besides, solid raw material is source of Al_2O_3, as well. So, the ratio of M_2SiO_3/MOH in alkali solution and solid/liquid ratio determine the M_2O/Al_2O_3, SiO_2/Al_2O_3 and H_2O/M_2O ratios.

Utilization of fly ash in the environmentally useful

product, deserve a special attention from the standpoint of environmental protection. Namely, fly ash may be successfully used as replacement for traditionally used cement in a construction. Moreover, especially attention is given to the geopolymer ability to immobilize toxic waste (heavy metals containing waste, nuclear waste, waste waters), (Jaarsvald et.al., 1998). In this sense, immobilization of electric arc furnace dust in the fly ash based geopolymers may be potential solution for utilization of metallurgical waste and prevention of environmental degradation.

Electric arc furnace dust (EAFD) is hazardous solid waste generated during remelting of iron and steel scrap in electric arc furnace. EAFD mainly contain Fe, but considerable amount of heavy metals is present, as well. According to the, Montenegrin environmental regulations, EAFD are classified as a hazardous waste due to the high leaching of heavy metals in the environment.

In this paper we have investigated the possibility of immobilization of Pb from EAFD in the fly ash based geopolymers with respect to the change of Na_2O/Al_2O_3 and SiO_2/Al_2O_3 ratios. Change of compressive strength of geopolymers with change of oxides ratios was investigated, as well.

Materials and Methods

Fly ash and EAFD were supplied from coal faired power station in Pljevlja and steel factory Niksic in Montenegro, respectively. Its chemical composition is given in the Table 1.

Table 1. Chemical composition of fly ash and heavy metals content in EAFD

FA		EAFD	
Content %		Heavy metals content, %	
SiO_2	49.45	Zn	26.6
Fe_2O_3	5.23	Pb	3.45
Al_2O_3	21.77	Cr	0.52
TiO_2	0.66	Cu	0.56
CaO	13.34	Cd	0.096
Na_2O	0.46		
ZnO	$4.5 \cdot 10^{-3}$		
MgO	1.29		
MnO	0.02		
P_2O_5	0.24		
K_2O	1.4		
LOI*	4.35		

*Loss on ignition.

Geopolymers were synthesized by mixing of alkali solutions with binder (fly ash) in a ratio of solid/liquid of 1. EAFD was mixed with fly ash in a ratio 1:9. Alkali solutions used in this experiments were concentrated NaOH solution and sodium water glass. Concentrated NaOH solutions (7,10 and 13 M) were prepared by dissolving of solid NaOH in water, the day prior to use. Commercial sodium water glass (Na_2O = 8.5%, SiO_2 = 28.5%, density of 1.4 kg/m^3), was used. The mass ratio between NaOH solution and water glass in a alkali mixture were 1, 1.5 and 2.

Composition of geopolymer mixtures is given in a Table 2.

Table 2. Details of geopolymer mixes.

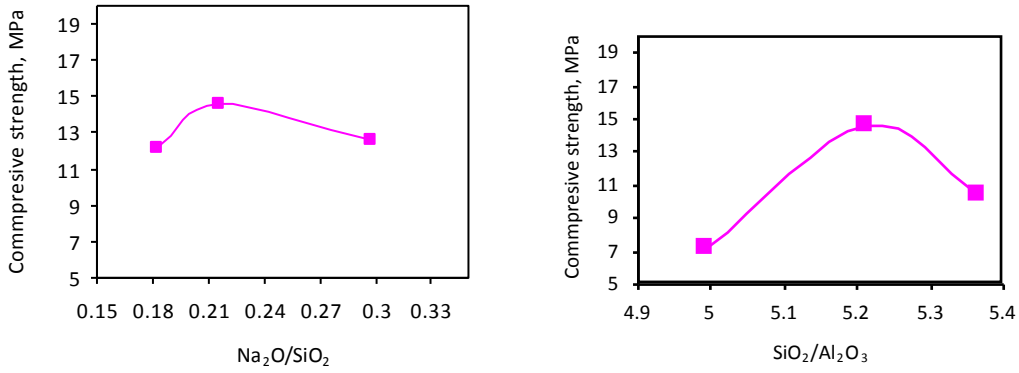
No	Na_2O/SiO_2	SiO_2/Al_2O_3
1	0.182	5.211
2	0.216	5.211
3	0.297	5.211
4	0.352	4.99
5	0.26	5.362

Geopolymers were synthesised in the way that EAFD was mixed with concentrated NaOH solution for 10 min. Than water glass was added and nixing was continued for few minutes. At the end, fly ash was adedded and mixing was prolongated for 10 min. As a result, geopolymer paste wes formed. Paste was casted in cylindrical mould and cured in the oven at the 65^{0}C for 48 h. After this time, the specimens were cooled down, removed from mould and left to stay additional 28 days at ambient temperature. Obtained geopolymer samples were tested on compressive strength Efficiency of Pb immobilization was evaluated using EN 12457-2 test. For the purpose of this test, geopolymer specimens were crushed and pulverized and mixed with a deionised water at the solid : liquid ratio 1: 10 for 24 h. After the tests, solid and liquid phases were separated by filtration and the eluates are acidified with nitric acid to the value of pH less than two. Metals content was determined using ICP-OES (Spectro Arcos).

Results and Discussion

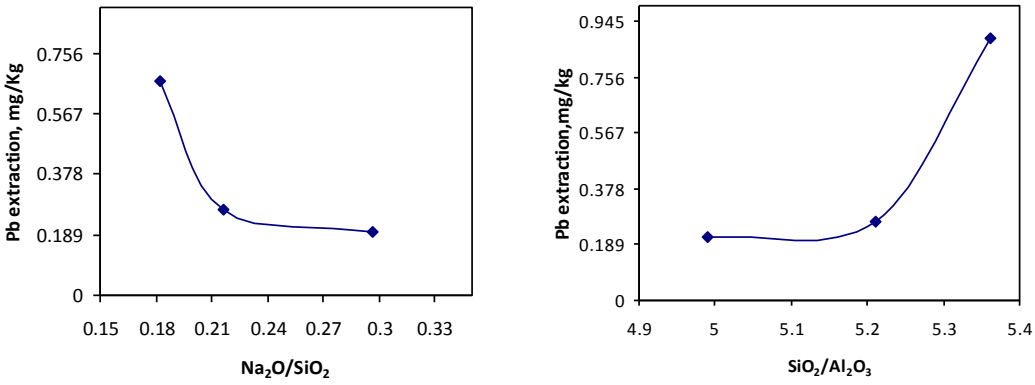
Changes of compressive strength of fly ash-EAFD geopolymers in dependence of Na_2O/SiO_2 and SiO_2/Al_2O_3 ratios are given in the Fig. 1(a) and (b), respectively. It is evident that compressive strength of geopolymers increase with increase of Na_2O/SiO_2 to the value of 0.216 and SiO_2/Al_2O_3 to the value of 0.21, while further increase of this ratios result in a decrease of compressive strength.

Possibility of Pb immobilization from EAFD in the fly ash based geopolymer depending on the Na_2O/SiO_2 and SiO_2/Al_2O_3 ratios changes was evaluated through Pb extraction in water eluate after the 12457-2 test. The results have shown that Na_2O/SiO_2 and SiO_2/Al_2O_3 ratios have influence on the Pb release from geopolymers (Fig. 3 and 4). It is evident that increase of Na_2O/SiO_2 ratio cause decrease of Pb extracted out. This could be associated with pH value of suspension during the EN 12457-2 test. Namely, increase of Na_2O/SiO_2 rate result in an increase of pH value of suspension from 10.8 to the 11.9 and decreasing of Pb dissolution from geopolymer matrix. In second case, increase of SiO_2/Al_2O_3 causes



(a) (b)

Fig. 1. Change of compressive strength of fly ash-EAFD geopolymers versus, Na_2O/SiO_2 ratio, (a) and SiO_2/Al_2O_3 ratio(b)



(a) (b)

Fig. 2. Change of Pb extraction versus Na_2O/SiO_2 ratio (a), and SiO_2/Al_2O_3 ratio (b), from fly ash-EAFD geopolymers

increase of Pb extraction from geopolymer matrix.

Besides, it is evident that at Na_2O/SiO_2 and SiO_2/Al_2O_3 ratios of 0.216 and 5.211, drastic changes in percentage of Pb leached out occur.

Conclusion

Compressive strength of fly ash-EAFD geopolymers is influenced by the change of Na_2O/SiO_2 and SiO_2/Al_2O_3 ratios. It increases with increase of Na_2O/SiO_2 and SiO_2/Al_2O_3 ratios to the values 0.216 and 5.211, respectively. Increase of this ratios above the mentioned values result in a decrease of compressive strength of both, fly ash-EAFD geopolymers.

Immobilization of Pb from EAFD is also strongly affected by the change of these ratios. Increase of Na_2O/SiO_2 ratio result in smaller Pb percentage leached out and better immobilization of Pb in the FA-EAFD geopolymer matrix. On the other side, increase of SiO_2/Al_2O_3 ratio causes the higher Pb percentage leached out and worse immobilization of Pb.

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