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# LEACHING OF THE POTENTIALLY TOXIC POLLUTANTS FROM COMPOSITES BASED ON WASTE RAW MATERIAL

The disposal of fly ash generated in coal based power-plants may pose a significant risk to the environment due to the possible leaching of hazardous pollutants, such as toxic metals. Also, there is a risk of leaching even when fly ash is built in the construction composites. Fly ashes from various landfills were applied in several composite samples (mortar, concrete and brick) without any physical or thermal pre-treatment. The leachability of the potentially toxic pollutants from the fly ash based products was investigated. The leaching behavior and potential environmental impact of 11 potentially hazardous elements was tracked: Pb, Cd, Zn, Cu, Ni, Cr, Hg, As, Ba, Sb and Se. A detailed study of physico-chemical characteristics of the fly ash, with an accent on trace elements and the chemical composition investigation is included. Physicochemical properties of fly ash were investigated by means of X-ray fluorescence, differential thermal analysis and X-ray diffraction methods. Scanning electron microscopy was used in microstructural analysis. The results show that most of the elements are more easily leachable from the fly ash in comparison with the fly ash based composites. The leaching of investigated pollutants is within allowed range thus investigated fly ashes can be reused in construction materials production.

Keywords: fly ash; construction materials; leaching test; toxic pollutants; possible reuse.

The properties of coal fly ash are strongly dependent on the geological origin and the combustion process of the coal in the power plant. It is important to characterize regional fly ash in detail to ascertain its potential uses as raw material in the production of high value products [1,2].

Over 40 million tons of fly ash is produced annually in Europe [3]. European Union regulations are focused on the recycling of coal combustion fly ash into added-value products [4]. One method of addressing this is the synthesis of economically low-cost and environmentally safe materials that offer mechanical properties close to those of classic building materials (cement [5], concrete [6], brick [7] and lime [8]).

According to the statistic data belonging to the "Javno preduzeće Elektro-privreda Srbije" (JP EPS),

utilization of the fly ash can be targeted at least to an extent of 40-50% in various fields worldwide. However, this percentage in Serbia is significantly lower. On the other hand, Serbian thermal plants electro-filters produce gases with extremely high solid particles concentrations: 1000-2000 mg/m³ [9]. These amounts are significantly higher than the minimal amount allowed by European Union (50 mg/m³) [3]. There is an ongoing project concerning innovation and restoration of electro-filters, originally introduced in 2003 and ought to be finished by the end of the 2012. Approximately 6-7 millions tons per year of fly ash are being produced in Serbia and the main goal is to find economic solution for reuse of the fly ash in the construction industry and in over-all mass-production of building materials.

As it was previously mentioned, fly ash represents a huge ecological and financial contemporary problem since the global generation of this by-product has already reached an alerting quantity of approximately 175 million tons per year [10]. For disposal of the fly ash, wide ground areas formerly decreed for

agrarian or building purpose are required. Fly ash depots are constantly increasing in size [11]. On one hand, there are large amounts of fly ash generated from thermal coal-fired power plants, and on the other hand, a small proportion of this waste material is used for a variety of purposes. Fly ash production leads to the problem of disposal as well as environmental damage by causing air and water pollution by leaching [12]. Namely, fly ash is regarded as a toxic material owing to its high concentration of leachable pollutants (usually metals) and, in some cases, to the presence of various organic compounds [13,14].

Since fly ash is often contaminated with the mentioned pollutants, the evaluation of the risk of leaching during fly ash application is of particular importance [15]. Laboratory leaching tests are common tools for evaluation of long-term impact of contaminated materials on the soil-groundwater pathway, as they determine the source term as an expression of release potential of water soluble contaminants during the use or disposal of waste materials [16,17].

Different leaching tests have been developed to characterize and assess the constituents that can be released from waste materials. Namely, there is a wide range of standardized leaching tests with variable complexity, reagents, temperature, liquid to solid ratio (L/S), contact time, along with some others non-standardized but appropriated for specific purposes: leaching tests using deionised water as leachant (no pH buffering) [18], column/flow-through tests provide valuable data on the release pattern of elements under uncontrolled pH conditions [19], toxicity characteristic leaching [20], etc. All of the procedures simulate specific scenarios of the leaching behavior of coal ash. The obtained results often cannot be directly compared; however they can depict a view of general mobility of toxic elements within fly ash [21,22].

This paper provides study on physicochemical characteristics and leachability of the 11 potentially hazardous elements (Pb, Cd, Zn, Cu, Ni, Cr, Hg, As, Ba, Sb and Se) of the fly ash samples originating from various landfills located besides Serbian coal-based power plants and fly ash based composites (concrete, mortar and brick).

## MATERIALS AND METHODS

The properties of fly ash from Serbian power plants were investigated, evaluated and discussed. A detailed study on the chemical composition of the appearing phases, trace elements and the possible leaching of the contaminant elements were accented along with comparison with leaching test performed on fly ash based building composites. Investigated fly

ashes have different origin and therefore their chemical and mineralogical compositions vary [23].

*Sampling of the material.* Five different types of investigated fly ash are here subsequently labeled as FA1, FA2, FA3, FA4 and FA5. The fly ash samples were landfilled without any previous treatment, but not mixed with household or similar type of waste. A sample of 500 kg of each fly ash was randomly taken from a landfill and re-sampled by the quarter method. Thus, 50 kg samples were obtained. The samples were preserved in hermetically sealed boxes until further analysis.

For further analyses, *i.e.*, leaching tests, fly ash based composites (fly ash mortar, fly ash concrete and fly ash brick) were prepared. Composites were prepared with five different fly ash types (FA1-5). Leaching properties of fly ash based composites were compared with "standard" composites. Fly ash replacement coefficient was 30% in concrete and mortar samples and 50% in brick samples.

The concrete samples (standard and fly ash based) were prepared with Portland cement PC 42.5R *Lafarge* as the bonding agent and river sand „*Moravac*" (grain size fractions used: 0/4, 4/8, 8/16, 16/32 mm) as the aggregate. Concretes were mixed for 8 min in a laboratory RILEM-cem mixer and, afterwards, shaped in 15 cm$^3$ moulds. After 7 days of curing in a climate chamber at 20 °C and humidity 60%, the samples were demoulded and stored for another 21 days under the same conditions as in the climate chamber.

The mortar samples (standard and fly ash based) were prepared with Portland cement PC 42.5R *Lafarge* as the bonding agent and river sand „*Moravac*" (grain size fraction used 0.4 mm) as the aggregate. The uring procedure was the same as in the case of the concrete specimens.

The brick samples (standard and fly ash based) were prepared with "brick" clay (containing following oxides: $SiO_2$ - 49.27%; $Al_2O_3$ - 29.31%; $Fe_2O_3$ - 3.2%; $TiO_2$ - 0.28%; CaO - 0.58%; MgO - 1.43%; $Na_2O$ - 0.2%; $K_2O$ - 1.85%). Preparation of each mixture was conducted in ball-mill with "wet" working regime. The mixture was grinded for 4-6 h until residue on sieve was 2-3% according to DIN 100. Afterwards, water/moist excess was removed. The composite samples were cylindrically shaped (radius 12 mm, length 250 mm). Samples were dried in a climate chamber at 105 °C before firing in the laboratory furnace, in oxide atmosphere, at 1150 °C with 1.5 h delay at maximum temperature.

*Chemical composition and trace elements analysis.* Analysis of chemical constituents of the fly ash was performed by X-ray fluorescence (XRF spectro-

photometer ED 2000 - Oxford). A representative sample of about 500 g was previously dried at 60 °C. The sample was used for determination of the major, minor and trace elements. The XRF resolution used was: for very light elements (Na-K) 170 keV; for medium elements (Ca-Mo) 185 keV; for heavy traces: 185 keV. The obtained results were averaged. The fly ash international reference material obtained from the National Institute of Standards & Technology, USA, was applied in verification of the accuracy of results of the analysis.

*Loss of ignition.* The loss of ignition (LOI) was determined by the weight difference between room temperature and 1000 °C. The maximum temperature was held constant for 4 h.

*Leaching test.* Potential mobility of trace elements from the fly ash samples was determined by means of the batch leaching test, procedure conducted according to Serbian Standard SRPS EN 12457 (1-4) (equivalent to EN 12457 (1-4)) [24]. Water was chosen as leachant after the assumption that atmospheric water (*i.e.*, rain or snow) was the main resource of penetration through open fly ash landfill, same as it will be one of main real-time leachants once investigated composites are built in exterior elements of structural objects. Samples were milled in order to provide greater contact surface between the media, and more likely leaching of present trace elements. The leaching test was performed at a liquid/solid ratio of 10 L/kg with a stirring time of 24 h and deionized water as the leachant. A blank sample was prepared using deionized water. At the end of the leaching process, the samples were filtered, and the content of the trace elements in the leachates were determined by means of inductively coupled plasma atomic emission spectrometer ICP-AES (Perkin-Elmer Optima 5300 DV). The leaching test was per-

formed on standard and fly ash based concretes, cement-mortars and brick samples. Results of the leaching test performed on the fly ash based composites were compared with results obtained on standard concretes/cement-mortars/bricks.

*Differential thermal analysis.* The differential thermal analysis (DTA) of the fly ash was performed with a *Shimadzu* DTA-50 apparatus. Approximately 30 mg of a sample, which was previously homogenized and then dried for 6 h at 105 °C, was used for a DTA testing along with $\alpha$-$Al_2O_3$ (corundum) powder as reference sample. The sample was heated under an air atmosphere from 20 up to 1100 °C at heating rate of 10 °C/min.

*X-ray diffraction analysis.* Homogenized and dried (for 6 h at 105 °C) fly ash samples were analyzed by X-ray powder diffraction (XRD). XRD Patterns of the fly ash samples were obtained using a Philips PW-1050 diffractometer with a graphite monochromator, NaI(Tl) detector and $\lambda CuK\alpha$ radiation and a step/time scan mode of 0.05°/1s.

*Microstuctural analysis.* The microstructure of the fly ash samples was characterized by scanning electron microscopy (SEM) using a *JEOL* JSM-6390 Lv microscope. SEM accelerating voltage was 15 kV. The original fly ash powder without further grinding was used as sample. The samples were covered with gold powder for better reflection to be obtained and measurements performed.

## RESULTS AND DISCUSSION

The chemical compositions of investigated fly ashes (FA1-5) originating from various power-plants were mutually compared and correlated with characteristics of European fly ash (EFA) [25]. The results are given in Table 1.

*Table 1. Chemical composition of the fly ashes*

| Oxide content, wt.% | FA1 | FA2 | FA3 | FA4 | FA5 | Standard deviation | EFA [25] |
|---|---|---|---|---|---|---|---|
| $SiO_2$ | 57.49 | 53.49 | 58.32 | 55.28 | 59.73 | 2.48 | 41.1-59.6 |
| $Al_2O_3$ | 17.72 | 21.28 | 18.08 | 17.43 | 20.97 | 1.87 | 17.6-35.6 |
| $Fe_2O_3$ | 10.48 | 6.20 | 6.85 | 10.31 | 5.99 | 2.24 | 2.6-16.0 |
| $TiO_2$ | 0.52 | 0.56 | 0.57 | 0.52 | 0.57 | 0.02 | 0.5-2.6 |
| CaO | 6.96 | 7.61 | 8.71 | 7.96 | 5.83 | 1.09 | 0.5-11.8 |
| MgO | 1.98 | 2.74 | 2.30 | 2.28 | 2.21 | 0.27 | 0.8-3.8 |
| $P_2O_5$ | 0.02 | 0.03 | 0.02 | 0.029 | 0.023 | 0.004 | 0.1-1.7 |
| $SO_3$ | 1.06 | 0.78 | 1.29 | 0.85 | 0.48 | 0.30 | 0.1-8.6 |
| $Na_2O$ | 0.36 | 0.44 | 0.50 | 0.40 | 0.41 | 0.05 | 0.1-1.2 |
| $K_2O$ | 0.59 | 1.21 | 1.16 | 0.55 | 1.18 | 0.34 | 0.4-4.0 |
| MnO | 0.03 | 0.03 | 0.03 | 0.026 | 0.030 | 0.002 | - |
| $CO_2$ | 0.09 | 0.25 | 0.11 | 0.28 | 0.07 | 0.09 | 0.6-7.6 |
| LOI | 2.94 | 4.91 | 1.84 | 4.51 | 2.92 | 1.26 | 1.1-8.1 |

The values given for the EFA were used both as a reference to check the quality of investigated fly ashes (FA1-5) and the accuracy of the analytical XRF method applied in this study. The obtained experimental results for FA1-5 were similar to the certified composition (EFA), namely the results are fitting in the result range given for EFA [25]. Therefore, the analysis of the major and minor elements verifies both usability of investigated fly ashes and gives positive results on testified reliability of XRF method for the given purpose.

The chemical analysis (Table 1) shows that, as in the case of EFA, all of investigated fly ashes consisted mainly of silica (53.49-59.73%), alumina (17.43--21.28%) and iron oxides (5.99-10.48%) depending on the origin of a fly ash.

Relatively low loss of ignition in weight (LOI) was noted when fly ash samples were heated at 1000 °C for 4 h. It was attributed to the fact that organic matters are not present in the investigated fly ash samples. FA2 had the highest LOI (4.91%). Results of LOI for FA1-5 were within the result range given for EFA [25,26].

$SiO_2$ content was higher than 50%, while content of $Al_2O_3$ was in the range 17-25% in all investigated fly ash samples. Thus, samples FA1-5 can be classified as alumino-silicate ashes and they show pozzolanic behavior. Calcium oxide content of sulfates was relatively low.

Concentrations of trace elements in FA1-5 are shown in Table 2. It was noted that all investigated fly ashes had either lower or "in range" trace elements concentrations when compared with EFA. Even though trace elements are present as a relatively small fraction in the fly ash samples, they are investigated due to their tendency of accumulation, long life and high toxicity to environment. Because trace elements can leach out and contaminate soil, as well as surface and groundwater, their study is important for environ-

mental protection [27,28].

Processes taking place during fly ash thermal treatment from 20 up to 1100 °C were identified by means of DTA method. DTA curves are given in Figures 1 and 2.

All DTA curves have a small peak at approximately 200 °C, which corresponds to the volatilization of the water mechanically bonded in form of $H_2O$ molecule [29]. Thus, the first dimensional change occurred between room temperature and 100 °C and it was related to the loss of humidity. In the temperature interval from 100 to 450 °C hydration water was altogether lost. The peak showing at approximately 500 °C is exothermic and characteristic for fly ash [30,31]. Exact peak values are as follows: (FA1) 533.85; (FA2) 522.45; (FA3) 522.54; (FA4) 503.80; (FA5) 533.26 °C. Exothermic hump corresponds to the transformation of organic matter present in the fly ash samples and to the decomposition of $CaCO_3$ and the burning of residual coal present in the fly ash [30]. The second peak is endothermic and distinctly visible for fly ashes FA1-4. This peak is at approximately 900 °C and it is induced by presence of alumino-silicates [31]. FA5 does not show second endothermic peak.

Differential thermal analysis showed a certain change above 900 °C in case of FA1-4 which pointed to the additional structural changes occurring with further increasing of the temperature and possible initiation of sintering.

Results of the mineralogical analysis are given in Figures 3 and 4.

Identified crystalline major phases present in the FA were aluminosilicate glass, quartz and mullite. The only defined peaks on diffractograms relate to quartz [32]. XRD Diffractograms highlighted high amount of amorphous matter within all investigated fly ash samples. The background hump between 10 and 40° in the X-ray spectrum provided additional evidence of the presence of an amorphous phase [33]. Magnetite,

*Table 2. Chemical analysis of trace elements in the fly ashes*

| Trace element, mg/kg | FA1 | FA2 | FA3 | FA4 | FA5 | Standard deviation | EFA[25] |
|---|---|---|---|---|---|---|---|
| Pb | 15.5 | 19.1 | 24.4 | 13.7 | 17.6 | 4.09 | 40-176 |
| Cd | 0.1 | 0.0 | 0.2 | 0.2 | 0.2 | 0.09 | 1-6 |
| Zn | 52.3 | 36.1 | 56.6 | 49.9 | 27.2 | 12.31 | 70-924 |
| Cu | 52.4 | 26.0 | 35.9 | 53.7 | 30.4 | 12.71 | 39-254 |
| Ni | 21.7 | 41.3 | 50.9 | 23.6 | 31.7 | 12.28 | 49-377 |
| Cr | 97.6 | 148.1 | 135.1 | 105.7 | 167.4 | 29.11 | 47-281 |
| Hg | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | <0.01-1.40 |
| As | 10.6 | 9.9 | 132.2 | 9.9 | 17.1 | 53.89 | 22-162 |
| Ba | 51.5 | 53.3 | 86.2 | 49.1 | 61.7 | 15.20 | 311-3134 |
| Sb | 0.5 | 0.1 | 1.1 | 1.6 | 0.5 | 0.59 | 1-120 |
| Se | 1.8 | 1.7 | 1.4 | 1.4 | 0.7 | 0.43 | 3-30 |
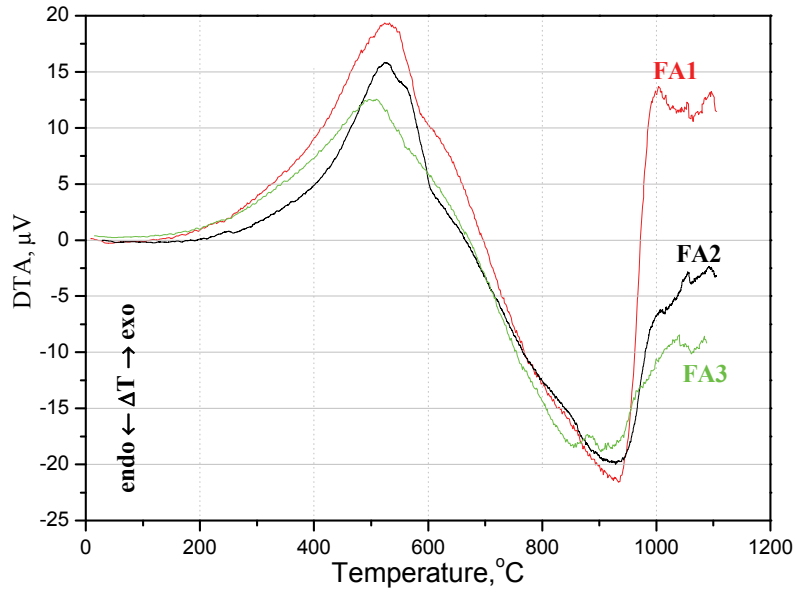
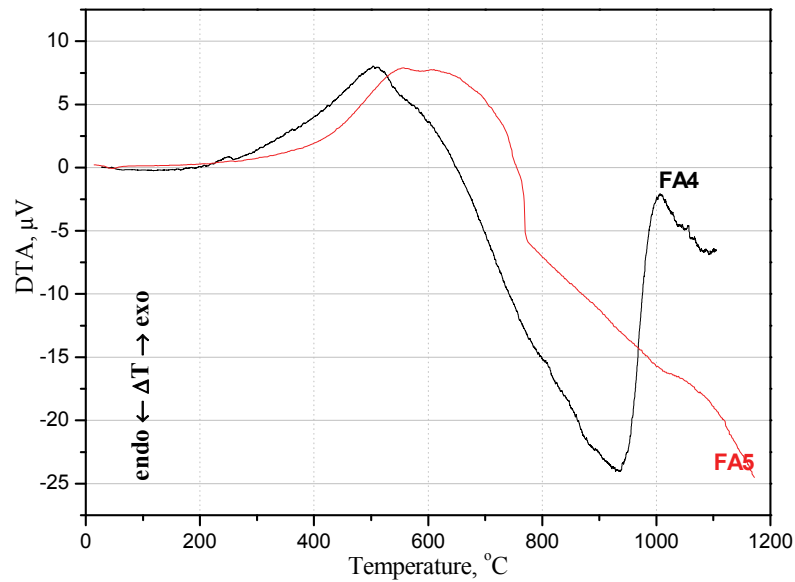Figure 1. DTA Curves of the FA1-3 fly ashes.



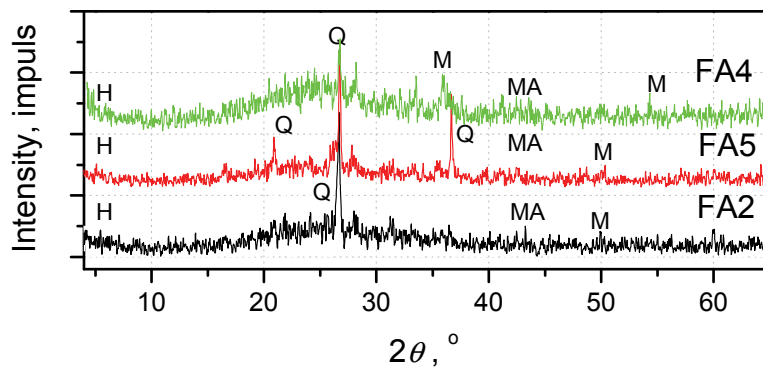Figure 2. DTA curves of the FA4-5 fly ashes.



Figure 3. XRD Diffractograms of FA2, FA4 and FA5 fly ashes (quartz - Q, magnetite - MA, hematite - H, mullite - M).
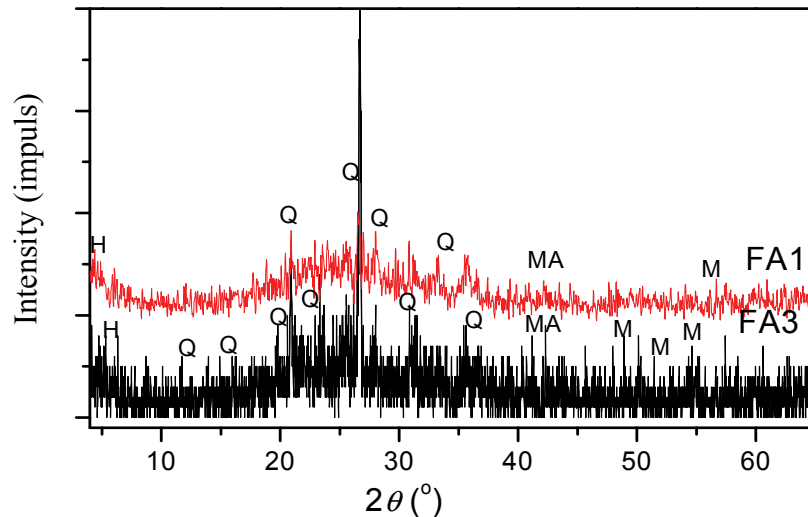
Figure 4. XRD Diffractograms of FA1 and FA3 fly ashes (quartz - Q, magnetite - MA, hematite - H, mullite - M).

hematite, fluorite and anhydrite were noted in negligible amounts. The very few differences observed in the mineralogical composition might be attributed to the heterogeneity of the fly ash composition. Namely, it is possible that fly ash contacted and merged with some other matter originating either from air or ground while lying on landfill [34,35]. Sample FA1 showed the lowest level of crystallinity and it was found to be one of the fly ashes with the highest amorphous material and lowest quartz and mullite contents, while the sample FA3 had the most noticeable quartz peak on diffractograms, *i.e.*, the highest level of crystallinity.

Figures 5 and 6 show SEM images of FA1 and cement mortar composite (FA1 cement). After visual examination of the fly ash samples it was concluded that FA1 is a mixture of various grains of different size and shape, *i.e.*, different inorganic phases and possibly certain quantity of unburned organic materials [36]. FA1 was mainly composed of spherical hollow particles, as Figure 5 shows. Increased surfacial porosity of this particular fly ash grain is evident. Irregularly shaped grains and agglomerations immersed in the fly ash mixtures. These irregular grains and agglomerations usually correspond to calcite (CaCO$_3$) [37]. Pseudospheres, *i.e.*, spherical particles composed of various layers or grains were noted. Such particles usually correspond to the presence of magnetite (detected by XRD method) [38]. Extremely porous particles might correspond to unburned coal [39]. Specific needle-shaped particles are either found isolated or merged with spheres in agglomerations. According to XRD analysis these needle-like shapes correspond to the mullite crystals [40]. Quartz was also found as irregularly shaped particles [41]. In cement mortar composite (Figure 6) fly ash particles are merged with bonding agent in a so-called "cementing

zone" between aggregate grains. In such correlation, fly ash behaves like a standard pozzolanic bonding agent.
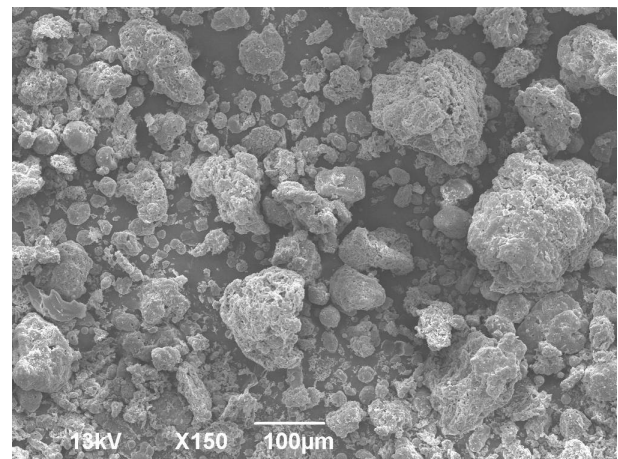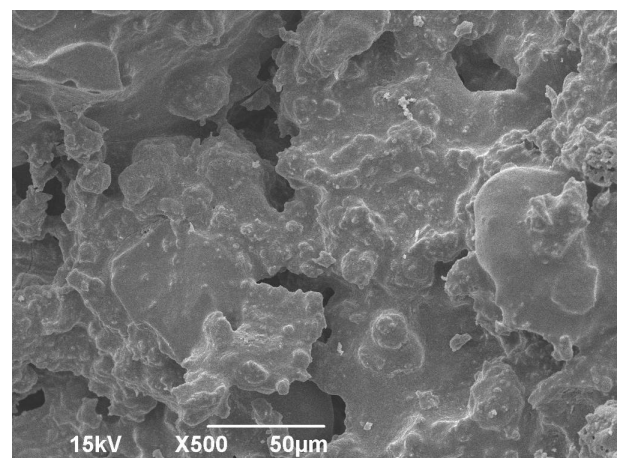


Figure 5. SEM Image of FA1; optical zoom: 150×.



Figure 6. SEM image of FA1-mortar; optical zoom: 500×.

Leaching test provided information on the leachability of various chemical constituents residing in the fly ash based composites (mortar, concrete and brick) in normal environment. Leachability of aluminium, calcium and iron was not investigated, although they are considered to be major constituents in the chemical composition of fly ash [42]. The reasons are as follows:

- Aluminium is poorly leached, particularly when considering its abundance in fly ash. Due to the slow dissolution rates of the glassy matrix and the crystalline aluminosilicate phases, low proportions of Al are leached regardless of the type of leaching test applied [42,43].

- Calcium is present in fly ash in multiple modes of occurrence (lime, anhydrite, calcite or within the glassy matrix). The leachable concentrations vary as a function of the mode of Ca occurrence in the fly ash, the proportion of each phase and the type of the leaching test performed. With only water, the amounts of Ca leached could easily fall in the 100-15,000 mg/kg range. However, calcium is not regarded as an element of concern. The presence and amount of Ca is particularly significant for the precipitation of ettringite and other secondary Ca-hydrated phases that are likely to incorporate and retain environmentally relevant elements such as As, Cr or Se. Ca controls the leachability of trace contaminants to a large extent [42,43].

- Iron becomes soluble at pH ≤1.5, but releases are of little significance in relation to the concentration in the ash. Proportions of Fe were found to be leached from fly ash are less than 10 mg/kg in near neutral and alkaline solutions, and concentrations in most cases are below the detection limit. Iron in fly ash is mainly present as magnetite mixed in various pro-portions with hematite, although a minor proportion can be assimilated in the glassy matrix. Iron oxyhydroxides are also present in fly ash. These chemical compounds are widely known to play a key role in the sorption of a number of elements of environmental concern, thus attenuating the concentrations in the leachates [42,43].

The water leaching trial showed potential mobility of elements existing in the fly ash composition: Cr, Ni, Cu, Zn, As, Se, Cd, Sb, Ba and Pb. Results of the leaching possibility of FA1-5 composites are given in Figures 7-9.

Leaching test performed on cement-mortar and fly-ash based cement mortar samples showed no presence of Cd, Hg and Ba. Concentration of Pb was higher in fly-ash based composites than in cement mortar. Overall, the leaching test showed no significant change in samples when 30% of cement was substituted with fly ash.

Leaching test performed on concrete and fly ash based concrete showed no presence of Cd and Hg in investigated samples. Ba was present in the sample FA5 concrete. The leaching test showed no significant change in samples when 30% of cement in concrete was substituted with fly ash.

Leaching test performed on brick and fly ash based bricks showed no presence of Pb, Zn, Ni, Cr in investigated samples. Hg was present in the samples FA1 and FA4 brick. Se was not present in sample FA2. The leaching test showed no significant change in samples when 50% of fly ash is added and mixed with clay in order to obtain fly ash based brick composite.

Cumulative results of leaching tests performed on various fly ash based samples are given in Figure 10.
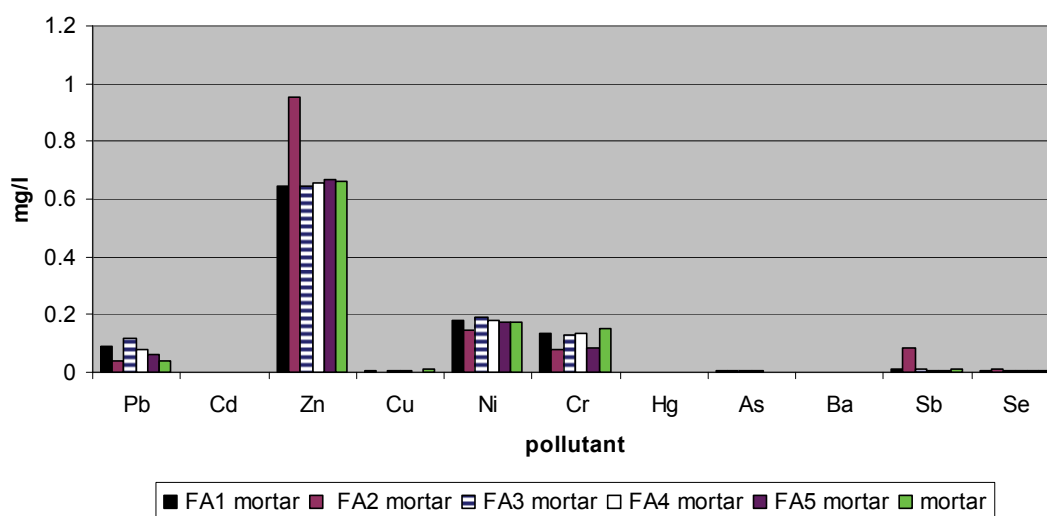


Figure 7. Leachable concentrations of elements in FA1-5 mortars and comparison with the reference (fly ash free) mortar sample.
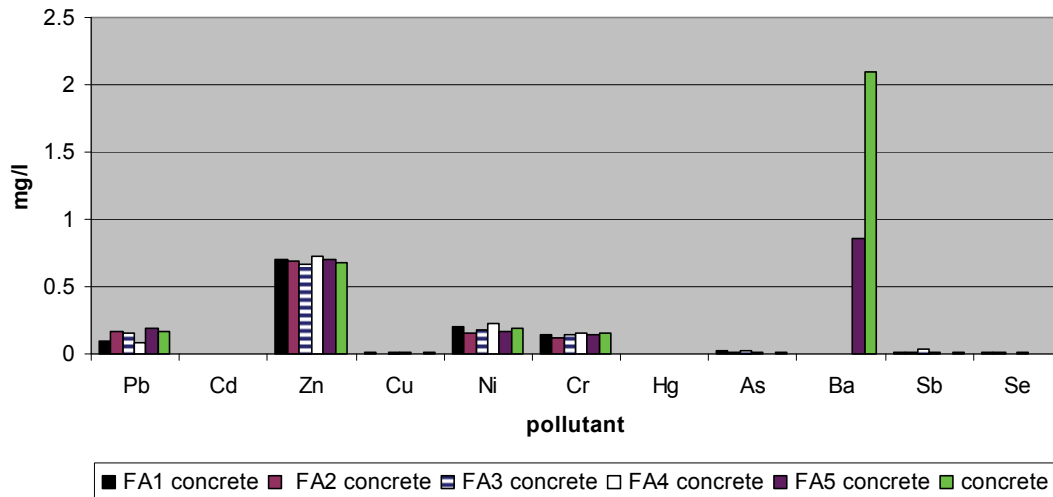
*Figure 8. Leachable concentrations of elements in FA1-5 concretes and comparison with the reference (fly ash free) concrete sample.*
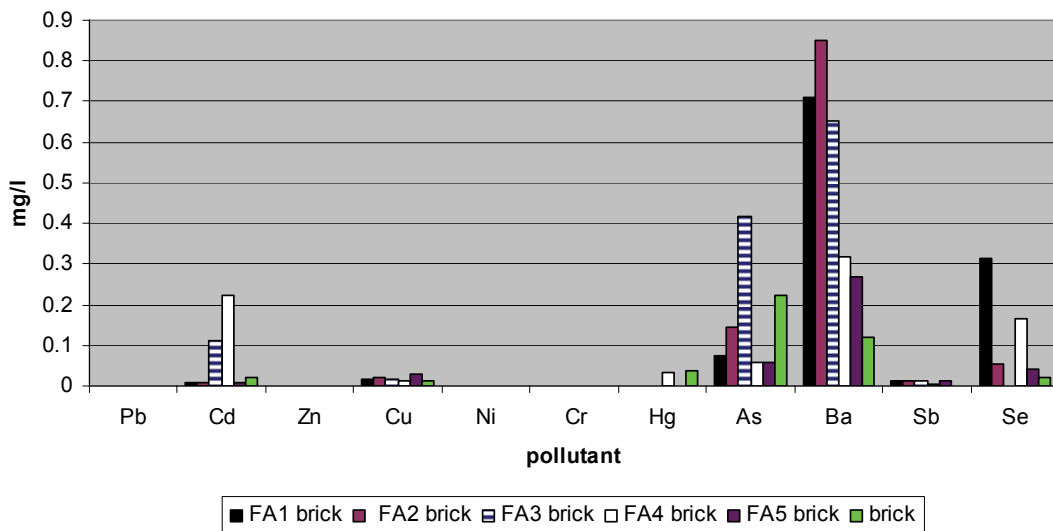


*Figure 9. Leachable concentrations of elements in FA1-5 bricks and comparison with the reference (fly ash free) brick sample.*

Lead (Pb) is associated with sulphides in coal [44]. The majority of Pb in fly ash is placed within the internal glassy matrix and therefore not readily leached. Pb is highly insoluble and immobile irrespective of the type of leaching test applied [44]. Thus, the leachable concentrations remain at very low levels, as the investigation confirmed. This provides evidence that Pb leaching does not pose a risk to the environment regardless of the total content in fly ash. Cadmium (Cd) present in fly ash is consistently immobile while in neutral and alkaline conditions. Although Cd is associated with the fly ash particles surface, the concentrations leached in water rarely surpass 0.01 mg/l [44,45]. In investigated fly ash based concretes and fly ash based mortars Cd was not found. Zinc (Zn) is in sulphide association in the parent coal and its mobility in fly ash is high [46]. The leachability of Zn in water for fly ash varies widely, as showed in the in-

vestigation: leachability for concrete and mortar composites is high, while at the other hand, Zn leachabilty for brick is negligible. Copper (Cu) is assimilated within the glassy phase and not easily released [46]. Leachability of Cu was low in all investigated samples. Nickel (Ni) is distributed between the silicate fraction and the magnetic fraction of fly ash during combustion. Most of the leachable Ni is solubilised from the non-magnetic fraction [47]. Leached concentrations of Ni were present in fly ash based concrete and mortar samples, but not present in investigated bricks. Although chromium (Cr) leaching from fly ash poses a high threat to the environment (Cr is widely recognized as potentially carcinogenic and highly soluble in aqueous media [44]), the obtained levels of Cr leachability in investigated samples do not impose danger to the environment nor humans. The condensation of mercury (Hg) on fly ash particles as the wa-
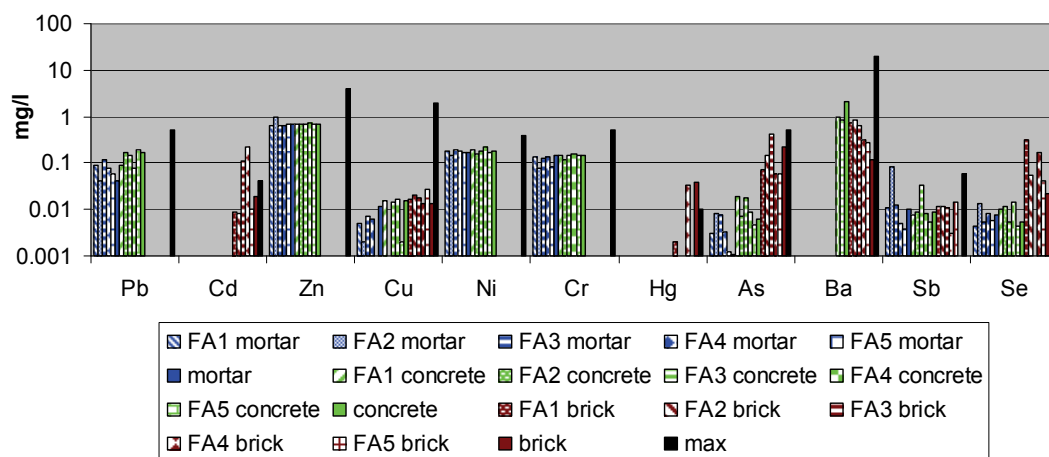
Figure 10. Cumulative results of leaching test ("max" - upper limit values of toxic metal in leachate from inert/non-harmful wastes, Standard EN 12457(1-4)2002).

ter soluble may pose a risk for of contaminating groundwater [48]. However, the concentrations of Hg in leachates are extremely low - here noticeable only in case of FA1-brick and FA3-brick. In most coals, Arsenic (As) is associated with As-bearing pyrite, which is decomposed during the combustion process. Arsenic condenses on the surface of fly ash as sparingly soluble arsenate species [44,48]. The leachability varies widely depending on the nature of the fly ash. For example FA3 brick shows noticeably higher content of As than other brick samples. Barium (Ba) in fly ash forms sparingly soluble compounds with carbonates and sulphates [48]. Investigated fly ash composites show high percentage of Ba in leachate. Antimony (Sb) in coal occurs in sulphides along with pyrite. These compounds dissociate during combustion and liberate Sb as volatile species. The leaching behaviour has been scarcely covered in the literature, although it is known that Sb is insoluble in all types of leachant [49]. Amout of Sb found in leachate of investigated composites is low. The solubility of selenium (Se) in water can be high which makes Se as an element of major concern in ash due to its mobility [50]. Level of Se in leachate is lower in concrete and mortar composites than in fly ash based brick, probably due to the additional Se imported with other brick compounds.

Leaching of investigated toxic elements was close to or below the detection limit with water used as leaching agent [51,52]. After comparing results of leaching test for fly ash composites and for original composites mortar, overall conclusion was that there is no significant difference in results and investigated fly ashes are secure for application in construction materials industry. Thus, there is equally low environmental risk posed by the presence of contaminant trace elements in the fly ash if it is used in a cons-

truction element and exposed to the action of rain and snow.

CONCLUSION

The mode of occurrence of trace elements in coal plays a primary role in the mode of occurrence in fly ash as well as the distribution of elements within ash particles. The elements enriched in the core of fly ash particles are not directly exposed to leaching, whilst surface-associated elements are more accessible to leaching in an aqueous environment. Therefore, the mode of occurrence in the parent coal controls to a large extent whether a given element will be immobile or will be easily released to the environment. Investigation of the fly ashes showed that they have negligible level of organic impurities and potentially toxic and/or leachable elements. The investigated elements - Pb, Cd, Zn, Cu, Ni, Cr, Hg, As, Ba, Sb and Se, can be regarded as being of low concern in fly ash under natural environmental conditions.

The leaching test carried out on the Serbian fly ashes denied possibility of potential transition of toxic elements if fly ash based building material is in contact with water. Therefore, atmospheric, ground or underground water contamination by migrating toxic fly ash constituents is unlikely.

The fly ash properties determined in this work could be regarded as representative of all coal fly ash produced in Serbia. Furthermore, the investigated fly ashes have excellent characteristics, which opens new frontiers for their utilization, not only in the region but worldwide. Also, this investigation opens the question of possible benefits of mechanical-activation of fly ash and economical aspect of its future application. The problem of fly ash is disclosed this study is but a first step of a larger scale investigation con-

cerning new possibilities of fly ash reapplication. The investigation will include implementation of other leaching tests, as well as various other investigation methods. The accent will be on mechanical and chemical activation of the fly ash and advantages of these methods in achieving advanced characteristics of new fly ash based composites.

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NAUČNI RAD

## LUŽENJE POTENCIJALNO TOKSIČNIH ELEMENATA IZ KOMPOZITA NA BAZI OTPADNIH SIROVINA

*Odlaganje letećeg pepela dobijenog u procesu sagorevanja uglja u termo-elektranama predstavlja veliki rizik za prirodnu sredinu zbog mogućnosti luženja opasnih čestica kao što su toksični metali. Takođe, opasnost od izluživanja je prisutna čak i kada je leteći pepeo ugrađen u konstukcioni kompozit. Leteći pepeli sa različitih deponija su upotrebljeni za spravljanje uzoraka različitih kompozita (malter, beton ili opeka) pri čemu na pepelu nije sproveden nikakav fizički ili termički pred-tretman. Ispitivano je luženje potencijalno toksičnih elemenata prisutnih u proizvodima na bazi letećeg pepela. Praćeno je izluživanje i mogući uticaj na prirodnu sredinu 11 potencijalno opasnih elemenata: Pb, Cd, Zn, Cu, Ni, Cr, Hg, As, Ba, Sb i Se. Sprovedeno je detaljno istraživanje fizičko-hemijskih svojstava letećeg pepela, sa posebnim naglaskom na ispitivanju hemijskog sastava i prisutnosti elemenata u tragovima. Fizičko-hemijska svojstva pepela su ispitivana sledećim metodama: X-ray fluorescenicija, difrakciona termička analiza i X-ray difrakciona metoda. Skenirajući elektronski mikroskop je primenjen u mikrostukturnoj analizi. Rezultati su pokazali da se većima elemenata lakše izlužuje iz pepela nego kompozita spravljenih na bazi pepela. Izluživanje svih ispitivanih toksičnih elemenata je bilo u dozvoljenim granicama što znači da se ispitivani leteći pepeo može upotrebiti u proizvodnji konstrukcionih materijala.*

*Ključne reči: leteći pepeo; konstrukcioni materijali; luženje; toksični elementi; mogućnost ponovnog korišćenja.*