# Building Materials Based on Fly Ash as Possible Solution for Reduction of Air Pollution Caused by Emissions from Power-plants

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*Most frequent environmental pollution sources are emissions caused by technological processes taking place at power-plants. High production of fly ash, the main residue in coal combustion, represents extreme hazard for the environment. Air pollution is taking proportions that require special attention, thus certain regulations are being imposed. The goal of the legislation is to reduce harmful emissions into the atmosphere by imposing standards for air quality and maximum emissions from various sources. Besides regulations, option of reapplication of fly ash in building materials industry is another solution for on-going problem. Fly ash can be used as component in cement, mortar, concrete, bricks, tiles, asphalt. Using produced fly ash capacities is sustainable solution for saving natural resources and environmental protection, but also a basis for designing new building materials from recycled nusproducts.*

**Key words**: *fly ash, pollution, regulations, building materials.*

## 1. INTRODUCTION

Industrial revolution is one of the main reasons for increasing trend of air pollution which has been recorded in the previous three centuries. Air pollution, as the most hazardous pollution, implies presence of one or more undesirable solid, liquid or gaseous substance in the air which causes threats for human kind and natural surrounding.

The type of particles emission in industrial zones depends on the industry type and the fuel used in combustion process [1].

Coal is one of the most frequently used fuels in energy plants. Smoke and sulfur dioxide from burning coal can conjugate with fog producing so-called "industrial smog". In high concentrations smog can be extremely toxic, both for humans and other living organisms.

Regulated concentrations of the main urban pollutants are given in Table 1.

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The European Union attempts to harmonize ecological legislation throughout Europe and, thus, to en sure a high level of environmental quality, especially in member countries, but also for other European citizens in the parts of Europe and beyond the EU borders. Countries are required to adopt the legislation in the field of environment and complete its application within about 10 years of post-accession [2].

The Directive of the Council 96/62 EC in assessing and managing air quality is a general legislative framework in this area. The Directive has established a group of atmospheric pollutants, which are supposed to define specific standards on air quality, within special directives-sisters. This list includes $CO_2$, $HO_2$, particular particles, lead, ozone, cadmium, arsenic, nickel and mercury. For areas where the actual level of any, one or more pollutants exceed the maximum value or limit of tolerance, obligation is to adopt and implement an action plan to achieve the maximum values in given time.

In Serbia, legislation norms for emission are being covered by the republic laws and regulations [3,4].

For pollution prevention, as well as remediation of already polluted air, it is necessary to develop appropriate strategies which must include local, regional and global factors. These strategies include reparation of electro-filters in power-plants, introduction of "pure" rather than "dirty" technologies, rational use of energy (thus reducing consumption of fossil fuels) and recycling of the waste materials obtained as nusproducts during technological processes.

*Table 1. Main air pollutants in urban areas.*

| Type of pollution | Source of pollution | Concentration, μg/m$^3$ |
|---|---|---|
| *Solid particles - ash* | Fuel combustion | 0.04-0.4 |
| Reducing of solar insolation and visibility, increasing cloudiness, appearance of fog. Possible lowering of the temperature of the Earth. | | |
| *Sulphur dioxide* | Fuel combustion | 0.5-1 |
| Chronic disease of plants, reducing yields in agriculture, deforestation. Respiratory disease. | | |
| *Nitrogen oxides* | Fuel combustion at high temperature | 0.05-0.2 |
| Absorption of sunlight, creation of photochemical fog-smog. Destruction of a number of materials, reducing yields, deforestation. Reducing the content of hemoglobin in the blood. | | |
| *Carbon monoxide* | Incomplete combustion | 1-50 |
| Reducing the content of hemoglobin in the blood. | | |
| *Soluble hydrocarbons* | Incomplete combustion | <3 |
| Disease of plants with higher concentrations of 0.2 μg/m$^3$. Exasperating effect on the eyes. | | |

Fly ash, originating from coal based power-plants, represents a huge ecological and financial contemporary problem. The generation of this by-product has already reached an alerting quantity of approximately 175 million tons per year [5]. Fly ash depots are constantly increasing in size. Fly ash production leads to the problems of disposal as well as environmental damage by causing air and water pollution on a large scale [6]. Namely, fly ash is regarded as a toxic material owing to its high concentration of leachable heavy metals and, in some cases, to the presence of various organic compounds. Thus, a significant risk to the environment threatens from the possible leaching of hazardous pollutants, such as toxic-metals [7, 8].

Disposal of the fly ash became an increasing economic and environmental burden. Finding disposal sites became increasingly more difficult. The environment is at constant and alerting risk of air, water and soil pollution, while the storage expenses, as well as expenses of water and air refining are perpetually increasing. As the consequence, there is a growing interest in looking for applications where the fly ash can be used as a potential resource for preparation of products with high-value. As per the estimation, utilization of fly ash can be targeted to an extent of 40–50 % in various fields. World-wide, this waste is utilized primarily in cementitious products, but remainder is directly discharged into ponds or landfills. These numbers have urged researchers to look for new and alternative applications of produced fly ash [9].

Approximately 6 - 7 millions of tones per year of fly ash are being produced in Serbia whilst an insignificant amount of it is being reused in the construction industry and in mass-production, generally. At the beginning of the 21$^{st}$ century, measured amount of solid fly ash particles emission, in Serbian power-plants, was at least 10 times higher than minimal amount allowed by European Union (50 mg/m$^3$). In certain thermal plants electro-filters produced gases with extremely high solid particles concentration: 1000 - 2000 mg/m$^3$. In year 2003, a project concerning innovation and restoration of electro-filters was introduced. The goal of the running project is to repair all electro-filters in Serbian thermal plants and to adjust their working-regime in accordance with allowed solid particles emission level [1].

Possible applications of fly ash can be grouped into following categories:

- construction materials - cement, concrete, ceramics, glass/glass–ceramics, light aerated concretes, building blocks, lime-silicate elements (gas-concrete);
- geotechnical applications - fly ash can be built in dams and embankments, used in road constructions, pavement, soil stabilization, protection of river banks, protection of plastic-clay soil layers, tailings and non-stable sand ground;
- other - fly ash can be used as filler in refractory concretes, for asphalt mixtures, for injection mixtures, as adsorbing material and for sludge conditioning [10].

Each application has own environmental impacts and benefits, but all of them are resulting in advantages in waste minimization. Application of fly ash in building industry is the only way to solve the ongoing global environmental problem.

## 2. EXPERIMENTAL

The properties of the fly ash are strongly dependent on its geological origin and the combustion process of the coal in energy plant. It is important to characterize fly ash in detail to ascertain its potential uses as raw material in the production of high value products within industry of building materials.

The physico-chemical properties of the fly ash originating from three different coal-fired power plants are presented, evaluated and discussed in this work. The fly ash samples were named subsequently: EFP1, EFP2 and EFP3. Each sample originated from different power-plant. The fly ash samples were landfilled without any previous treatment and have not been mixed with household or similar type of waste

Chemical composition analysis of the fly ash samples was performed by means of X-ray fluorescence method (XRF spectrophotometer ED 2000 - *Oxford*). The loss of ignition (LOI) content was determined by the weight difference between room temperature and 1000 °C. The maximum temperature was held constant for 4 h.

Differential thermal analysis (DTA) of the fly ash samples 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 α-$Al_2O_3$ (corundum) powder as reference sample. The sample was heated under an air atmosphere from 20 up to 1100 ºC at heating rate of 10 ºC/min.

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 λCu –Kα radiation and a step/time scan mode of 0.05 º/1s.

The microstructure of the fly ash samples was characterized by scanning electron microscopy method (SEM) using a JEOL JSM-6390 Lv microscope. The original fly ash powder (without further grinding) was used as sample. The samples were covered with gold powder. SEM microphotographs were recorded at different magnifications: 100x, 1000x and 1500x.

Potential mobility of trace elements from the fly ash samples was determined by means of leaching test [11]. 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. The content of major and trace elements in the leachates were determined by means of inductively coupled plasma atomic emission spectrometer ICP-AES (Perkin-Elmer Optima 5300 DV).

The bulk density and the mechanical strength of the fly ash samples were determined in accordance with classic laboratory procedure which is analog to the procedure applied in cement samples investigation. Samples of cement-mortar with fly ash replacement factor 30 % were shaped into dimensions 40x40x160 mm and submitted to the testing. Pouzzolanic activity was estimated by mechanical characteristics determination method.

## 3. RESULTS

The chemical compositions of the fly ashes (EFP1-3) were mutually compared and correlated with standard European fly ash (EEFP) [12]. Results and comparison are given in Table 2.

The obtained experimental results for fly ashes EFP1-3 were similar to the certified composition given for EEFP. Thus, analysis of the main oxides in the EFP1-3 verifies possible usability of investigated fly ashes. As in the case of European fly ash, the investigated fly ashes consisted mainly of silica, alumina and iron oxides depending on the origin of a fly ash. Relatively low loss of ignition in weight (LOI) was attributed to the fact that organic matters were not present in the investigated fly ash samples. All three fly ash samples EFP1, EFP2 and EFP3 can be classified as alumo-slicate ashes. Therefore, all investigated fly ash samples show pouzzolanic behavior and can be used as substitution for bonding agent in concrete or mortar.

*Table 2. Chemical composition of the fly ashes.*

| Constituents, weight % | EEFP | EFP1 | EFP2 | EFP3 |
|---|---|---|---|---|
| $SiO_2$ | 41.1–59.6 | 57.49 | 53.49 | 58.32 |
| $Al_2O_3$ | 17.6–35.6 | 17.72 | 21.28 | 18.08 |
| $Fe_2O_3$ | 2.6–16 | 10.48 | 6.2 | 6.85 |
| $TiO_2$ | 0.5–2.6 | 0.52 | 0.56 | 0.57 |
| CaO | 0.5–11.8 | 6.96 | 7.61 | 8.71 |
| MgO | 0.8–3.8 | 1.98 | 2.74 | 2.3 |
| $P_2O_5$ | 0.1–1.7 | 0.02 | 0.03 | 0.02 |
| $SO_3$ | 0.1–8.6 | 1.06 | 0.78 | 1.29 |
| $Na_2O$ | 0.1–1.2 | 0.36 | 0.44 | 0.5 |
| $K_2O$ | 0.4–4 | 0.59 | 1.21 | 1.16 |
| $CO_2$ | 0.6–7.6 | 0.09 | 0.25 | 0.11 |
| LOI | 1.1–8.1 | 2.94 | 4.91 | 1.84 |

Trace element concentrations of EFP1-3 are shown in Table 3. It was noted that EFP1-3 had lower concentrations of a number of trace elements, namely Pb, Cd, Zn, Hg, Ba and Se than European fly ashes. Even though trace elements are present as a relatively small fraction in fly ash, they are, nevertheless, investigated due to their tendency of cumulative build-up, long life and high toxicity to humans and environment in general. Relatively low level of mentioned trace elements, usually results in low leachability of toxic elements and heavy metals. 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 construction element and exposed to the action of rain and air.

The leaching test provided information on the leaching potential of various chemical constituents appearing within investigated fly ash and fly ash based cement mortar (with 30 % replacement factor) in normal environment. 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 are given in Figure 1. Transition of the elements leached was close to or below the detection limit with water as leaching agent. After comparing results of leaching test for EFP1-3 and for fly ash based cement mortar, overall conclusion was that there is no significant difference in results and investigated fly ashes are secure for application in construction materials industry.

*Table 3. Chemical analysis of trace elements.*

| Trace element, ppm | EEFP | EFP1 | EFP2 | EFP3 |
|---|---|---|---|---|
| Pb | 40-176 | 15.5 | 19.1 | 24.4 |
| Cd | 1-6 | 0.1 | 0.00 | 0.2 |
| Zn | 70-924 | 52.3 | 36.1 | 56.6 |
| Cu | 39-254 | 52.4 | 26 | 35.9 |
| Ni | 49-377 | 21.7 | 41.3 | 50.9 |
| Cr | 47–281 | 97.6 | 148.1 | 135.1 |
| Hg | <0.01–1.4 | 0.00 | 0.00 | 0.00 |
| As | 22–162 | 10.6 | 9.9 | 132.2 |
| Ba | 311–3134 | 51.5 | 53.3 | 86.2 |
| Sb | 1-120 | 0.5 | 0.1 | 1.1 |
| Se | 3-30 | 1.8 | 1.7 | 1.4 |

Results of the mineralogical analysis of the flay ash are given in Figure 2. Clay components within fly ash contain coal particles and as result of its phase transformation on elevated temperatures, amorphous and crystalline phases were formed. These phases have different chemical compositions which, as well as phase ratio, influence fly ash final characteristics.

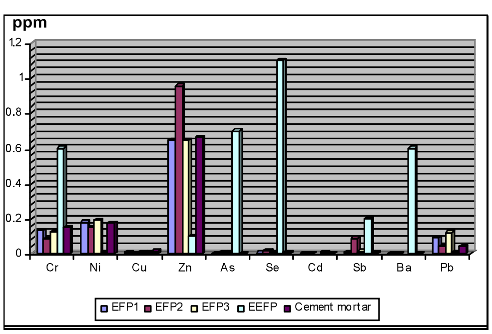


*Figure 1 – Leachability of fly ash.*

XRD diffractograms highlighted high amount of amorphous matter within all investigated fly ash samples. Some of the identified crystalline major phases present in the fly ash were aluminosilicate glass, quartz ($SiO_2$) and mullite ($Al_6Si_2O_{13}$). The only defined peaks on diffractogams relate to quartz. The background hump between 10 and 40° in the X-ray spectrum provided additional evidence of the presence of an amorphous phase. Calcite, magnetite, 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 a landfill. Sample EFP1 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 EFP3 had the most noticeable quartz peak on diffractograms, i.e. the highest level of crystallinity.

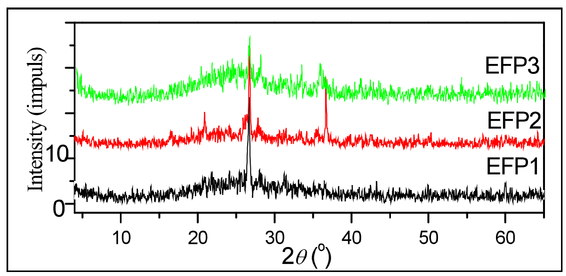


*Figure 2 – XRD diffractogram of EFP1-3.*

Processes taking place during fly ash thermal treatment were identified by means of DTA method. DTA curves are given in Figures 3. All DTA curves have a small peak at approximately 200 ºC which corresponds to the volatilization of the water mechanically bonded in form of $H_2O$ molecule. Thus, the first dimensional change occurred between room temperature and 100 °C and it was related to the loss of humidity. During temperature interval from 100 to 450°C hydration water was altogether lost. Peak showing at approximately 500 ºC is exothermic and characteristic for fly ash. 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.

The second peak is endothermic and distinctly visible and located at approximately 900 ºC. This peak was induced by presence of alumo-silicates. The results of DT analysis led to the conclusion that investigated fly ashes were thermally stable at temperatures around 900 °C and could be directly used in processes carried out at or below this temperature, which means that they could be applied as component for high-temperature construction materials (thermo insulation mortar or refractory concrete). Differential thermal analysis also showed a certain change above 900 °C which pointed to the additional structural changes in fly ash occurring with further increasing of the temperature, i.e. initiation of the sintering process.

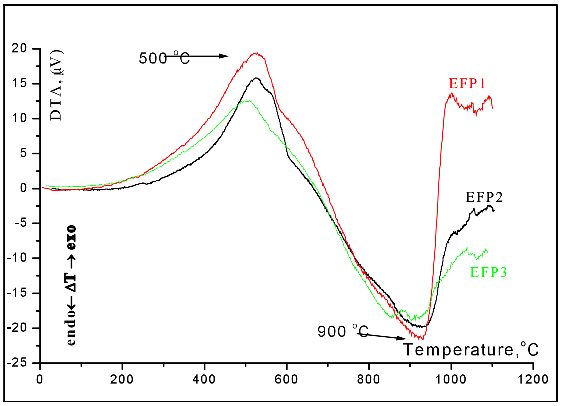


*Figure 3 – DTA diagram of EFP1-3.*

All of the fly ash samples studied revealed very similar particle size distribution. The small particle size of the fly ash is advantageous factor for obtaining better reactivity of the material in transformation processes, especially when it comes to fly ash „building-in" into 3-dimensional structure of concrete and other cementitous materials. Average grain-size is 0.089, 0.118 and 0.143 mm for EFP1-3, respectively. Further milling would only improve characteristic of the fly ash.

Investigated mechanical characteristics of the fly ash samples are given in Table 4.

*Table 4. Mechanical characteristics of the fly ash.*

| Mechanical characteristic / Sample | Bulk density, g/cm3 | Compressive strength, MPa | Flexural strength, MPa |
|---|---|---|---|
| EFP1 | 2.18 | 4.50 | 1.50 |
| EFP2 | 2.20 | 4.60 | 1.50 |
| EFP3 | 2.17 | 7.70 | 2.20 |

The values of the bulk density are approximately same for all investigated fly ash samples. EFP3 sample has the highest compressive and flexural strength, which could be correlated to its intense crystallinity and better developed structure than the structure of EFP2 and EFP1.

In addition, pozzolanic activity was estimated and presented in Fig.4 As expected from chemical analysis, all investigated fly ashes have high pozzolanic activity. The highest value of pozzolanic activity was obtained for EFP3 sample. As previously concluded, mechanical activation of fly ash would induce rise in both flexural and compressive strength, but also in pozzolanic activity. This phenomenon can be explainned by increasing of the specific area of fly ash due to process of grinding and milling.

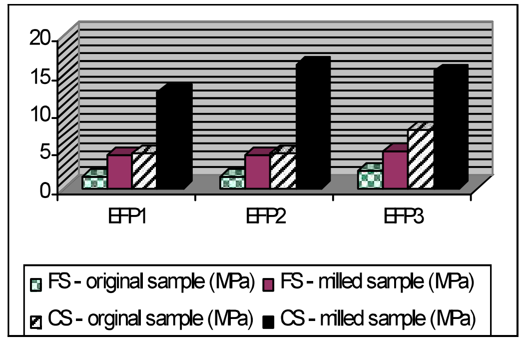


*Figure 4 - Pozzolanic activity of fly ash samples: FS – flexural strength, CS - compressive strength.*

Figures 5. and 6. show SEM microphotographs of some of investigated fly ashes recorded with different magnifications. As SEM microphotographs show, fly ash is a mixture of various grains of different size and shape, i.e. different inorganic phases and possibly certain quantity of unburned organic materials.

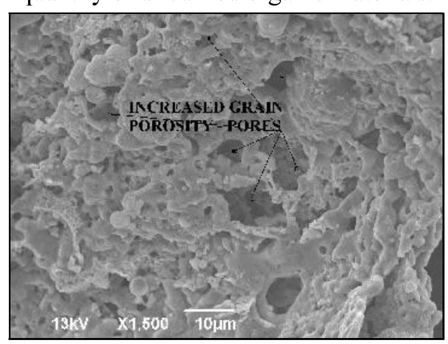


*Figure 5 - SEM of EFP1 (magnification 1500 x).*

Fly ash was mainly composed of spherical hollow particles. Increased superficial porosity of fly ash grain is evident. Visible pores are regularly shaped and round.

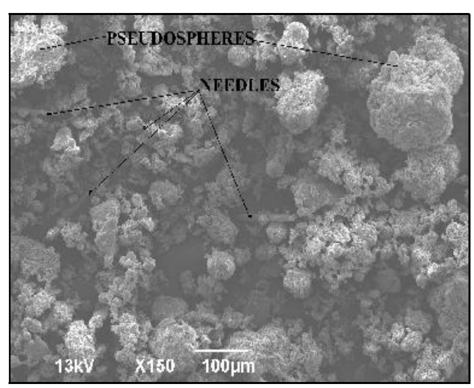


*Figure 6 - SEM of EFP3 (magnification 150 x).*

Other particles with different shape, size and texture were identified by SEM method. Namely, it can be seen that each fly ash sample is a composition of various grains of different sizes, shapes and morphology. Such diverse composition of fly ash is in relation with varieties in its chemical and mineralogical composition previously explained. Most of the grains in the mixture were spherical or rounded, although irregularly shaped grains were also present

Irregularly shaped grains and agglomerations immersed in the fly ash mixtures. These irregular grains and agglomerations usually correspond to calcite ($CaCO_3$) which was identified with XRD method. Pseudospheres, i.e. spherical particles composed of various layers or grains were noted. Such particles usually correspond to the presence of magnetite. Specific needle-shaped particles are visible and they are either found isolated or merged with spheres in agglomerations. According to XRD analysis these needle-like shapes correspond to the mullite crystals. Quartz was also found as irregularly shaped particles

Grain porosity was evidently present as it can be seen in Fig.5. Smaller, internal pores are, also, visible within superficial pores, which lead to the conclusion that the fly ash grains are intersected with pore channels which might increase water absorption. Extremely porous particles might correspond to unburned coal.

## 4. CONCLUSION

Air pollution originating from energy-plants threatens to damage environment, both visible and invisible parts of nature. The consequences affect the general climate, because destruction of vegetation reduces the possibility of restoration of oxygen. Pollution of air, as the basic condition of life, threatens human health and life, as well as the whole above-ground bioceonosis (forests and plants).

Republic of Serbia in its process of integration into the family of the European Union must harmonize the regulations in the field of ecology. As it is necessary to harmonize the regulations it is also necessary to direct the population to develop awareness of the protection of the environment in which they live and necessity of reapplication of waste materials. It has already been common practice in the European Union, but in Serbia this is on-going procedure. The state must regulate, through the system of education, basic introduction into ecology, environment, human share in the pollution and the ways to prevent it all. Only the timely education and informing with consequences and the ways of their prevention can develop awareness.

Since the fly ash is potentially hazardous for living environment, it is necessary to recycle it and to find its reapplication instead of leaving it on open landfills. Most economic manner of the fly ash recycling is its re-application in the industry of construction materials.

Investigation of the fly ashes showed that they have negligible level of organic impurities and potentially toxic and/or leachable elements. The investigated fly ashes, due to extent chemical analysis, were classified into alumo-silicate group, i.e. the group of ashes accompanied with well manifested pozzolanic-/bonding properties which is of importance for fly ash application as cement component or substitution. Investigated fly ashes showed satisfying grain-size distribution and excellent physico-chemical, thermical and mechanical characteristic, with accent on significant compressive strength which is of huge importance for fly ash application as a substitution for small fraction aggregate in concrete or mortar. Because of its morphological properties and structural and thermal stability, fly ash could be used as additive or filler for thermal insulation materials, as well as refractory and other high-temperature building products.

The fly ash showed high pozzolanic activity which makes is useful as a raw material to make cement or cement replacement in concretes and mortars. Milling and even possible mechanical-activation of fly ash would result in further advance of characterristics.

On the negative side, SEM analysis pointed out on increased fly ash grain porosity which could cause higher water necessity during the preparation cementitous mixtures. Such thing can be avoided by application of water-reducing additives or admixtures.

Leaching test carried out on the fly ashes denied possibility of potential transition of toxic elements if fly-ash based building material is in contact with water. Thus ground or underground water contamination by migrating toxic fly ash constituents is also denied.

The fly ash properties determined in this work could be regarded as representative for most of coal fly ash produced in energy-plants. Thus, investigated fly ashes have excellent characteristics which opens new frontiers for its utilization, not only in the region but world-wide.

## 5. ACKNOWLEDGMENT

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## SUMMARY

GRAĐEVINSKI MATERIJALI NA BAZI ELEKTROFILTERSKOG PEPELA KAO REŠENJE ZA SMANJENJE ZAGAĐENJA VAZDUHA PROUZROKOVANO EMISIJOM IZ TERMOELEKTRANA

*Najčešći uzročnici zagađenja životne sredine su emisije uzrokovane različitim tehnološkim procesima koji se odvijaju u termoelektranama . Velika produkcija elektrofilterskog pepela, koji je glavni ostatak pri sagorevanju uglja, predstavlja ogroman rizik i opasnost za životnu sredinu. Zagađenje vazduha je poprimilo tolike razmere da se ovom problemu obraća specijalna pažnja i sa tim u vezi uvode mnoge regulative. Cilj legislacije je da se smanji opasna emisija u atmosferu, a to se positže uvođenjem standarda vezanih za kvalitet vazduha i maksimume emisije raznih zagađujućih čestica. Pored uvođenja regulative, moguće rešenje za problem zagađenja i njegovog smanjenja jeste reaplikacija letećeg pepela u industriji građevinskih materijala. Elektrofilterski pepeo se može upotrebiti kao komponenta pri proizvodnji cementa, maltera, betona, opeke, pločica, asfalta. Korišćenje elektrofilterskog pepela je održivo rešenje za očuvanje pripodnih resursa i zaštitu životne sredine, ali i osnova za dizajniranje novih građevinskih materijala na principu reciklaže nusprodukata.*

**Ključne reči:** *leteći pepeo, zagađenje, regulative, građevinski materijali.*