Recycling of Power Plants Wastes - Potential Water Pollutants

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Abstract

Fly ash and bottom ash are major by-products of the coal combustion process in thermal power plants. They are composed of oxides of Si, Al, Fe, Ca, Mg, Na and K. Along with oxides, fly ash and bottom ash contain trace elements (Sb, As, F, Cr, Cu, Co, Ni, Zn, Cd, Mn, Pb, Hg, V etc.) which represent the potential water pollutants.
Usually, fly ash and bottom ash are disposed of using the wet method (slurry form) into disposal sites. Consequently, potential ground water and surface water contamination by the toxic elements - ions from the ash disposal sites is predictable.
A combination of suitable disposal technique and increased utilization is required to combat the environmental problem associated with coal ashes generation.
Solidification/stabilization (S/S) is a widely used technique for disposal of wastes which inhibits the migration of waste ions into the surrounding environment.
Now a days, the utilization of coal ashes in road construction is very interest, because this utilization ensure consumption of fly ash and bottom ash in bulk.
The possibilities of recycling of fly ash and bottom ash from the "Nicola Tesla" power plant (Serbia) for road construction were investigated in this study. The results showed that three mixtures could be utilized as sub-base mixtures for road construction:
1. 34.18 % bottom ash : 25.64 % fly ash : 8.55 % Portland cement : 31.63 % water ,
2. 34.18 % bottom ash : 27.34 % fly ash : 6,85 % Portland cement : 31.63 % water and
3. 41.14 % bottom ash : 20.57 % fly ash : 6.85 % Portland cement : 31.44 % water.
The compressive strength values of these mixtures after 7 days of hardening (1.62 MPa, 1.57 and 1.61 MPa, respectively) were higher than the low limitation (1.5 MPa) recommended by JUS (Yugoslav Standard) for sub-base mixtures.
On the basis of X-ray diffraction analysis (XRD) and thermogravimetric analysis (TGA) of mentioned hardened mixtures it was evident that the products of hydration reactions: ettringite ($3CaO.3CaSO_4.Al_2O_3.32H_2O$) and hydrated calcium silicates (C-S-H) were formed. Their formation is important because of their possibility to immobilize the trace elements and to minimize the adverse environmental impact of recycled materials in road construction.
The combination of cementitious properties (compressive strengths) and the potential for immobilization of hazardous trace elements could result in utilization of mentioned three mixtures in road construction.
However, long-term examinations are necessary .

Keywords: *fly ash, bottom ash, ettringite, C-S-H, immobilization of ions*

Intodruction

Fly ash and bottom ash are by-products, of the coal combustion process in thermal power plants.
The "Nicola Tesla" power plant (the biggest power plant in Serbia) produces about 5 million tones of coal ashes per annum (fly ash and bottom ash, a mass ratio 8 : 2, respectively).
These ashes are disposed of using the wet method (slurry form) into disposal sites. The disposal sites are located in the area reach in ground and surface waters. The water to ash ratio in a slurry is 9:1. Because of the high water requirement, "Nicola Tesla" power plant is located near river Sava. Consequently, the contamination of the surrounding waters and the water of river Sava (which is final recipient of waters from the power plant disposal sites) could be realized. Namely, the trace elements

present in the fly ash and bottom ash from the “Nicola Tesla “ power plant (Cr, Cu, As, Pb, Zn, B, Cd, Ni, Mn, Ba, Sr, V) may leach from them and migrate into surface and ground waters.

Leaching of trace elements and their environmental impacts on surface water, groundwater and soil is a concern when coal ashes deposit. Because of that it is always preferable to utilize wastes
than dispose of them. The Ministry of Science and Environmental Protection of Serbia and the Serbian Power Plant Nicola Tesla encourage and financially support the project about the reuse of by products -coal ashes. Road construction is of particular interest, because road construction has potential for large volume use of coal ashes.

In this study (supported by The Ministry of Science and Environmental Protection of Serbia and the “Nicola Tesla" power plant) the possibilities of the application of bottom ash and fly ash for base and sub-base in road construction were investigated.

The use of fly ash as a component of base and sub-base mixtures is based on its pozzolanic properties. A pozzolan is defined as “a siliceous or siliceous and aluminous material which, by itself possess little or no cementing value but, in the presence of moisture, chemically reacts with calcium hydroxide at ambient temperatures to form compounds that possess cementing properties”. The major products of pozzolanic reactions are ettringite ($3CaO{\cdot}Al_2O_3{\cdot}3CaSO_4{\cdot}32H_2O$), monosulphate ($3CaO{\cdot}Al_2O_3{\cdot}CaSO_4{\cdot}12H_2O$), calcium silicate hydrate (C-S-H) and calcium aluminate hydrat (C-A-H). These products (cementing material) cause hardening in base and sub-base mixtures, by binding the particles of the aggregate (bottom ash). Also, the ettringite and C-S-H phase provide the waste ion immobilization.

The structure of ettringite consists of columns of $\{Ca_6[Al(OH)_6]_2{\cdot}24H_2O\}^{6+}$ with the intercolumn space (channels) occupied by anions $[(SO_4)_3{\cdot}2H_2O]^{6-}$. Substitution of Ca^{2+} and Al^{3+} can occur in the crystal lattice. Structural Ca^{2+} can be replaced by other divalent metal cations such as Zn^{2+}, Cd^{2+}, Cu^{2+}, Ni^{2+}, Pb^{2+}, etc., and structural Al^{3+} can be replaced by Cr^{3+}, Mn^{3+}, Fe^{3+}, Si^{4+}, Ti^{4+}, etc.. Furthermore, SO_4^{2-} and H_2O can be replaced (channel substitution) by anions such as CrO_4^{2-}, AsO_4^{3-}, ZnO_2^{2-}, CO_3^{2-}, $B(OH)_4^{-}$, etc.. (Poellmann and Kuzel ,1990; Poellmann et al.,1993; Vempati et al.,1995; Solem-Tishmack et al., 1995; Dermatas and Meng, 2003).

Semi-crystalline C-S-H gel isolates ions of the waste speciies through a variety of mechanisms, including sorption and substitution at interlayer sites (Gaugar et al., 1996).

Bottom ash uses as aggregate in base and sub-base mixtures for road construction. It is a more or less inert material, i.e., normally, bottom ash does not exhibit pozzolanic properties. The coarse, fused, glassy texture of the particles of bottom ash make them an ideal substitute for natural aggregates, such as sand, gravel, or limestone.

The development of the potential use of fly and bottom ashes will be carried out taking into account two aspects of these materials:

1. chemical properties (chemical composition, mineralogical composition) and
2. physical and engineering properties (grain size distribution, density, bulk density, moisture, standard Proctor density, compressive strength, etc.).

The utilization of ashes is the subject of controversy and misunderstanding. They are thought by some to be harmful to human health from their radioactivity. On the other hand, some specialists emphasize that they are safe. Therefore, it is reasonable to investigate whether the ashes have acceptable radioactivity for their utilization in civil engineering (Wyszomirski and Beylska,1996).

Experimental

The radioactive contamination of the ashes from the “Nicola Tesla” power plant (the specific activity of the radioactive nuclides present in the fly ash and bottom ash) was study by gamma spectrometric analysis. The obtained results were compared with the maximum permissible specific activity of radioactive nuclides in building materials, according to the standard used in our country (Yugoslav Official Register No 8/87).

The chemical composition of materials utilized in this work: fly ash, bottom ash and Portland cement (presented in the oxide equivalents) was determined by classic chemical analysis.

The mineralogical composition of the fly ash and bottom ash was investigated by X-ray diffraction analysis, using a Philips PW 1729 X-ray generator, and a Philips PW 1710 diffractometer.

The physical properties of the fly and bottom ash from the “Nicola Tesla “ power plant (moisture, density, pour bulk density and compact bulk density) were determined by the methods proposed in the

Yugoslav standard. The grain size distribution of these materials was determined by sieving through Tyler sieves.

The compaction test was performed by the Standard Proctor method. This test was conducted on fly ash-water, bottom ash-water and fly ash + bottom ash-water mixtures of different compositions. The aim of this investigation was to establish the content compactability of these materials, in other words to determine the moisture-dry density relationship and the optimum moisture at which the materials have the maximum dry density, applicable for road construction. These investigations where curried out using Yugoslav standards for soil (JUS U.B1.012 and JUS U.B1.038) and literature data [7, 8, 9].
Once the content of fly ash-bottom ash and the optimum moisture content had been determined, the ratio of activator to fly ash had also to be determined. For determining the most suitable proportion of activator-Portland cement to fly ash, three mixtures with a ratio Portland cement : fly ash = 1 : 3, 1 : 4 and 1 : 5 were tested. Namely, in the mixture fly ash-bottom ash with the maximum dry density and optimum moisture content, the fly ash was partially replaced by Portland cement (25 %, 20 % and 16.7 % of fly ash). Then the compressive strength of Proctor-sized specimens (10.2 cm in diameter and 11.7 cm in height) of these mixtures were measured, after 7 days. The samples with the high compressive strength present the mixtures with the suitable Portland cement: fly ash ratio (Larrimore and Pike; Kumar et al., 2005).
Finally, specimens obtained by the hardening of mixtures (with the optimum content of the components and the optimum moisture content): fly ash , Portland cement and bottom ash were studied by X-ray diffraction (XRD) and thermogravimetric (TGA) analysis. The TGA examinations were performed on a STA 1000 instrument (Stanton Redcroft, England).

Results and discussion

The radioactivity of the coal ashes

The results obtained for the specific activity of the radioactive nuclides present in the ashes from the “Nicola Tesla” power plant are presented in Table 1.

Table 1. Gamma spectrometric analysis of the ashes

Radioactive nuclides	Fly ash	Bottom ash	Permissible values
^{238}U (Bq/kg)	129 $\pm$ 36	161 $\pm$ 42	-
^{235}U (Bq/kg)	10 $\pm$ 1	6.3 $\pm$ 0.8	-
^{40}K (Bq/kg)	396 $\pm$ 40	358 $\pm$ 36	5000 *
^{226}Ra (Bq/kg)	126 $\pm$ 13	86 $\pm$ 8	400 *
^{232}Th (Bq/kg)	86 $\pm$ 9	63 $\pm$ 6	300*

* According to Yugoslav Official Register No 8/87

The results in Table 1. show that the radioactive contamination of the ashes from the “Nicola Tesla” power plant do not exceed the permissible limits and that these ashes can be used for road construction.

Chemical composition of the materials

The chemical compositions of the materials used in this work: fly ash, bottom ash and Portland cement are shown in Table 2. The requirements for the potential use of fly ash as a pozzolan (according to ASTM 618) are also shown in Table 2..
On the basis of the results presented in Table 2, it evident that fly ash from the “Nicola Tesla” power plant can be classified as a class F fly ash and that it satisfies the chemical requirements for use as a pozzolon, because the content of pozzolan oxides (SiO_2+Al_2O_3+Fe_2O_3) in this fly ash is greater (80.66 %) than the minimum content for these oxides required by ASTM 618. Also, the contents of SO_3 and Na_2O, as well as the L.O.I. are in accordance with ASTM 618.
The high carbon content, based on the high value of the loss on ignition (L.O.I. = 28.39 %), in bottom ash may have a negative influence on the strength development in base and sub-base mixtures.

Table 2. Chemical compositions of materials

	Fly ash (%)	Fly ash specifications (ASTM 618) (%)	Bottom ash (%)	Portland cement (%)
SiO_2	52.27	-	42.09	22.03
Al_2O_3	22.34	-	14.72	3.87
Fe_2O_3	6.05	-	5.56	3.25
$SiO_2+Al_2O_3+Fe_2O_3$	80.66	50.0 minimum, Class C 70.0 minimum, Class F	62.37	-
CaO	6.64	less than 10%, Class F more than 10%, Class C	2.64	64.37
MgO	4.41	-	2.69	1.43
SO_3	2.74	50.0 maximum	1.98	1.98
P_2O_5	0.08	-	0.08	-
TiO_2	1.07	-	0.70	-
Na_2O	0.41	1.5 maximum	0.33	0.21
K_2O	1.36	-	0.90	0.63
CaO free	-	-	-	1.29
Loss on ignition (L.O.I.)	2.34	6.0 maximum	28.39	1.00

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Mineralogical composition of the coal ashes

An X-ray diffraction study was carried out to identify the mineral phases in fly ash and bottom ash from the “Nicola Tesla” power plant. On the basis of these results, it can be stated that the major crystalline phase in the fly ash is quartz-SiO_2. The other crystalline phase in fly ash, present in small amounts, are: mulite-$Al_6Si_2O_{13}$, anhydite-$CaSO_4$, feldspar-$NaAlSi_3O_8$, diopside-$CaMg(SiO_3)_2$, and hematite-Fe_2O_3. In the bottom ash, the major phase is quartz, then calcite-$CaCO_3$ and small amounts of anhydrite, feldspar, diopsid and hematite.
Besides the mentioned crystalline phases, a significant amount of amorphous aluminosilicates (glass) are represented by the broad “ hump” in the XRD pattern of fly ash. The “hump” in the XRD pattern of bottom ash is smaller than that of fly ash, from which it can be concluded that the amount of amorphous aluminosilicates is smaller in bottom than in fly ash. The SiO_2 present as quartz or in the crystalline aluminosilucates phase is inert. Similarly, the part of the alumina in the crystalline phase is inactive. Both the silica and alumina in the amorphous aluminosilcates (glass) are reactive.
On the basis of the results of X-ray analysis, it is evident that fly ash is more reactive than bottom ash, which is in accordance with the chemical composition of the oxides of these materials (Table 2.).

Physical properties of coal ashes

Moisture content of coal ashes

The determined average moisture of fly ash, was 0.57 % and that of bottom ash 32.47 %. On the basis of these results, it was concluded that fly ash does not need drying, but bottom ash does, before their use as components for the mixtures: fly ash-activator-bottom ash. Therefore, the bottom ash was dried for 2 day at 105^{o} C, before use.

Density, poured bulk density and compacted bulk density

The density and the bulk densities of the fly ash and bottom ash from the "Nicola Tesla" power plant are presented in Table 3.

Table 3. Some physical properties of the coal ashes

	Fly ash	Bottom ash
Density (g/cm^3)	2.10	1.92
Poured bulk density (kg/m^3)	652.40	503.80
Compacted bulk density (kg/m^3)	795.10	560.10

It can be seen (Table 3) that coal ashes have lower values for the density, pored bulk density and compacted bulk density than soil. This means that hollow particles are present in significant portion in coal ashes, which would usually result in low dry densities (determined by the Proctor test) (Cherial et al., 1999). The high carbon content (L.O.I.) in the bottom ash contributed to the lower density, pour bulk density and compact bulk density compared to fly ash.

Grain size distribution of the coal ashes

The results of sieving of the fly ash and bottom ash from the "Nicola Tesla" power plant through Tyler sieves show that the fly ash consisted of fine powdery particles: 41.21 % smaller than 0.074 mm and 58.79 % between 0.074 and 1.651 mm.

The bottom ash is consisted of coarser particles: 4.28 % greater than 4.699mm, 29.91 % between 1.651 and 4.699 mm, 64.85 % between 0.074 and 1.651 mm, and only 0.96 % smaller than 0.074 mm.

There is no standard in Serbia about grain size distribution requirements for application of coal ashes as components of base and sub-base mixtures. The fineness requirement in ASTM C 593 specify that 98 % of the fly ash should be finer then 0.6 mm. On the basis of this requirement, the fly ash from the "Nicola Tesla" power plant is almost satisfactory (97.4 % particles finer than 0.589 mm) and should be investigated regarding its potential application as a component of the mentioned mixtures. For cement stabilized base mixtures, the American Portland Cement Association recommends the following gradation of bottom ash particles: 100 % smaller than 19 mm and 0-30 % smaller than 0.074 mm (Portland cement Association, 1979). The bottom ash from the "Nicola Tesla" power plant satisfies these recommendations.

Compaction tests

On the basis of the results of standard Proctor tests the plots of the dry density versus the moisture content were constructed for the following samples: 1. 100 % bottom ash, 2. 30% fly ash and 70 % bottom ash, 3. 40 % fly ash and 60 % bottom ash, 4. 50 % fly ash and 50 % bottom ash , 5. 100 % fly ash.

The maximum dry density and the corresponding moisture (optimum moisture content) for the samples (read from the mentioned plots) are presented in Table 4.

Table 4. Maximum dry density and optimum moisture content of samples

Percent of fly ash, %	0.00	30.00	40.00	50.00	100.00
Maximum dry density, t/m^3	0.69	0.83	0.83	0.85	0.88
Optimum moisture content, %	48.24	50.06	45.86	46.27	50.91

The maximum dry densities for bottom ash, fly ash and mixtures of bottom and fly ash are lower than those of naturally occurring granular materials. The optimum moisture content values of these materials is higher than that of naturally occurring granular materials.

This is known and explained in the literature (Andian, 2004) by the lower density of coal ashes compared to naturally occurring granular materials (soil).
On the basis of the results presented in Table 5., it can be seen that the dry density of bottom ash is lower than that of fly ash and that the augment of dry densities of the mixtures of fly ash-bottom ash follow the increase of the content of fly ash in the mixture. The mixture fly ash : bottom ash=50% : 50 % has the highest dry density-0.85 t/m^3(at the optimum moisture content of 46.27 %).

Compressive strength behavior

Effect of the fly ash-Portland cement ratio on the compressive strength

The development of the compressive strengths in mixtures fly ash + Portland cement-bottom ash: 1. 50 % : 50 % and 2. 40 : 60 with different contents of Portland cement, at the optimum moisture content 46.27 % and 45.85 %, respectively, are presented in Table 5. The compressive strengths for base and sub-base mixtures of natural materials proposed by JUS U. E9.024 are given in Table 6..

Table 5. Compressive strength of the mixtures in dependence on the ratio fly ash to Portland cement

No.	Composition of mixture	Compressive strength after 7 days (MPa)
1.	Fly ash : Portland cement : Bottom ash = 37.5% : 12.5% : 50% Fly ash + Portland cement : Bottom ash = 1 : 1 Fly ash : Portland cement = 3 : 1	1.62
2.	Fly ash : Portland cement : Bottom ash = 40 % : 10% : 50% Fly ash + Portland cement : Bottom ash = 1 : 1 Fly ash : Portland cement = 4 : 1	1.57
3.	Fly ash : Portland cement : Bottom ash = 8.3 % : 41.7 % : 50% Fly ash + Portland cement : Bottom ash = 1 : 1 Fly ash : Portland cement = 5 : 1	1.19
4,	Fly ash : Portland cement : Bottom ash = 30 % : 10 % : 60% Fly ash + Portland cement : Bottom ash = 1 : 1.5 Fly ash : Portland cement = 3 : 1	1.61
5.	Fly ash : Portland cement : Bottom ash = 32 % : 8 % : 60% Fly ash + Portland cement : Bottom ash = 1 : 1.5 Fly ash : Portland cement = 4 : 1	1.39
6.	Fly ash : Portland cement : Bottom ash = 33.33 % : 6.66 % : 60% Fly ash + Portland cement : Bottom ash = 1 : 1.5 Fly ash : Portland cement = 5 : 1	1.25

Table 6. Compressive strengths according to JUS U. E9.024

	Compressive strength after 7 days, MPa
Base mixtures	2-5.5
Sub-base mixtures	1.5-4.5

On the basis of the results presented in Table 5, it is evident that the compressive strength in the mixture fly ash + Portland cement - bottom ash increases with increasing content of Portland cement. Comparing the results in Table 5 with those in Table 6, it can be concluded that the mixtures 1. and 2. (with fly ash : Portland cement ratio equal to 3 : 1, and 4 : 1, respectively), as well as the mixture 4 (with fly ash : Portland cement ratio equal to 3 : 1) satisfy the requirements of JUS U. E9.024 for sub-base mixtures, which was not the case with the mixture 3, 5 and 6.

Identification of hydrate phases

The results of XRD analysis of the three samples (samples no. 1., 2. and 4. in Table 5.) showed the presence of quartz, feldspar, mulite and calcite, which are inherent in coal ash, as well as a new crystal phase resulting from the hydration reaction-ettringite. Diffraction of unhydrated clinker minerals

(alite and belite|) and amorphous hydrated calcium silicate (C-S-H) coincide with diffraction of calcite. Therefore, their identification is not possible by X-ray diffraction analysis.
The results of TGA (temperature intervals and mass loss in these intervals) of the specimens are presented in Table 7.

Table 7. Results of TGA of the specimens

Number of specimen*	Mass loss (%)		
	20 – 200 ^{0}C	200 – 600 ^{0}C	600 – 1000 ^{0}C
1.	2.67	4.81	1.89
2.	1.90	4.16	1.64
4.	1.80	4.82	1.07

*the hardened mixtures from Table 6., cracked and pulverized after 7 days

The mass loss in the temperature interval from 20 to 200 ^oC corresponds to ettringite decomposition, the mass loss in the temperature interval between 200 to 600 ^oC to the decomposition of C-S-H and the mass loss in the temperature interval from 600 to1000 ^oC to decomposition of carbonates (Blondin.et al., 1999).
On the basis of the results of XRD and TGA, it is evident that the products of hydration reaction in the mixtures: fly ash, Portland cement and bottom ash were ettringite and hydrated calcium silicates.

Conclusion

1) The investigation carried out on fly ash from the “Nicola Tesla” power plant shows that this material has good potential for use as a component in base and sub-base mixtures for road construction. This is feasible from the point of view of both environmental safety (low radioactive contamination) and the properties determining its good pozzolanic activity (high content of pozzolanic oxides – SiO_2 and Al_2O_3, high amount of amorphous aluminosilicates, very fine particles and low loss on ignition).
2) Regarding the bottom ash from the “Nicola Tesla” power plant B, several deficiencies were found (high loss on ignition, low density, low bulk density and low compactability). Therefore, limitation on its application as an aggregate could be expected, i.e. only its employment as component in sub-base mixtures. Very probably, the separation of unburned coal (carbon particles) from this bottom ash, by some physical method, could improve its properties and expand its utilization.
3) The results obtained in this work showed that three mixtures prepared from by-products of the “Nicola Tesla” power plant could be utilized as sub-base mixtures for road construction:.
1. 50 % bottom ash : 37.5 % fly ash : 12.5 % Portland cement,
2. 50 % bottom ash: 40 % fly ash : 10 % Portland cement,
3. 60 % bottom ash : 30 % fly ash : 10 % Portland cement.
(The composition of these mixtures with water contents are:
1. 34.18 % bottom ash : 25.64 % fly ash : 8.55 % Portland cement : 31.63 % water ,
2. 34.18 % bottom ash : 27.34 % fly ash : 6,85 % Portland cement : 31.63 % water and
3. 41.14 % bottom ash : 20.57 % fly ash : 6.85 % Portland cement : 31.44 % water).
The compressive strength values of these mixtures (1.62 MPa, 1.57 and 1,61 MPa, respectively) are higher than the low limitation recommended by JUS (1.5-4.5 MPa) for sub-base mixtures.
4) The formations of ettringite and C-S-H during hydration reactions in the mixtures bottom ash-fly ash-Portland cement promise the immobilization of the waste ion migration to the surface and groundwater (Gaugar et al., 1996; Chrysochoou and Dermatas, 2006).

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