

Laboratory investigation of hydraulic characteristics of fly ash as a fill material from the aspects of pollutant transport

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ABSTRACT

Depending on the usage of fly ash, it is necessary to determine its environmental parameters, such as the potential for pollutant transport/leaching after its built-in. This study presents a methodology for determination of transport parameters (filtration coefficient, effective porosity, longitudinal dispersivity, and the mean residence time) from experimental data collected from column experiments with a conservative tracer on different mixtures of fly ash with stabilizers (4.8% lime and 5% cement). The transport parameters are determined using (1) numerical model results and (2) an adapted analytical solution results against measured outflow tracer concentrations. The study shows that the addition of stabilizers decreases the filtration coefficient by an order of magnitude and the effective porosity by half. The longitudinal dispersivity is not influenced by the addition of lime to the mixture, and is increased by 40% by the addition of cement. The pollutant contact time with fly ash increases by six or nine times with the addition of lime and cement, respectively. The adaptation of the analytical solution agrees well with both the numerical solution and the experimental results, and it is anticipated to be of high value for determination of transport parameters for practitioners not familiar with numerical methods.

Key words | conservative tracer test, fly ash, transport parameters

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INTRODUCTION

Coal is a dominant commercial fuel in Serbia, as it is in many parts of the world. It is estimated that around 70% of all electricity in Serbia is produced by means of coal burning (EPS 2014). This is why an increasing amount of valuable land (mostly rich agricultural) needs to be dedicated for the deposition of coal burning byproducts, mainly fly ash. In addition, the toxicity of fly ash (Borm 1997) and the environmental concern it presents, particularly due to possible leaching of hazardous materials (Izquierdo & Querol 2012), inspire efforts to find alternative ways for fly ash utilization. Additives such as braite, zeolite, ferric oxide, gypsum and high alumina are used for absorbing radioactive pollutants contained in fly ash (He *et al.* 2010). The good pozzolanic properties of fly ash make it usable for concrete production, as a retardant additive and a raw material in cement production (Ahmaruzzaman 2010). Additionally, fly ash is used as a replacement for stone filler for asphalt concrete. Fly ash is used for construction of embankments or as

filler instead of natural materials, since its properties correspond to properties of well compacted soils (Santos *et al.* 2011). Fly ash has also proved to be a good material for stabilization of soils with low mechanical characteristics, and it is used with or without activators – cement or lime. Lime is used mostly as a fly ash additive when used with crushed granular soils for road construction and earthwork applications, since these mixtures increase soil strength properties (Kalkan 2011).

The main characteristic of fly ash is its local variety of physical and chemical properties, depending mainly on the coal composition. However, in order to use fly ash, it is necessary to determine its geotechnical and environmental parameters. Depending on the purpose for which fly ash is used, the environmental parameters may include the potential for leaching of toxic materials, but also the potential for transport of pollutants, particularly when the intention is to use fly ash for embankments, landfill liners, and road bases.

These particular uses require the definition of pollutant transport properties of fly ash, due to the contact of these structures with water (rivers, urban runoff, stormwater, etc.).

The aim of this study was to develop a methodology for determination of the transport characteristics of multiple fly ash samples using the experimental data. Through a series of experiments, fly ash and different mixtures of fly ash with stabilization ingredients, such as lime or cement, were analyzed from the aspects of potential pollutant transport/leaching after it is built-in. The main parameters of interest were the filtration coefficient, the effective porosity and the longitudinal dispersivity of the fly ash mixtures. The parameters of interest were determined through calibration of a mathematical model against experimental data. In addition to the numerical model, an adaptation of the analytical solution of Ogata & Banks (1961) is presented, that can help practitioners not familiar with numerical methods to determine model parameters.

MATERIALS AND METHODS

Experimental setup

The experimental setup consisted of up-flow columns for determination of pollutant transport characteristics of samples made of fly ash and fly ash – additive mixtures. The system was equipped with an upstream reservoir with a constant level controlled by a valve. The transport experiments consisted of dosing a specific concentration of conservative tracer (NaCl) at the upstream reservoir, and measuring concentration breakthrough at the columns' outlet pipe (detection reservoir, Figure 1).

The columns used for samples were identical and had a diameter (D) of 110 mm and a length (height) (L) of 105 mm

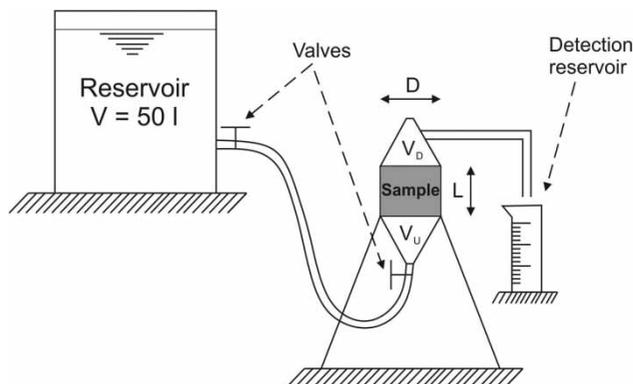


Figure 1 | The experimental setup scheme.

(Figure 1). A total of three types of samples were used for the experiment, and their description is given in Table 1. The first sample consisted of pure fly ash, which was taken from the Thermal Power Plant (TPP) Kostolac. The second and the third sample had a small percentage of lime or cement as additives (stabilizers) to the TPP Kostolac fly ash that improved both mechanical and hydraulic properties of the mixture. The particle size distribution of the analyzed fly ash was similar to silt, with 60–71% grains smaller than 0.075 mm. The specific gravity of fly ash was 2.22. This is considered a class 'F' fly ash according to ASTM C618 (2008) with $\text{SiO}_2 + \text{Al}_2\text{O}_3 + \text{Fe}_2\text{O}_3$ content above 70% and SO_3 less than 1%.

All samples were compacted in the same manner to simulate *in situ* conditions as realistically as possible (a standard compaction energy of 600 kN/m^2 was used). The percentage of stabilizers was adopted based on previous geotechnical and mechanical experiments that resulted in selection of optimal characteristics of such mixtures (Vukicević et al. 2016).

At the beginning of the experiment, all samples were saturated with clean water. Once the samples were saturated, the valve that controls the flow from the upstream reservoir was opened and salted water was introduced into the columns. The concentration of NaCl tracer was monitored frequently at the outflow, and this was done until the outflow NaCl concentration was equal with the inflow NaCl concentration (measured at the upstream reservoir). In the second part of the experiment, once the two concentrations were equalized, the clean water was introduced in the columns. This so-called fresh water forced the 'contamination,' i.e. salty solution, to filtrate towards the outflow end (Figure 1).

During the conducted experiments, inflow NaCl concentration was not constant in time, due to the existence of a small reservoir upstream from the sample (marked as V_u in Figure 1). The effect of this small upstream reservoir was that it diluted the introduced NaCl solution and therefore reduced the NaCl concentration at the upstream cross-section. Additionally, there was a small reservoir downstream from the sample (marked as V_D in Figure 1) that also influenced the breakthrough curve. The effect of the downstream small reservoir included a time lag in the measured outflow NaCl concentration.

The aim of the experiments was to determine the flow properties of samples (the filtration coefficient and the effective porosity), as well as the pollutant transport properties (the advection and dispersion parameters, and the mean residence time). Since it was not possible to measure directly the

Table 1 | Characteristics of material mixtures

Fly ash source	Sample	Stabilizer	% of stabilizer in the mixture	Compaction energy (kN/m ²)	Porosity (-)	Compaction wettability (%)
TPP Kostolac	1	–	–	600	0.45	43.9
	2	Lime	4.8	600	0.42	41.8
	3	Cement	5.0	600	0.42	43.4

effective porosity, nor the longitudinal dispersivity, it was necessary to develop a methodology for their estimation. The flow and transport parameters were determined using a mathematical model of transport of dissolved substance with the experimental data. The mathematical model was solved in two ways: (1) by adapting a form of the analytical solution and (2) by a numerical method, as described in sections below. The mean residence time was estimated from the experimental data transformed as per Shook *et al.* (2004) into a residence time distribution curve.

Mathematical model

One dimensional (1D) transport equation for substance, that is considered an ideal tracer, can be described as follows:

$$\frac{\partial C}{\partial t} = -v_{eff} \frac{\partial C}{\partial x} + D \frac{\partial^2 C}{\partial x^2} \quad (1)$$

where v_{eff} is the effective velocity of the stream (filtration flow), C is the concentration of the pollutant in water (NaCl in this case), D is the dispersion coefficient, t is the time and x is the space coordinate. The first part of the right side of the equation presents the advection term, while the second part presents the effect of the hydrodynamic dispersion. The dispersion coefficient can be presented using the longitudinal dispersivity, α_D , as $D = \alpha_D \cdot v_{eff}$.

Analytical solution

Equation (1) can be rewritten to the form of Equation (3) by transforming the coordinate system into the one that moves with the same effective velocity as the filtration stream, presented by Equation (2).

$$s = x - v_{eff}t \quad (2)$$

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial s^2} \quad (3)$$

Equation (3) is a parabolic differential equation that has an analytical solution when the flow through a uniform and

homogeneous sample is steady and the substance concentration at the upstream boundary (inflow) is constant (C_0). This analytical solution was developed by Ogata & Banks (1961), and is presented as Equation (4), used for the calculation of the substance concentration at $x = L$, which is the cross section at the outflow, i.e. the breakthrough curve.

$$C(x = L, t) = \frac{C_0}{2} \operatorname{erfc} \left(\frac{L - v_{eff}t}{2\sqrt{Dt}} \right) \quad (4)$$

As previously stated, the NaCl concentration was not constant at the upstream boundary, due to the existence of a small reservoir, V_u . This effect required an adjustment of the constant-inflow-boundary analytical solution (Ogata & Banks 1961). The modified analytical solution, developed to fit the boundary conditions of the conducted experiment, included a discretization of the upstream boundary concentration as depicted in Figure 2(a). Once the varying upstream boundary concentration is presented as a sequence of multiple steps of constant concentrations, each increased by $\Delta C_{in,i}$, the solution is obtained by successive additions of separate analytical solutions valid for the constant inflow concentration, i.e. by the superposition of analytical solutions relevant to each time step Δt_i , as described by Equations (5) and (6). The t_i in Equation (5) is the time when the inflow concentration increases for $\Delta C_{in,i}$. The effective velocity, v_{eff} , was calculated using Darcy's law. This type of a step-wise adaptation can be seen in contaminant and heat transport problems (e.g. Kwak 2008; Wang *et al.* 2011).

$$C(x = L, t) = \sum_i \frac{\Delta C_{in,i}}{2} \operatorname{erfc} \left(\frac{L - v_{eff}(t - t_i)}{2\sqrt{D(t - t_i)}} \right) \quad (5)$$

$$\Delta C_{in,i} = C_{in,i+1} - C_{in,i} \quad (6)$$

Numerical solution

A finite differences numerical method used for solving the 1D transport Equation (1) is given in two different forms

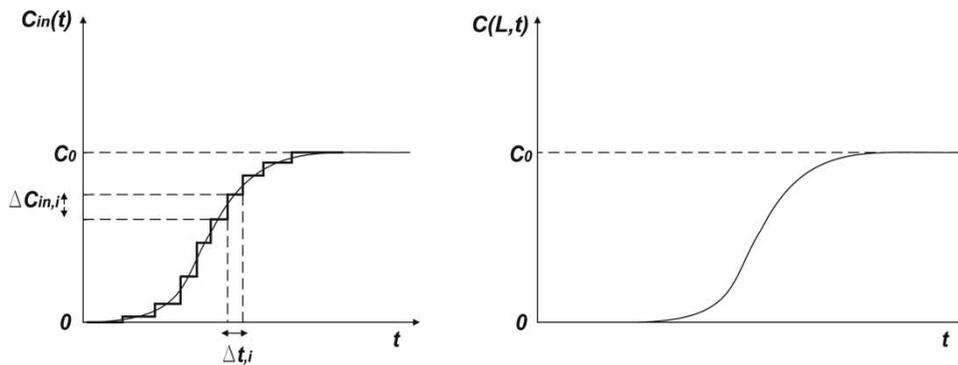


Figure 2 | The discretization of the varying concentration at the upstream boundary: (a) diagram of the concentration at the upstream boundary, (b) calculated concentration at the downstream boundary as a result of the superposition of constant-inflow-boundary analytical solutions.

that depend on the Péclet number (Equations (7) and (8)).

$$C_i^{n+1} = C_i^n + \Delta t \left(-v_{\text{eff}} \frac{C_{i+1}^n - C_{i-1}^n}{2\Delta x} + D \frac{C_{i+1}^n - 2C_i^n + C_{i-1}^n}{\Delta x^2} \right) \quad (7)$$

... ($Pe < 2$)

$$C_i^{n+1} = C_i^n + \Delta t \left(-v_{\text{eff}} \frac{C_i^n - C_{i-1}^n}{\Delta x} + D \frac{C_{i+1}^n - 2C_i^n + C_{i-1}^n}{\Delta x^2} \right) \quad (8)$$

... ($Pe > 2$)

i and n in Equations (7) and (8) are indices that describe the space and the time coordinates, respectively, while $Pe = \frac{\Delta x \cdot v_{\text{eff}}}{D}$ is the Péclet number, which provides a ratio of advective to diffusive transport rates (Hyakorn & Pinder 1983).

The numerical solution of the transport Equations (7) and (8) complements a 1D numerical model for long term simulation of water filtration in the porous media, based on either Richard's or Darcy's equations (depending on the porous media saturation). Since the samples in the experiment were fully saturated, the effective velocity was calculated using Darcy's law.

Boundary conditions

Since the volumes of the two reservoirs at the upstream and the downstream side, V_u and V_D , affected the final concentration of the salt in both the inflow and the outflow, it was necessary to include them in the mass balance calculation. The inflow and the outflow salt concentrations were determined by Equations (9) and (10) that describe the process of mixing the salted and the fresh water during every time interval, Δt .

$$C_{in}^n = (C_0 - C_{in}^{n-1}) \frac{Q\Delta t}{V_u} + C_{in}^{n-1} \quad (9)$$

$$C_{out}^n = (C_{out}^{n-1} - C_{out}^{n-1}) \frac{Q\Delta t}{V_D} + C_{out}^{n-1} \quad (10)$$

C_{in} and C_{out} in Equations (9) and (10) are concentrations of the pollutant in the inflow and the outflow (small upstream and downstream reservoirs), respectively, C_0 is the constant concentration in the large (main) upstream reservoir, Q is the flow through the sample, and V_u and V_D are volumes of small reservoirs upstream and downstream from the sample.

Hydraulic and transport parameter estimation

The filtration coefficient of the sample material, K , was determined using the least squares regression line on Darcy's law, i.e. by plotting the average velocity through the sample (v , Figure 3) versus the hydraulic gradient ($\Delta\Pi/L$, Π – piezometric head, Figure 3). The effective porosity and the longitudinal dispersivity were determined by calibrating the modified analytical and the numerical solution against the measured outflow concentration. The coefficient of determination, R^2 , was used as a measure of the agreement between the modeled and the measured outflow concentrations.

RESULTS AND DISCUSSION

Figure 3 presents the determination of the filtration coefficient, K , for the three samples, while numerical values can be found in Table 2. It can be seen that samples 2 and 3, which included a mixture of fly ash and stabilizers, had a lower filtration coefficient than the sample made of fly ash only. The addition of 4.8% of lime decreases the filtration coefficient by almost an order of magnitude, while addition

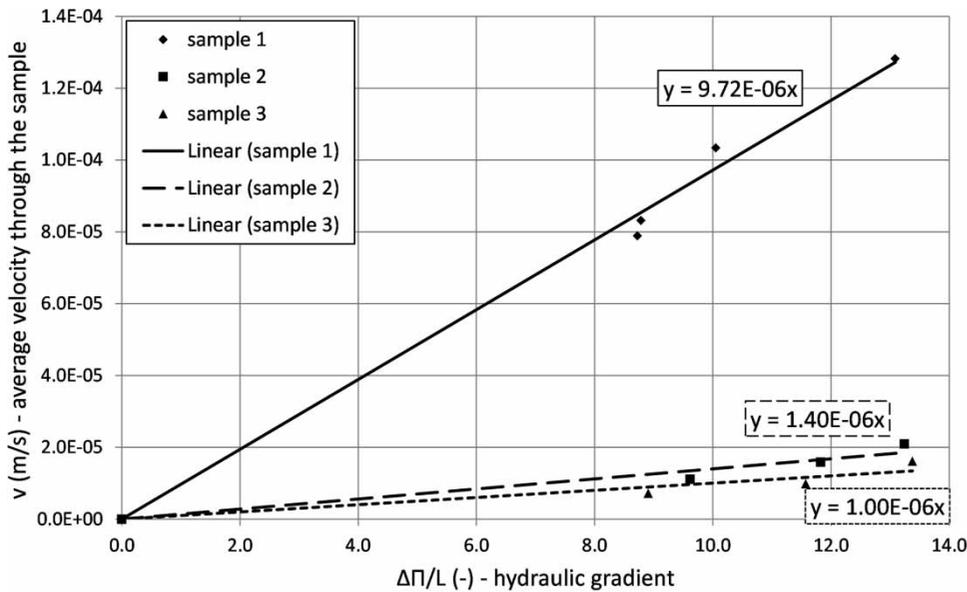


Figure 3 | Determination of the filtration coefficient.

Table 2 | Results of laboratory experiments: hydraulic and transport parameters of fly ash with effects of stabilizers

Fly ash source	Smp.	Stabilizer	% of stabilizer in the mixture	Porosity (-)	Filtr. coeff. (m/s)	Eff. porosity (-)	Long. dispersivity (m)	Mean residence time (min)
TPP Kostolac	1	-	-	0.45	9.7×10^{-6}	0.30	0.005	17
	2	Lime	4.8	0.42	1.4×10^{-6}	0.13	0.005	105
	3	Cement	5.0	0.42	1.0×10^{-6}	0.18	0.007	158

of 5.0% of cement decreases filtration coefficient by an additional 35% when compared with the addition of lime.

The adapted analytical solution and numerical model results were compared with measured data on Figure 3, while the estimated values of the model parameters (effective porosity and longitudinal dispersivity) are shown in Table 2. The coefficient of determination, R^2 , for each sample was higher than 0.98 (0.993, 0.978, 0.991 for samples 1, 2, 3, respectively), which can be considered an acceptable agreement level for the estimation of model parameters.

The results in Table 2 indicate that the additives/activators significantly reduce the effective porosity of fly ash. The effective porosity is found to be 0.30 for the 'pure' fly ash, while the activators decreased the effective porosity to 0.13 (mixture with lime), and 0.18 (mixture with cement). It can be concluded that the addition of stabilizers (lime or cement) as 5% of mass decreases the effective porosity by half. It is hypothesized that the decrease in the filtration coefficient is due to the conversion of lime and cement into pozzolanic compounds that block the pores of fly ash (Sivapullaiah & Baig 2011). The mentioned pozzolanic reactions

can take place only if an adequate content of free silica and free lime are contained in fly ash (Throne & Watt 1965). Also, it is proved that a combination of proper quantities of additives such as lime and gypsum (4% of lime and 4% of gypsum) gives the maximum strength of fly ash (Pandian 2004).

The estimated values of the longitudinal dispersivity (Table 2) indicate that the addition of lime does not influence the value of the longitudinal dispersivity, while the addition of cement increases it by 40% when compared with the fly ash only sample. However, it should be noted that the value of longitudinal dispersivity for all three samples was found to be low when compared to values compiled by Gelhar *et al.* (1992). Since the longitudinal dispersivity value depends highly on the scale of the problem (Gelhar *et al.* 1992), the findings of the conducted experiment cannot be accepted as general values.

Interestingly, the mean residence time differs substantially between the samples: 17 min for the 'pure' fly ash, compared to 105 and 158 min for mixtures with lime and cement, respectively. It is hypothesized that the mixture

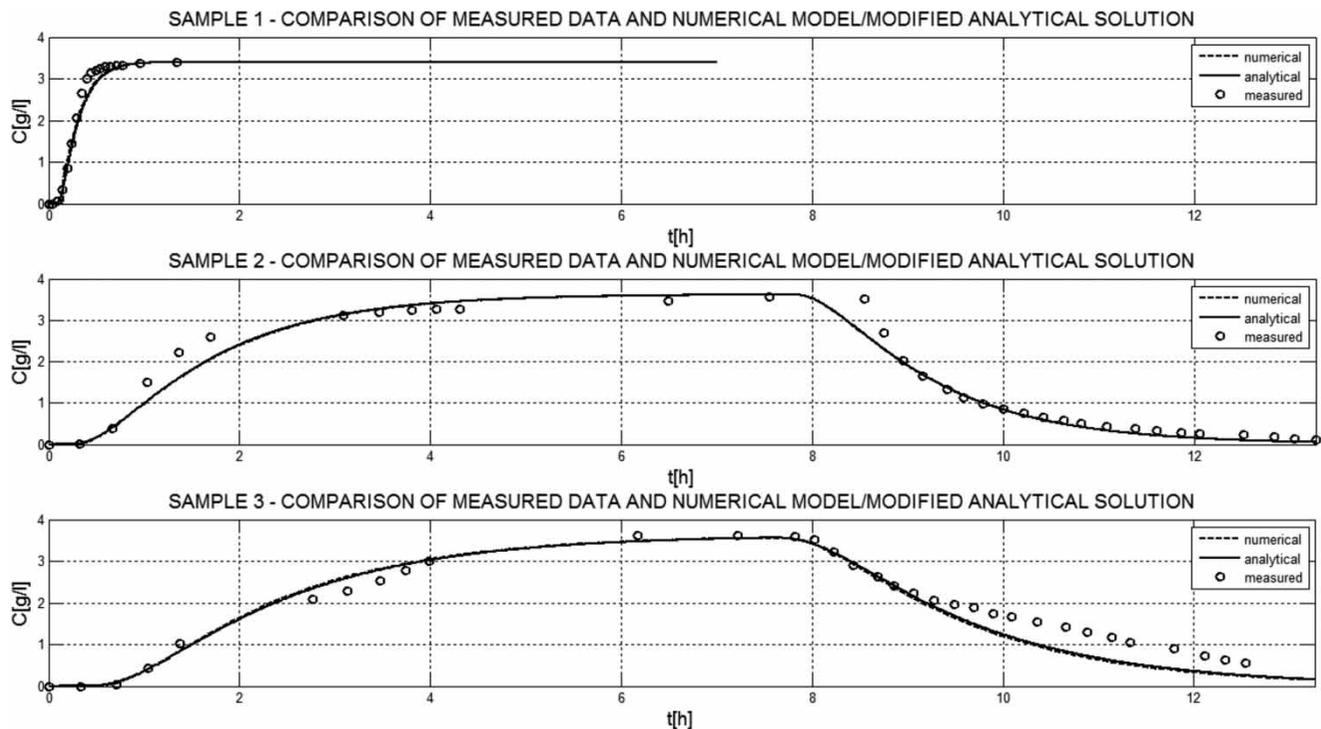


Figure 4 | The comparison of measured data and numerical/modified analytical solution for each sample.

with cement will be the most favourable in terms of preventing the pollution from passing through, as the contact time with fly-ash would be the longest.

The adapted analytical solution was found to agree quite well with the numerical model results (Figure 4), which enables it to be used as an alternative method for model parameter estimation. This is particularly of interest for practitioners who are not familiar with numerical methods, and need a quick solution.

CONCLUSIONS

This study included the analysis of the hydraulic/transport characteristics of fly ash and effects of stabilizers on its properties. The clean fly ash characteristics were compared with two mixtures: one with the lime mass content of 4.8%, and the other with 5.0% mass content of cement. These contents were previously determined as optimal by various geotechnical testings. The results indicate that both activators, lime and cement, reduced the filtration coefficient nearly by an order of magnitude, while the effective porosity was reduced by half. The longitudinal dispersivity was found not to be influenced by the addition of lime, while the addition of cement increased this parameter by 40%. The

effect of the addition of stabilizers, particularly cement, increased the pollutant mean residence time up to nine times, meaning that there was an increase in the contact time of pollutants with the fly-ash. These transport parameters are important and need to be taken into consideration in fly ash utilization, particularly in respect of its potential impact and interaction with the environment. Additionally, the study presented an adaptation of the analytical solution, which agrees well with both the numerical solution and the experimental results. This approach is anticipated to be of high value for practitioners who are not familiar with numerical methods that can be effectively used for determination of transport parameters.

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