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Effect of Bentonite Addition on the Properties of Fly Ash as a Material for Landfill Sealing Layers

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Abstract: Landfill sealing layers protect groundwater and soil against contamination from waste leachate. Fly ash, which is a coal combustion by-product, might be used as a substitute material for natural soils to build sealing layers. To improve its properties, different additives can be used. In this study, bentonite in the amounts of 5%, 10% and 15% of the dry mass of fly ash was used as an addition. Compacted fly ash and fly ash-bentonite mixtures were tested in order to verify how bentonite and the amount of addition affect the properties of the materials. Therefore, hydraulic conductivity, as the most important parameter, which determines the suitability of a material for construction landfill sealing layers, were tested. Results indicate that bentonite changes the properties of fly ash and allows the achievement of smaller hydraulic conductivity values. To provide a comparison to the natural soil, sandy silty clay that meets the criteria for natural soils for sealing layers was tested and the differences are presented. Fly ash with 15% of bentonite addition reached a value of hydraulic conductivity for the material suitable to build compacted landfill sealing layers.

Keywords: fly ash; bentonite; hydraulic conductivity; landfill sealing layers

1. Introduction

Sealing layers are one of the most important components of municipal and industrial landfill construction. They provide protection for groundwater and soil against the seepage of harmful substances present in leakage from the waste. Landfill sealing layers can be distinguished into sealing the base and slopes, cover layer, and vertical cutoff walls. It is assumed that each landfill should have sealing barriers. Soils rich in clay minerals are generally used as compacted liner materials. Natural sealing layers should be characterized by tightness, the capability of self-sealing, ease of molding and resistance to filtration deformations [1,2]. If there is no opportunity to place landfill construction in the area where geological barriers are present, artificial sealing layers should be made of compacted soils (which contain a significant quantity of clayey particles) or of other materials.

The most important parameter of materials used for landfill liner construction is its hydraulic conductivity, which is defined as the velocity of flow of water per unit cross-sectional area under a unit hydraulic gradient. Hydraulic conductivity required by common regulations for landfill barriers has to be equal to or less than value of 1×10^{-9} m/s [1,3–6]. These requirements for landfills of hazardous and non-hazardous waste are given by Council Directive 1999/31/EC [6]. According to Reference [6] hydraulic conductivity for sealing layers in landfill of inert waste should have the value of 10^{-7} m/s or lower. In order to check the suitability of the material for the construction of landfill sealing layers, a series of tests should be carried out to determine chemical, physical, and mechanical properties. The application of natural and other materials should be verified with regard to the technical guidelines and local legal standards [2,7].

Burning coal in power plants leads to the production of combustion by-products such as fly ash and bottom ash. The most common way of disposing of combustion by-products is landfilling.

Test results of properties of fly ash presented in the literature [8–14] indicate that it is a material that might be used instead of natural soils in construction engineering.

Recycling combustion by-products is a good alternative to disposal and has economic and technical benefits as well. According to References [15,16], unburned carbon presented in fly ash adsorb ions from the leachate and the alkaline nature of the fly ash helps to precipitate ions presented in leakage. Edil et al. [17] mentioned that precipitates can block the pores of compacted fly ash. Also, the pozzolanic properties of fly ashes [18] affect hydraulic conductivity with time. That is why researchers test fly ash with different additions including lime [19], cement [20] or bentonite [21,22]. Fly ash is a material where particles are predominately silt size, but it is much easier to compact than cohesive soils. However, bentonite clay is difficult to work in the field and prone to desiccation under dry conditions which leads to an increase in leakage rates [23]. The addition of bentonite to soil [24,25], fly ash and fly ash-soil mixtures reduces its hydraulic conductivity as confirmed by test results [26–28]. As mentioned, the most important property in case of sealing layers is hydraulic conductivity of the material, however other properties of fly ash and its mixtures such as compressibility, tensile strength, shear strength are no less important, as presented in the literature [29,30].

The objective of this study was to determine the impact of bentonite addition on the properties of compacted fly ash which is required in the construction of waste landfill liners. In present research, bentonite has been added in various, small and cost-effective percentages to investigate how the amount of the addition affects saturated hydraulic conductivity and other parameters of compacted fly ash. The hydraulic conductivity tests were performed under conditions with control of the sample saturation, under different effective stresses, and various hydraulic gradients. Additionally, tests on cohesive soil were conducted in order to correlate the properties of natural soil with anthropogenic material. Compaction characteristics, specific surface area, and hydraulic conductivity were tested.

2. Materials and Methods

2.1. Materials

Hydraulic conductivity tests were carried out on fly ash (FA) and fly ash and bentonite mixtures (FA+B). Additionally, to provide a comparison, tests on sandy silty clay were conducted. Fly ash originated as a hard coal combustion by-product in Bialystok Heat and Power Station, was obtained from the dry disposal site. Commercially available bentonite powder was added to fly ash in the amount of 5 (FA+15%B), 10 (FA+10%B) and 15% (FA+15%B) by dry weight of the sample. The given percentage of the bentonite represents the mass of dry bentonite per mass of fly ash in a given sample. Cohesive soil properties were also investigated. Grain size distribution curves of tested materials are presented in Figure 1.

According to the grain size distribution curves, it can be seen that fly ash corresponds to sandy silt saSi [31], whereas fly ash with different amount of bentonite addition is as follows: with 5%B to sandy silt (saSi), with 10%B to clayey silt with sand (saclSi), with 15%B to silty clay with sand (sasiCl) and bentonite to clay (Cl). Natural soil, which met requirements for cohesive soils that might be used as liner material [7,32] was sandy silty clay (sasiCl), of which liquid limit (LL) = 39% and plasticity index (PI) = 24%.

Scanning electron microscope (SEM) images of the tested materials are shown in Figures 2–4, and its chemical composition is shown in Table 1.

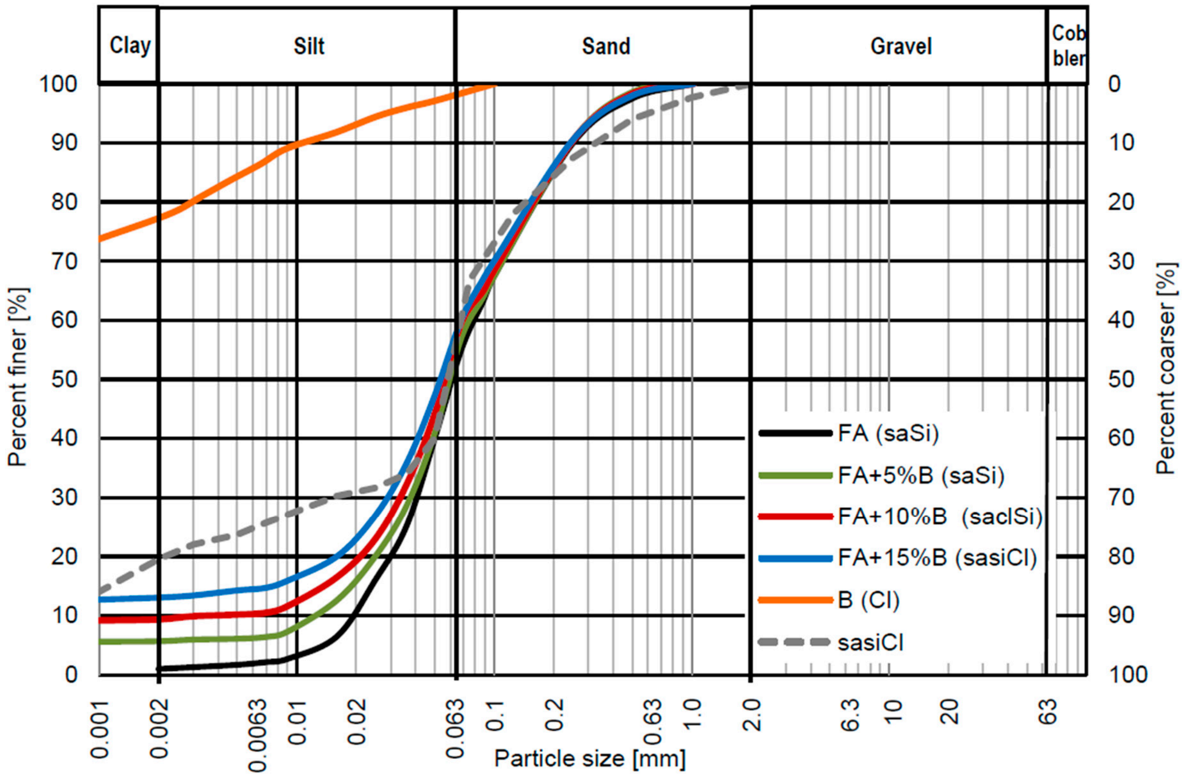
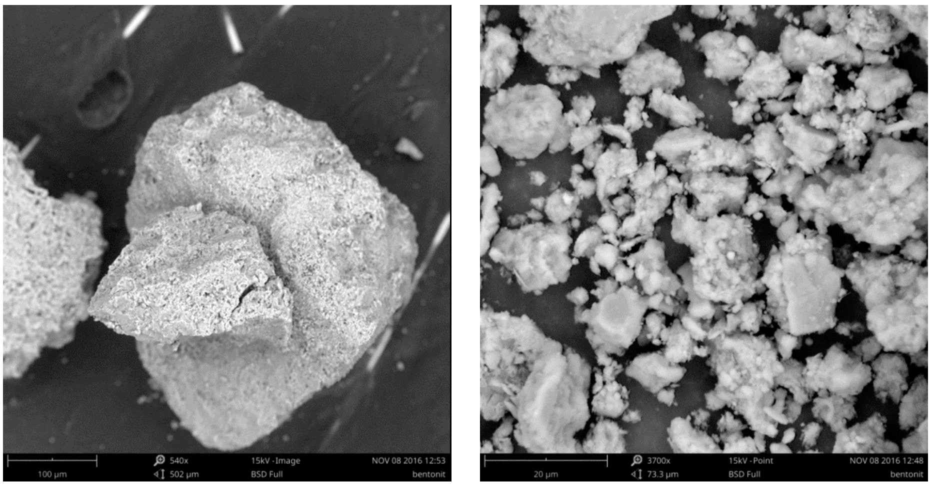
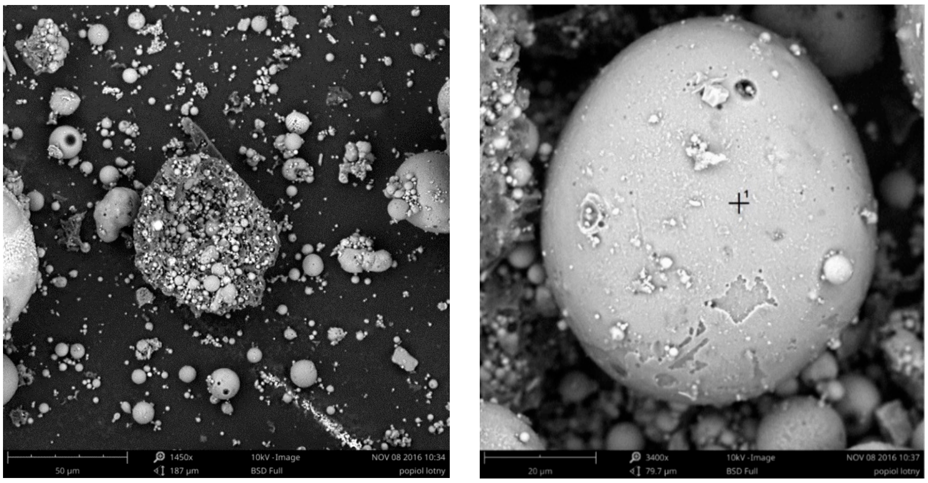


Figure 1. Grain size distribution curves of tested materials.



(**a**) (**b**)

Figure 2. Scanning electron microscope (SEM) images of bentonite: (**a**) magnification of 540×, (**b**) magnification of 3700×.



(**a**) (**b**)

Figure 3. SEM images of fly ash: (**a**) magnification of 1400×, (**b**) magnification of 3400×.

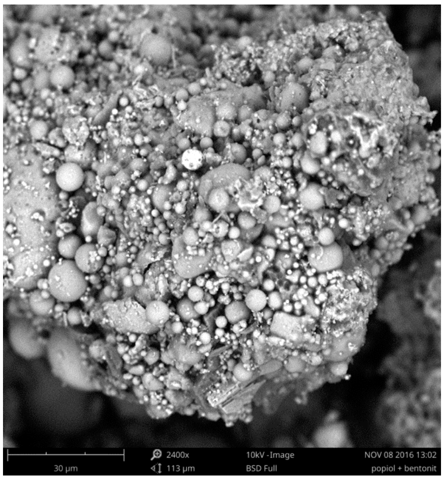


Figure 4. SEM images of fly ash-bentonite mixture—magnification of 2400×.

Table 1. Chemical composition of bentonite (B) and fly ash (FA).

Element	FA	B	Chemical Compound	FA	B
Si	22.4%	31.1%	SiO_2	43.3%	71.6%
Al	15.8%	7.7%	Al_2O_3	31.8%	18.4%
Ca	0.8%	1.9%	C	15.8%	–
An	–	1.5%	K_2O	3.4%	–
Mg	1.0%	1.0%	Fe_2O_3	1.8%	–
K	2.5%	–	MgO	2.2%	2.6%
Fe	1.9%	–	CaO	1.1%	3.1%
C	3.5%	–	TiO_2	0.6%	–
Ti	0.6%	–	Na_2O	–	4.3%

From Figures 2–4, the spherical shape of fly ash particles, the flocculated structure of irregular bentonite particles, and compact aggregates of particles in the case of the fly ash-bentonite mixture can be observed. In the micrograph of the mixture (Figure 4), the bentonite particles stuff and seal the voids between fly ash particles. During the contact with water, bentonite swells, so the pores are smaller. With higher bentonite content in the mixture, there are fewer pores. According to Table 1 and chemical compositions, the tested fly ash is classified as a Class F fly ash by ASTM C618 [33]. The results are similar to those presented in Ref [14]. Chemical composition of tested bentonite (Table 1) has higher content of silica and aluminum. Bentonite consists mainly of montmorillonite, which is a clay mineral. Tested bentonite chemical composition is similar to the results presented in Ref [34].

Values of specific soil density, optimum moisture content (OMC), maximum dry density (MDD) and specific surface area S_s of tested materials are given in Table 2.

Table 2. Properties of tested materials.

Tested Material	Specific Density ρ_s (Mg/m^3)	Compaction Parameters		Specific Surface Area S_s (m^2/g)
		Optimum Moisture Content (OMC) (%)	Maximum Dry Density (MDD) (Mg/m^3)	
FA	2.18	40.0	1.073	21.01
FA+5%B	2.18	39.0	1.100	42.24
FA+10%B	2.22	36.3	1.118	57.90
FA+15%B	2.24	33.0	1.134	67.73
sasiCl	2.68	14.0	1.950	102.58

According to Table 2, specific densities of fly ash and fly ash-bentonite mixture are lower than for natural soil—the bentonite addition in a higher amount than 5% caused an increase of specific density. Lower specific gravity and the porous nature of the coal ash particles result in lower maximum dry density and higher optimal moisture content than natural soils. As the amount of bentonite addition increases, the optimal moisture content decreases and the maximum dry density increases. Specific surface area is higher when the addition of bentonite is larger—for 5% of addition, specific surface area increased twice, whereas for 10% almost three times in the relation to the fly ash alone. The amount of 15% of bentonite in the mixture caused an increase in specific surface area of more than three times compared to fly ash. The sandy silty clay has the highest specific surface area—approximately 30% higher than for FA+15%B, which indicates higher clay particles' content.

2.2. Methods

Laboratory tests of the hydraulic conductivity of fly ash, fly ash-bentonite mixture, and cohesive soil were carried out in Rowe-Barden consolidation cell. In this type of apparatus, a sample under conditions of no lateral strain is hydraulically loaded by the membrane with a floating ring, which transfers the load to the rigid porous disk. In the Rowe-Barden cell, the saturation of the sample can be controlled by means of back pressure, constant measurements of vertical displacement and pore water pressure.

Specimens, in the form of a disc with 70 mm in diameter and 25 mm in height, were prepared by the compaction using the standard Proctor (SP) method at the optimal moisture content in accordance with the European standard [35]. It must be pointed out here that in order to establish compaction parameters of fly ash and its mixtures, the sample can be compacted only once, the importance of which is emphasized in Zabielska-Adamska [36]. It has been found that re-compaction of fly ash causes crushing of the spherical grains, which are filled by smaller grains, resulting in changes in the physical properties of the fly ash. The maximum dry density of the material compacted several times increases while optimum moisture content decreases.

To test the hydraulic conductivity, the required quantity of water was added to the dry fly ash, then the material was thoroughly mixed and placed in an airtight container to keep the moisture content. After 24 h fly ash was mixed with various quantities of bentonite immediately before compaction, then the material was mixed thoroughly. The appropriate mass of each mixture was compacted in the consolidation ring to get its respective maximum dry density at optimum moisture content. The quantity of the mixture used for the test was dependent on the volume of the sample and compaction parameters. The filter paper was then placed on the top and on the bottom of the sample to prevent finer particles from clogging the pores of porous stones. Then, the saturated porous stones were placed and the entire assembly was mounted in the consolidation cell. The cell cover was fitted and the seating pressure (5 kPa) was applied in accordance with British Standard 1377-6:1990 [37]. At first, the sample was saturated and when the saturation stage finished, the load increment was applied to the sample.

Saturation of the soil is assessed from the change of pore water pressure Δu, which occurs under changes in principal stresses $\Delta\sigma_1$ and $\Delta\sigma_3$, described by Skempton's equation [38]:

$$\Delta u = B[\Delta\sigma_3 + A(\Delta\sigma_1 - \Delta\sigma_3)] \tag{1}$$

where A and B are pore-pressure coefficients; $\Delta\sigma_1$ and $\Delta\sigma_3$ are changes in principal stresses. Coefficients A and B are measured in the undrained triaxial test. Saturation of soil tested under conditions of no lateral strain can be determined based on the value of B parameter:

$$B = \frac{\Delta u}{\Delta\sigma} \tag{2}$$

where Δu is pore water pressure corresponding to an increase of total vertical stress $\Delta\sigma$.

For fully saturated soils $B = 1$, while for dry soils $B = 0$. In laboratory conditions, saturation of the soil takes place by the back pressure saturation method. The higher the plasticity of cohesive soil, the higher the value of back pressure that is needed to reach the value of $B = 1$ [39]. In the case of some soil types, it might be unjustified to consider full saturation when B reaches value equal to or near 1. Therefore, from the relation of B parameter to degree of saturation S_r, minimum value of B can be determined [39]. The cohesive soils might be saturated to a degree that the water voids are continuous and some of the air voids are occluded. Under such conditions, it can be assumed that the soil is in a quasi-saturated state [40]. For the compacted fly ash, the minimum value of $B = 0.8$ was established for describing sufficient saturation–quasi-saturation [14]. In Figure 5, the relation between B parameter and degree of saturation of the tested materials is presented.

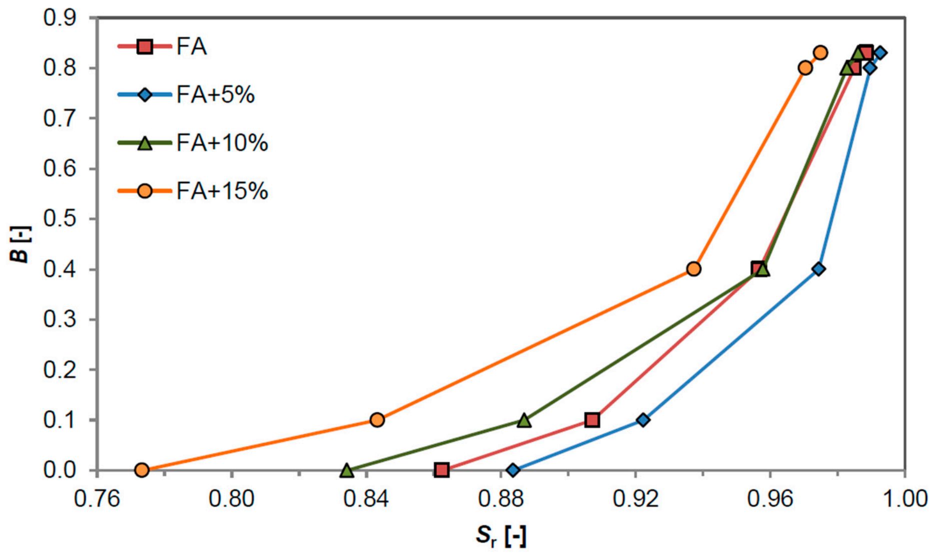


Figure 5. The relation between B parameter and degree of saturation S_r for tested fly ash and fly ash-bentonite mixture.

It can be observed in Figure 5 that when the B parameter achieved the value around 0.8 and higher, the degree of saturation S_r changed slightly. Therefore, $B = 0.8$ can be considered as a minimum value of sufficient saturation (quasi-saturation) of fly ash and fly ash-bentonite mixture. The consolidation cell was modified to measure the hydraulic conductivity at the end of each load increment. The hydraulic conductivity tests were carried out with vertical drainage condition from base to top of the tested specimens. The schematic for the test in the consolidometer is shown in Figure 6.

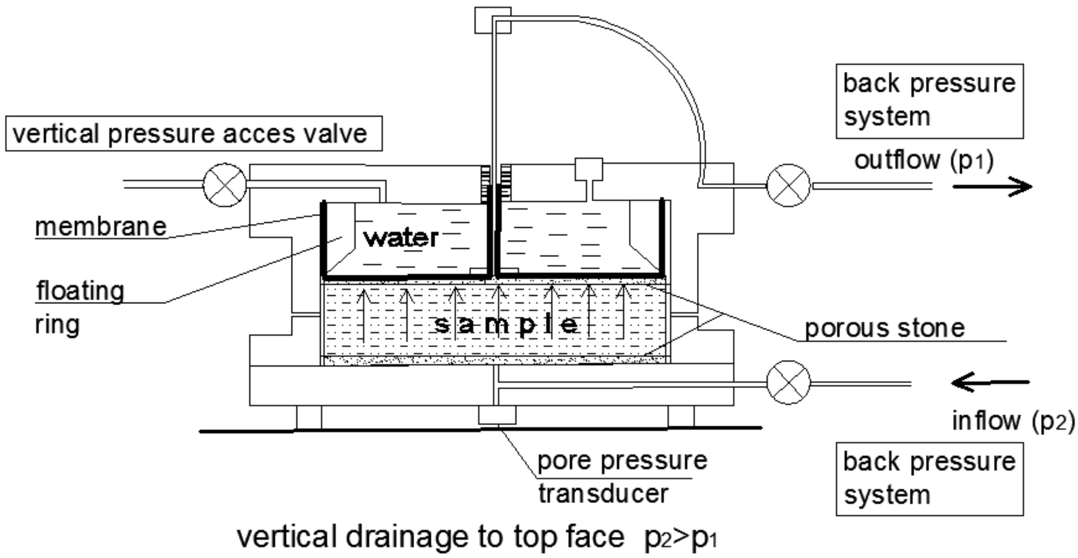


Figure 6. The Rowe-Barden consolidation cell set up for hydraulic conductivity test with vertical drainage to the top face.

The flow of water through a fully saturated soil has been described by Darcy's law according to empirical Equation (3):

$$q = Aki \tag{3}$$

or

$$v = \frac{q}{A} = ki \tag{4}$$

where q is the volume of water flowing per unit time, k is the coefficient of permeability, A is the cross-sectional area, v is the discharge velocity and i is the hydraulic gradient $i = \Delta H/l$, where ΔH is the difference in elevation of water levels in the piezometers and l is the length of the flow path. In Reference [41], k is described as hydraulic conductivity, which depends on features and properties of the material and the fluid flowing through the material. These parameters include density and viscosity of the fluid, grain-size distribution and porosity of the soil.

There is a deviation from Darcy's law at low hydraulic gradients [42]. Seepage through the cohesive soils is initiated when the hydraulic gradient exceeds a certain value called the initial gradient i_0. The fluid flow can be described by nonlinear (non-Darcian) relationship determined by Hansbo [43,44]:

$$v = \kappa i^n, \text{ when } i < i_1 \tag{5}$$

$$v = k(i - i_0), \text{ when } i \geq i_1, \tag{6}$$

where v is flow velocity, k is the coefficient of permeability of the linear flow at high gradient; k is the permeability coefficient described by an exponential relationship; i_1 is the hydraulic gradient needed to overcome the maximum binding energy of mobile pore water $i_1 = \frac{i_0 n}{(n-1)}$, n—constant dependent on soil type, void ratio and temperature.

According to classical geotechnical approaches [45], factors that affect hydraulic conductivity of fine-grained soils are: soil composition, characteristic of the fluid flows through the soil, structure, and degree of saturation during the permeation.

Tests of hydraulic conductivity were conducted on quasi-saturated samples, where the B parameter had values from 0.79 to 0.84. Afterwards the B parameter reached the required value, the consolidation tests (lasting 24 h) were carried out. The loading steps for fly ash specimens were as follows: 25, 50, 100, 200 and 400 kPa; for fly ash-bentonite mixtures: 50, 100, 200 and 400 kPa, required by the higher value of effective stress during the saturation process. According to the compression tests of municipal solid wastes in Reference [46], the range of loading pressures is predominantly from 50 to 1000 kPa. For cover systems, range of vertical stress is about 15–50 kPa [47], while for the bottom system, it is dependent on the waste level. The flow of water (distilled) through the samples was tested after each consolidation step at the initial value of the hydraulic gradient: 4, 8, 12, 16, 24, 32, 40. Different hydraulic gradients were established from material behaviour under various conditions, which may take place in the landfill conditions.

3. Results and Discussion

3.1. Hydraulic Conductivity

In Figure 7, the test results of hydraulic conductivity of fly ash and its mixtures with bentonite are shown. Charts are plotted as a relationship of flow velocity to hydraulic gradient. As can be observed in Figure 7, the value of flow velocity decreased when the higher load was attached in case of every sample. At a higher hydraulic gradient, the flow velocity increased, as expected and reported for clay and other materials like sand-bentonite mixture in Reference [48]. At the lowest hydraulic gradient, the flow of water is often irregular. It might be caused by the occurrence of an initial gradient i_0 = 4–8 which, when exceeded, will result in reliable results of hydraulic conductivity. The initial gradients were not observed in previous fly ash studies in the literature. Relationships presented in Figure 7 can be described by exponential curves according to Hansbo's recommendations [43,44], especially in the case of the FA+15%B mixture, while others follow the linear relationship. According to the presence of the initial gradient, the i_1 can be calculated, and its value varies from 12 to 24.

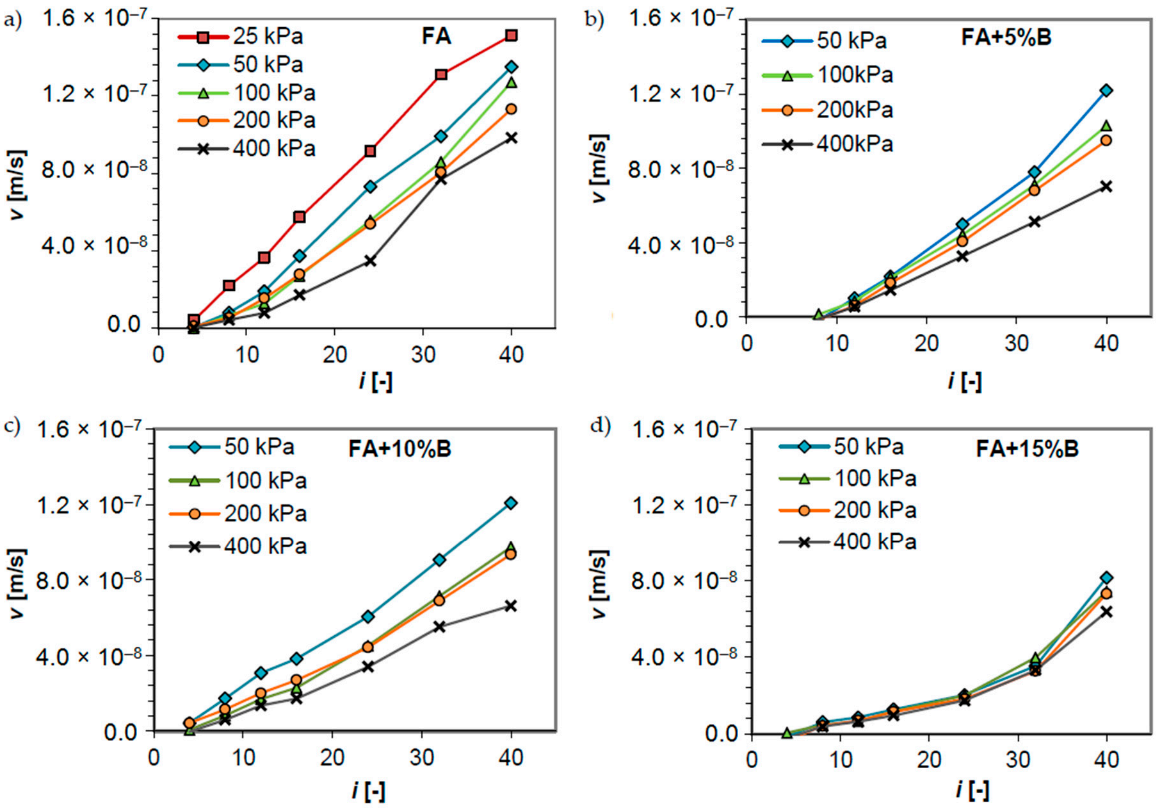


Figure 7. Flow velocity at a hydraulic gradient under variable effective stresses for the samples of: (**a**) fly ash, (**b**) fly ash with 5% of bentonite, (**c**) fly ash with 10% of bentonite, and (**d**) fly ash with 15% of bentonite.

Figure 8 presents the dependency of hydraulic conductivity on effective stress tested at hydraulic gradient i = 32. It is a gradient value close to one recommended in constant gradient method i = 30 [49] for testing materials intended to be used in sealing layers.

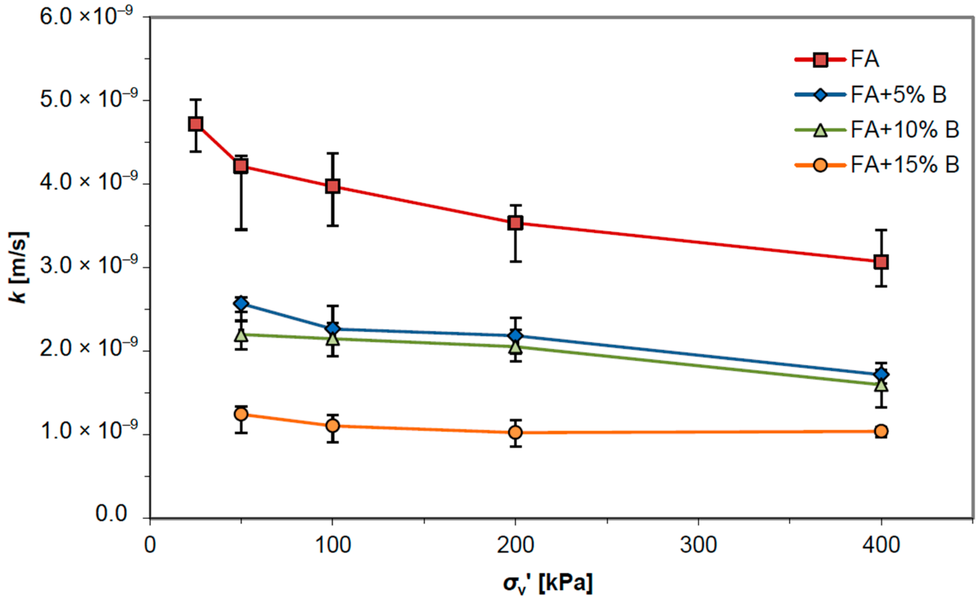


Figure 8. The hydraulic conductivity of tested materials in relation to effective stress at hydraulic gradient i = 32.

In the case of samples tested at i = 32 and $\sigma_v{}'$ = 400 kPa it has been concluded, that 5% of bentonite addition causes a decrease of hydraulic conductivity value almost twofold, 10% also twofold and 15% almost three times, compared to fly ash. Figure 8 shows the dependence of hydraulic conductivity on the value of effective stress and its largest decrease in the case of fly ash, where a decrease in the value of k from 4.7×10^{-9} to 3.1×10^{-9} m/s was observed. It can be seen that hydraulic conductivity decreases with increasing bentonite content. The values range from 3.1×10^{-9} m/s to 4.7×10^{-9} m/s for 0% bentonite, from 1.7×10^{-9} m/s to 2.6×10^{-9} m/s for 5% bentonite, from 1.6×10^{-9} m/s to 2.2×10^{-9} m/s for 10% bentonite and from 1×10^{-9} m/s to 1.2×10^{-9} m/s for 15% of bentonite content in fly ash-bentonite mixtures.

Figure 9 presents hydraulic conductivity for bentonite addition contents tested under different effective stresses at hydraulic gradient i = 32. From the figure, it can be seen that the hydraulic conductivity of fly ash decreased with increasing bentonite content. It also can be observed that the variation in the k value under load is smaller when the bentonite content is higher in the mixture.

As mentioned above, hydraulic conductivity test conditions are indispensable when describing the permeability characteristics of the material. Present results show saturated hydraulic conductivity. The saturation degree of the sample affects the value of hydraulic conductivity. The author conducted the test in the consolidometer for the partially saturated sample, without a former saturation process, at the initial degree of saturation. The volume of water inflow to the partially saturated sample was different from the volume of water outflow from the sample, therefore the flow velocity cannot be reliably assessed. As the water flows, the sample degree of saturation increases, but it is not certain that the back pressure value used to test partially saturated sample is sufficient to fully saturate the sample. It is shown that the condition of full and partial saturation is also important in determining compression and settlement characteristics of the compacted fly ash [50].

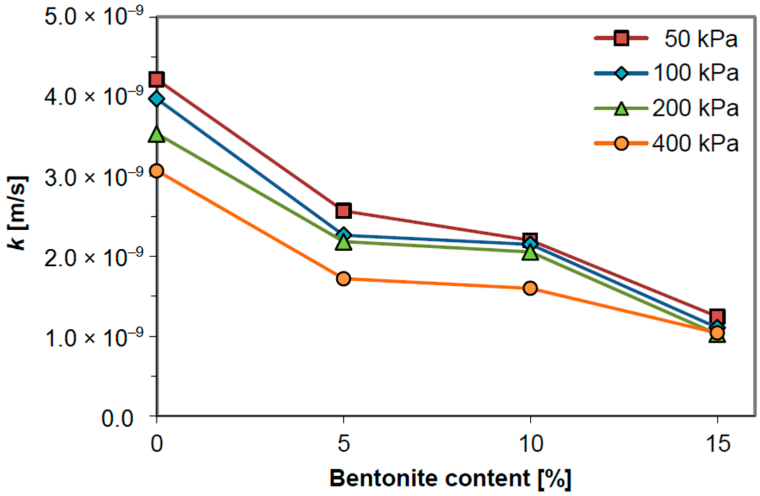


Figure 9. Hydraulic conductivity for bentonite content of materials tested at hydraulic gradient i = 32.

3.2. Cohesive Soil

Figure 10 shows the dependency of hydraulic conductivity on effective stress at hydraulic gradient i = 32 of fly ash, fly ash-bentonite mixes and cohesive soil.

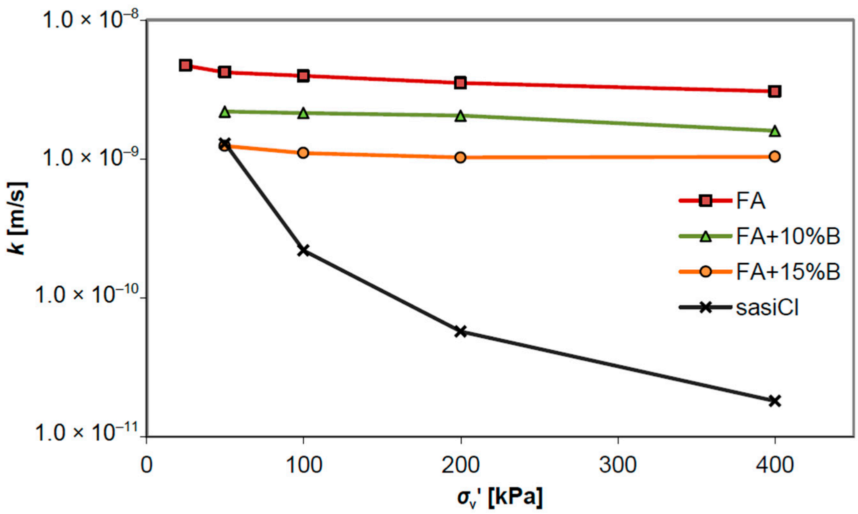


Figure 10. Hydraulic conductivity of sandy silty clay, fly ash and fly ash-bentonite mixes in the relation to effective stress at hydraulic gradient i = 32.

It can be observed that sandy silty clay achieved the smallest values of the hydraulic conductivity, from 1.3×10^{-9} m/s to 1.8×10^{-11} m/s. The value obtained at load 400 kPa was lower by more than two orders of magnitude than for FA+15%B. In the case of sasiCl, an increase of load from 50 to 100 kPa, and from 100 to 200 kPa, caused a decrease of k value by one order of magnitude. At the 50 kPa load, sasiCl had a hydraulic conductivity value slightly higher than FA+15%B (1.2×10^{-9} m/s). Hydraulic conductivity of sasiCl at 100 kPa load had the value of 2.2×10^{-10} m/s—lower by one order of magnitude than FA+15%B (1.1×10^{-9} m/s), at 200 kPa $k = 5.7 \times 10^{-11}$ m/s according to Figure 10 for sasiCl and $k = 1.0 \times 10^{-9}$ m/s for FA+15%B, and at 400 kPa for sasiCl $k = 1.8 \times 10^{-11}$ m/s, which is lower by more than two orders of magnitude than $k = 1.0 \times 10^{-9}$ m/s for FA+15%B. Tested cohesive soil was characterized by finer grain size than fly ash-bentonite mixes (FA+15%B) and greater specific surface area. The specific surface area of FA+15%B has a value of 67.79 m^2/g, while sandy silty clay is 102.58 m^2/g (Table 2). According to Santamarina et al. [51] in the case of soils which have specific surface area greater than 1 m^2/g, a significant role is played by the physico-chemical phenomena in the soil. When the $S_s > 1$ m^2/g, the surface-related forces have an impact on, among others, changes in internal hydraulic and electrical conduction as well as chemical diffusion processes within the soil. The results obtained—the specific surface area (Table 2) and the hydraulic conductivity (Figure 10) of tested materials—confirm this statement. With the change of specific surface area, the plasticity index changes [52]. The influence of the specific surface area on hydraulic conductivity of cohesive soil has been described in References [53–56], where it can be seen that the higher the amount of clayey particle, the lower the hydraulic conductivity. The same trend can be observed in tested material—the higher the amount of bentonite, the lower the hydraulic conductivity of the fly ash, which is related to the increasing value of the specific surface area (Table 2).

4. Conclusions

1. Bentonite addition to fly ash causes a decrease in optimal moisture content (OMC) and an increase in maximum dry density (MDD) of the mixes. The specific density of fly ash and fly ash mixes is lower than for natural soils.
2. The specific surface area increased with the amount of bentonite addition to the fly ash.
3. With the addition of bentonite to fly ash, hydraulic conductivity of the mixture is decreased compared to that of fly ash alone. Samples with bentonite addition in the amount of 5%, 10% and 15% by dry weight tested at $i = 32$ and $\sigma' = 400$ kPa resulted in the decrease in hydraulic conductivity value: about twice, twice and three times in relation to the fly ash, respectively. The hydraulic conductivity values of fly ash may be reduced to the values $\leq 1.0 \times 10^{-9}$ m/s with 15% of bentonite addition and it meets the criteria of a compacted liner and cover materials in terms of hydraulic conductivity.
4. Fly ash and fly ash-bentonite mixtures with a lower amount of bentonite meets the requirement for the sealing layers of landfill of inert wastes, where the hydraulic conductivity k should have a value equal or lower than 1.0×10^{-7} m/s.
5. Hydraulic conductivity of fly ash and fly ash-bentonite mixtures should be tested under conditions of a fully saturated sample in order to produce reliable results.
6. The lowest values of hydraulic conductivity were obtained for sandy silty clay, which was caused by its higher specific surface area in relation to fly ash-bentonite mixtures.

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