Hydraulic conductivity of fly ash as a barrier material: some problems in determination

Katarzyna Zabielska-Adamska¹

Received: 17 January 2019 / Accepted: 17 June 2020 / Published online: 26 June 2020
© The Author(s) 2020

Abstract

Recently, researchers have conducted investigations with the possibility of utilising fly ash as a barrier material, which is justified by its chemical, physical and mechanical properties. The mean necessary characteristic, which should be tested is coefficient of permeability. The diversification of test methods and sample preparation techniques can significantly change test results. The purpose of the article is to describe the fly ash permeability in dependence on compaction and moisture content at compaction, as a material embedded into the sealing layer of storage yards. The impact of sample saturation on hydraulic conductivity is shown too. The hydraulic conductivity was investigated for unsaturated and fully saturated fly ash samples, which had been compacted by the Standard and Modified Proctor methods at various water contents, ranging over optimum water content $\pm 5\%$. The lowest values of hydraulic conductivity were obtained at the highest moisture contents $w = w_{\text{opt}} + 5\%$, for both compaction methods. It is proven that compacted fly ash, in spite of its likeness to non-cohesive soils, must be assessed while considering water content during compaction, as in the case of compacted cohesive soils.

Keywords Hydraulic conductivity · Fly ash · Sealing layer · Landfill

Introduction

Significant element of the municipal landfill and hazardous waste disposal site construction, or existing storage yard modernisation and development, is storage yard leak-proof assurance, which reduces the negative influence of waste on the environment. Impermeability of the waste disposal site is achieved by means of independent protective barriers in the form of geological barriers, artificial sealing layers, mineral soil liners and covers, and sidewall sealing. Soil liners and covers consist of appropriately built-in cohesive soil layers, with a coefficient of permeability (k) less than 10^{-9} m/s, that are characterised by the long-lasting ability to bond and interrupt chemical compounds from waste leachate (Daniel 1997; Van Impe 1998; Rowe et al. 2004; Rowe 2005; Koda and Osinski 2017; Stark 2017; Saadeldin and Froc 2017). Mineral layers are the cheapest and the most stable element of storage yard sealants, which is due to its properties such

as a low value of the coefficient of permeability, swelling ability, as well as the ability to self-sealing, the possibility to form layers of unrestricted volume, and resistance to temperature and chemical compounds existing in the leachate. Mineral sealing layers are commonly built from clay, and boulder clay improved by the addition of bentonite, cement, lime or silica.

In the recent past, many researchers have conducted investigations with the possibility of using fly ash as a material for the mineral sealing layer, which is justified by its chemical, physical and mechanical properties. Shear strength and resistance to failure, tested on compacted fly ash, are significantly greater than those of mineral soils corresponding to fly ash in terms of graining. Although the results of compressibility tests are similar (Zabielska-Adamska and Sulewska 2015; Zabielska-Adamska 2018, 2020).

Fly ash is characterised by the capacity to absorb and reduce leakage, because of its significant water-absorption qualities (up to 80%) and reduction of filtering off (2–16%), which depend on the depth of the tested layer and its density (Ewertowska-Madej 1993). Fly ash built-in sealing layer has the ability to retain several contaminants and heavy metals (Edil et al. 1992; Ewertowska-Madej 1993; Nhan et al. 1996; Dertmatas and Meng 2003; Qureshi et al. 2016;

✉ Katarzyna Zabielska-Adamska
kadamska@pb.edu.pl

¹ Faculty of Civil Engineering and Environmental Sciences,
Białystok University of Technology, Wiejska Str. 45A,
15-351 Białystok, Poland

Moghal 2017; Rubinos and Spagnoli 2018). Palmer et al. (2000) clarified this phenomenon by sorption of contamination by unburnt organic coal that remains in fly ash. Zhu et al. (2013) proved that unburned carbon content, as well as specific surface area and methylene blue sorption capacity were strongly correlated to one another, enhancing the fly ash sorption behaviour. Lieberman et al. (2015) suggested that silicon fly ash utilises multiple fixation mechanisms in parallel (e.g. cation exchange interactions), and it can serve as a substitute for the fixation of radioactive nucleotides. According to fly ash leaching research, the content of chemical compound in the aqueous solution of fly ash does not exceed the accumulation of these compounds in natural soil solutions; however, the tested concentration of trace element is greater in the case of copper, lead, nickel and manganese (Zabielska-Adamska 2006). 5-year-application of fly ash as a sealing layer confirms that none of the elements released from the field-applied material exceeded the EU limits of inert materials; they were far below the limits for hazardous materials (Jia et al. 2018). Coefficient of permeability values published in a number of papers range from 10^{-3} to 10^{-10} m/s, but the test methods and sample preparation techniques have not been detailed. Thus, hydraulic conductivity results for fly ashes from numerous electric-power plants show that permeability is low enough and decreases with time (Edil et al. 1992).

The permeability of fly ash depends on the total content of CaO in the waste mass. The hydraulic conductivity of calcium fly ashes (class C fly ash according to ASTM C618 2015) is far less than that of silicon ashes (class F fly ash after ASTM C618) for ashes with a similar grain size distribution, owing to their self-setting properties. This relationship is also visible for class F fly ashes alone with a variety of chemical compositions (Porbaha et al. 2000) or even for a mixture of class F fly ash with lime and calcium bentonite (Nhan et al. 1996). Fly ash with a higher content of CaO (such as 2.39%) is characterised by permeability decreasing with time, while the permeability of fly ash with a CaO content of 1.39% is greater and steady. This phenomenon was justified by different fly ash grain structures: fly ash grains with higher CaO content are covered by crystallised, needle-shaped protuberances, which are responsible for reducing permeability over time. The gradually increasing hydraulic conductivity of class C fly ashes is explained by their slow cementation during research, both for the flow of water and leachate from the storage yard (Edil et al. 1992).

Kaniraj and Gayathri (2004) tested fly ash (class F) with smooth, equal grains smaller than 0.1 mm. They conducted hydraulic conductivity test under falling head conditions in consolidation cell on samples that were gravitationally saturated under a pressure of 5 kPa (field-saturated). Samples had been compacted at optimum water content according to the Modified Proctor method. Research was

carried out at consolidation stress ranging from 10 to 1250 kPa. The insignificant influence of normal stress (σ) on permeability values was stated for hydraulic conductivity in the range of 6×10^{-8} to 4.7×10^{-8} m/s. The permeability of fly ash from the Bialystok Thermal-Electric Power Station, which was tested using the same method as in Kaniraj's and Gayathri's research in the stress range of $\sigma = 12.5\text{--}300$ kPa, depends on the consolidation stress. For fly ash (class F) with sandy silt grains, k values in the range of $10^{-7}\text{--}4 \times 10^{-10}$ m/s were obtained (Zabielska-Adamska 2006; Wasil 2020). The dry density of fly ash also affects the permeability value. Interesting study of hydraulic conductivity for mixtures of class F fly ash and bottom ash was conducted by Palmer et al. (2000). Tests were executed with the falling head method under consolidation stress of 10–12 kPa, in a cell with a diameter of 10 cm and flexible sides. Samples had been saturated gravitationally. Test results obtained for class F fly ash showed the relationship between permeability and fly ash moisture content at compaction. The highest values were obtained at the moisture content (w) greater than optimal. The lowest values were obtained at the same moisture content, which is greater 5% than the optimum water content by the Standard Proctor method, and 10% by the Modified method. The border value of hydraulic conductivity below 10^{-9} m/s was not obtained for all water content and compaction combinations. The addition of 20–30% class C fly ash (after 7-day-curing) causes the permeability to be closer to the required value.

The permeability of class C fly ash compacted at optimum water content is half as much as the value obtained for samples compacted at lower water contents (Vesperman et al. 1985). Permeability test results for class F fly ash described in the literature are not very advanced. A majority of the papers discuss only research on samples tested at optimum water content (w_{opt}); other tests were merely conditional on the compaction degree. Palmer et al. (2000) indicated the dependence of hydraulic conductivity on fly ash moisture content, nevertheless, all of the tests were performed without controlling the sample saturation.

In a classic paper (Mitchell et al. 1965), the influence of compaction effort and water content at compaction on the hydraulic conductivity of silty clay were determined. It was proven that the permeability decreases as both the compaction effort and the moisture content at compaction increase. The permeability increased insignificantly at water content greater than the optimum 2–5%, after reaching the minimum. The moisture content at compaction on hydraulic conductivity of clays was only influential for fully or field-saturated soils (Meerdink et al. 1996). While testing the water flow through unsaturated clay samples, the influence of moisture content at compaction was not stated, only the impact of compaction effort was noted.

The aim of the study is to present fly ash hydraulic conductivity in dependence on its compaction method, moisture content at compaction and saturation during the tests. The physical properties and chemical composition of tested fly ash are described too. The hydraulic conductivity is investigated on saturated and unsaturated samples compacted by the Standard and Modified Proctor methods at various water contents, ranging over optimum water content $\pm 5\%$. The range of water content was determined to demonstrate the differences in the value of hydraulic conductivity of fly ash compacted at moisture contents on both sides of the compaction curve.

Materials and methods

Tested fly ash

Hydraulic conductivity research was carried out on fly ash and bottom ash mixture from a dry storage yard; which is referred to as fly ash because there is only a vestige of bottom ash in the mix. This fly ash was a by-product of bituminous coal combustion at the Bialystok Thermal-Electric Power Station. The basic chemical composition for tested fly ash is shown in Table 1. A relatively high CaO content should be noted, present within the range of 3.04–4.48%.

Figure 1 shows SEM images and elemental spectra (EDS) for chosen grains of tested fly ash with visible surface hydration. X-ray diffraction (XRD) patterns of the fly ash indicate basic mineralogical composition as quartz SiO_2 , mullite $3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$ and calcite CaCO_3 . Mineral composition of fly ash is related mainly to the operating condition of combustion and type of coal (Shreya et al. 2015).

Table 1 The basic chemical composition for tested fly ash

Chemical	Content in tested fly ash (%)
Si as SiO_2	44.28–47.44
Al as Al_2O_3	17.85–20.86
Fe as Fe_2O_3	5.18–5.43
Ca as CaO	3.04–4.48
Mg as MgO	0.73–2.01
S as SO_3	0.496–0.585
P as P_2O_5	0.082–0.430
Ti as TiO_2	1.04–1.40
Mn as Mn_3O_4	0.035–0.110
Na as Na_2O	0.093–0.202
K as K_2O	0.078–0.604
C as a loss of ignition	7.6–15.0

Grain size distribution of the tested Class F fly ash (Fig. 2), obtained on the basis of the sieve and hydrometer analysis, corresponds in terms of graining to sandy silt with a median particle diameter D_{50} of 0.07 mm.

According to the criterion for assessing mineral soils based on their coefficients of uniformity and curvature, the tested fly ash classifies as a material that responds poorly to compaction, because its C_U is 4.00 and C_C —1.26. The unit weight (γ_s) obtained for an average sample was 20.5 kN/m³. The specific surface of tested fly ash, measured by the methylene blue spot test, was 21.01 m²/g. This value is comparable to the specific surface of kaolinite (Santamarina et al. 2002). It should be noted that fly ash is fine non-plastic material. It consists of silt-sized particles and is characterised by kaolinite-like specific surface, but a fly ash thread cannot be rolled out down to about 3 mm at any moisture possible.

Fly ash was compacted with water contents (w) ranging over optimum water contents $w = w_{\text{opt}} \pm 5\%$, using the Standard (SP) and Modified Proctor (MP) Tests, up to densities corresponding to the values from the compaction curves. Optimum water content was equal to 46% and 37%, respectively, for the SP and MP compaction method. Each point of the compaction curve was determined for separately prepared fly ash sample because fly ash re-compaction causes partial crumbling of dynamically rammed grains. Spherical ash grains, crushed during compaction, can be stuffed with smaller grains, which improves their packing (Zabielska-Adamska 2008). The range of water content was determined to demonstrate the differences in the value of hydraulic conductivity of fly ash compacted at moisture contents on both sides of the compaction curve.

Class F fly ash is pozzolanic in nature but requires a cementing agent, e.g. Portland cement, mixed with water to involve pozzolanic reaction and produce cementitious compounds. In the study minimum quantity of cement—2% by weight—was used to improve sealing property without producing a rigid material. Fly ash mixed with cement additions was compacted, at moisture content closed to the optimum, up to densities corresponding to the maximum for compacted samples by the Modified (MP) and Standard Proctor (SP) method. Optimum water content of fly ash–cement mix was equal to 44.5% and 35.5%, respectively, for the SP and MP compaction.

Hydraulic conductivity tests

The main hydraulic conductivity result presentation was focused to values carried out in a consolidation cell, which is generally known as the Rowe cell, which is 150 mm in diameter and 50-mm high, at vertical drainage condition. Fly ash samples were dynamically compacted directly in the cell body or compacted in a CBR mould and carefully

Fig. 1 SEM micrographs of tested fly ash: **a** general view, **b** SEM/EDS analysis for chosen grains

relocated to the cell. The tested samples were compacted by two methods—the Proctor Standard and Modified methods—at five moistures within the range of $w_{\text{opt}} \pm 5\%$ for each compaction method. Fly ash with 2% cement addition, compacted at optimum water content, was examined too. To prevent uncontrolled leaks between the sample side

surface and cell wall, silicone fluid was used in addition to the typical seals.

Hydraulic conductivity at a constant hydraulic gradient in one-dimensional flow is calculated based on Darcy's law, as:

Fig. 2 Fly ash grain size curve obtained on the base of sieve and hydrometer analysis

$$k = \frac{Q}{Ait} \quad (1)$$

where Q is the volumetric discharge of fluid flowing through the given cross-section area A , t is the time-duration; and i is the hydraulic gradient, which is defined as the head loss per unit length:

$$i = \frac{\Delta h}{L} = \frac{\Delta p C}{L} \quad (2)$$

where L is the sample height, Δh is the difference of total pressure head (fluid levels), Δp is the pressure difference measured from the bottom to the top of the sample face, and C is the constant that depends on the apparatus and equipment parameters, for the Rowe cell $C = 102$ (Head 1986).

According to Head's recommendations samples, after removing intact from the cell, were split in two along the diameter by partially cutting and then breaking open. Each of the samples was examined to reveal any preferential drainage paths directly after the test and after being exposed to air-dry for 24 h.

The tested samples were compacted at moistures within the range of $w_{\text{opt}} \pm 5\%$ for each compaction method, standard and modified. Table 2 presents values of moisture contents and dry densities, which are referred to as initial values, obtained for compacted fly ash samples.

Saturation of tested samples

The Rowe cell allows the tested samples to be saturated by applying increments of back pressure and controlling it at every stage of cell pressure raising. Degree of saturation of soil pore with water is assessed on the basis of parameter B , called "pore-pressure coefficient" (Skempton 1954). In

Table 2 Values of moisture contents and dry densities obtained for compacted fly ash samples

Compaction method	Sample symbol	Initial moisture (%)	Initial dry density (g/cm ³)
Standard proctor (SP)	$w_{\text{opt}} - 5\%$	41.0	0.963
	$w_{\text{opt}} - 2.5\%$	43.5	0.971
	w_{opt}	46.0	0.984
	$w_{\text{opt}} + 2.5\%$	48.5	0.967
	$w_{\text{opt}} + 5\%$	51.0	0.949
Modified proctor (MP)	$w_{\text{opt}} - 5\%$	32.0	1.045
	$w_{\text{opt}} - 2.5\%$	34.5	1.056
	w_{opt}	37.0	1.090
	$w_{\text{opt}} + 2.5\%$	39.5	1.064
	$w_{\text{opt}} + 5\%$	42.0	1.035

one-dimensional compression test, parameter B is calculated based on simplified Eq. 3:

$$B = \frac{\Delta u}{\Delta \sigma} \quad (3)$$

where Δu and $\Delta \sigma$ are the increment in pore water pressure and stress.

According to Skempton, the soil is fully saturated with water ($S_r = 1$), when parameter $B = 1$. For of partially saturated soils, B gains values $0 < B < 1$. Thus, in the case of soils compacted by means of Proctor's method at w_{opt} the values of B range typically from about 0.1 to 0.5.

The course of saturation for soils with different cohesion and state is various, and pore pressure values necessary to gain required value of Skempton's parameter B , grow with soil cohesion. The value of back pressure, which is theoretically needed to raise initial values of degree of saturation (S_{r0}) to required value of S_r , is given by Lowe and Johnson (1960) in Eq. 4:

$$\Delta u = p_0 \frac{(S_r - S_{r0})(1 - H)}{1 - S_r(1 - H)} \quad (4)$$

where p_0 is the atmospheric pressure, assumed as 101.33 kPa, H is the Henry law constant, which is a temperature-dependent ratio of the appropriate molar concentrations in the gas and water phases assumed as 0.02 cm³ of gas per 1 cm³ of water in a temperature of 20 °C.

It is considered that applying for all soils rigorous criteria fully saturation ($B = 1$) is unfounded (Shahu et al. 1999; Lipiński and Wdowska 2010). Shahu et al. (1999) labelled saturation to the minimum sufficient value of Skempton's parameter as the quasi-saturated state.

Saturation impact on hydraulic conductivity was shown by means a steady-controlled flow technique, also called the gradient stabilisation method (Olsen et al. 1991), which was

executed with a flow pump that cooperated with the sample placed in the chamber of the triaxial apparatus. Constant flow velocity is forced: water is pumped from the bottom to the top of the sample. Pore-water pressure dissipation is controlled during the test. Water discharge is pumped through the sample (one-dimensional flow), and the time and the pressure difference between the sample bottom and top are recorded (Lipiński and Wdowska 2010).

Results

Saturation fly ash samples

Fly ash samples with an initial degree of saturation in the range of 0.75–0.90 (at $w_{\text{opt}} \pm 5\%$ for each compaction method) were saturated in the Rowe cell before starting consolidation. Skempton's parameter (B) versus increment in back pressure (Δu) for consecutive stages of saturation in tested samples is presented in Fig. 3. Transforming Eq. 4 and taking into consideration values of back pressure obtained during sample saturation in the Rowe cell, values S_r in saturation process can be calculated (see Fig. 4). The minimum value of parameter B can be estimated based on the correlation between Skempton's parameter B and degree of saturation S_r . After averaging values presented in Fig. 4, the value of Skempton's parameter B , which defines minimum sufficient saturation of compacted fly ash, was recognised as $B = 0.8$.

Hydraulic conductivity in the Rowe cell

Hydraulic conductivity tests were performed for the vertical drainage condition after full sample saturation ($B = 0.8$) by applying increments of back pressure and then

Fig. 3 Skempton's parameter (B) versus increment in back pressure (Δu) for consecutive stages of saturation in tested samples

Fig. 4 Skempton's parameter (B) versus degree of saturation (S_r) for consecutive stages of saturation in tested samples

its consolidation under normal effective stress (σ') equal to 50, 100, 150, and 200 kPa (at standard compaction) or 20, 50, 100, 150 and 200 kPa (at modified compaction), keeping by airdraulic system. None of the preferential drainage path was stated, so flow had been uniformed through cross-section area in all the tested samples. Moreover, there were no crushing fly ash grains during the consolidation process, which can cause a reduction in hydraulic conductivity (Feia et al. 2016).

The dependence of the water-flow velocity on the hydraulic gradient is presented in Fig. 5 for determining values of water content at compaction. In most cases, the relationships $v(i)$ can be generally approximated by straight lines. In some research, curvilinearity of the graph was observed, which was stated for low values of the hydraulic gradient $i = 1.2\text{--}5$. Graphs are close to linearity at gradient $i \geq 10$, as has been stated for clays (Hansbo 2001, 2003). The initial gradient i_0 , which is often necessary in cohesive soils to initiate flow by overcoming the resistance of water viscosity, was not reported.

The flow velocity (v) determined for fly ash compacted at a moisture content ranging over $w_{\text{opt}} \pm 5\%$ (for both compaction methods) depending on effective stress is shown in Fig. 6. The results were presented for tests at a gradient value of $i = 30$, which is recommended for the constant-gradient method (ETC 8 1993).

Hydraulic conductivity at effective stress of $\sigma' = 50\text{--}200$ kPa was $1.4 \times 10^{-7}\text{--}3.7 \times 10^{-10}$ m/s or $7.3 \times 10^{-8}\text{--}1.3 \times 10^{-9}$ m/s, respectively, for fly ash compacted by the Standard or the Modified Proctor method. Hydraulic conductivity of fly ash samples compacted by means of the Modified Proctor method may be greater than those compacted by the Standard method. Samples are compacted by means of higher energy, so they are less

Fig. 5 Water-flow velocity for compacted fly ash that depends on the hydraulic gradient, which is obtained at effective stress of σ' : 20, 50, 100, 150 and 200 kPa: **a** standard compaction (SP) at $w = w_{\text{opt}} - 2.5\%$, **b** modified compaction (MP) at $w = w_{\text{opt}} + 2.5\%$

Fig. 6 Water-flow velocity for fly ash compacted at moisture contents at a compaction $w_{\text{opt}} \pm 5\%$ and with a hydraulic gradient $i = 30$: **a** compaction by SP method, **b** compaction by MP method

compressible, with the result that the final permeability at higher stress is greater.

Fly ash hydraulic conductivity that depends on moisture content at compaction, for both compaction methods, standard and modified, is presented in Fig. 7. Test results are shown for the hydraulic gradient $i = 30$. The visible impact of moisture content at compaction is reduced under higher effective stress of σ' , 150 and 200 kPa and at moisture contents greater than the optimum water content and it is more perceptible for the standard compaction. Thus, the statement regarding the relationship between hydraulic conductivity and fly ash moisture content should be applied only for the

determined fly ash compaction method, and it must be compared with the suitable optimum water content.

Saturation influence on hydraulic conductivity test results

Hydraulic conductivity results achieved by the flow pump for fly ash from the Bialystok Thermal-Electric Power Station are presented in Figs. 8 and 9. Research was performed on cylinder samples placed in the chamber of the triaxial apparatus after stabilising the compression under the determined

Fig. 7 Hydraulic conductivity (at a hydraulic gradient $i = 30$) of fly ash compacted at moisture contents $w_{opt} \pm 5\%$, obtained at various effective stress of $\sigma' = 20 - 200$ kPa

effective stress. Compacted fly ash was tested as either fully saturated or unsaturated.

Water volumes entering the tested sample and flowing out in the same period were established to characterise the water-flow condition. The test results show differences between the flow characteristics of saturated and unsaturated fly ash, as in the case of the cohesive soils (Lipiński and Wdowska 2010).

Hydraulic conductivity of fly ash with cement addition

Fly ash with 2% cement addition was compacted in a CBR mould at optimum water content, which was determined for a fly ash and cement mix. It was stored for 7 days in a humidity-controlled chamber at a constant temperature (20 °C) and a relative humidity greater than 95%, because increasing the curing time of mixtures from 7 to 28 days did not significantly reduce their hydraulic conductivity (Bowders et al. 1987). Then the sample was relocated to the Rowe cell, consolidated, and subjected to the filtration test. Hydraulic conductivity of stabilised fly ash that depends on the effective stress is compared to the test results of fly ash without binder addition in Fig. 10. Test results are presented for a hydraulic gradient $i = 30$.

The 2% cement addition significantly affected the fly ash hydraulic conductivity tested under effective stress lower than 150 kPa. This effect was visible for fly ash compacted by both compaction methods: standard and modified. During the permeability test performed with higher values of σ' , the differences in the hydraulic conductivity of fly ash and fly ash–cement mix were observed only for samples compacted by the Standard Proctor method. It should be stated that the compaction method only influenced stabilised fly ash hydraulic conductivity in tests with lower values of effective stress, like those for non-stabilised samples. While samples

were tested at values of $\sigma' \geq 150$ kPa, changing the compaction method only caused an insignificant change in the hydraulic conductivity. The 2% cement addition was more effective for samples compacted by the Standard method.

Discussion

Fly ash from the Białystok Thermal-Electric Power Station compacted by the Standard Proctor method achieved values of $k \leq 1 \times 10^{-9}$ m/s (required for sealing layers) at a moisture content greater than the optimum water content by at least 2.5% and effective stress of $\sigma' \geq 150$ kPa. Fly ash compacted by means of the Modified method only approach the limit value at a moisture content of $w = w_{opt} + 5\%$ and effective stress of $\sigma' \geq 150$ kPa.

Hydraulic conductivity of compacted fly ash, despite its macroscopic resemblance to non-cohesive soils, should be assessed considering water content during compaction (see Fig. 7), as in the case of compacted cohesive soils. For fly ash compacted with greater effort (the Modified method), the water content at compaction and effective stress had a smaller effect on the permeability than for samples compacted by the Standard method. The evident effect of moisture content at compaction decreased under effective stress greater than $\sigma' \geq 150$ kPa and at moisture contents on the wet side of the compaction curve (which is more noticeable for the lower energy of compaction). A distinct increase in the value of hydraulic conductivity at high water contents, which is known for cohesive soils (Mitchell et al. 1965), was not found for compacted fly ash.

The dependence of $k(w)$ can be explained by microstructural observations of compacted fly ash samples using ESEM mode, which allows to assess moisture impact during dynamic compaction on microstructure (Zabińska-Adamśka et al. 2019). The fly ash compacted at water content

Fig. 8 Test results of flow through the saturated fly ash sample: **a** stabilisation of pore-water pressure in a saturated fly ash sample, **b** comparison of water coming in and flowing out of the tested sample

$w = w_{\text{opt}} - 5\%$ show a structure composed of aggregates with well-defined large macro-pores and the small pores inside the aggregates, as in case of the cohesive soil. Structure of sample compacted at optimum water content is the most uniform. Pore spaces are rather small, and they occur in small quantities. In the case of sample compacted at $w = w_{\text{opt}} + 5\%$ a great number of small pore spaces can be observed. It is visible that the fly ash structure is loosened by a higher water content during the compaction process. Samples compacted wet of optimum presents a dispersed structure with continuous water phase in the pores and air phase in the mostly occluded state. According to Burton et al. (2015)

macropore space decreases as the moisture content or the compaction energy increases, while the micropore space remains unchanged by compaction. Thus, smaller pore space in samples compacted by the Modified method triggers the capillary effects and suction, which has influence on results of hydraulic conductivity. Additionally, compressibility of fly ash samples compacted by Standard method and saturated has differentiated characteristics in comparison modified compaction, which affects hydraulic conductivity—measurements are carried out after stabilising the settlement under specified effective stress. The relative vertical deformation ranges 3.8–12.8% for samples compacted by the

Fig. 9 Test results of flow through an unsaturated fly ash sample: **a** stabilisation of pore-water pressure in an unsaturated fly ash sample, **b** comparison of water coming in and flowing out of the tested sample

Standard method and 7.8–13.6% for the Modified method in the 5% w_{opt} moisture range for both compaction methods (Zabielska-Adamska 2018). Compressibility of saturated specimens decreases along with their water content during compaction.

Saturation influence on hydraulic conductivity results was shown by means a steady-controlled flow technique. During testing of saturated samples, the increase in pore-water pressure stabilised quickly, regardless of the amount of forced flow, and the volumes of inflowing and outgoing water were balanced. This condition indicates that steady-flow condition was obtained for fully saturated fly ash

sample (see Fig. 8) with Skempton's parameter $B = 0.8$. Parameter $B = 0.8$ expresses the minimum sufficient saturation of compacted fly ash, which was recognised in the study. In the case of unsaturated sample ($B = 0.2$) water volumes inflowing and outgoing were not balanced and a pore-water pressure was not stabilised despite extending the test period; consequently, the flow conditions remained non-steady (see Fig. 9). This phenomenon motivates a proper assessment of the maximum hydraulic conductivity of fly ash, which can be found only by methods that enable full saturation of the samples.

Fig. 10 Hydraulic conductivity of fly ash and fly ash–cement mix compacted at optimum moisture content tested at a hydraulic gradient $i = 30$, which depends on the effective stress σ'

Fly ash with 2% cement addition compacted at an optimum water content was characterised by hydraulic conductivity below 10^{-8} m/s. It should be admitted that tests were investigated after 7 days of storing, and hydraulic conductivity decreases over time. Lav et al. (2006) stated, for cement stabilised fly ash (class F), that Calcium Silicate Hydrate (C–S–H) gel densified and was able to cover fly ash spheres, including the gaps between particles, after 6 months. In addition, it is necessary to expect that samples with a greater cement content or samples compacted at a moisture content greater than the optimum obtain lower hydraulic conductivity values. Kalinski and Yerra (2006) stated that the fly ash–cement mix water content at compaction affected hydraulic conductivity values, as in the case of fly ash itself. Increasing the water content during compaction decreased the fly ash–cement mix permeability.

Conclusions

On the basis of the test results of the hydraulic conductivity of compacted fly ash or fly ash with cement addition the following can be stated:

1. Hydraulic conductivity of fly ash depends on the test method and sample preparation technique. Sample saturation significantly affects permeability test results; maximum value of fly ash permeability can be achieved only by the test method that enables full sample saturation. In the case of unsaturated sample water volumes flowing in and out are not balanced and the flow conditions remained non-steady.

2. In most cases, the linear relationship between water-flow velocity and hydraulic gradient (Darcian flow) was observed. However, the curvilinearity of graph $v(i)$ for low values of hydraulic gradients ($i = 1.2-5$) was stated. The relationship between the fluid velocity and hydraulic gradient is generally linear for gradients of $i \geq 10$.
3. Fly ash hydraulic conductivity closely depends on water content during compaction. It decreases as the moisture content increases. Moisture content at compaction $w_{opt} \pm 5\%$ a greater extent influences on coefficients of permeability of samples compacted by the Standard Proctor method (about three orders of magnitude) than by the Modified method (about two orders of magnitude). In both cases, the lowest values of hydraulic conductivity were obtained at the highest moisture contents $w = w_{opt} + 5\%$. Because effective stress affects hydraulic conductivity, the test conditions must be taken into consideration when designing a fly ash sealing layer.
4. Hydraulic conductivity at effective stress of $\sigma' = 50-200$ kPa was $1.4 \times 10^{-7}-3.7 \times 10^{-10}$ m/s or $3 \times 10^{-8}-1.3 \times 10^{-9}$ m/s, respectively, for fly ash compacted by the Standard or the Modified method. Obtaining the required coefficient of permeability $k \leq 10^{-9}$ m/s for a fly ash built-in sealing layer is possible at high effective stress but only for fly ash compacted at high moisture content. Fly ash used for a mineral cover must be improved by the binder addition.
5. The 2% cement addition significantly (about one order of magnitude) decreases the fly ash coefficient of permeability in comparison to the fly ash itself. The addition of cement considerably affects the fly ash hydraulic conductivity tested at lower effective stress ($\sigma' < 150$ kPa), independently of the sample compaction method, standard or modified. This impact is visible at greater effective stress only for samples compacted by the Standard Proctor method.

Acknowledgements The work, carried out at Białystok Technical University in Poland, was supported by Polish financial resources on science under project nos. 5 T07E 051 24 and S/WBiIS/2/2018. The author is grateful to Prof. Mirosław J. Lipiński for his cooperation in conducting laboratory flow pump tests at the Warsaw University of Life Science.

Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons licence, and indicate if changes were made. The images or other third party material in this article are included in the article's Creative Commons licence, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons licence and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will

need to obtain permission directly from the copyright holder. To view a copy of this licence, visit <http://creativecommons.org/licenses/by/4.0/>.

References

- ASTM (2015) C 618: Specification for coal fly ash and raw or calcined natural pozzolan for use in concrete. ASTM International, West Conshohocken
- Bowders J, Usmen M, Gidley J (1987) Stabilized fly ash for use as low-permeability barriers. In: Geotechnical practice for Waste disposal, ASCE GSP No. 13, New York
- Burton GJ, Pineda JA, Sheng D, Airey D (2015) Microstructural changes of an undisturbed, reconstituted and compacted high plasticity clay subjected to wetting and drying. *Eng Geol* 193:363–373
- Daniel DE (1997) Clay liners. In: Daniel DE (ed) Geotechnical practice for waste disposal. Chapman & Hall, London, pp 137–163
- Dertmatas D, Meng X (2003) Utilization of fly ash for stabilization/solidification of heavy metal contaminated soils. *Eng Geol* 70(2–4):377–394
- Edil TB, Sandstrom LK, Berthouex PM (1992) Interaction of inorganic leachate with compacted pozzolanic fly ash. *J Geotech Eng* 118(9):1410–1430
- European Technical Committee No. 8 (1993) Geotechnics of landfill design and remedial works—Technical Recommendation GLR. Ernst & Sohn, Berlin
- Ewertowska-Madej Z (1993) Model test of pollution and liquid retaining process in a fly ash layer. In: Proceedings of 4th international symposium on the reclamation, treatment and utilisation of coal mining wastes, Krakow, pp 229–238
- Feia S, Sulem J, Canou J, Ghabezloo S, Clain X (2016) Changes in permeability of sand during triaxial loading: effect of fine particles production. *Acta Geotech* 11(1):1–19
- Hansbo S (2001) Consolidation equation valid for both Darcian and non-Darcian flow. *Géotechnique* 51(1):51–54
- Hansbo S (2003) Deviation from Darcy's law observed in one dimensional consolidation. *Géotechnique* 53(6):601–605
- Head KH (1986) Manual of soil laboratory testing. In: Vol. 2: Permeability, shear strength and compressibility tests; Vol. 3: Effective stress tests. Pentech Press Ltd, London
- Jia Y, Stahre N, Maurice C, Öhlander B (2018) Geotechnical and chemical characterization of field-applied fly ash as sealing material over mine tailings. *Int J Environ Sci Technol* 16(3):1701–1710
- Kalinski ME, Yerra PK (2006) Hydraulic conductivity of compacted cement-stabilized fly ash. *Fuel* 85(16):2330–2336
- Kaniraj SR, Gayathri V (2004) Permeability and consolidation characteristics of compacted fly ash. *J Energy Eng* 130(1):18–43
- Koda E, Osinski P (2017) Bentonite cut-off walls: solution for landfill remedial works. *Environ Geotech* 4(4):223–232
- Lav A, Lav MA, Goktepe AB (2006) Analysis and design of a stabilized fly ash as pavement base material. *Fuel* 85(16):2359–2370
- Lieberman RN, Green U, Segev G, Polat M, Mastai Y, Cohen H (2015) Coal fly ash as a potential fixation reagent for radioactive wastes. *Fuel* 153:437–444
- Lipiński MJ, Wdowska MK (2010) Saturation criteria for heavy over-consolidated cohesive soils. *Ann Warsaw Univ Life Sci SGGW Land Reclam* 42(2):295–302
- Lowe J and Johnson TC (1960) Use of back pressure to increase degree of saturation of triaxial test specimens. In: Proceedings of research conference on shear strength of cohesive soils. Boulder, Colorado, pp 819–836
- Meerdink JS, Benson CH, Khire MV (1996) Unsaturated hydraulic conductivity of two compacted barrier soils. *J Geotech Eng* 122(7):565–576
- Mitchell JK, Hooper DR, Campanella RG (1965) Permeability of compacted clay. *J Soil Mech Found Div* 91(SM4):41–63
- Moghal AAB (2017) A state-of-the-art review on the role of fly ashes in geotechnical and geoenvironmental applications. *J Mater Civ Eng*. [https://doi.org/10.1061/\(ASCE\)MT.1943-5533.0001897](https://doi.org/10.1061/(ASCE)MT.1943-5533.0001897)
- Nhan CT, Graydon JW, Kirk DW (1996) Utilizing coal fly ash as a landfill barrier material. *Waste Manag* 16(7):587–595
- Olsen HW, Gill JD, Wilden AT and Nelson KR (1991) Innovations in hydraulic-conductivity measurements. In: Transportation research record 1309, transportation research board, Washington, pp 9–17
- Palmer BG, Edil TB, Benson CH (2000) Liners for waste containment constructed with class F and C fly ashes. *J Hazard Mater* 76(2–3):193–216
- Porbaha A, Pradhan TBS, Yamane N (2000) Time effect on shear strength and permeability of fly ash. *J Energy Eng* 126(1):15–31
- Qureshi AS, Jia Y, Maurice C, Öhlander B (2016) Potential of fly ash for neutralization of acid mine drainage. *Environ Sci Pollut Res* 23(17):17083–17094
- Rowe RK (2005) Long-term performance of contaminant barrier system. *Géotechnique* 55(9):631–678
- Rowe RK, Quigley RM, Brachman RWI, Broker JR (2004) Barrier systems for waste disposal facilities. Taylor & Francis Books Ltd., London
- Rubinos DA, Spagnoli G (2018) Utilization of waste products as alternative landfill liner and cover material—critical view. *Crit Rev Environ Sci Technol* 48(4):376–435
- Saadeldin R, Froc G (2017) Hydraulic conductivity of a glacial clay till liner. *Bull Eng Geol Environ* 76:553–560
- Santamarina JC, Klein KA, Wang YH, Preneke E (2002) Specific surface: determination and relevance. *Can Geotech J* 39:233–241
- Shahu JT, Yudhbir, Kameswara Rao NSV (1999) Effective stress behavior of quasi-saturated compacted cohesive soils. *J Geotech Geoenviron Eng* 125(4):322–329
- Shreya N, Valentim B, Paul B, Guedes A, Pinho S, Ribeiro J, Ward CR, Flores D (2015) Multi-technique study of fly ash from the Bokaro and Jharia coalfields (Jharkhand state, India): a contribution to its use as a geoliner. *Int J Coal Geol* 152:25–38
- Skempton AW (1954) The pore pressure coefficients A and B. *Géotechnique* 4(4):143–147
- Stark TD (2017) Evaluation of a four-component composite landfill liner system. *Environ Geotech* 4(4):257–273
- Van Impe WF (1998) Environmental geotechnics. ITC 5 activities. State of the art. In: Sêcoe P (ed) Environmental geotechnics. A. A. Balkema, Rotterdam, pp 121–126
- Vesperman KD, Edil TB and Berthouex PM (1985) Permeability of fly ash and fly ash-sand mixtures. In: Johnson AI, Frobel RK, Cavalli NJ, Petterson CB (eds) Proceedings of symposium “hydraulic barriers in soil and rock”. ASTM STP 874, Philadelphia, pp 289–298
- Wasil M (2020) Effect of bentonite addition on the properties of fly ash as a material for landfill sealing layers. *Appl Sci* 10(4):1488
- Zabielska-Adamska K (2006) Shear strength parameters of compacted fly ash–HDPE geomembrane interfaces. *Geotex Geomem* 24(2):91–102
- Zabielska-Adamska K (2008) Laboratory compaction of fly ash and fly ash with cement additions. *J Hazard Mater* 151(2–3):481–489
- Zabielska-Adamska K (2018) One-dimensional compression and swelling of compacted fly ash. *Geotech Res* 5(2):96–105
- Zabielska-Adamska K (2020) Characteristics of compacted fly ash as a transitional soil. *Mater* 13(6):1387
- Zabielska-Adamska K, Sulewska MJ (2015) Dynamic CBR test to assess the soil compaction. *J Test Eval* 53(5):1028–1036
- Zabielska-Adamska K, Małaszkiwicz D, Konopko M (2019) Microstructure of compacted fly ash. In: Proceedings of the XVII

ECSMGE “geotechnical engineering foundation of the future”, 1–6 September 2019, Reykjavik. <http://doi.org/10.32075/17ECSMGE-2019-0465>

Zhu Z, Wang X, Dai S, Huang B, He Q (2013) Fractional characteristics of coal fly ash for beneficial use. *J Mater Civ Eng* 25(1):63–69

Publisher's Note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.